# Hardy Spaces $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ Associated to Operators Satisfying $k$-Davies-Gaffney Estimates 

Jun Cao and Dachun Yang*


#### Abstract

Let $L$ be a one to one operator of type $\omega$ having a bounded $H_{\infty}$ functional calculus and satisfying the $k$-Davies-Gaffney estimates with $k \in \mathbb{N}$. In this paper, the authors introduce the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ with $p \in(0,1]$ associated to $L$ in terms of square functions defined via $\left\{e^{-t^{2 k} L}\right\}_{t>0}$ and establish their molecular and generalized square function characterizations. Typical examples of such operators include the $2 k$-order divergence form homogeneous elliptic operator $L_{1}$ with complex bounded measurable coefficients and the $2 k$-order Schrödinger type operator $L_{2} \equiv(-\Delta)^{k}+V^{k}$, where $\Delta$ is the Laplacian and $0 \leq V \in L_{\mathrm{loc}}^{k}\left(\mathbb{R}^{n}\right)$. Moreover, as applications, for $i \in\{1,2\}$, the authors prove that the associated Riesz transform $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ is bounded from $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ to $H^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(n /(n+k), 1]$ and establish the Riesz transform characterizations of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(r n /(n+k r), 1]$ if $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimates with $r \in(1,2]$. These results when $k \equiv 1$ and $L \equiv L_{1}$ are known.


## 1 Introduction

The Hardy spaces, as a suitable substitute of Lebesgue spaces $L^{p}\left(\mathbb{R}^{n}\right)$, play an important role in various fields of analysis and partial differential equations. It is well known that the Hardy spaces $H^{p}\left(\mathbb{R}^{n}\right)$ are essentially related to the Laplacian operator $\Delta \equiv \sum_{j=1}^{n} \frac{\partial^{2}}{\partial x_{j}^{2}}$, which have been intensively studied; see, for example, [42, 23, 15, 43, 41, 25] and the references therein.

In recent years, the study of Hardy spaces associated to different differential operators inspires great interests; see, for example, $[4,7,8,12,19,20,18,21,22,28,30,31,32]$ and their references. In particular, in [4], when the operator $L$ satisfies a pointwise Poisson

[^0]upper bound, Auscher, McIntosh and Duong introduced the Hardy space $H_{L}^{1}\left(\mathbb{R}^{n}\right)$ associated to $L$ in terms of area integral functions. Later, in [19, 20], Duong and Yan introduced the BMO-type space $\mathrm{BMO}_{L}\left(\mathbb{R}^{n}\right)$ associated to such an $L$ and proved the dual space of $H_{L}^{1}\left(\mathbb{R}^{n}\right)$ is $\mathrm{BMO}_{L^{*}}\left(\mathbb{R}^{n}\right)$, where $L^{*}$ denotes the adjoint operator of $L$ in $L^{2}\left(\mathbb{R}^{n}\right)$. Yan [46] further generalized these results to the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ with $p \in(0,1]$ close to 1 and its dual space. Also, the Orlicz-Hardy space and its dual space associated to such an $L$ were studied in $[38,35]$.

Auscher and Russ [8] studied the Hardy space $H_{L}^{1}$ on strongly Lipschitz domains associated with a second order divergence form elliptic operator $L$ whose heat kernels have the Gaussian upper bounds and regularity. Very recently, Auscher, McIntosh and Russ [7] treated the Hardy space $H^{1}$ associated with the Hodge Laplacian on a Riemannian manifold with doubling measure; Hofmann-Mayboroda in [30,31] and Hofmann-MayborodaMcIntosh in [32] introduced the Hardy and Sobolev spaces associated to a second order divergence form elliptic operator $L$ on $\mathbb{R}^{n}$ with bounded measurable complex coefficients and these operators may not have the pointwise heat kernel bounds, while a theory of the Orlicz-Hardy space and its dual space associated to $L$ was independently developed in [36, 37].

Moreover, a theory of Hardy spaces associated to the Schrödinger operators $-\Delta+V$ was well developed, where the nonnegative potential $V$ satisfies the reverse Hölder inequality; see, for example, Dziubański and Zienkiewicz [21, 22] and Yang and Zhou [48] and their references. More generally, for nonnegative self-adjoint operators $L$ satisfying the DaviesGaffney estimates, Hofmann et al. [28] introduced a new Hardy space $H_{L}^{1}\left(\mathbb{R}^{n}\right)$, which was extended to the Orlicz-Hardy space by Jiang and Yang [34]. Recently, the Hardy space $H_{(-\Delta)^{2}+V^{2}}^{1}\left(\mathbb{R}^{n}\right)$ associated to the Schrödinger type operators $(-\Delta)^{2}+V^{2}$ was also studied in [12].

From now on, in what follows of this paper, we always let $L$ be a one to one operator of type $\omega$ having a bounded $H_{\infty}$ functional calculus and satisfying the $k$-Davies-Gaffney estimates with $k \in \mathbb{N}$ (see (2.6) below). Motivated by [32, 28], in this paper, we introduce the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ with $p \in(0,1]$ associated to $L$ in terms of the square function defined via $\left\{e^{-t^{2 k} L}\right\}_{t>0}$ (see (4.1) below) and establish their molecular and generalized square function characterizations. Typical examples of such operators include the $2 k$ order divergence form homogeneous elliptic operator $L_{1}$ with complex bounded measurable coefficients and the $2 k$-order Schrödinger type operator $L_{2} \equiv(-\Delta)^{k}+V^{k}$, where $\Delta$ is the Laplacian and $0 \leq V \in L_{\text {loc }}^{k}\left(\mathbb{R}^{n}\right)$. Moreover, as applications, for $i \in\{1,2\}$, we prove that the associated Riesz transform $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ is bounded from $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ to $H^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(n /(n+k), 1]$ and establish the Riesz transform characterizations of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(r n /(n+k r), 1]$ if $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimates with $r \in(1,2]$ (see Definition 6.1 below for the definition). These results when $k \equiv 1$ and $L \equiv L_{1}$ were already obtained recently by Hofmann-Mayboroda [30, 31], Jiang-Yang [36, 34], and Hofmann-Mayboroda-McIntosh [32].

A new ingredient appearing in this paper is the introduction of the $k$-Davies-Gaffney estimates with $k \in \mathbb{N}$, which is naturally satisfied by $2 k$-order Schrödinger operators $(-\Delta)^{k}+V^{k}$. Via the perturbation technique (see, for example, $[10,11]$ ) and some ideas
from the proof of [16, Lemma 2], and using the elliptic condition, we further show that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ also satisfies the $k$-Davies-Gaffney estimates.

Another new observation of this paper is that the nonnegative self-adjoint property of operators in $[28,34]$ can be weakened into the assumption that $L$ has a bounded $H_{\infty}$ functional calculus. We point out that when this manuscript was in preparation, we learned from Anh and Li [1] that this was also observed by Duong and Li [17].

The paper is organized as follows. In Section 2, we first recall some results on the $H_{\infty}$ functional calculus and describe some assumptions on operators considered in this paper. In particular, we introduce the notion of $k$-Davies-Gaffney estimates with $k \in \mathbb{N}$ in (2.6) below. Some examples satisfying these assumptions are also given in this section.

Let $L$ be an operator satisfying assumptions in Section 2. In Section 3, using some ideas from [28, 30, 31, 32], we establish some off-diagonal estimates for some families of operators related to $L$. More precisely, we show that if $\left\{e^{-t L}\right\}_{t>0}$ satisfies the $k$-DaviesGaffney estimates, then the family $\left\{(z L)^{m} e^{-z L}\right\}_{z \in S_{\ell(\pi / 2-\omega)}^{0}}$ of operators for any $m \in \mathbb{N} \cup\{0\}$ also satisfies the $k$-Davies-Gaffney estimates in $z$ (see Lemma 3.1), the $k$-Davies-Gaffney estimates are stable under compositions (see Lemma 3.2) and the family $\{\psi(t L) f(L)\}_{t>0}$ of operators satisfies the $k$-Davies-Gaffney estimates of order $\sigma$ (see (3.7) below for the definition), where $\psi$ belongs to the decaying function class $\Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$ as in (2.2) below (see Lemma 3.3 below). Let $L_{1}$ be the $2 k$-order divergence form homogeneous elliptic operator with complex bounded measurable coefficients and $L_{2}$ the $2 k$-order Schrödinger type operator. In this section, we also prove that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ and the family $\left\{\sqrt{t} \nabla^{k} e^{-t L_{i}}\right\}_{t>0}$ of operators for $i \in\{1,2\}$ satisfy the $k$-Davies-Gaffney estimates, respectively, in Propositions 3.1 and 3.2.

In Definition 4.1 of Section 4, we first introduce the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(0,1]$ in terms of the square function $S_{L}$ defined via $\left\{e^{-t^{2 k} L}\right\}_{t>0}$ and, in Definition 4.3, the molecular Hardy space $H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ with $M \in(n(1 / p-1 / 2) /(2 k), \infty)$. Then, by using Lemma 3.1, we prove that for each $\left(H_{L}^{p}, \epsilon, M\right)$-molecule $m,\left\|S_{L}(m)\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}$ is uniformly bounded (see (4.6) below), which together with a boundedness criteria from [32] (see also Lemma 4.1 below) implies that $H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right) \subset H_{L}^{p}\left(\mathbb{R}^{n}\right)$. On the other hand, using the atomic decomposition of the tent space $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ and the $k$-Davies-Gaffney estimate, we obtain that the operator $\pi_{M, L}$ in (4.15) maps any $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atom into an $\left(H_{L}^{p}, \epsilon, M\right)$-molecule up to a harmless positive constant multiple in Lemma 4.2 below. Then, by a Calderón reproducing formula, we establish a molecular decomposition of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ which yields another inclusion $H_{L}^{p}\left(\mathbb{R}^{n}\right) \subset H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$. Thus, we obtain the molecular characterization of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 4.1 below.

Section 5 is devoted to the generalized square function characterization of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$. Motivated by [32], we first introduce the generalized square function Hardy space $H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(0,1]$ and some $\psi \in \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$ in Definition 5.1 below. Then, for any $\psi \in \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$ and all $f \in H_{\infty}\left(S_{\mu}^{0}\right)$ (see (2.1) for the definition), we introduce the operators $Q_{\psi, L}, \pi_{\psi, L}$ and their composition $Q^{f}$ (see (5.1), (5.4) and (5.5) for their definitions). Using the $k$ -Davies-Gaffney estimates of order $\sigma$ for $\{\psi(t L) f(L)\}_{t>0}$ in Lemma 3.3 below, we prove that the operator $Q^{f}$ is bounded on the tent space $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ (see Lemma 5.2), $Q_{\psi, L}$ is bounded from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ and $\pi_{\psi, L}$ is bounded from $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ to $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ for
some $\psi$ (see Lemma 5.3 below). Combining these boundedness and using a Calderón reproducing formula in (5.14), we then obtain the generalized square function characterization of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 5.1, which is used in obtaining the Riesz transform characterization of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ in Section 6. For all $\alpha \in(0, \infty)$, let $L^{\alpha}$ be the fractional power with exponent $\alpha$ of $L$ and the Hardy space $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)$ be defined as in (5.3) below via the square function $S_{L^{\alpha}}$ as in (5.2). As another application of Theorem 5.1, we then obtain in Corollary 5.1 that $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)=H_{L}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms, in particular, $H_{(-\Delta)^{k}}^{p}\left(\mathbb{R}^{n}\right)=H^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms for all $k \in \mathbb{N}$, where $H^{p}\left(\mathbb{R}^{n}\right)$ is the classical Hardy space in [42, 23].

Finally, in Section 6, we concentrate on the behavior of the Riesz transforms $\nabla^{k} L_{i}^{-1 / 2}$ on $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ for $i \in\{1,2\}$. By the gradient estimates of the semigroup $\left\{e^{-t L_{i}}\right\}_{t>0}$ in Proposition 3.2 and the composition rule of $k$-Davies-Gaffney estimates in Lemma 3.2, we first show that the two families of operators, $\left\{\nabla^{k} L_{i}^{-1 / 2}\left(I-e^{-t L_{i}}\right)^{M}\right\}_{t>0}$ and $\left\{\nabla^{k} L_{i}^{-1 / 2}\left(t L_{i} e^{-t L_{i}}\right)^{M}\right\}_{t>0}$ for all $M \in \mathbb{N}$, satisfy some estimates similar to the $k$-DaviesGaffney estimates of order $M$ (see Lemma 6.1 below). Then, using these estimates, we prove that for each $\left(H_{L_{i}}^{p}, \epsilon, M\right)$-molecule $m$ with $p \in(n /(n+k), 1]$ and

$$
M \in(n(1 / p-1 / 2) /(2 k), \infty)
$$

$\nabla^{k}\left(L_{i}^{-1 / 2}\right)(m)$ is a classical $H^{p}\left(\mathbb{R}^{n}\right)$-molecule up to a harmless constant multiple, which further implies that Riesz transforms $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ are bounded from $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ to the classical Hardy space $H^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 6.1 below. In the remaining part of this section, motivated by [32], by assuming that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-offdiagonal estimates for $r \in(1,2]$, we then establish the Riesz transform characterization of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$. To this end, we first show in Lemma 6.2 below that $\left\{t L_{1} e^{-t L_{1}}\right\}_{t>0}$ also satisfy the $L^{r}-L^{2} k$-off-diagonal estimates. We then recall some known results concerning the homogeneous Triebel-Lizorkin space $\dot{F}_{p, q}^{\alpha}\left(\mathbb{R}^{n}\right)$ and their atomic characterizations from [44, 27, 13] and [45, Proposition 4.3]. Let $W^{k, 2}\left(\mathbb{R}^{n}\right)$ be the homogenous Sobolev space of order $k$. With the help of these results, we show that if $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ when $p \in(0,1]$, then its atomic decomposition converges in both $\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)$ and $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ (see Lemma 6.3 below). Moreover, by the $L^{r}-L^{2} k$-off-diagonal estimates for $\left\{t L_{1} e^{-t L_{1}}\right\}_{t>0}$, we prove that for each $H^{k, p}\left(\mathbb{R}^{n}\right)$-atom $b, S_{1} \sqrt{L_{1}}(b)$ is uniformly bounded on $L^{p}\left(\mathbb{R}^{n}\right)$ (see (6.13) below), which, together with the generalized square function characterization of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 5.1 and Lemma 6.3, shows that $S_{1} \sqrt{L_{1}}$ is bounded from the Hardy-Sobolev space $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ to $L^{p}\left(\mathbb{R}^{n}\right)$. This combined with the boundedness of Riesz transforms on $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 6.1 yields the Riesz transform characterization of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ in Theorem 6.2 below. We point out in the proof of the estimate (6.13), we use the embedding result (6.15) below on the homogeneous Triebel-Lizorkin space from [44] and another key fact from [6, Theorem 1.1] that $\left\|\sqrt{L_{1}} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}$. The latter fact may not be true for $L_{2}$; see Remark 6.1 below. Thus, it seems that one needs some new ideas to obtain the Riesz characterization of $H_{L_{2}}^{p}\left(\mathbb{R}^{n}\right)$.

We now make some conventions on the notation. Throughout the whole paper, we always let $\mathbb{N} \equiv\{1,2, \cdots\}$ and $\mathbb{Z}_{+} \equiv \mathbb{N} \cup\{0\}$. Denote the differential operator $\frac{\partial^{|\alpha|}}{\partial x_{1}^{\alpha_{1}} \ldots \partial x_{1}^{\alpha_{1}}}$ simply by $\partial^{\alpha}$, where $\alpha \equiv\left(\alpha_{1}, \cdots, \alpha_{n}\right)$ and $|\alpha| \equiv \alpha_{1}+\cdots+\alpha_{n}$. We also denote the $2 k$-order
divergence form homogenous elliptic operator with complex bounded measurable coefficients $(-1)^{k} \sum_{|\alpha|=|\beta|=k} \partial^{\alpha}\left(a_{\alpha, \beta} \partial^{\beta}\right)$ by $L_{1}$ and the $2 k$-order Schrödinger type operator $(-\Delta)^{k}+V^{k}$ by $L_{2}$. We use $C$ to denote a positive constant, that is independent of the main parameters involved but whose value may differ from line to line, and $C(\alpha, \cdots)$ to denote a positive constant depending on the parameters $\alpha, \cdots$. Constants with subscripts, such as $C_{0}$, do not change in different occurrences. If $f \leq C g$, we then write $f \lesssim g$; and if $f \lesssim g \lesssim f$, we then write $f \sim g$. For all $x \in \mathbb{R}^{n}$ and $r \in(0, \infty)$, let $B(x, r) \equiv\left\{y \in \mathbb{R}^{n}:|x-y|<r\right\}$. Also, for any set $E \in \mathbb{R}^{n}$, we use $E^{\complement}$ to denote $\mathbb{R}^{n} \backslash E$ and $\chi_{E}$ the characteristic function of $E$.

## 2 Preliminaries

We first collect some basic results on the theory of $H_{\infty}$ functional calculus, developed by McIntosh in [39], that we need in the sequel. For more details and further references about functional calculus, we refer the reader to $[2,26,39]$ and the references therein.

For $\theta \in[0, \pi)$, the open and closed sectors, $S_{\theta}^{0}$ and $S_{\theta}$, of angle $\theta$ in the complex plane $\mathbb{C}$ are defined as follows:

$$
S_{\theta}^{0} \equiv\{z \in \mathbb{C} \backslash\{0\}:|\arg z|<\theta\}
$$

and

$$
S_{\theta} \equiv\{z \in \mathbb{C} \backslash\{0\}:|\arg z| \leq \theta\} \cup\{0\} .
$$

Let $\omega \in[0, \pi)$. A closed operator $T$ in $L^{2}\left(\mathbb{R}^{n}\right)$ is called of type $\omega$, if the spectrum of $T$, $\sigma(T)$, is contained in $S_{\omega}$, and for each $\theta \in(\omega, \pi)$, there exists a nonnegative constant $C$ such that for all $z \in \mathbb{C} \backslash S_{\theta},\left\|(T-z I)^{-1}\right\|_{\mathcal{L}\left(L^{2}\left(\mathbb{R}^{n}\right)\right)} \leq C|z|^{-1}$, where and in what follows, $\|S\|_{\mathcal{L}(\mathcal{H})}$ denotes the operator norm of the linear operator $S$ on the normed linear space $\mathcal{H}$.

For $\mu \in[0, \pi)$ and $\sigma, \tau \in(0, \infty)$, we need the following spaces of functions:

$$
\begin{align*}
H\left(S_{\mu}^{0}\right) & \equiv\left\{f: f \text { is holomorphic on } S_{\mu}^{0}\right\} \\
H_{\infty}\left(S_{\mu}^{0}\right) & \equiv\left\{f \in H\left(S_{\mu}^{0}\right):\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}<\infty\right\} \tag{2.1}
\end{align*}
$$

and

$$
\begin{equation*}
\Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right) \equiv\left\{f \in H\left(S_{\mu}^{0}\right):|f(\xi)| \leq C \inf \left\{|\xi|^{\sigma},|\xi|^{-\tau}\right\} \text { for all } \xi \in S_{\mu}^{0}\right\} \tag{2.2}
\end{equation*}
$$

It is known that every one to one operator $T$ of type $\omega$ in $L^{2}\left(\mathbb{R}^{n}\right)$ has a unique holomorphic functional calculus which is consistent with the usual definition of polynomials of operators; see, for example, [39]. More precisely, let $T$ be a one to one operator of type $\omega$, with $\omega \in[0, \pi), \mu \in(\omega, \pi), \sigma, \tau \in(0, \infty)$, and $f \in \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$. The function of the operator $T, f(T)$ can be defined by the $H_{\infty}$ functional calculus in the following way,

$$
\begin{equation*}
f(T) \equiv \frac{1}{2 \pi i} \int_{\gamma}(\xi I-T)^{-1} f(\xi) d \xi \tag{2.3}
\end{equation*}
$$

where $\gamma \equiv\left\{r e^{i \nu}: \infty>r>0\right\} \cup\left\{r e^{-i \nu}: 0<r<\infty\right\}, \nu \in(\omega, \mu)$, is a curve consisting of two rays parameterized anti-clockwise. It is well known that the above definition is independent of the choice of $\nu \in(\omega, \mu)$ and the integral in (2.3) is absolutely convergence in $\mathcal{L}\left(L^{2}\left(\mathbb{R}^{n}\right)\right.$ ) (see [39, 26]).

In what follows, we always assume $\omega \in[0, \pi / 2)$. Then, it follows from [26, Proposition 7.1.1] that for every operator $T$ of type $\omega$ in $L^{2}\left(\mathbb{R}^{n}\right),-T$ generates a holomorphic $C_{0}{ }^{-}$ semigroup $\left\{e^{-z L}\right\}_{z \in S_{\pi / 2-\omega}^{0}}$ on the open sector $S_{\pi / 2-\omega}^{0}$ such that $\left\|e^{-z L}\right\|_{\mathcal{L}\left(L^{2}\left(\mathbb{R}^{n}\right)\right)} \leq 1$ for all $z \in S_{\pi / 2-\omega}^{0}$ and, moreover, every nonnegative self-adjoint operator is of type 0 .

Let $\Psi\left(S_{\mu}^{0}\right) \equiv \cup_{\sigma, \tau>0} \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$. By the relationship between the associated semigroup and the resolvent of $T$, for all $f \in \Psi\left(S_{\mu}^{0}\right), f(T)$ can further be represented as

$$
\begin{equation*}
f(T) \equiv \int_{\Gamma_{+}} e^{-z T} \eta_{+}(z) d z+\int_{\Gamma_{-}} e^{-z T} \eta_{-}(z) d z \tag{2.4}
\end{equation*}
$$

where

$$
\begin{equation*}
\eta_{ \pm}(z) \equiv \frac{1}{2 \pi i} \int_{\gamma \pm} e^{\xi z} f(\xi) d \xi, \quad z \in \Gamma_{ \pm} \tag{2.5}
\end{equation*}
$$

$\Gamma_{ \pm} \equiv \mathbb{R}^{+} e^{ \pm i(\pi / 2-\theta)}, \gamma_{ \pm} \equiv \mathbb{R}^{+} e^{ \pm i \nu}$ and $0 \leq \omega<\theta<\nu<\mu<\pi / 2$. Here and in what follows, $\mathbb{R}^{+} \equiv(0, \infty)$.

It is well known that the above holomorphic functional calculus defined on $\Psi\left(S_{\mu}^{0}\right)$ can be extended to $H_{\infty}\left(S_{\mu}^{0}\right)$ via a limit process (see [39]). Recall that for $\mu \in(0, \pi)$, the operator $T$ is said to have a bounded $H_{\infty}\left(S_{\mu}^{0}\right)$ functional calculus in the Hilbert space $\mathcal{H}$, if there exists a positive constant $C$ such that for all $\psi \in H_{\infty}\left(S_{\mu}^{0}\right),\|\psi(T)\|_{\mathcal{L}(\mathcal{H})} \leq C\|\psi\|_{L^{\infty}\left(S_{\mu}^{0}\right)}$ and $T$ is called to have a bounded $H_{\infty}$ functional calculus in the Hilbert space $\mathcal{H}$ if there exists $\mu \in(0, \pi)$ such that $T$ has a bounded $H_{\infty}\left(S_{\mu}^{0}\right)$ functional calculus.

Now, we describe our assumptions of operators $L$ considered in this paper. Throughout the whole paper, we always assume that $L$ satisfies the following assumptions:
$\left(\mathrm{A}_{1}\right)$ The operator $L$ is a one to one operator of type $\omega$ in $L^{2}\left(\mathbb{R}^{n}\right)$ with $\omega \in[0, \pi / 2)$;
$\left(\mathrm{A}_{2}\right)$ The operator $L$ has a bounded $H_{\infty}$ functional calculus in $L^{2}\left(\mathbb{R}^{n}\right)$;
$\left(\mathrm{A}_{3}\right)$ Let $k \in \mathbb{N}$. The operator $L$ generates a holomorphic semigroup $\left\{e^{-t L}\right\}_{t>0}$ which satisfies the $k$-Davies-Gaffney estimate, namely, there exist positive constants $\widetilde{C}$ and $C_{1}$ such that for all closed sets $E$ and $F$ in $\mathbb{R}^{n}, t \in(0, \infty)$ and $f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$,

$$
\begin{equation*}
\left\|e^{-t L} f\right\|_{L^{2}(F)} \leq \widetilde{C} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{1} t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \tag{2.6}
\end{equation*}
$$

where and in what follows, $\operatorname{dist}(E, F) \equiv \inf _{x \in E, y \in F}|x-y|$ is the distance between $E$ and $F$.

Remark 2.1. We point out that when $k=1$, the $k$-Davies-Gaffney estimate is usually called the Davies-Gaffney estimate (or the $L^{2}$ off-diagonal estimate or just the Gaffney estimate); see, for example, [30, 31, 28, 34, 32].

Let $k \in \mathbb{N}$. Examples of operators, satisfying the above assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$, include the following $2 k$-order divergence form homogeneous elliptic operator:

$$
L_{1} \equiv(-1)^{k} \sum_{|\alpha|=|\beta|=k} \partial^{\alpha}\left(a_{\alpha, \beta} \partial^{\beta}\right)
$$

with complex bounded measurable coefficients $a_{\alpha, \beta}$ for all multi-indices $\alpha, \beta$ and the $2 k$ order Schrödinger type operator $L_{2} \equiv(-\Delta)^{k}+V^{k}$ with $0 \leq V \in L_{\mathrm{loc}}^{k}\left(\mathbb{R}^{n}\right)$. More precisely, let $W^{k, 2}\left(\mathbb{R}^{n}\right)$ be the Sobolev space of order $k$ endowed with the norm

$$
\|\cdot\|_{W^{k, 2}\left(\mathbb{R}^{n}\right)} \equiv \sum_{0 \leq|\alpha| \leq k}\left\|\partial^{\alpha}(\cdot)\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} .
$$

Denote by $\mathfrak{a}$ the sesquilinear form given by

$$
\begin{equation*}
\mathfrak{a}(f, g) \equiv \int_{\mathbb{R}^{n}} \sum_{|\alpha|=|\beta|=k} a_{\alpha, \beta}(x) \partial^{\beta} f(x) \overline{\partial^{\alpha} g(x)} d x \tag{2.7}
\end{equation*}
$$

with domain $D(\mathfrak{a}) \equiv W^{k, 2}\left(\mathbb{R}^{n}\right)$. We further assume that $\mathfrak{a}$ satisfies the ellipticity condition, that is, there exist positive constants $0<\lambda \leq \Lambda<\infty$ such that

$$
\begin{equation*}
\left\|a_{\alpha, \beta}\right\|_{L^{\infty}\left(\mathbb{R}^{n}\right)} \leq \Lambda \quad \text { for all } \alpha, \beta \text { with }|\alpha|=k=|\beta| \tag{2.8}
\end{equation*}
$$

and

$$
\begin{equation*}
\Re \mathfrak{a}(f, f) \geq \lambda\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \quad \text { for all } f \in W^{k, 2}\left(\mathbb{R}^{n}\right) \tag{2.9}
\end{equation*}
$$

where and in what follows, $\Re z$ for any $z \in \mathbb{C}$ denotes the real part of $z$. The $2 k$-order divergence form homogeneous elliptic operator $L_{1}$ with complex bounded measurable coefficients is then defined to be the operator associated to the form $\mathfrak{a}$.

Let $\omega \in[0, \pi / 2]$. Recall that an operator $T$ in the Hilbert space $\mathcal{H}$ is called $m-\omega$ accretive if
(i) the range of the operator $T+I, R(T+I)$, is dense in $\mathcal{H}$;
(ii) for all $u \in D(T),|\arg (T u, u)| \leq \omega$,
where $D(T)$ denotes the domain of $T$ and $\arg (T u, u)$ the argument of $(T u, u)$. It is known by [26, Proposition 7.1.1] that every closed $m$ - $\omega$-accretive operator is of type $\omega$ (see [26, p. 173]).

From [6], it follows that $L_{1}$ is closed and maximal accretive (see [26, p. 327] for the definition), which further yields that $R\left(L_{1}+I\right)$ is dense in $L^{2}\left(\mathbb{R}^{n}\right)$; see, for example [26, Proposition C.7.2]. Moreover, by the ellipticity condition (2.8) and (2.9), we obtain that for all $f \in W^{k, 2}\left(\mathbb{R}^{n}\right)$,

$$
\left|\tan \left(\arg \left(L_{1} f, f\right)\right)\right|=\left|\frac{\Im\left(L_{1} f, f\right)}{\Re\left(L_{1} f, f\right)}\right| \leq \frac{\Lambda}{\lambda},
$$

where and in what follows, $\Im z$ for any $z \in \mathbb{C}$ denotes the imaginary part of $z$. Thus, $\left|\arg \left(L_{1} f, f\right)\right| \leq \arctan \frac{\Lambda}{\lambda}$, which together with the fact that $R\left(L_{1}+I\right)$ is dense in $L^{2}\left(\mathbb{R}^{n}\right)$ shows that $L_{1}$ is an $m$-arctan $\frac{\Lambda}{\lambda}$-accretive operator in $L^{2}\left(\mathbb{R}^{n}\right)$ with the angle $\arctan \frac{\Lambda}{\lambda} \in$ $[\pi / 4, \pi / 2)$. Thus, $L_{1}$ is an operator of type $\arctan \frac{\Lambda}{\lambda}$.

Now, we show that $L_{1}$ is one to one. Let $N\left(L_{1}\right) \equiv\left\{f \in D\left(L_{1}\right): L_{1} f=0\right\}$ be the null space of $L_{1}$. For any fixed $f \in N\left(L_{1}\right)$, by the elliptic condition (2.8) and (2.9), we have

$$
\begin{equation*}
\int_{\mathbb{R}^{n}}\left|\nabla^{k} f(x)\right|^{2} d x \sim\left|\left(L_{1} f, f\right)\right|=0 \tag{2.10}
\end{equation*}
$$

which implies that $\nabla^{k} f=0$ almost everywhere in $\mathbb{R}^{n}$. In what follows, denote by $C_{c}^{\infty}\left(\mathbb{R}^{n}\right)$ the space of all $C^{\infty}$ functions with compact support in $\mathbb{R}^{n}$. For all $f \in W^{k, 2}\left(\mathbb{R}^{n}\right)$, by the density of $C_{c}^{\infty}\left(\mathbb{R}^{n}\right)$ in $W^{k, 2}\left(\mathbb{R}^{n}\right)$, there exists a sequence $\left\{f_{j}\right\}_{j \in \mathbb{N}}$ of functions in $C_{c}^{\infty}\left(\mathbb{R}^{n}\right)$ such that $\lim _{j \rightarrow \infty} f_{j}=f$ in $W^{k, 2}\left(\mathbb{R}^{n}\right)$. Denote the Fourier transform and the inverse Fourier transform of $f$, respectively, by $\hat{f}$ and $\check{f}$. If $f \in N\left(L_{1}\right)$, by (2.10), the fact that $f_{j} \in C_{c}^{\infty}\left(\mathbb{R}^{n}\right)$, the multiplication formula of Fourier transform and Plancherel's theorem (see, for example, [24, Theorem 2.2.14]), we have that for all $\varphi \in C_{c}^{\infty}\left(\mathbb{R}^{n}\right)$,

$$
\begin{aligned}
0 & =\left(\nabla^{k} f, \hat{\varphi}\right)=\lim _{j \rightarrow \infty}\left(\nabla^{k} f_{j}, \hat{\varphi}\right)=\lim _{j \rightarrow \infty}(-1)^{k}\left(f_{j}, \nabla^{k} \hat{\varphi}\right)=\lim _{j \rightarrow \infty}(-1)^{k}\left(\hat{f}_{j},\left(\nabla^{k} \hat{\varphi}\right)\right) \\
& =\lim _{j \rightarrow \infty} i^{k}\left(\hat{f}_{j},(\cdot)^{k} \varphi(\cdot)\right)=i^{k}\left(\hat{f},(\cdot)^{k} \varphi(\cdot)\right),
\end{aligned}
$$

which implies that $\operatorname{supp} \hat{f} \subset\{0\}$. By [24, Corollary 2.4.2], we have that $f$ is a polynomial, which, together with the fact that $f \in L^{2}\left(\mathbb{R}^{n}\right)$, implies that $f=0$. Hence, $N\left(L_{1}\right)=\{0\}$ and $L_{1}$ is one to one.

Since $L_{1}$ is maximal accretive, from [2], it follows that $L_{1}$ has a bounded holomorphic functional calculus. Finally, in Proposition 3.1 below, we will show that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $k$-Davies-Gaffney estimate. Thus, the $2 k$-order divergence form homogenous elliptic operator $L_{1}$ with complex bounded measurable coefficients satisfies the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$.

Let $k \in \mathbb{N}, \Delta \equiv \sum_{j=1}^{n} \frac{\partial^{2}}{\partial x_{j}^{2}}$ be the Laplace operator and $0 \leq V \in L_{\text {loc }}^{k}\left(\mathbb{R}^{n}\right)$. The $2 k$-order Schrödinger type operator $L_{2} \equiv(-\Delta)^{k}+V^{k}$ is the associated operator of the following sesquilinear form

$$
\begin{equation*}
\mathfrak{b}(f, g) \equiv \int_{\mathbb{R}^{n}} \nabla^{k} f(x) \overline{\nabla^{k} g(x)} d x+\int_{\mathbb{R}^{n}}[V(x)]^{k} f(x) \overline{g(x)} d x \tag{2.11}
\end{equation*}
$$

with domain $D(\mathfrak{b}) \equiv\left\{f \in W^{k, 2}\left(\mathbb{R}^{n}\right): \int_{\mathbb{R}^{n}}[V(x)]^{k}|f(x)|^{2} d x<\infty\right\}$ which is also dense in $L^{2}\left(\mathbb{R}^{n}\right)$, since $C_{c}^{\infty}\left(\mathbb{R}^{n}\right) \subset D(\mathfrak{b})$.

It is easy to see that the $2 k$-order Schrödinger type operator $L_{2}$ is a nonnegative selfadjoint operator. From [26], it follows that $L_{2}$ is $m$ - 0 -accretive. Thus, by [26, Proposition 7.1.1], $L_{2}$ is a one to one operator of type 0 . Therefore, $L_{2}$ has a bounded $H_{\infty}$ functional calculus. Moreover, by [9], the semigroup $\left\{e^{-t L_{2}}\right\}_{t>0}$ satisfies a Gaussian type estimate, that is, the integral kernel $e^{-t L_{2}}(x, y)$ of $e^{-t L_{2}}$ has the property that there exist positive
constant $C_{2}$ and $C_{3}$ such that for all $t \in(0, \infty)$ and $x, y \in \mathbb{R}^{n}$,

$$
\begin{equation*}
\left|e^{-t L_{2}}(x, y)\right| \leq C_{2} t^{-n /(2 k)} \exp \left\{-C_{3} \frac{|x-y|^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\} \tag{2.12}
\end{equation*}
$$

which implies that the semigroup $\left\{e^{-t L_{2}}\right\}_{t>0}$ satisfies the $k$-Davies-Gaffney estimate immediately. Thus, the $2 k$-order schrödinger type operator $L_{2}$ also satisfies the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$.

## $3 k$-Davies-Gaffney estimates

In this section, we prove some properties about the $k$-Davies-Gaffney estimates. We point out that when $k=1$ and $L$ is a non-negative self-adjoint operator or a second order divergence form elliptic operator with complex bounded measurable coefficients, these properties are already well known; see, for example, [5, 30, 31, 28, 34, 32].

Let $\theta \in[0, \pi / 2)$ and $E, F$ be two closed sets in $\mathbb{R}^{n}$. A family $\{T(z)\}_{z \in S_{\theta}^{0}}$ of operators is called to satisfies the $k$-Davies-Gaffney estimate in $z$ if there exist positive constants $C_{4}$ and $C_{5}$ such that for all $f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$ and $z \in S_{\theta}^{0}$,

$$
\begin{equation*}
\|T(z) f\|_{L^{2}(F)} \leq C_{5} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \tag{3.1}
\end{equation*}
$$

For any operator satisfying the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2, we have the following property.

Lemma 3.1. Assume that the operator $L$ defined in $L^{2}\left(\mathbb{R}^{n}\right)$ satisfies the assumptions $\left(\mathrm{A}_{1}\right)$, $\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. Then for all $\ell \in(0,1), m \in \mathbb{Z}_{+}$, the family of operators, $\left\{(z L)^{m} e^{-z L}\right\}_{z \in S_{\ell\left(\frac{\pi}{2}-\omega\right)}^{0}}$, satisfy the $k$-Davies-Gaffney estimate in $z$, (3.1), with positive constants $C_{4}$ and $C_{5}$ depending only on $m, \ell, n, k, \omega, \widetilde{C}$ and $C_{1}$.

Proof. We prove this lemma by using some ideas from [28]. Since $L$ is of type $\omega$, we know that the semigroup $\left\{e^{-t L}\right\}_{t>0}$ can be extended to a holomorphic semigroup $\left\{e^{-z L}\right\}_{z \in S_{\pi / 2-\omega}^{0}}$. Thus, for all $z \in S_{\pi / 2-\omega}^{0}$, closed sets $E, F \subset \mathbb{R}^{n}$ and $f, g \in L^{2}\left(\mathbb{R}^{n}\right)$ supported respectively in $E$ and $F$, the function $G(z): z \longmapsto\left(e^{-z L} f, g\right)$ is holomorphic on $S_{\pi / 2-\omega}^{0}$. Moreover, $G$ satisfies the following properties:
(i) there exists a nonnegative constant $C$ such that for all $z \in S_{\pi / 2-\omega}^{0}$,

$$
|G(z)| \leq C\|f\|_{L^{2}(E)}\|g\|_{L^{2}(F)},
$$

(ii) there exist nonnegative constants $C$ and $C_{1}$ such that for all $t \in(0, \infty)$,

$$
|G(t)| \leq C \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{1} t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)}\|g\|_{L^{2}(F)}
$$

In [40, Lemma 6.18], letting $\psi \equiv \pi / 2-\omega, a \equiv C\|f\|_{L^{2}(E)}\|g\|_{L^{2}(F)}, \beta \equiv 0, b \equiv$ $\frac{1}{C_{1}}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}, r \equiv t$ and $\alpha \equiv \frac{1}{2 k-1}$, we then obtain that for any $z \equiv r e^{i \theta} \in$ $S_{\ell(\pi / 2-\omega)}^{0}$,

$$
\begin{align*}
|F(z)| & \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{2(2 k-1) C_{1} r^{1 /(2 k-1)}} \sin (\pi / 2-\omega-|\theta|)\right\}\|f\|_{L^{2}(E)}\|g\|_{L^{2}(F)}  \tag{3.2}\\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4} r^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)}\|g\|_{L^{2}(F)},
\end{align*}
$$

where $C_{4} \equiv \frac{2 C_{1}(2 k-1)}{\sin \left((1-\ell)\left(\frac{\pi}{2}-\omega\right)\right)}$. From the analytic property of semigroups and the Cauchy integral formula, it follows that for all $m \in \mathbb{N}$ and $z \in S_{\ell\left(\frac{\pi}{2}-\omega\right)}^{0}$

$$
\begin{equation*}
(z L)^{m} e^{-z L}=(-z)^{m} \frac{m!}{2 \pi i} \int_{|\xi-z|=\eta|z|} e^{-\xi L} \frac{d \xi}{(\xi-z)^{m+1}}, \tag{3.3}
\end{equation*}
$$

where $\eta \in(0, \sin ((1-\ell)(\pi / 2-\omega)))$. Thus, for any $z \in S_{\ell\left(\frac{\pi}{2}-\omega\right)}^{0}$, the ball $B(z, \eta|z|) \subset$ $S_{\pi / 2-\omega}^{0}$. Combining (3.2) and (3.3), by Minkowski's inequality, we obtain

$$
\begin{aligned}
\left\|(z L)^{m} e^{-z L} f\right\|_{L^{2}(F)} & \lesssim|z|^{m} \int_{|\xi-z|=\eta|z|}\left|\frac{1}{(\xi-z)^{m+1}}\right|\left\|e^{-\xi L} f\right\|_{L^{2}(F)}|d \xi| \\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)},
\end{aligned}
$$

which implies that $\left\{(z L)^{m} e^{-z L}\right\}_{S_{\ell\left(\frac{\pi}{2}-\omega\right)}^{0}}$ satisfies the $k$-Davies-Gaffney estimate in $z$. This finishes the proof of Lemma 3.1.

Lemma 3.2. Let $\left\{A_{t}\right\}_{t>0},\left\{B_{s}\right\}_{s>0}$ be two families of linear operators, $C_{6}$ and $C_{7}$ two positive constants. Assume that for all closed sets $E, F \subset \mathbb{R}^{n}, f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$ and $t>0$, the following estimates hold:

$$
\begin{equation*}
\left\|A_{t} f\right\|_{L^{2}(F)} \leq C_{6} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{7} t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)}, \tag{3.4}
\end{equation*}
$$

and

$$
\begin{equation*}
\left\|B_{s} f\right\|_{L^{2}(F)} \leq C_{6} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{7} s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \tag{3.5}
\end{equation*}
$$

Then, there exists a positive constant $C$ such that for all $t, s>0$, all closed sets $E, F \subset \mathbb{R}^{n}$ and $f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$,

$$
\begin{equation*}
\left\|A_{t} B_{s} f\right\|_{L^{2}(F)} \leq C \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{\widetilde{C}_{7}(\max \{t, s\})^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \tag{3.6}
\end{equation*}
$$

where $\widetilde{C}_{7} \equiv C_{7} 2^{2 k /(2 k-1)}$.

Proof. If $\operatorname{dist}(E, F)=0$, then (3.6) is a simple corollary of (3.4) and (3.5). Now, we assume that $\operatorname{dist}(E, F)>0$. As in [29], let $\rho \equiv \operatorname{dist}(E, F)$ and $G \equiv\left\{x \in \mathbb{R}^{n}: \operatorname{dist}(x, F)<\rho / 2\right\}$. Denote by $\bar{G}$ the closure of $G$. It is clear that $\operatorname{dist}(E, \bar{G}) \geq \rho / 2$. Moreover, by (3.4) and (3.5), we have

$$
\begin{aligned}
\left\|A_{t}\left(\chi_{G} B_{s} f\right)\right\|_{L^{2}(F)} & \leq\left\|A_{t}\left(\chi_{G} B_{s} f\right)\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\left\|B_{s} f\right\|_{L^{2}(\bar{G})} \\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, \bar{G})]^{2 k /(2 k-1)}}{C_{7} s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \\
& \sim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{7} 2^{2 k /(2 k-1)} s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} .
\end{aligned}
$$

Let $\widetilde{C}_{7} \equiv C_{7} 2^{2 k /(2 k-1)}$. Similarly, by (3.4) and (3.5), we obtain

$$
\begin{aligned}
\left\|A_{t}\left(\chi_{\mathbb{R}^{n} \backslash G} B_{s} f\right)\right\|_{L^{2}(F)} & \lesssim \exp \left\{-\frac{\left[\operatorname{dist}\left(\mathbb{R}^{n} \backslash G, F\right)\right]^{2 k /(2 k-1)}}{C_{7} t^{1 /(2 k-1)}}\right\}\left\|B_{s} f\right\|_{L^{2}(E)} \\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{\widetilde{C}_{7} t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} .
\end{aligned}
$$

Combining the above estimates, we have

$$
\begin{aligned}
& \left\|A_{t} B_{s} f\right\|_{L^{2}(F)} \\
& \leq\left\|A_{t}\left(\chi_{G} B_{s} f\right)\right\|_{L^{2}(F)}+\left\|A_{t}\left(\chi_{\mathbb{R}^{n} \backslash G} B_{s} f\right)\right\|_{L^{2}(F)} \\
& \\
& \lesssim\left[\exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{\widetilde{C}_{7} 5^{1 /(2 k-1)}}\right\}+\exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{\widetilde{C}_{7} t^{1 /(2 k-1)}}\right\}\right]\|f\|_{L^{2}(E)} \\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{\widetilde{C}_{7} \max \{t, s\}^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)},
\end{aligned}
$$

which completes the proof of Lemma 3.2.
Let $\sigma \in[0, \infty)$. As in [32], a family $\left\{T_{t}\right\}_{t>0}$ of operators is called to satisfy the $k$ -Davies-Gaffney estimate of order $\sigma$, if there exists a positive constant $C_{8}$, depending on $\sigma$, such that for all closed sets $E, F \subset \mathbb{R}^{n}, g \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$ and $t \in(0, \infty)$,

$$
\begin{equation*}
\left\|T_{t} g\right\|_{L^{2}(F)} \leq C_{8} \min \left\{1, \frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right\}^{\sigma}\|g\|_{L^{2}(E)} \tag{3.7}
\end{equation*}
$$

Lemma 3.3. Let $\mu \in(\omega, \pi / 2), \psi \in \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right)$ for some $\sigma \in(0, \infty), \tau \in(1, \infty)$, and $f \in H_{\infty}\left(S_{\mu}^{0}\right)$. Then the family $\{\psi(t L) f(L)\}_{t>0}$ of operators satisfy the $k$-Davies-Gaffney estimate of order $\sigma$, (3.7), with the positive constant $C_{8}$ controlled by $\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}$.

Proof. For any fixed $\psi \in \Psi_{\sigma, \tau}\left(S_{\mu}^{0}\right) \subset \Psi\left(S_{\mu}^{0}\right)$ and $f \in H_{\infty}\left(S_{\mu}^{0}\right)$, by (2.4) and (2.5), we have

$$
\begin{equation*}
\psi(t L) f(L)=\int_{\Gamma_{+}} e^{-z L} \eta_{+}(z) d z+\int_{\Gamma_{-}} e^{-z L} \eta_{-}(z) d z \tag{3.8}
\end{equation*}
$$

where $\Gamma_{ \pm} \equiv \mathbb{R}^{+} e^{ \pm i(\pi / 2-\theta)}$ and for all $z \in \Gamma_{ \pm}$,

$$
\eta_{ \pm}(z)=\frac{1}{2 \pi i} \int_{\gamma_{ \pm}} e^{\xi z} \psi(t \xi) f(\xi) d \xi
$$

$\gamma_{ \pm} \equiv \mathbb{R}^{+} e^{ \pm i \nu}$ and $0 \leq \omega<\theta<\nu<\mu<\pi / 2$. It was proved in [32, (2.32)] that for all $z \in \Gamma_{ \pm}$,

$$
\begin{equation*}
\left|\eta_{ \pm}(z)\right| \lesssim \frac{\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}}{t} \min \left\{1,\left(\frac{t}{|z|}\right)^{\sigma+1}\right\} \tag{3.9}
\end{equation*}
$$

Thus, by (3.8) and Minkowski's inequality, we have that for all $g \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$,

$$
\begin{aligned}
\|\psi(t L) f(L) g\|_{L^{2}(F)} & \leq \int_{\Gamma_{+}}\left\|e^{-z L} g\right\|_{L^{2}(F)}\left|\eta_{+}(z)\right||d z|+\int_{\Gamma_{-}}\left\|e^{-z L} g\right\|_{L^{2}(F)}\left|\eta_{-}(z)\right||d z| \\
& \equiv \mathrm{J}_{+}+\mathrm{J}_{-} .
\end{aligned}
$$

Since $\pi / 2-\theta<\pi / 2-\omega$, there exists a positive number $\ell \in(0,1)$ such that $\pi / 2-\theta<$ $\ell(\pi / 2-\omega)$, which immediately yields that $S_{\pi / 2-\theta}^{0} \subset S_{\ell(\pi / 2-\omega)}^{0}$. Thus, by Lemma 3.1, the family $\left\{e^{-z L}\right\}_{z \in S_{\pi / 2-\theta}^{0}}$ satisfy the $k$-Davies-Gaffney estimate in $z$, which, together with (3.9), implies that

$$
\begin{aligned}
\mathrm{J}_{ \pm} \lesssim & \|g\|_{L^{2}(E)} \int_{\Gamma_{ \pm}} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\}\left|\eta_{ \pm}(z)\right||d z| \\
\lesssim & \|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\|g\|_{L^{2}(E)} \int_{\Gamma_{ \pm}} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\} \min \left\{1,\left(\frac{t}{|z|}\right)^{\sigma+1}\right\} \frac{1}{t}|d z| \\
\lesssim & {\left[\int_{\left\{z \in \Gamma_{ \pm}:|z| \leq t\right\}} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\} \min \left\{1,\left(\frac{t}{|z|}\right)^{\sigma+1}\right\} \frac{1}{t}|d z|\right.} \\
& \left.+\int_{\left\{z \in \Gamma_{ \pm}:|z|>t\right\}} \cdots\right]\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\|g\|_{L^{2}(E)} \equiv\left[\mathrm{O}_{1}+\mathrm{O}_{2}\right]\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\|g\|_{L^{2}(E)} .
\end{aligned}
$$

We estimate $\mathrm{O}_{1}$ by

$$
\begin{aligned}
\mathrm{O}_{1} & \lesssim \int_{\left\{z \in \Gamma_{ \pm}:\right.} \exp \left\{-\frac{[\operatorname{dist}(E t\})}{\left.C_{4}|z|^{1 /(2 k-1)}\right]^{2 k /(2 k-1)}}\right\} \frac{1}{t}|d z| \\
& \lesssim \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|t|^{1 /(2 k-1)}}\right\} .
\end{aligned}
$$

On the other hand, $\mathrm{O}_{2}$ can be written into

$$
\mathrm{O}_{2} \lesssim \int_{\left\{z \in \Gamma_{ \pm}:|z|>t\right\}} \exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{4}|z|^{1 /(2 k-1)}}\right\}\left(\frac{t}{|z|}\right)^{\sigma+1} \frac{1}{t}|d z| .
$$

If $t \geq[\operatorname{dist}(E, F)]^{2 k}$, in this case, we trivially have

$$
\mathrm{O}_{2} \lesssim \int_{\left\{z \in \Gamma_{ \pm}:|z|>t\right\}}\left(\frac{t}{|z|}\right)^{\sigma+1} \frac{1}{t}|d z| \sim 1
$$

If $t<[\operatorname{dist}(E, F)]^{2 k}$, by choosing $N \in[\sigma, \infty)$, we obtain

$$
\begin{aligned}
\mathrm{O}_{2} \lesssim & \int_{\left\{z \in \Gamma_{ \pm}: t<|z| \leq[\operatorname{dist}(E, F)]^{2 k}\right\}}\left(\frac{|z|}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{N}\left(\frac{t}{|z|}\right)^{\sigma+1} \frac{1}{t}|d z| \\
& +\int_{\left\{z \in \Gamma_{ \pm}:|z|>[\operatorname{dist}(E, F)]^{2 k}\right\}}\left(\frac{t}{|z|}\right)^{\sigma+1} \frac{1}{t}|d z| \\
\lesssim & \left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{N}+\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{\sigma} \sim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{\sigma} .
\end{aligned}
$$

Combining the estimates of $\mathrm{O}_{1}$ and $\mathrm{O}_{2}$, we obtain that $\{\psi(t L) f(L)\}_{t>0}$ satisfies the $k$ -Davies-Gaffney estimate of order $\sigma$.

Now, we turn to some properties of the operators $L_{1}$ and $L_{2}$ given in Section 2. First, we introduce the definition of the Legendre transform. Let $h$ be a real valued function defined on $[0, \infty)$. The Legendre transform $h^{\sharp}$ of $h$ is defined by setting, for all $s \in \mathbb{R}$,

$$
\begin{equation*}
h^{\sharp}(s) \equiv \sup _{t \geq 0}\{s t-h(t)\} . \tag{3.10}
\end{equation*}
$$

We have the following proposition about the operator $L_{1}$.
Proposition 3.1. Let $L_{1}$ be the $2 k$-order divergence form homogeneous elliptic operator defined as in Section 2. Then, the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $k$-Davies-Gaffney estimate.

Proof. We prove Proposition 3.1 by borrowing some ideas from [10, 11, 16]. In [11, Theorem 1.2], letting $(\Omega, \mathscr{U}, \mu, d)$ be the usual Euclidean space $\mathbb{R}^{n}$, endowed with the Lebesgue measure $d x$ and the Euclidean distant $d$, and with the set class $\mathscr{U}$ being the set of all Lebesgue measurable sets, and also letting $\mathcal{A} \equiv\left\{\phi \in C^{\infty}\left(\mathbb{R}^{n}\right) \cap L^{\infty}\left(\mathbb{R}^{n}\right):\left\|D^{\alpha} \phi\right\|_{L^{\infty}\left(\mathbb{R}^{n}\right)} \leq\right.$ $1,1 \leq|\alpha| \leq k\}, p \equiv q \equiv 2, \alpha \equiv \beta \equiv \gamma=0, r \equiv t^{1 /(2 k)}, h(x) \equiv x^{2 k /(2 k-1)}$ for all $x \in[0, \infty)$ and $\mathcal{R} \equiv e^{-t L_{1}}$, we then obtain the following two equivalent statements:
(i) There exists a positive constant $C(k)$, depending on $k$, such that for all $\phi \in \mathcal{A}$, $\rho \in[0, \infty)$ and $t \in(0, \infty)$,

$$
\begin{equation*}
\left\|e^{-\rho \phi} e^{-t L_{1}} e^{\rho \phi}\right\|_{\mathcal{L}\left(L^{2}\left(\mathbb{R}^{n}\right)\right)} \leq e^{C(k) h^{\sharp}\left(\rho t^{1 /(2 k)}\right)}, \tag{3.11}
\end{equation*}
$$

where, by (3.10),

$$
h^{\sharp}\left(\rho t^{1 /(2 k)}\right) \equiv \sup _{s \geq 0}\left\{\rho t^{1 /(2 k)} s-s^{2 k /(2 k-1)}\right\}=\left[\frac{(2 k-1)^{2 k-1}}{(2 k)^{2 k}}\right] \rho^{2 k} t ;
$$

(ii) There exists a positive constant $C_{1}$ such that for all the closed sets $E$ and $F$ of $\mathbb{R}^{n}$ and $t \in(0, \infty)$,

$$
\begin{equation*}
\left\|\chi_{E} e^{-t L_{1}} \chi_{F}\right\|_{\mathcal{L}\left(L^{2}\left(\mathbb{R}^{n}\right)\right)} \leq \exp \left(-\frac{\operatorname{dist}(E, F)}{C_{1} t^{1 /(2 k)}}\right)^{2 k /(2 k-1)} \tag{3.12}
\end{equation*}
$$

Recall that in this case, by [16, Lemma 4], $d(E, F)$ defined in $[11,(1.4)]$ is equivalent to $\inf _{x \in E, y \in F}|x-y|$ and, moreover, $C(k)$ in [11, Theorem 1.2(ii)] is assumed to be 1 . However, by the change of variables, we easily see that the above equivalent statements are simply corollary of the equivalence of (ii) and (iii) of Theorem 1.2 in [11].

Notice that by the density of the simple functions in $L^{2}\left(\mathbb{R}^{n}\right),(3.12)$ is equivalent to the $k$-Davies-Gaffney estimate. Thus, to prove that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $k$-Davies-Gaffney estimate, by the equivalence of (i) and (ii), it suffices to prove (3.11).

To this end, let $\mathfrak{a}$ be the sesquilinear form as in (2.7) associated with $L_{1}$. Recall that its twisted form is defined by setting, for all $\rho \in[0, \infty), \phi \in \mathcal{A}$ and $f, g \in W^{k, 2}\left(\mathbb{R}^{n}\right)$,

$$
\mathfrak{a}_{\rho \phi}(f, g) \equiv \mathfrak{a}\left(e^{\rho \phi} f, e^{-\rho \phi} g\right),
$$

which, together with the Leibniz formula, further yields that there exist positive constant $C(\alpha, \gamma)$ and $C(\beta, \widetilde{\gamma})$ with $|\alpha|=|\beta|=k, 0<\gamma \leq \alpha$ and $0<\widetilde{\gamma} \leq \beta$,

$$
\begin{aligned}
\mathfrak{a}_{\rho \phi}(f, f)= & \sum_{|\alpha|=|\beta|=k} \int_{\mathbb{R}^{n}} a_{\alpha, \beta}(x) \partial^{\alpha}\left(e^{\rho \phi} f\right)(x) \overline{\partial^{\beta}\left(e^{-\rho \phi} f\right)(x)} d x \\
= & \sum_{|\alpha|=|\beta|=k} \int_{\mathbb{R}^{n}} a_{\alpha, \beta}(x)\left\{\left[\sum_{0<\gamma \leq \alpha} C(\alpha, \gamma) \partial^{\gamma} e^{\rho \phi(x)} \partial^{\alpha-\gamma} f(x)+e^{\rho \phi(x)} \partial^{\alpha} f(x)\right]\right. \\
& +\left[\sum_{0<\tilde{\gamma} \leq \beta} C\left(\beta, \widetilde{\gamma} \overline{\partial^{2} \tilde{\gamma}} e^{-\rho \phi(x)} \partial^{\beta-\tilde{\gamma} f(x)}+\overline{e^{-\rho \phi(x)} \partial^{\beta} f(x)}\right]\right\} d x \\
= & \sum_{|\alpha|=|\beta|=k} \int_{\mathbb{R}^{n}} a_{\alpha, \beta}(x) \\
& \times\left\{\left[\sum_{0<\gamma \leq \alpha, 0<\tilde{\gamma} \leq \beta} C(\alpha, \gamma) C(\beta, \widetilde{\gamma}) \partial^{\gamma} e^{\rho \phi(x)} \partial^{\alpha-\gamma} f(x) \overline{\partial_{\tilde{\gamma}} e^{-\rho \phi(x)} \partial^{\beta-\tilde{\gamma}} f(x)}\right]\right. \\
& +\left[\overline{e^{-\rho \phi(x)} \partial^{\beta} f(x)} \sum_{0<\gamma \leq \alpha} C(\alpha, \gamma) \partial^{\gamma} e^{\rho \phi(x)} \partial^{\alpha-\gamma} f(x)\right]
\end{aligned}
$$

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$$
\left.+\left[\overline{e^{\rho \phi(x)} \partial^{\alpha} f(x)} \sum_{0<\tilde{\gamma} \leq \beta} C(\beta, \widetilde{\gamma}) \partial^{\widetilde{\gamma}} e^{-\rho \phi(x)} \partial^{\beta-\widetilde{\gamma}} f(x)\right]\right\} d x+\mathfrak{a}(f, f) .
$$

Let $C(\alpha, 0) \equiv 1 \equiv C(\beta, 0)$ and $\widetilde{C}(k) \equiv \Lambda \sum_{|\alpha|=|\beta|=k}\left[\sum_{0 \leq \gamma \leq \alpha, 0 \leq \tilde{\gamma} \leq \beta} C(\alpha, \gamma) C(\beta, \widetilde{\gamma})\right]$, where $\Lambda$ is as in (2.8). By this estimate, $\phi \in \mathcal{A}$, (2.8) and Hölder's inequality, we further have

$$
\begin{aligned}
&\left|\mathfrak{a}_{\rho, \phi}(f, f)-\mathfrak{a}(f, f)\right| \\
& \leq \Lambda \sum_{|\alpha|=|\beta|=k} \int_{\mathbb{R}^{n}}\left\{\sum_{0 \leq \gamma \leq \alpha, 0 \leq \tilde{\gamma} \leq \beta} C(\alpha, \gamma) C(\beta, \widetilde{\gamma})\left|\rho^{|\gamma|} \partial^{\alpha-\gamma} f(x) \rho^{|\tilde{\gamma}|} \overline{\partial^{\beta-\tilde{\gamma}} f(x)}\right|\right\} d x \\
& \leq \Lambda \sum_{|\alpha|=|\beta|=k} \sum_{0 \leq \gamma \leq \alpha, 0 \leq \tilde{\gamma} \leq \beta} C(\alpha, \gamma) C(\beta, \widetilde{\gamma})\left\{\int_{\mathbb{R}^{n}}\left|\rho^{|\gamma|} \partial^{\alpha-\gamma} f(x)\right|^{2} d x\right\}^{1 / 2} \\
& \times\left\{\int_{\mathbb{R}^{n}}\left|\rho^{\tilde{\gamma} \mid} \partial^{\beta-\widetilde{\gamma}} f(x)\right|^{2} d x\right\}^{1 / 2} \\
& \equiv \Lambda \sum_{|\alpha|=|\beta|=k} \sum_{0 \leq \gamma \leq \alpha, 0 \leq \tilde{\gamma} \leq \beta} C(\alpha, \gamma) C(\beta, \widetilde{\gamma}) \mathrm{I}_{1} \times \mathrm{I}_{2} .
\end{aligned}
$$

Applying Plancherel's theorem, (2.9) and Young's inequality with $\epsilon \in\left(0, \frac{\lambda}{4 \tilde{C}(k)}\right)$, we obtain that there exists a positive constant $C(\epsilon)$ such that for all $\widetilde{\lambda} \in(C(\epsilon) \widetilde{C}(k), \infty)$,

$$
\begin{aligned}
\left(\mathrm{I}_{1}\right)^{2} & \leq \int_{\mathbb{R}^{n}}\left[\rho^{|\gamma|}|\xi|^{k-|\gamma|}|\hat{f}(\xi)|\right]^{2} d \xi \leq \int_{\mathbb{R}^{n}}\left[C(\epsilon) \rho^{2 k}+\epsilon|\xi|^{2 k}\right]|\hat{f}(\xi)|^{2} d \xi \\
& \leq C(\epsilon) \rho^{2 k}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2}+\epsilon\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \leq C(\epsilon) \rho^{2 k}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2}+\frac{\epsilon}{\lambda} \Re \mathfrak{a}(f, f),
\end{aligned}
$$

which, together with a similar estimate for $\mathrm{I}_{2}$, shows that

$$
\begin{equation*}
\left|\mathfrak{a}_{\rho \phi}(f, f)-\mathfrak{a}(f, f)\right| \leq \frac{1}{4} \Re \mathfrak{a}(f, f)+\widetilde{\lambda} \rho^{2 k}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \tag{3.13}
\end{equation*}
$$

Denote by $L_{\rho \phi}\left(=e^{-\rho \phi} L_{1} e^{\rho \phi}\right)$ the operator associated with $\mathfrak{a}_{\rho \phi}$. Let $f_{t} \equiv e^{-t L_{\rho \phi}} f$. By (3.13), we have

$$
\begin{aligned}
\frac{d}{d t}\left\|f_{t}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} & =-\left(L_{\rho \phi} f_{t}, f_{t}\right)-\left(f_{t}, L_{\rho \phi} f_{t}\right)=-2 \Re \mathfrak{a}_{\rho \phi}\left(f_{t}, f_{t}\right) \\
& =2\left[\Re\left(\mathfrak{a}\left(f_{t}, f_{t}\right)-\mathfrak{a}_{\rho \phi}\left(f_{t}, f_{t}\right)\right)-\Re \mathfrak{a}\left(f_{t}, f_{t}\right)\right] \\
& \leq 2 \mid \mathfrak{a}_{\rho \phi}\left(f_{t}, f_{t}\right)-\mathfrak{a}\left(f_{t}, f_{t}\right)-2 \Re \mathfrak{a}\left(f_{t}, f_{t}\right) \\
& \leq \frac{1}{2} \Re \mathfrak{a}\left(f_{t}, f_{t}\right)+2 \widetilde{\lambda} \rho^{2 k}\left\|f_{t}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2}-2 \Re \mathfrak{a}\left(f_{t}, f_{t}\right) \leq 2 \widetilde{\lambda} \rho^{2 k}\left\|f_{t}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} .
\end{aligned}
$$

Thus,

$$
\begin{aligned}
\left\|f_{t}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} & =\left\|e^{-t L_{\rho \phi}} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \leq \exp \left\{2 \tilde{\lambda} \rho^{2 k} t\right\}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \\
& \leq \exp \left\{2 C(k) h^{\sharp}\left(\rho t^{1 /(2 k)}\right)\right\}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} .
\end{aligned}
$$

That is, (3.11) holds. Therefore, $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $k$-Davies-Gaffney estimate, which completes the proof of Proposition 3.1.

Remark 3.1. In the proof of Proposition 3.1, we obtain the estimate (3.11) by following the proof of [16, Lemma 2]. The same method should also work for the proof of [10, Proposition 3.1]. Notice that the scaling method mentioned in the last two lines of [10, p. 143] may not be valid to be used to remove the factor $e^{(\alpha w+\epsilon) t}$ appearing in the proof of [10, Proposition 3.1], as the authors claimed therein.

We also have the following gradient estimate for $L_{1}$ and $L_{2}$.
Proposition 3.2. Let $k \in \mathbb{N}$, $L_{1}$ be the $2 k$-order divergence form elliptic operator and $L_{2}$ the $2 k$-order Schrödinger type operator defined as in Section 2. Then, $\left\{\sqrt{t} \nabla^{k} e^{-t L_{i}}\right\}_{t>0}$ for $i \in\{1,2\}$ satisfy the $k$-Davies-Gaffney estimate.
Proof. For any Hilbert space $\mathcal{H}$, let $(\cdot, \cdot)_{\mathcal{H}}$ be the inner product of $\mathcal{H}$. By Hölder's inequality and the fact that $\left\{t L_{i} e^{-t L_{i}}\right\}_{t>0}$ and $\left\{e^{-t L_{i}}\right\}_{t>0}$ satisfy the $k$-Davies-Gaffney estimate which are deduced respectively from Proposition 3.1 and Lemma 3.1, we have that for all closed sets $E, F \subset \mathbb{R}^{n}, f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$, and $t \in(0, \infty)$,

$$
\begin{aligned}
\left\|\sqrt{t} \nabla^{k} e^{-t L_{i}} f\right\|_{L^{2}(F)}^{2} & \lesssim\left|\left(t L_{i} e^{-t L_{i}} f, e^{-t L_{i}} f\right)_{L^{2}(F)}\right| \\
& \lesssim\left\|t L_{i} e^{-t L_{i}} f\right\|_{L^{2}(F)}\left\|e^{-t L_{i}} f\right\|_{L^{2}(F)} \\
& \lesssim\left(\exp \left\{-\frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{C_{1} t^{1 /(2 k-1)}}\right\}\right)^{2}\|f\|_{L^{2}(E)}^{2}
\end{aligned}
$$

which implies that $\left\{\sqrt{t} \nabla^{k} e^{-t L_{i}}\right\}_{t>0}$ also satisfies the $k$-Davies-Gaffney estimate. This finishes the proof of Proposition 3.2.

## 4 Molecular characterizations of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$

Assume that the operator $L$ satisfies the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. In this section, we introduce the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ in means of the $L$-adapted square function and characterize these Hardy spaces by the molecular decomposition. First, we recall some notions.

Let $\Gamma(x) \equiv\left\{(y, t) \in \mathbb{R}^{n} \times(0, \infty):|x-y|<t\right\}$ be the cone with vertex $x \in \mathbb{R}^{n}$. For all $f \in L^{2}\left(\mathbb{R}^{n}\right)$ and $x \in \mathbb{R}^{n}$, the $L$-adapted square function $S_{L} f$ is defined by

$$
\begin{equation*}
S_{L} f(x) \equiv\left\{\iint_{\Gamma(x)}\left|t^{2 k} L e^{-t^{2 k} L} f(y)\right|^{2} \frac{d y d t}{t^{n+1}}\right\}^{1 / 2} \tag{4.1}
\end{equation*}
$$

Definition 4.1. Let $p \in(0,1]$ and $L$ satisfy the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. A function $f \in L^{2}\left(\mathbb{R}^{n}\right)$ is said to be in $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$ if $S_{L} f \in L^{p}\left(\mathbb{R}^{n}\right)$; moreover, define $\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)} \equiv\left\|S_{L} f\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}$. The Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ is then defined to be the completion of $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$ with respect to the quasi-norm $\|\cdot\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)}$.

Remark 4.1. Since both the $2 k$-order divergence form homogenous elliptic operator $L_{1}$ with complex bounded measurable coefficients and the $2 k$-order Schrödinger type operator $L_{2}$ satisfy the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$, we then define the Hardy spaces $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ and $H_{L_{2}}^{p}\left(\mathbb{R}^{n}\right)$, respectively, associated to $L_{1}$ and $L_{2}$ as in Definition 4.1. In particular, when $k=1, H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$ is just the Hardy space $H_{-\operatorname{div}(\mathrm{AD})}^{p}\left(\mathbb{R}^{n}\right)$ associated to the second order divergence form elliptic operator $-\operatorname{div}(\mathrm{A} \nabla)$ with complex bounded measurable coefficients in $[30,31,32,36]$ and $H_{L_{2}}^{1}\left(\mathbb{R}^{n}\right)$ appears in $[21,22,28,48]$; when $k=2, H_{L_{2}}^{1}\left(\mathbb{R}^{n}\right)$ was also studied in [12].

In what follows, a cube always means a closed cube whose sides are parallel to the coordinate axes. Let $Q \subset \mathbb{R}^{n}$ be a cube with the side length $l(Q)$. For $i \in \mathbb{Z}_{+}$, denote by $S_{i}(Q)$ the dyadic annuli based on $Q$, namely,

$$
\begin{equation*}
S_{0}(Q) \equiv Q \text { and } S_{i}(Q) \equiv 2^{i} Q \backslash\left(2^{i-1} Q\right) \text { for } i \in \mathbb{N} \tag{4.2}
\end{equation*}
$$

where $2^{i} Q$ is the cube with the same center as $Q$ and the side length $2^{i} l(Q)$.
Definition 4.2. Let $p \in(0,1], \epsilon \in(0, \infty), M \in \mathbb{N}$ and $L$ satisfy the assumptions ( $\mathrm{A}_{1}$ ), $\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. A function $m \in L^{2}\left(\mathbb{R}^{n}\right)$ is called an $\left(H_{L}^{p}, \epsilon, M\right)$-molecule if there exists a cube $Q \subset \mathbb{R}^{n}$ such that
(i) for each $\ell \in\{1, \cdots, M\}$, $m$ belongs to the range of $L^{\ell}$ in $L^{2}\left(\mathbb{R}^{n}\right)$;
(ii) for all $i \in \mathbb{Z}_{+}$and $\ell \in\{0,1, \cdots, M\}$,

$$
\begin{equation*}
\left\|\left([l(Q)]^{-2 k} L^{-1}\right)^{\ell} m\right\|_{L^{2}\left(S_{i}(Q)\right)} \leq\left[2^{i} l(Q)\right]^{n\left(\frac{1}{2}-\frac{1}{p}\right)} 2^{-i \epsilon} \tag{4.3}
\end{equation*}
$$

Assume that $\left\{m_{j}\right\}_{j=0}^{\infty}$ is a sequence of $\left(H_{L}^{p}, \epsilon, M\right)$-molecules and $\left\{\lambda_{j}\right\}_{j=0}^{\infty} \in l^{p}$. For any $f \in L^{2}\left(\mathbb{R}^{n}\right)$, if $f=\sum_{j=0}^{\infty} \lambda_{j} m_{j}$ in $L^{2}\left(\mathbb{R}^{n}\right)$, then $\sum_{j=0}^{\infty} \lambda_{j} m_{j}$ is called a molecular ( $\left.H_{L}^{p}, 2, \epsilon, M\right)$-representation of $f$.

We now introduce the notion of a molecular Hardy space $H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ generated by $\left(H_{L}^{p}, \epsilon, M\right)$-molecules.

Definition 4.3. Let $p \in(0,1], \epsilon \in(0, \infty), M \in \mathbb{N}$ and $L$ satisfy the assumptions $\left(\mathrm{A}_{1}\right)$, $\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. The molecular Hardy space $H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ is defined to be the completion of the space

$$
\mathbb{H}_{L, \text { mol }, M}^{p}\left(\mathbb{R}^{n}\right) \equiv\left\{f: f \text { has a molecular }\left(H_{L}^{p}, 2, \epsilon, M\right) \text { - representation }\right\}
$$

with respect to the quasi-norm

$$
\|f\|_{H_{L, \mathrm{~mol}, M}^{p}}\left(\mathbb{R}^{n}\right) \equiv \inf \left\{\left(\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right)^{1 / p}: f=\sum_{j=0}^{\infty} \lambda_{j} m_{j}\right. \text { is a molecular }
$$

$$
\left.\left(H_{L}^{p}, 2, \epsilon, M\right)-\text { representation }\right\},
$$

where the infimum is taken over all the molecular $\left(H_{L}^{p}, 2, \epsilon, M\right)$-representations of $f$ as above.

Now, we establish the molecular characterization of the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)$.
Theorem 4.1. Let $p \in(0,1], \epsilon \in(0, \infty)$, L satisfy the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2 and $M \in \mathbb{N}$ such that $M>\frac{n}{2 k}\left(\frac{1}{p}-\frac{1}{2}\right)$. Then, $H_{L}^{p}\left(\mathbb{R}^{n}\right)=H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms.

To prove Theorem 4.1, by Definitions 4.1 and 4.3 , it suffices to prove that

$$
\begin{equation*}
\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)=\mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right), \quad M>\frac{n}{2 k}\left(\frac{1}{p}-\frac{1}{2}\right), \tag{4.4}
\end{equation*}
$$

with equivalent norms. We divide this proof into two parts: (i) $\mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$; (ii) $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$.

To prove the inclusion $\mathbb{H}_{L, \text { mol, } M}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$, we need the following key lemma which is just [32, Lemma 3.8]. Recall that a nonnegative sublinear operator $T$ means that $T$ is sublinear and $T f \geq 0$ for all $f$ in the domain of $T$.

Lemma 4.1. Let $p \in(0,1], M \in \mathbb{N}$ and $T$ be a linear operator, or a nonnegative sublinear operator, which is of weak type (2,2), that is, there exists a positive constant $C$ such that for all $\eta \in(0, \infty)$ and $f \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\left|\left\{x \in \mathbb{R}^{n}:|T f(x)|>\eta\right\}\right| \leq C \eta^{-2}\|f\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2}
$$

Assume further that there exists a positive constant $C$ such that for all ( $\left.H_{L}^{p}, \epsilon, M\right)$ molecules $m,\|T m\|_{L^{p}\left(\mathbb{R}^{n}\right)} \leq C$. Then the operator $T$ is bounded from $H_{L, \mathrm{~mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ to $L^{p}\left(\mathbb{R}^{n}\right)$.

Proof of Theorem 4.1: the inclusion $\mathbb{H}_{L, \text { mol, } M}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$. Recall that $L$ is a one to one operator of type $\omega$ having a bounded $H_{\infty}$ functional calculus. For all $x \in \mathbb{R}^{n}, \psi \in$ $\Psi\left(S_{\mu}^{0}\right)$ defined as in Section 2, set $\psi_{t}(x) \equiv \psi(t x)$ for all $t \in(0, \infty)$. The quadratic norm $\|g\|_{T, \psi}$, associated with the operator $L$ in $L^{2}\left(\mathbb{R}^{n}\right)$ and $\psi$, is defined by

$$
\|g\|_{T, \psi} \equiv\left\{\int_{0}^{\infty}\left\|\psi_{t}(T) g\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d t}{t}\right\}^{1 / 2}
$$

for all $g \in\left[L^{2}\left(\mathbb{R}^{n}\right)\right]_{T, \psi}$ which is a subspace of $L^{2}\left(\mathbb{R}^{n}\right)$ such that the above integral is finite. Since $L$ has a bounded $H_{\infty}$ functional calculus on $L^{2}\left(\mathbb{R}^{n}\right)$, it follows from [2] that $\left[L^{2}\left(\mathbb{R}^{n}\right)\right]_{T, \psi}=L^{2}\left(\mathbb{R}^{n}\right)$ and for all $g \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\begin{equation*}
\|g\|_{T, \psi} \lesssim\|g\|_{L^{2}\left(\mathbb{R}^{n}\right)} \tag{4.5}
\end{equation*}
$$

By Fubini's theorem, we have that $\left\|S_{L} g\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \sim\|g\|_{T, \psi_{0}}$, where $\psi_{0}(z) \equiv z e^{-z} \in \Psi\left(S_{\mu}^{0}\right)$ for all $\mu \in(0, \pi / 2)$. Thus, $S_{L}$ is bounded on $L^{2}\left(\mathbb{R}^{n}\right)$. By Lemma 4.1, to prove the inclusion $\mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$, it suffices to prove that for all $\left(H_{L}^{p}, \epsilon, M\right)$-molecules $m$ with $M>\frac{n}{2 k}\left(\frac{1}{p}-\frac{1}{2}\right)$,

$$
\begin{equation*}
\left\|S_{L} m\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} \lesssim 1 \tag{4.6}
\end{equation*}
$$

Let $Q$ be the cube associated with $m$ as in Definition 4.2. Let $j_{0} \in \mathbb{N}$ be such that $2^{j_{0}-1}<$ $\sqrt{n} \leq 2^{j_{0}}$. By Minkowski's inequality, Hölder's inequality and the $L^{2}\left(\mathbb{R}^{n}\right)$-boundedness of $S_{L}$, we have

$$
\begin{aligned}
\left\|S_{L} m\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} & \leq\left\|S_{L} m\right\|_{L^{p}\left(2^{j_{0}+4} Q\right)}+\sum_{j=j_{0}+5}^{\infty}\left\|S_{L} m\right\|_{L^{p}\left(S_{j}(Q)\right)} \\
& \lesssim\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)}\left|2^{j_{0}+4} Q\right|^{\frac{1}{p}-\frac{1}{2}}+\sum_{j=j_{0}+5}^{\infty}\left\|S_{L} m\right\|_{L^{2}\left(S_{j}(Q)\right)}\left|S_{j}(Q)\right|^{\frac{1}{p}-\frac{1}{2}} .
\end{aligned}
$$

For $\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)}$, by Minkowski's inequality and the size condition (4.3) of $m$, we have

$$
\begin{equation*}
\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)} \leq \sum_{j=0}^{\infty}\|m\|_{L^{2}\left(S_{j}(Q)\right)} \leq \sum_{j=0}^{\infty}\left[2^{j} l(Q)\right]^{n\left(\frac{1}{2}-\frac{1}{p}\right)} 2^{-j \epsilon} \lesssim[l(Q)]^{n\left(\frac{1}{2}-\frac{1}{p}\right)} . \tag{4.7}
\end{equation*}
$$

For $j \in\left\{j_{0}+5, \cdots\right\}$, let $\mathrm{I}_{j} \equiv\left\|S_{L} m\right\|_{L^{2}\left(S_{j}(Q)\right)}$. Then,

$$
\begin{aligned}
\left(\mathrm{I}_{j}\right)^{2}= & \int_{S_{j}(Q)}\left|S_{L} m\right|^{2} d x=\int_{S_{j}(Q)} \int_{0}^{\infty} \int_{|y-x|<t}\left|t^{2 k} L e^{-t^{2 k} L} m(y)\right|^{2} \frac{d y d t}{t^{n+1}} d x \\
= & \int_{S_{j}(Q)} \int_{0}^{2^{\theta(j-5)} l(Q)} \int_{|y-x|<t}\left|t^{2 k} L e^{-t^{2 k} L} m(y)\right|^{2} \frac{d y d t}{t^{n+1}} d x \\
& +\int_{S_{j}(Q)} \int_{2^{\theta(j-5) l(Q)}}^{\infty} \int_{|y-x|<t} \cdots \equiv \mathrm{~B}_{j}+\mathrm{D}_{j},
\end{aligned}
$$

where $\theta \in(0,1)$ is determined later.
For $\mathrm{D}_{j}$, let $b \equiv L^{-M} m$. By Fubini's theorem, Lemma 3.1 and the size condition (4.3) of $m$, we have

$$
\begin{aligned}
\mathrm{D}_{j} & =\int_{S_{j}(Q)} \int_{2^{\theta(j-5)} l(Q)}^{\infty} \int_{|y-x|<t}\left|t^{2 k} L e^{-t^{2 k} L} L^{M} b(y)\right|^{2} \frac{d y d t}{t^{n+1}} d x \\
& =\int_{S_{j}(Q)} \int_{2^{\theta(j-5)} l(Q)}^{\infty} \int_{|y-x|<t}\left|t^{2 k(M+1)} L^{M+1} e^{-t^{2 k} L} b(y)\right|^{2} \frac{d y d t}{t^{n+1+4 k M}} d x \\
& \lesssim \int_{2^{\theta(j-5) l(Q)}}^{\infty}\left\|t^{2 k(M+1)} L^{M+1} e^{-t^{2 k} L} b\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d t}{t^{4 k M+1}} \lesssim \int_{2^{\theta(j-5)} l(Q)}^{\infty}\|b\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d t}{t^{4 k M+1}} \\
& \sim\|b\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2}\left[2^{\theta(j-5)} l(Q)\right]^{-4 k M} \sim\left[\sum_{i=0}^{\infty}\|b\|_{L^{2}\left(S_{i}(Q)\right)}^{2}\right]\left[2^{\theta(j-5) l(Q)}\right]^{-4 k M}
\end{aligned}
$$

$$
\lesssim[l(Q)]^{4 k M+2 n\left(\frac{1}{2}-\frac{1}{p}\right)}\left[2^{\theta(j-5)} l(Q)\right]^{-4 k M} \sim 2^{-j\left[4 k M \theta+n\left(1-\frac{2}{p}\right)\right]}\left[2^{j} l(Q)\right]^{n\left(1-\frac{2}{p}\right)}
$$

Recall that $M>\frac{n}{2 k}(1 / p-1 / 2)$. Letting $\theta$ be sufficiently close to 1 such that $\alpha_{0} \equiv$ $2 k M \theta-n(1 / p-1 / 2)>0$, we then obtain

$$
\begin{equation*}
\mathrm{D}_{j} \lesssim 2^{-2 j \alpha_{0}}\left|S_{j}(Q)\right|^{1-\frac{2}{p}} \tag{4.8}
\end{equation*}
$$

To estimate $\mathrm{B}_{j}$, let $\widetilde{S}_{j}(Q) \equiv 2^{j+j_{0}+1} Q \backslash\left(2^{j-j_{0}-2} Q\right)$ and $\widehat{S}_{j}(Q) \equiv 2^{j+j_{0}+2} Q \backslash\left(2^{j-j_{0}-3} Q\right)$. By Fubini's theorem, we have

$$
\begin{aligned}
\mathrm{B}_{j} \lesssim & \int_{0}^{2^{\theta(j-5)} l(Q)} \int_{\widetilde{S}_{j}(Q)}\left|t^{2 k} L e^{-t^{2 k} L} m(y)\right|^{2} \frac{d y d t}{t} \\
\lesssim & \int_{0}^{2^{\theta(j-5)} l(Q)} \int_{\widetilde{S}_{j}(Q)}\left|t^{2 k} L e^{-t^{2 k} L}\left(\chi_{2^{j-j_{0}-3} Q} m\right)(y)\right|^{2} \frac{d y d t}{t} \\
& +\int_{0}^{2^{\theta(j-5)} l(Q)} \int_{\widetilde{S}_{j}(Q)}\left|t^{2 k} L e^{-t^{2 k} L}\left(\chi_{\widehat{S}_{j}(Q)} m\right)(y)\right|^{2} \frac{d y d t}{t} \\
& +\int_{0}^{2^{\theta(j-5)} l(Q)} \int_{\widetilde{S}_{j}(Q)}\left|t^{2 k} L e^{-t^{2 k} L}\left(\chi_{\mathbb{R}^{n} \backslash 2^{j+j_{0}+2} Q} m\right)(y)\right|^{2} \frac{d y d t}{t} \equiv \mathrm{~B}_{j, 1}+\mathrm{B}_{j, 2}+\mathrm{B}_{j, 3} .
\end{aligned}
$$

By the $k$-Davies-Gaffney estimate, (4.7) and choosing $\alpha \in(2 n(1 / p-1 / 2) /(1-\theta), \infty)$, we obtain

$$
\begin{aligned}
\mathrm{B}_{j, 1}+\mathrm{B}_{j, 3} & \lesssim \int_{0}^{2^{\theta(j-5)} l(Q)} \exp \left\{-\widetilde{C}\left[\frac{2^{j} l(Q)}{t}\right]^{2 k /(2 k-1)}\right\}\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d t}{t} \\
& \lesssim\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \int_{0}^{2^{\theta(j-5)} l(Q)}\left[\frac{t}{2^{j} l(Q)}\right]^{\alpha} \frac{d t}{t} \sim[l(Q)]^{2 n\left(\frac{1}{2}-\frac{1}{p}\right)}\left[2^{\theta(j-5)-j}\right]^{\alpha}
\end{aligned}
$$

where $\widetilde{C}$ denotes a positive constant. Let $\alpha_{1} \equiv(1-\theta) \alpha / 2-n(1 / p-1 / 2)$. Then $\alpha_{1}>0$ and we have

$$
\begin{equation*}
\mathrm{B}_{j, 1}+\mathrm{B}_{j, 3} \lesssim\left[2^{j} l(Q)\right]^{2 n(1 / 2-1 / p)} 2^{-2 j \alpha_{1}} \tag{4.9}
\end{equation*}
$$

Finally, by (4.5) and the size condition (4.3) of $m$, we obtain

$$
\begin{equation*}
\mathrm{B}_{j, 2} \lesssim\|m\|_{L^{2}\left(\widehat{S}_{j}(Q)\right)}^{2} \lesssim \sum_{\ell=j-j_{0}-2}^{j+j_{0}+2}\|m\|_{L^{2}\left(S_{\ell}(Q)\right)}^{2} \lesssim 2^{-2 j \epsilon}\left[2^{j} l(Q)\right]^{2 n(1 / 2-1 / p)} \tag{4.10}
\end{equation*}
$$

which, together with (4.8) and (4.9), shows that there exists a positive constant $\alpha_{2} \equiv$ $\min \left\{\alpha_{0}, \alpha_{1}, \epsilon\right\}$ such that for all $j \in\left\{j_{0}+5, \cdots\right\}$,

$$
\begin{equation*}
\mathrm{I}_{j} \lesssim\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \alpha_{2}} \sim\left|S_{j}(Q)\right|^{1 / 2-1 / p} 2^{-j \alpha_{2}} \tag{4.11}
\end{equation*}
$$

Combining (4.7) and (4.11), we have

$$
\begin{equation*}
\left\|S_{L} m\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} \lesssim[l(Q)]^{n\left(\frac{1}{2}-\frac{1}{p}\right)}\left|2^{j_{0}+4} Q\right|^{\frac{1}{p}-\frac{1}{2}}+\sum_{j=j_{0}+5}^{\infty} 2^{-j \alpha_{2}} \lesssim 1, \tag{4.12}
\end{equation*}
$$

from which we deduce (4.6). Thus, the inclusion $\mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$ holds, which completes the proof of part one of Theorem 4.1.

Now, we prove the inclusion $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$. To this end, we need use some results concerning the tent space from [14]. Let $F$ be a function on $\mathbb{R}_{+}^{n+1} \equiv \mathbb{R}^{n} \times(0, \infty)$. The $\mathcal{A}$-functional of $F$ is defined by setting, for all $x \in \mathbb{R}^{n}$,

$$
\mathcal{A}(F)(x) \equiv\left\{\iint_{\Gamma(x)}|F(y, t)|^{2} \frac{d y d t}{t^{n+1}}\right\}^{\frac{1}{2}}
$$

For $p \in(0, \infty)$, the tent space $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ is defined by

$$
T^{p}\left(\mathbb{R}_{+}^{n+1}\right) \equiv\left\{F: \mathbb{R}_{+}^{n+1} \rightarrow \mathbb{C}:\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \equiv\|\mathcal{A}(F)\|_{L^{p}\left(\mathbb{R}^{n}\right)}<\infty\right\}
$$

For any cube $Q$, denote by $R_{Q} \equiv Q \times(0, l(Q))$ the Carleson box of $Q$. A measurable function $A$ on $\mathbb{R}_{+}^{n+1}$ is called a $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atom associated with $Q$ with $p \in(0,1]$, if $A$ satisfies the following properties:

$$
\begin{equation*}
\operatorname{supp} A \subset R_{Q} \tag{4.13}
\end{equation*}
$$

and

$$
\begin{equation*}
\left\{\iint_{R_{Q}}|A(x, t)|^{2} \frac{d x d t}{t}\right\}^{1 / 2} \leq|Q|^{\frac{1}{2}-\frac{1}{p}} \tag{4.14}
\end{equation*}
$$

For the tent space $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ with $p \in(0,1]$, we have the following atomic decomposition from [14] (see also [32, Proposition 3.25]).

Theorem $4.2([14])$. Let $p \in(0,1]$. For all $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$, there exist a numerical sequence $\left\{\lambda_{j}\right\}_{j=0}^{\infty}$ and a sequence $\left\{A_{j}\right\}_{j=0}^{\infty}$ of $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atoms such that for almost every $(x, t) \in \mathbb{R}_{+}^{n+1}$,

$$
F(x, t)=\sum_{j=0}^{\infty} \lambda_{j} A_{j}(x, t)
$$

Moreover,

$$
\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p} \sim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}^{p}
$$

where the implicit equivalent positive constants depend only on the dimension n. Finally, if $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$, then the decomposition also converges in $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$.

Let $M \in \mathbb{N}$. For all $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$, define the operator $\pi_{M, L}$ by setting, for all $x \in \mathbb{R}^{n}$,

$$
\begin{equation*}
\pi_{M, L} F(x) \equiv \int_{0}^{\infty}\left(t^{2 k} L\right)^{M+1} e^{-t^{2 k} L} F(x, t) \frac{d t}{t} \tag{4.15}
\end{equation*}
$$

For this operator, we have the following useful lemma on its properties.
Lemma 4.2. Let $M \in \mathbb{N}, p \in(0,1], \epsilon \in(0, \infty)$ and the operator $L$ satisfy the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. Let $\pi_{M, L}$ be as in (4.15). Then
(i) The operator $\pi_{M, L}$ is bounded from $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ to $L^{2}\left(\mathbb{R}^{n}\right)$;
(ii) For any $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atom $A, \pi_{M, L} A$ is an $\left(H_{L}^{p}, \epsilon, M\right)$-molecule up to a harmless positive constant multiple.
(iii) If $M \in(n(1 / p-1 / 2) /(2 k), \infty)$, then the operator $\pi_{M, L}$ is bounded from the tent space $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ to the molecular Hardy space $H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$.

Proof. We first show (i). Let $L^{*}$ be the adjoint operator of $L$ in $L^{2}\left(\mathbb{R}^{n}\right)$. Observe that $L^{*}$ also satisfies the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2. By Fubini's theorem, Hölder's inequality and the quadratic estimate (4.5) with $L$ replaced by $L^{*}$, we have that for all $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ and $g \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\begin{aligned}
\left|\left(\pi_{M, L} F, g\right)\right|= & \left|\int_{\mathbb{R}^{n}} \int_{0}^{\infty}\left(t^{2 k} L\right)^{M+1} e^{-t^{2 k} L} F(x, t) \overline{g(x)} \frac{d t}{t} d x\right| \\
= & \left|\int_{0}^{\infty} \int_{\mathbb{R}^{n}} F(x, t) \overline{\left(t^{2 k} L^{*}\right)^{M+1} e^{-t^{2 k} L^{*}} g(x)} d x \frac{d t}{t}\right| \\
& \lesssim\left\{\int_{0}^{\infty} \int_{\mathbb{R}^{n}}|F(x, t)|^{2} d x \frac{d t}{t}\right\}^{1 / 2} \\
& \times\left\{\int_{0}^{\infty} \int_{\mathbb{R}^{n}}\left|\left(t^{2 k} L^{*}\right)^{M+1} e^{-t^{2 k} L^{*}} g(x)\right|^{2} d x \frac{d t}{t}\right\}^{1 / 2} \\
& \lesssim\|F\|_{T^{2}\left(\mathbb{R}_{+}^{n+1}\right)}\|g\|_{L^{2}\left(\mathbb{R}^{n}\right)},
\end{aligned}
$$

which further implies that the operator $\pi_{M, L}$ is bounded from $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ to $L^{2}\left(\mathbb{R}^{n}\right)$. Thus, (i) holds.

To prove (ii), let $A$ be a $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atom $A$ associated with the cube $Q$. From (4.15), it follows that for all $\ell \in\{0, \cdots, M\}$ and $x \in \mathbb{R}^{n}$,

$$
\begin{align*}
\pi_{M, L} A(x) & =\int_{0}^{\infty}\left(t^{2 k} L\right)^{M+1} e^{-t^{2 k} L} A(x, t) \frac{d t}{t}  \tag{4.16}\\
& =L^{\ell} \int_{0}^{\infty} t^{2 k(M+1)} L^{M+1-\ell} e^{-t^{2 k} L} A(x, t) \frac{d t}{t}
\end{align*}
$$

Observe that

$$
\int_{0}^{\infty} t^{2 k(M+1)} L^{M+1-\ell} e^{-t^{2 k} L} A(x, t) \frac{d t}{t}=\int_{0}^{\infty} t^{2 k(M+1)}\left(L^{1-\frac{\ell}{M+1}}\right)^{M+1} e^{-t^{2 k} L} A(x, t) \frac{d t}{t},
$$

which belongs to $L^{2}\left(\mathbb{R}^{n}\right)$ via a dual argument similar to that used in the proof of (i). This, combined with (4.16), implies that $\pi_{M, L}(A)$ satisfies Definition 4.2(i).

For all $x \in \mathbb{R}^{n}$, letting

$$
b(x) \equiv \int_{0}^{\infty} t^{2 k(M+1)} L e^{-t^{2 k} L} A(x, t) \frac{d t}{t},
$$

we then have $\pi_{M, L} A(x)=L^{M} b(x)$. For all $g \in L^{2}\left(\mathbb{R}^{n}\right)$, by Hölder's inequality, (4.13), (4.14) and the quadratic estimate (4.5) with $L$ replaced by $L^{*}$, we obtain

$$
\begin{align*}
& \left|\int_{\mathbb{R}^{n}}\left([l(Q)]^{2 k} L\right)^{\ell} b(x) \overline{g(x)} d x\right|  \tag{4.17}\\
& \quad=\left|\int_{\mathbb{R}^{n}}\left\{\int_{0}^{\infty}[l(Q)]^{2 k \ell} L^{\ell+1} t^{2 k(M+1)} e^{-t^{2 k} L} A(x, t) \frac{d t}{t}\right\} \overline{g(x)} d x\right| \\
& \quad=[l(Q)]^{2 k \ell}\left|\iint_{R_{Q}} A(x, t) \overline{\left(L^{*}\right)^{\ell+1} t^{2 k(M+1)} e^{-t^{2 k} L^{*}} g(x)} \frac{d x d t}{t}\right| \\
& \left.\left.\quad \lesssim[l(Q)]^{2 k l}\left|\iint_{R_{Q}}\right| A(x, t)\right|^{2} \frac{d x d t}{t}\right]^{1 / 2} \\
& \quad \times\left[\iint_{R_{Q}}\left|\overline{\left(L^{*}\right)^{\ell+1} t^{2 k(M+1)} e^{-t^{2 k} L^{*}} g(x)}\right|^{2} \frac{d x d t}{t}\right]^{1 / 2} \\
& \quad \lesssim[l(Q)]^{n(1 / 2-1 / p)+2 k M}\left\{\iint_{R_{Q}}\left|\left(t^{2 k} L^{*}\right)^{\ell+1} e^{-t^{2 k} L^{*}} g(x)\right|^{2} \frac{d x d t}{t}\right\}^{1 / 2} \\
& \quad \lesssim[l(Q)]^{n(1 / 2-1 / p)+2 k M}\|g\|_{L^{2}\left(\mathbb{R}^{n}\right)},
\end{align*}
$$

which further implies that for all $\ell \in\{0, \cdots, M\}$,

$$
\left\|\left([l(Q)]^{2 k} L\right)^{\ell} b\right\|_{L^{2}(2 Q)} \lesssim[l(Q)]^{2 k M}|Q|^{\frac{1}{2}-\frac{1}{p}} .
$$

Thus, by this, we obtain that for all $\tilde{\ell} \in\{0, \cdots, M\}$ and $j \in\{0,1\}$,

$$
\begin{align*}
& \left\|\left([l(Q)]^{-2 k} L^{-1}\right)^{\tilde{\ell}}\left(\pi_{M, L} A\right)\right\|_{L^{2}\left(S_{j}(Q)\right)}  \tag{4.18}\\
& \quad=\left\|\left([l(Q)]^{-2 k} L^{-1}\right)^{\tilde{\ell}} L^{M} b\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \quad \lesssim\left\|\left([l(Q)]^{2 k(M-\tilde{\ell})} L^{M-\tilde{\ell}}\right) b\right\|_{L^{2}(2(Q))}|l(Q)|^{-2 k M} \lesssim[l(Q)]^{n(1 / 2-1 / p)},
\end{align*}
$$

which is desired.
Moreover, for all $\tilde{\ell} \in\{0, \cdots, M\}$ and $j \in\{2,3, \cdots\}$, letting $g \in L^{2}\left(\mathbb{R}^{n}\right)$ with supp $g \subset$ $S_{j}(Q)$, choosing $\alpha \in(n(1 / p-1 / 2)(2-1 / k), \infty)$ and using Lemma 3.1, similarly to the
estimate for (4.17), we have

$$
\begin{aligned}
& \left|\int_{\mathbb{R}^{n}}\left([l(Q)]^{2 k} L\right)^{\ell} b(x) \overline{g(x)} d x\right| \\
& \quad \lesssim[l(Q)]^{n(1 / 2-1 / p)+2 k M}\left\{\int_{0}^{l(Q)} \int_{Q}\left|\left(t^{2 k} L^{*}\right)^{\ell+1} e^{-t^{2 k} L^{*}} g(x)\right|^{2} \frac{d x d t}{t}\right\}^{1 / 2} \\
& \quad \lesssim[l(Q)]^{n(1 / 2-1 / p)+2 k M}\left[\int_{0}^{l(Q)} \exp \left\{-C\left[\frac{\operatorname{dist}\left(Q, S_{j}(Q)\right)}{t}\right]^{2 k /(2 k-1)}\right\} \frac{d t}{t}\right]^{1 / 2} \\
& \quad \times\|g\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \quad \lesssim[l(Q)]^{n(1 / 2-1 / p)+2 k M} 2^{-j k \alpha /(2 k-1)}\|g\|_{L^{2}\left(S_{j}(Q)\right)},
\end{aligned}
$$

which further implies that for all $\tilde{\ell} \in\{0, \cdots, M\}$ and $j \in\{2,3, \cdots\}$,

$$
\begin{align*}
& \left\|\left([l(Q)]^{-2 k} L^{-1}\right)^{\tilde{\ell}}\left(\pi_{M, L} A\right)\right\|_{L^{2}\left(S_{j}(Q)\right)}  \tag{4.19}\\
& \quad \leq\left\|\left([l(Q)]^{-2 k} L^{-1}\right)^{\tilde{\ell}} L^{M} b\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \quad \lesssim\left\|\left([l(Q)]^{2 k(M-\widetilde{\ell})} L^{M-\tilde{\ell}}\right) b\right\|_{L^{2}\left(S_{j}(Q)\right)}|l(Q)|^{-2 k M} \\
& \quad \lesssim\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j[k \alpha /(2 k-1)-n(1 / p-1 / 2)]} \sim\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \epsilon},
\end{align*}
$$

where $\epsilon \equiv k \alpha /(2 k-1)-n(1 / p-1 / 2) \in(0, \infty)$.
Combining (4.18) and (4.19), we know that $\pi_{M, L} A$ satisfies Definition 4.2(ii) up to a harmless positive constant multiple. Thus, $\pi_{M, L} A$ is an $\left(H_{L}^{p}, \epsilon, M\right)$-molecule up to a harmless positive constant multiple, which completes the proof of (ii).

To show (iii), by density, we only need show that for all $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$,

$$
\left\|\pi_{M, L} F\right\|_{H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)} \lesssim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}
$$

To this end, by Theorem 4.2, there exist a sequence $\left\{A_{i}\right\}_{i=0}^{\infty}$ of $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atoms and $\left\{\lambda_{i}\right\}_{i=0}^{\infty} \in l^{p}$ such that $F=\sum_{i=0}^{\infty} \lambda_{i} A_{i}$ in both pointwise and $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$, and

$$
\left(\sum_{i=0}^{\infty}\left|\lambda_{i}\right|^{p}\right)^{1 / p} \sim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}
$$

By (i) of this lemma, we know that

$$
\pi_{M, L} F=\sum_{i=0}^{\infty} \lambda_{i} \pi_{M, L} A_{i}
$$

in $L^{2}\left(\mathbb{R}^{n}\right)$, which, combined with (ii) of this lemma, shows that $\sum_{i=0}^{\infty} \lambda_{i} \pi_{M, L} A_{i}$ is a molec$\operatorname{ular}\left(H_{L}^{p}, 2, \epsilon, M\right)$-representation of $\pi_{M, L} F$. Thus, $\pi_{M, L} F \in H_{L, \mathrm{~mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ and

$$
\left\|\pi_{M, L} F\right\|_{H_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)} \lesssim\left\{\sum_{i=0}^{\infty}\left|\lambda_{i}\right|^{p}\right\}^{1 / p} \sim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}
$$

which completes the proof of (iii) and hence Lemma 4.2.
Proof of Theorem 4.1: the inclusion $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L, \text { mol, } M}^{p}\left(\mathbb{R}^{n}\right)$. For all $f \in \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$, $t \in$ $(0, \infty)$ and $x \in \mathbb{R}^{n}$, let

$$
\begin{equation*}
F(x, t) \equiv t^{2 k} L e^{-t^{2 k} L} f(x) . \tag{4.20}
\end{equation*}
$$

By $S_{L} f \in L^{p}\left(\mathbb{R}^{n}\right)$ and $f \in L^{2}\left(\mathbb{R}^{n}\right)$ together with the fact that $S_{L}$ is bounded on $L^{2}\left(\mathbb{R}^{n}\right)$, we know that $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$. Moreover, by the $H_{\infty}$ functional calculus in $L^{2}\left(\mathbb{R}^{n}\right)$, we have the following Calderón reproducing formula that for all $g \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
g=C_{9} \int_{0}^{\infty}\left(t^{2 k} L\right)^{M+2} e^{-2 t^{2 k}} L_{g} \frac{d t}{t}
$$

where $C_{9}$ is a positive constant such that $C_{9} \int_{0}^{\infty} t^{2 k(M+2)} e^{-2 t^{2 k}} \frac{d t}{t}=1$. Thus, for all $f \in \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$, if letting $F$ be as in (4.20), then $f=C_{9} \pi_{M, L} F$ and, by Lemma 4.2 (iii) and its proof, we further know that $f \in \mathbb{H}_{L, \operatorname{mol}, M}^{p}\left(\mathbb{R}^{n}\right)$ and $\|f\|_{H_{L, \text { mol }, M}}^{p}\left(\mathbb{R}^{n}\right) \lesssim\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)}$. Therefore, $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L, \text { mol, } M}^{p}\left(\mathbb{R}^{n}\right)$, which completes the proof Theorem 4.1.

## 5 Generalized square function characterizations of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$

This section is devoted to the generalized square function characterization of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$. We first introduce the notion of the Hardy space $H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ defined via the generalized square function. Let $\omega \in[0, \pi / 2), \alpha \in(0, \infty), \beta \in(n(1 / p-1 / 2) /(2 k), \infty)$ and $\psi \in$ $\Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$ with $\mu \in(\omega, \pi / 2)$. For all $f \in L^{2}\left(\mathbb{R}^{n}\right)$ and $(x, t) \in \mathbb{R}_{+}^{n+1}$, define the operator $Q_{\psi, L} f$ by,

$$
\begin{equation*}
Q_{\psi, L} f(x, t) \equiv \psi\left(t^{2 k} L\right) f(x) . \tag{5.1}
\end{equation*}
$$

Definition 5.1. Let $p \in(0,1], \omega \in[0, \pi / 2), L$ be the operator of type $\omega$ satisfying the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section $2, \alpha \in(0, \infty), \beta \in(n(1 / p-1 / 2) /(2 k), \infty)$, $\mu \in(\omega, \pi / 2)$ and $\psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$. The generalized square function Hardy space $H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ is defined to be the completion of the space

$$
\mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right) \equiv\left\{f \in L^{2}\left(\mathbb{R}^{n}\right): Q_{\psi, L} f \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right)\right\}
$$

with respect to the quasi-norm $\|f\|_{H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)} \equiv\left\|Q_{\psi, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}$.
The following theorem, which establishes the generalized square function characterization of $H_{L}^{p}\left(\mathbb{R}^{n}\right)$, is the main result of this section.

Theorem 5.1. Let $p \in(0,1], \omega \in[0, \pi / 2)$, L be the operator of type $\omega$ satisfying the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2, $\alpha \in(0, \infty), \beta \in(n(1 / p-1 / 2) /(2 k), \infty)$, $\mu \in(\omega, \pi / 2)$ and $\psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$. Then the Hardy space $H_{L}^{p}\left(\mathbb{R}^{n}\right)=H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms.

Before proving Theorem 5.1, we first give an application of this theorem. Let $\alpha \in(0, \infty)$ and $L^{\alpha}$ be the fractional power with exponent $\alpha$ of $L$ defined by the $H_{\infty}$ functional calculus in $L^{2}\left(\mathbb{R}^{n}\right)$ (see, for example, $\left.[39,26]\right)$. More precisely, choose $m \in \mathbb{N}$ such that $m>\alpha$. Then, $z^{\alpha}(1+z)^{-m} \in \Psi_{\alpha, m-\alpha}\left(S_{\mu}^{0}\right)$ for all $\mu \in[0, \pi / 2)$ and $L^{\alpha}$ is defined by setting

$$
L^{\alpha} \equiv\left(z^{\alpha}\right)(L) \equiv(1+L)^{m}\left(\frac{z^{\alpha}}{(1+z)^{m}}\right)(L)
$$

For more details about $L^{\alpha}$, we refer the reader to [39, 26] and the references therein.
Assume that $-L^{\alpha}$ generates a bounded holomorphic semigroup $\left\{e^{-t L^{\alpha}}\right\}_{t>0}$. From $[26$, Example 3.4.6], it follows that this is true when $\alpha \in(0,1]$, and in this case, $\left\{e^{-t L^{\alpha}}\right\}_{t>0}$ is called the subordinated semigroup (see $[26$, p. 80$]$ for more details). For all $f \in L^{2}\left(\mathbb{R}^{n}\right)$, define the $L^{\alpha}$-adapted square function $S_{L^{\alpha}}$ by setting, for all $x \in \mathbb{R}^{n}$,

$$
\begin{equation*}
S_{L^{\alpha}} f(x) \equiv\left\{\iint_{\Gamma(x)}\left|t^{2 k \alpha} L^{\alpha} e^{-t^{2 k \alpha} L^{\alpha}} f(y)\right|^{2} \frac{d y d t}{t^{n+1}}\right\}^{1 / 2} \tag{5.2}
\end{equation*}
$$

For $p \in(0,1]$, we also define the Hardy space $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)$ associated to $L^{\alpha}$ to be the completion of the set

$$
\begin{equation*}
\mathbb{H}_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right) \equiv\left\{f \in L^{2}\left(\mathbb{R}^{n}\right):\left\|S_{L^{\alpha}} f\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}<\infty\right\} \tag{5.3}
\end{equation*}
$$

with respect to the quasi-norm $\|f\|_{H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)} \equiv\left\|S_{L^{\alpha}} f\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}$.
With the help of Theorem 5.1, we immediately obtain the following interesting corollary.
Corollary 5.1. Let $p \in(0,1]$ and $L$ satisfy the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$. Assume further that when $\alpha \in(1, \infty),-L^{\alpha}$ generates a bounded holomorphic semigroup. Then, for all $\alpha \in(0, \infty)$, the Hardy spaces $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)=H_{L}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms.

Proof. Let $\omega \in[0, \pi / 2)$. Recall that $L$ is an operator of type $\omega$. For all $\alpha \in(0, \infty)$, $\mu \in(\omega, \pi / 2)$ and $\xi \in S_{\mu}^{0}$, set $\psi(\xi) \equiv \xi^{\alpha} e^{-\xi^{\alpha}}$. Then, for all $\beta \in(n(1 / p-1 / 2) /(2 k), \infty)$, $\psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$ and hence, by Theorem 5.1, we have that for all $f \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\|f\|_{H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)}=\left\|S_{L^{\alpha}} f\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}=\left\|Q_{\psi, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}=\|f\|_{H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)} \sim\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)}
$$

which together with the density of $L^{2}\left(\mathbb{R}^{n}\right)$ in $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ and $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)$ shows that $H_{L}^{p}\left(\mathbb{R}^{n}\right)=$ $H_{L^{\alpha}}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms. This finishes the proof of Corollary 5.1.

Let $\omega \in[0, \pi / 2)$ be as in Section 2 and $\mu \in(\omega, \pi / 2)$. To prove Theorem 5.1 , we introduce two operators as follows:
(i) For all $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ and $\psi \in \Psi\left(S_{\mu}^{0}\right)$, the operator $\pi_{\psi, L}$ is defined by setting, for all $x \in \mathbb{R}^{n}$,

$$
\begin{equation*}
\pi_{\psi, L} F(x) \equiv \int_{0}^{\infty} \psi\left(t^{2 k} L\right) F(x, t) \frac{d t}{t} \tag{5.4}
\end{equation*}
$$

(ii) For all $\psi, \tilde{\psi} \in \Psi\left(S_{\mu}^{0}\right), f \in H_{\infty}\left(S_{\mu}^{0}\right)$ and $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$, the operator $Q^{f}$ is defined by setting, for all $x \in \mathbb{R}^{n}$ and $s \in(0, \infty)$,

$$
\begin{align*}
Q^{f} F(x, s) & \equiv Q_{\psi, L} \circ f(L) \circ \pi_{\tilde{\psi}, L} F(x, s)  \tag{5.5}\\
& =\int_{0}^{\infty} \psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right) F(x, t) \frac{d t}{t}
\end{align*}
$$

where the operator $Q_{\psi, L}$ is defined as in (5.1).
Observe that by (4.5), $Q_{\psi, L}$ is bounded from $L^{2}\left(\mathbb{R}^{n}\right)$ to $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ and so is $Q_{\bar{\psi}, L^{*}}$. By Fubini's theorem and Hölder's inequality, we have that for all $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ and $g \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\begin{aligned}
\int_{\mathbb{R}^{n}} \pi_{\psi, L} F(x) \overline{g(x)} d x & =\int_{\mathbb{R}^{n}} \int_{0}^{\infty} \psi\left(t^{2 k} L\right) F(x, t) \frac{d t}{t} \overline{g(x)} d x \\
& =\int_{0}^{\infty} \int_{\mathbb{R}^{n}} F(x, t) \overline{Q_{\bar{\psi}, L^{*}}(g)(x)} d x \frac{d t}{t} .
\end{aligned}
$$

Thus, $Q_{\bar{\psi}, L^{*}}$ is the adjoint operator of $\pi_{\psi, L}$, which, together with the above observation, shows that $\pi_{\psi, L}$ is bounded from $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ to $L^{2}\left(\mathbb{R}^{n}\right)$. From these facts and (5.5) together with that $L$ has a bounded $H_{\infty}$ functional calculus in $L^{2}\left(\mathbb{R}^{n}\right)$, it follows that $Q^{f}$ is bounded on $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$.

Let $\sigma_{1}, \sigma_{2}, \tau_{1}, \tau_{2} \in(0, \infty)$. Assume that $\psi \in \Psi_{\sigma_{1}, \tau_{1}}\left(S_{\mu}^{0}\right)$ and $\tilde{\psi} \in \Psi_{\sigma_{2}, \tau_{2}}\left(S_{\mu}^{0}\right)$. We now consider the operator $\psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right)$ in (5.5). Let $a \in\left(0, \min \left\{\sigma_{1}, \tau_{2}\right\}\right)$ and $b \in\left(0, \min \left\{\sigma_{2}, \tau_{1}\right\}\right)$. For $s, t \in(0, \infty)$, when $s \leq t$, we write

$$
\begin{align*}
\psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right) & =\left(\frac{s^{2 k}}{t^{2 k}}\right)^{a}\left(s^{2 k} L\right)^{-a} \psi\left(s^{2 k} L\right) f(L)\left(t^{2 k} L\right)^{a} \widetilde{\psi}\left(t^{2 k} L\right)  \tag{5.6}\\
& \equiv\left(\frac{s^{2 k}}{t^{2 k}}\right)^{a} T_{s^{2 k}, t^{2 k}}
\end{align*}
$$

while when $s>t$, we write

$$
\begin{align*}
\psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right) & =\left(\frac{t^{2 k}}{s^{2 k}}\right)^{b}\left(s^{2 k} L\right)^{b} \psi\left(s^{2 k} L\right) f(L)\left(t^{2 k} L\right)^{-b} \widetilde{\psi}\left(t^{2 k} L\right)  \tag{5.7}\\
& \equiv\left(\frac{t^{2 k}}{s^{2 k}}\right)^{b} T_{s^{2 k}, t^{2 k}}
\end{align*}
$$

Then, we have the following useful estimate on $\left\{T_{s, t}\right\}_{s, t>0}$.

Lemma 5.1. Let $k \in \mathbb{N}$ be as in (2.6), $\sigma_{1}, \sigma_{2}, \tau_{1}, \tau_{2} \in(0, \infty), \omega \in[0, \pi / 2), \mu \in$ $(\omega, \pi / 2), \psi \in \Psi_{\sigma_{1}, \tau_{1}}\left(S_{\mu}^{0}\right), \widetilde{\psi} \in \Psi_{\sigma_{2}, \tau_{2}}\left(S_{\mu}^{0}\right), a \in\left(0, \min \left\{\sigma_{1}, \tau_{2}\right\}\right)$ and $b \in\left(0, \min \left\{\sigma_{2}, \tau_{1}\right\}\right)$. Let $f \in H_{\infty}\left(S_{\mu}^{0}\right)$. Let $\left\{T_{s, t}\right\}_{s, t>0}$ be as in (5.6) and (5.7) with $s^{2 k}$ and $t^{2 k}$ replaced, respectively, by $s$ and $t$. Then, there exists a positive constant $C$ such that for all $M \in$ $\left(0, \min \left\{\sigma_{2}+a, \tau_{1}+b\right\}\right)$, $s, t \in(0, \infty)$, closed sets $E, F \subset \mathbb{R}^{n}$ and $g \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$,

$$
\begin{equation*}
\left\|T_{s, t} g\right\|_{L^{2}(F)} \leq C\|f\|_{L^{\infty}\left(\mathbb{R}^{n}\right)} \min \left\{1, \frac{\max \{t, s\}}{[\operatorname{dist}(E, F)]^{2 k}}\right\}^{M}\|g\|_{L^{2}(E)} \tag{5.8}
\end{equation*}
$$

Proof. We prove this lemma by considering two cases. If $s \leq t$, since $a \in\left(0, \min \left\{\sigma_{1}, \tau_{2}\right\}\right)$, we have that for all $\xi \in S_{\mu}^{0}$,

$$
\left|(s \xi)^{-a} \psi(s \xi) f(\xi)\right| \lesssim \frac{|s \xi|^{\sigma_{1}-a}}{1+|s \xi|^{\sigma_{1}+\tau_{1}}}\|f\|_{L^{\infty}\left(\mathbb{R}^{n}\right)} \lesssim 1
$$

and

$$
\left|(t \xi)^{a} \widetilde{\psi}(t \xi)\right| \lesssim \frac{|t \xi|^{\sigma_{2}+a}}{1+|t \xi|^{\sigma_{2}+\tau_{2}}},
$$

which, together with Lemma 3.3 with $\psi$ and $f$ therein respectively replaced by $(t \xi)^{a} \widetilde{\psi}(t \xi)$ and $(s \xi)^{-a} \psi(s \xi) f(\xi)$, implies that the family $\left\{T_{s, t}\right\}_{s \leq t}$ of operators satisfy the $k$-DaviesGaffney estimate of order $\sigma_{2}+a$ in $t$.

Similarly, if $s>t$, since $b \in\left(0, \min \left\{\sigma_{2}, \tau_{1}\right\}\right)$, we obtain that for all $\xi \in S_{\mu}^{0}$,

$$
\left|f(\xi)(t \xi)^{-b} \widetilde{\psi}(t \xi)\right| \leq \frac{|t \xi|^{\sigma_{2}-b}}{1+|t \xi|^{\sigma_{2}+\tau_{2}}}\|f\|_{L^{\infty}\left(\mathbb{R}^{n}\right)} \lesssim 1
$$

and

$$
\left|(s \xi)^{b} \psi(s \xi)\right| \leq \frac{|s \xi|^{\tau_{1}+b}}{1+|s \xi|^{\sigma_{1}+\tau_{1}}}
$$

which, together with Lemma 3.3 with $\psi$ and $f$ therein respectively replaced by $(s \xi)^{b} \psi(s \xi)$ and $f(\xi)(t \xi)^{-b} \widetilde{\psi}(t \xi)$, implies that the family $\left\{T_{s, t}\right\}_{s>t}$ of operators satisfy the $k$-DaviesGaffney estimate of order $\tau_{1}+b$ in $s$.

Thus, for all $M \in\left(0, \min \left\{\sigma_{2}+a, \tau_{1}+b\right\}\right)$, we immediately obtain (5.8), which completes the proof of Lemma 5.1.

Lemma 5.2. Let $p \in(0,1], L$ be the operator of type $\omega$ satisfying the assumptions $\left(\mathrm{A}_{1}\right)$, $\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2, $\alpha \in(0, \infty), \beta \in(n(1 / p-1 / 2) /(2 k), \infty), \omega \in[0, \pi / 2)$, $\mu \in(\omega, \pi / 2), \psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$ and $\widetilde{\psi} \in \Psi_{\beta, \alpha}\left(S_{\mu}^{0}\right)$. Then the operator $Q^{f}$ originally defined in (5.5) on $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ can be continuously extended to a bounded linear operator on $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$. Moreover, there exists a positive constant $C$ such that for all $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ and $f \in$ $H_{\infty}\left(S_{\mu}^{0}\right)$,

$$
\begin{equation*}
\left\|Q^{f} F\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \leq C\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \tag{5.9}
\end{equation*}
$$

Proof. By the density of $T^{2}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ in $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ (see [14]), it suffices to prove (5.9) for all $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$. To this end, by borrowing some idea from the proof of Theorem 1.1 in [47], we only need show that for all $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atoms $A$,

$$
\begin{equation*}
\left\|Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \lesssim\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)} . \tag{5.10}
\end{equation*}
$$

Indeed, if (5.10) holds, then from Theorem 4.2 and the $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$-boundedness of $Q^{f} A$, it follows that for any $F \in T^{2}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$, there exist a sequence $\left\{A_{j}\right\}_{j=0}^{\infty}$ of $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atoms and $\left\{\lambda_{j}\right\}_{j=0}^{\infty} \in l^{p}$ such that $F \equiv \sum_{j=0}^{\infty} \lambda_{j} A_{j}$ with the sum converges in both pointwise and $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$, and $\left\{\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right\}^{1 / p} \sim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}$. We claim that for $\frac{d x d t}{t}$-almost every $(x, t) \in \mathbb{R}_{+}^{n+1}$,

$$
\begin{equation*}
\left|Q^{f}\left(\sum_{j=0}^{\infty} \lambda_{j} A_{j}\right)\right| \leq \sum_{j=0}^{\infty}\left|\lambda_{j} Q^{f} A_{j}(x, t)\right| . \tag{5.11}
\end{equation*}
$$

Assume this claim for the moment. By (5.11) and $p \in(0,1]$ together with the monotonicity of $l^{p}$, we have

$$
\begin{aligned}
\left\|Q^{f} F\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} & \leq\left\{\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\left\|Q^{f} A_{j}\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}^{p}\right\}^{1 / p} \\
& \leq \sup _{j \in \mathbb{Z}_{+}}\left\{\left\|Q^{f}\left(A_{j}\right)\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}\right\}\left\{\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right\}^{1 / p} \\
& \lesssim\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\left\{\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right\}^{1 / p} \sim\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} .
\end{aligned}
$$

That is, $Q^{f}$ is bounded on $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$. To show the claim (5.11), for simplicity of the notation, let $d \mu(x, t) \equiv \frac{d x d t}{t}$ for all $(x, t) \in \mathbb{R}_{+}^{n+1}$. By the $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$-boundedness of $Q^{f}$ and the $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$-convergence of $F=\sum_{j=0}^{\infty} \lambda_{j} A_{j}$, we obtain that for any $\eta \in(0, \infty)$,

$$
\lim _{N \rightarrow \infty} \mu\left(\left\{x \in \mathbb{R}^{n}:\left|Q^{f}\left(\sum_{i=N+1}^{\infty} \lambda_{j} A_{j}\right)\right|>\eta\right\}\right) \lesssim \lim _{N \rightarrow \infty} \frac{1}{\eta^{2}}\left\|\sum_{i=N+1}^{\infty} \lambda_{j} A_{j}\right\|_{T^{2}\left(\mathbb{R}_{+}^{n+1}\right)}^{2}=0
$$

This, combined with the Riesz theorem, implies that there exists a subsequence

$$
\left\{Q^{f}\left(\sum_{j=N_{\ell}+1}^{\infty} \lambda_{j} A_{j}\right)\right\}_{l \in \mathbb{N}}
$$

of $\left\{Q^{f}\left(\sum_{j=N+1}^{\infty} \lambda_{j} A_{j}\right)\right\}_{N \in \mathbb{N}}$ such that for $\mu$-almost every $(x, t) \in \mathbb{R}_{+}^{n+1}$,

$$
\lim _{\ell \rightarrow \infty} Q^{f}\left(\sum_{j=N_{\ell}+1}^{\infty} \lambda_{j} A_{j}\right)(x, t)=0
$$

where $\left\{N_{\ell}\right\}_{\ell \in \mathbb{N}} \subset \mathbb{N}$ and $\lim _{\ell \rightarrow \infty} N_{\ell}=\infty$. Therefore, for $\mu$-almost every $(x, t) \in \mathbb{R}_{+}^{n+1}$ and all $\ell \in \mathbb{N}$,

$$
\left|Q^{f}\left(\sum_{j=0}^{\infty} \lambda_{j} A_{j}\right)(x, t)\right| \leq \sum_{j=0}^{N_{\ell}}\left|\lambda_{j} Q^{f} A_{j}(x, t)\right|+\left|Q^{f}\left(\sum_{j=N_{\ell}+1}^{\infty} \lambda_{j} A_{j}\right)(x, t)\right|
$$

which, together with letting $\ell \rightarrow \infty$, shows the claim (5.11).
To finish the proof of Lemma 5.2, we still need prove (5.10). By the homogeneity of the norm $\|\cdot\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}$, without loss of generality, we may assume that $\|f\|_{L^{\infty}\left(S_{\mu}^{0}\right)}=1$. Let $Q$ be the cube associated with the $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atom $A$ and $R_{Q} \equiv Q \times(0, l(Q))$, where $l(Q)$ denotes the side length of $Q$. For all $i \in \mathbb{N}$, set $2^{i} R_{Q} \equiv 2^{i} Q \times\left(0,2^{i} l(Q)\right) \subset \mathbb{R}_{+}^{n+1}$ and $S_{i}\left(R_{Q}\right) \equiv 2^{i} R_{Q} \backslash\left(2^{i-1} R_{Q}\right)$.

For $i=1$, by Hölder's inequality and the $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$-boundedness of $Q^{f}$ and the size condition (4.14) of $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$-atoms, we have

$$
\begin{align*}
\left\|\chi_{2 R_{Q}} Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} & =\left\|\mathcal{A}\left(\chi_{2 R_{Q}} Q^{f} A\right)\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}  \tag{5.12}\\
& \leq\left\|\mathcal{A}\left(\chi_{2 R_{Q}} Q^{f} A\right)\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}|2(\sqrt{n}+2) Q|^{1 / p-1 / 2} \\
& \lesssim\left\{\iint_{R_{Q}}|A(x, t)|^{2} \frac{d x d t}{t}\right\}^{1 / 2}|Q|^{1 / p-1 / 2} \lesssim 1 .
\end{align*}
$$

For $i \geq 2$, using Hölder's inequality and Fubini's theorem, we then obtain

$$
\begin{aligned}
& \left\|\chi_{S_{i}\left(R_{Q}\right)} Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \\
& \quad=\left\|\mathcal{A}\left(\chi_{S_{i}\left(R_{Q}\right)} Q^{f} A\right)\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\mathcal{A}\left(\chi_{S_{i}\left(R_{Q}\right)} Q^{f} A\right)\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}\left|2^{i}(2+\sqrt{n}) Q\right|^{\frac{1}{p}-\frac{1}{2}} \\
& \sim\left\{\left[\int_{0}^{2^{i-1} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s)\left|Q^{f} A(x, s)\right|^{2} \frac{d x d s}{s}\right]^{1 / 2}\right. \\
& \left.\quad+\left[\int_{2^{i-1} l(Q)}^{2^{i} l(Q)} \int_{\mathbb{R}^{n}} \cdots\right]^{1 / 2}\right\}\left|2^{i} Q\right|^{\frac{1}{p}-\frac{1}{2}} \equiv\{\mathrm{I}+\mathrm{O}\}\left|2^{i} Q\right|^{\frac{1}{p}-\frac{1}{2}} .
\end{aligned}
$$

To estimate O, from (5.5), Minkowski's inequality, Fubini's inequality, Lemma 5.1 and Hölder's inequality, we deduce that

$$
\begin{aligned}
\mathrm{O} & \sim\left\{\int_{2^{i-1} l(Q)}^{2^{i} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s)\left|Q^{f} A(x, s)\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2} \\
& \sim\left\{\int_{2^{i-1} l(Q)}^{2^{i} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{j}\left(R_{Q}\right)}(x, s)\left|\int_{0}^{\infty} \psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right) A(x, t) \frac{d t}{t}\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2}
\end{aligned}
$$

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$$
\begin{aligned}
& \lesssim\left\{\int_{2^{i-1} l(Q)}^{2^{i} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s)\left|\int_{0}^{\infty}\left(\frac{t}{s}\right)^{2 k b} T_{s^{2 k}, t^{2 k}} A(x, t) \frac{d t}{t}\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2} \\
& \lesssim \int_{0}^{\infty}\left[\frac{t}{2^{i} l(Q)}\right]^{2 k b}\left[\int_{2^{i-1} l(Q)}^{2^{i} l(Q)} \int_{\mathbb{R}^{n}}\left|T_{s^{2 k}, t^{2 k}} A(x, t)\right|^{2} \chi_{S_{i}\left(R_{Q}\right)}(x, s) d x \frac{d s}{s}\right]^{1 / 2} \frac{d t}{t} \\
& \lesssim \int_{0}^{l(Q)}\left[\frac{t}{2^{i} l(Q)}\right]^{2 k b}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)} \frac{d t}{t} \\
& \lesssim\left\{\int_{0}^{l(Q)}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d t}{t}\right\}^{1 / 2}\left\{\int_{0}^{l(Q)}\left[\frac{t}{2^{i} l(Q)}\right]^{4 k b} \frac{d t}{t}\right\}^{1 / 2} \\
& \lesssim 2^{-2 i k b}|Q|^{1 / 2-1 / p} \sim 2^{-i[2 k b-n(1 / p-1 / 2)]}\left|2^{i} Q\right|^{1 / 2-1 / p} \sim 2^{-i \gamma_{1}}\left|2^{i} Q\right|^{1 / 2-1 / p}
\end{aligned}
$$

where $b \in(n(1 / p-1 / 2) /(2 k), \beta)$ and $\gamma_{1} \equiv 2 k b-n(1 / p-1 / 2)>0$.
Let $a \in(0, \alpha)$. To estimate I, by Fubini's theorem and Minkowski's inequality, we write

$$
\begin{aligned}
\mathrm{I} & \sim\left\{\int_{0}^{2^{i-1} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s)\left|Q^{f} A(x, s)\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2} \\
& \sim\left\{\int_{0}^{2^{i-1} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s)\left|\int_{0}^{\infty} \psi\left(s^{2 k} L\right) f(L) \widetilde{\psi}\left(t^{2 k} L\right) A(x, t) \frac{d t}{t}\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2} \\
& \sim\left\{\int_{0}^{2^{i-1} l(Q)} \int_{\mathbb{R}^{n}} \chi_{S_{i}\left(R_{Q}\right)}(x, s) \left\lvert\, \int_{0}^{\infty} \min \left\{\left(\frac{s}{t}\right)^{2 k a},\left(\frac{t}{s}\right)^{2 k b}\right\}\right.\right. \\
& \left.\times\left. T_{s^{2 k}, t^{2 k}} A(x, t) \frac{d t}{t}\right|^{2} d x \frac{d s}{s}\right\}^{1 / 2} \\
& \lesssim \int_{0}^{l(Q)}\left\{\int_{0}^{t} \int_{\mathbb{R}^{n}}\left(\frac{s}{t}\right)^{4 k a}\left|T_{s^{2 k}, t^{2 k}} A(x, t)\right|^{2} \chi_{S_{i}\left(R_{Q}\right)}(x, s) d x \frac{d s}{s}\right\}^{1 / 2} \frac{d t}{t} \\
& +\int_{0}^{l(Q)}\left\{\int_{t}^{2^{i-1} l(Q)} \int_{\mathbb{R}^{n}}\left(\frac{t}{s}\right)^{4 k b}\left|T_{s^{2 k}, t^{2 k}} A(x, t)\right|^{2} \chi_{S_{i}\left(R_{Q}\right)}(x, s) d x \frac{d s}{s}\right\}^{1 / 2} \frac{d t}{t} \\
\equiv & \mathrm{I}_{1}+\mathrm{I}_{2} .
\end{aligned}
$$

Let $M \in(n(1 / p-1 / 2) /(2 k), \min \{\alpha+b, \beta+a\})$. It follows from Lemma 5.1 and Hölder's inequality that

$$
\begin{aligned}
\mathrm{I}_{1} & \lesssim \int_{0}^{l(Q)}\left[\int_{0}^{t}\left(\frac{s}{t}\right)^{4 k a}\left\{\frac{t^{2 k}}{\left[\operatorname{dist}\left(R_{Q}, S_{i}\left(R_{Q}\right)\right)\right]^{2 k}}\right\}^{2 M}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d s}{s}\right]^{1 / 2} \frac{d t}{t} \\
& \lesssim \int_{0}^{l(Q)}\left[\int_{0}^{t}\left(\frac{s}{t}\right)^{4 k a}\left\{\frac{t^{2 k}}{\left[2^{i} l(Q)\right]^{2 k}}\right\}^{2 M}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{2} \frac{d s}{s}\right]^{1 / 2} \frac{d t}{t}
\end{aligned}
$$

$$
\begin{aligned}
& \sim \frac{1}{\left[2^{i} l(Q)\right]^{2 k M}} \int_{0}^{l(Q)}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)} t^{2 k M}\left[\int_{0}^{t}\left(\frac{s}{t}\right)^{4 k a} \frac{d s}{s}\right]^{1 / 2} \frac{d t}{t} \\
& \lesssim \frac{1}{\left[2^{i} l(Q)\right]^{2 k M}}\left\{\iint_{R_{Q}}|A(x, t)|^{2} \frac{d x d t}{t}\right\}^{1 / 2}\left\{\int_{0}^{l(Q)} t^{4 k M} \frac{d t}{t}\right\}^{1 / 2} \\
& \sim 2^{-2 i k M}\left\{\iint_{R_{Q}}|A(x, t)|^{2} \frac{d x d t}{t}\right\}^{1 / 2} \lesssim 2^{-i[2 k M-n(1 / p-1 / 2)]}\left|2^{i} Q\right|^{1 / p-1 / 2} \\
& \sim 2^{-i \gamma_{2}}\left|2^{i} Q\right|^{1 / p-1 / 2},
\end{aligned}
$$

where $\gamma_{2} \equiv 2 k M-n(1 / p-1 / 2)>0$.
For $\mathrm{I}_{2}$, via some similar calculations to the estimate of $\mathrm{I}_{1}$, we obtain

$$
\begin{aligned}
\mathrm{I}_{2} & \lesssim \int_{0}^{l(Q)}\left[\int_{t}^{2^{i} l(Q)}\left(\frac{t}{s}\right)^{4 k b}\left\{\frac{s^{2 k}}{\left[2^{i} l(Q)\right]^{2 k}}\right\}^{2 M} \frac{d s}{s}\right]^{1 / 2}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)} \frac{d t}{t} \\
& \lesssim \int_{0}^{l(Q)}\left\{\left[\frac{t}{2^{i} l(Q)}\right]^{2 k b}+\left[\frac{t}{2^{i} l(Q)}\right]^{2 k M}\right\}\|A(\cdot, t)\|_{L^{2}\left(\mathbb{R}^{n}\right)} \frac{d t}{t} \\
& \lesssim\left\{\iint_{R_{Q}}|A(x, t)|^{2} \frac{d x d t}{t}\right\}^{1 / 2}\left\{\int_{0}^{l(Q)}\left(\left[\frac{t}{2^{i} l(Q)}\right]^{4 k b}+\left[\frac{t}{2^{i} l(Q)}\right]^{4 k M}\right) \frac{d t}{t}\right\}^{1 / 2} \\
& \lesssim\left(2^{-2 i k b}+2^{-2 i k M}\right)|Q|^{1 / 2-1 / p} \sim\left(2^{-i \gamma_{1}}+2^{-i \gamma_{2}}\right)\left|2^{i} Q\right|^{1 / 2-1 / p}
\end{aligned}
$$

Combining the estimates of $\mathrm{I}_{1}$ and $\mathrm{I}_{2}$, we obtain

$$
\begin{equation*}
\mathrm{O} \lesssim\left(2^{-i \gamma_{1}}+2^{-i \gamma_{2}}\right)\left|2^{i} Q\right|^{1 / 2-1 / p} \tag{5.13}
\end{equation*}
$$

By (5.12) and (5.13), we have

$$
\begin{aligned}
\left\|Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}^{p} & \lesssim\left\|\chi_{2 R_{Q}} Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}^{p}+\sum_{i=2}^{\infty}\left\|\chi_{S_{i}\left(R_{Q}\right)} Q^{f} A\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}^{p} \\
& \lesssim 1+\sum_{i=2}^{\infty}\left(2^{-i \gamma_{1} p}+2^{-i \gamma_{2} p}\right) \lesssim 1
\end{aligned}
$$

Thus, (5.10) holds, which completes the proof of Lemma 5.2.
As an application of Lemma 5.2, we obtain the following boundedness of $Q_{\psi, L}$ and $\pi_{\psi, L}$.
Lemma 5.3. Let $p \in(0,1], \omega \in[0, \pi / 2), L$ be the operator of type $\omega$ satisfying the assumptions $\left(\mathrm{A}_{1}\right),\left(\mathrm{A}_{2}\right)$ and $\left(\mathrm{A}_{3}\right)$ in Section 2, $\alpha \in(0, \infty), \beta \in(n(1 / p-1 / 2) /(2 k), \infty)$ and $\mu \in(\omega, \pi / 2)$. Then
(i) the operator $Q_{\psi, L}$, originally defined on $L^{2}\left(\mathbb{R}^{n}\right)$ as in (5.1) with $\psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$, can be extended to a bounded linear operator from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$,
(ii) the operator $\pi_{\psi, L}$, originally defined on $T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$ as in (5.4) with $\psi \in \Psi_{\beta, \alpha}\left(S_{\mu}^{0}\right)$, can be extended to a bounded linear operator from $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ to $H_{L}^{p}\left(\mathbb{R}^{n}\right)$.

Proof. The proof of Lemma 5.3 is quite similar to that of [32, Proposition 4.9]. For the convenience of the reader, we present the details. We first recall a Calderón reproducing formula from [32, (4.12)]. For all $\psi \in \Psi\left(S_{\mu}^{0}\right)$, there exists a function $\widetilde{\psi} \in \Psi\left(S_{\mu}^{0}\right)$ such that

$$
\int_{0}^{\infty} \psi(t) \widetilde{\psi}(t) \frac{d t}{t}=1
$$

Moreover, we have

$$
\begin{equation*}
\pi_{\psi, L} \circ Q_{\tilde{\psi}, L}=\pi_{\widetilde{\psi}, L} \circ Q_{\psi, L}=I \text { in } L^{2}\left(\mathbb{R}^{n}\right) \tag{5.14}
\end{equation*}
$$

In particular, let $\psi_{0}(z) \equiv z e^{-z}$ for all $z \in S_{\mu}^{0}$. We then choose $\widetilde{\psi}_{0}(z) \equiv C(M) z^{M} e^{-z}$ for all $z \in S_{\mu}^{0}$ such that $\widetilde{\psi}_{0} \in \Psi_{M, N}\left(S_{\mu}^{0}\right)$ for any $N \in(0, \infty)$, where $M$ is the smallest positive integer larger than $n(1 / p-1 / 2) /(2 k)$ and $C(M) \int_{0}^{\infty} t^{M} e^{-2 t} d t=1$.

By Definition 5.1 and (5.1), we have that for all $f \in H_{L}^{p}\left(\mathbb{R}^{n}\right) \cap L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\left\|Q_{\psi_{0}, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}=\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)}
$$

which implies that $Q_{\psi_{0}, L}$ is bounded from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$. For all $\psi \in \Psi_{\alpha, \beta}\left(S_{\mu}^{0}\right)$, by this, together with the Calderón reproducing formula (5.14) and Lemma 5.2 with $f \equiv 1$ therein, we obtain that for all $f \in H_{L}^{p}\left(\mathbb{R}^{n}\right)$,

$$
\begin{aligned}
\left\|Q_{\psi, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} & \sim\left\|Q_{\psi, L} \circ \pi_{\tilde{\psi}_{0}, L} \circ Q_{\psi_{0}, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \lesssim\left\|Q_{\psi_{0}, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \\
& \sim\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)} .
\end{aligned}
$$

That is, $Q_{\psi, L}$ is bounded from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$, which completes the proof of (i).
On the other hand, for all $\psi \in \Psi_{\beta, \alpha}\left(S_{\mu}^{0}\right)$, since $\psi_{0} \in \Psi_{1, \beta}\left(S_{\mu}^{0}\right)$, it follows from Lemma 5.2 with $f \equiv 1$ therein that for all $F \in T^{p}\left(\mathbb{R}_{+}^{n+1}\right) \cap T^{2}\left(\mathbb{R}_{+}^{n+1}\right)$,

$$
\left\|\pi_{\psi, L} F\right\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)}=\left\|Q_{\psi_{0}, L} \circ \pi_{\psi, L} F\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \lesssim\|F\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}
$$

which shows that $\pi_{\psi, L}$ is bounded from $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$ to $H_{L}^{p}\left(\mathbb{R}^{n}\right)$. This finishes the proof of (ii) and hence Lemma 5.3.

Proof of Theorem 5.1. By Definitions 4.1 and 5.1, to show Theorem 5.1, it suffices to prove that $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)=\mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ with equivalent norms.

The inclusion $\mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$ is an easy consequence of the boundedness of $Q_{\psi, L}$ from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to $T^{p}\left(\mathbb{R}_{+}^{n+1}\right)$, which is true by Lemma $5.3(\mathrm{i})$. We now prove $\mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right) \subset$ $\underset{\sim}{\mathbb{H}_{L}^{p}}\left(\mathbb{R}^{n}\right)$. Let $\psi_{0}(z) \equiv z e^{z}$ for all $z \in S_{\mu}^{0}$. Observe that for any $\psi \in \underset{\sim}{\Psi}\left(\underset{\alpha}{ }\left(S_{\mu}^{0}\right)\right.$, we can choose $\widetilde{\psi}(z) \equiv \widetilde{C}(M) z^{M} e^{-z}$ for all $z \in S_{\mu}^{0}$ such that (5.14) holds, where $\widetilde{C}(M)$ is a constant such
that $\widetilde{C}(M) \int_{0}^{\infty} t^{M-1} e^{-t} \psi(t) d t=1$. By (5.14), Lemma 5.2 with $f \equiv 1$ therein, and Lemma 5.3(i), we obtain that for all $f \in \mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)$,

$$
\begin{aligned}
\|f\|_{H_{L}^{p}\left(\mathbb{R}^{n}\right)} & =\left\|Q_{\psi_{0}, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)}=\left\|Q_{\psi_{0}, L} \circ \pi_{\tilde{\psi}, L} \circ Q_{\psi, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \\
& \lesssim\left\|Q_{\psi, L} f\right\|_{T^{p}\left(\mathbb{R}_{+}^{n+1}\right)} \sim\|f\|_{H_{\psi, L}^{p}\left(\mathbb{R}^{n}\right)},
\end{aligned}
$$

which implies that $\mathbb{H}_{\psi, L}^{p}\left(\mathbb{R}^{n}\right) \subset \mathbb{H}_{L}^{p}\left(\mathbb{R}^{n}\right)$. This finishes the proof of Theorem 5.1.

## 6 Riesz transforms on $H_{L^{i}}^{p}\left(\mathbb{R}^{n}\right)$ for $i \in\{1,2\}$

In this section, for the $2 k$-order divergence form homogeneous elliptic operator $L_{1}$ with complex bounded measurable coefficients and the $2 k$-order Schrödinger type operator $L_{2}$, we consider the behavior of their Riesz transforms $\nabla^{k} L_{i}^{-1 / 2}$ on the Hardy space $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$, respectively for $i \in\{1,2\}$. First, we study the boundedness of $\nabla^{k} L_{i}^{-1 / 2}$ on $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ for $i \in\{1,2\}$. To this end, we need the following useful estimates.

Lemma 6.1. Let $p \in(0,1], M, k \in \mathbb{N}$, $L_{1}$ be the $2 k$-order divergence form homogenous elliptic operator with complex bounded measurable coefficients and $L_{2}$ the $2 k$-order Schrödinger type operator. Then, there exists a positive constant $C$ such that for all $i \in\{1,2\}$, closed sets $E, F$ in $\mathbb{R}^{n}$ with $\operatorname{dist}(E, F)>0, f \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$ and $t \in(0, \infty)$,

$$
\begin{equation*}
\left\|\nabla^{k} L_{i}^{-1 / 2}\left(I-e^{-t L_{i}}\right)^{M} f\right\|_{L^{2}(F)} \leq C\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)} \tag{6.1}
\end{equation*}
$$

and

$$
\begin{equation*}
\left\|\nabla^{k} L_{i}^{-1 / 2}\left(t L_{i} e^{-t L_{i}}\right)^{M} f\right\|_{L^{2}(F)} \leq C\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)} \tag{6.2}
\end{equation*}
$$

Proof. We prove this lemma by borrowing some ideas from [29]. Let $i \in\{1,2\}$. From [6, Theorem 1.1] and [40, Theorem 8.1], we deduce that $\nabla^{k} L_{i}^{-1 / 2}$ is bounded on $L^{2}\left(\mathbb{R}^{n}\right)$. Thus, it suffices to prove Lemma 6.1 in the case that $t<[\operatorname{dist}(E, F)]^{2 k}$. By the $H_{\infty}$ functional calculus in $L^{2}\left(\mathbb{R}^{n}\right)$, we obtain that for all $f \in L^{2}\left(\mathbb{R}^{n}\right)$,

$$
\begin{equation*}
L_{i}^{-1 / 2} f=\frac{1}{2 \sqrt{\pi}} \int_{0}^{\infty} e^{-s L_{i}} s^{-1 / 2} f d s \tag{6.3}
\end{equation*}
$$

which together with the change of variables yields that

$$
\begin{aligned}
& \nabla^{k} L_{i}^{-1 / 2}\left(I-e^{-t L_{i}}\right)^{M} f \\
& \quad=\frac{1}{2 \sqrt{\pi}} \int_{0}^{\infty} \nabla^{k} e^{-s L_{i}}\left(I-e^{-t L_{i}}\right)^{M} f \frac{d s}{\sqrt{s}} \\
& \quad=\frac{\sqrt{M+2}}{2 \sqrt{\pi}} \int_{0}^{\infty} \nabla^{k} e^{-(M+2) s L_{i}}\left(I-e^{-t L_{i}}\right)^{M} f \frac{d s}{\sqrt{s}}
\end{aligned}
$$

$$
\begin{aligned}
= & \frac{\sqrt{M+2}}{2 \sqrt{\pi}} \int_{0}^{t} \sqrt{s} \nabla^{k} e^{-(M+2) s L_{i}}\left[\sum_{j=0}^{M}\binom{M}{j}(-1)^{j} e^{-j t L_{i}}\right] f \frac{d s}{s} \\
& +\frac{\sqrt{M+2}}{2 \sqrt{\pi}} \int_{t}^{\infty} \sqrt{s} \nabla^{k} e^{-(M+2) s L_{i}}\left(I-e^{-t L_{i}}\right)^{M} f \frac{d s}{s} \equiv \mathrm{I}+\mathrm{O},
\end{aligned}
$$

where $\binom{M}{j}$ denotes the binomial coefficient.
To estimate I, we write

$$
\begin{aligned}
\mathrm{I}= & \frac{\sqrt{M+2}}{2 \sqrt{\pi}} \int_{0}^{t} \sqrt{s} \nabla^{k} e^{-s L_{i}} e^{-(M+1) s L_{i}} f \frac{d s}{s} \\
& +\sum_{j=1}^{M} \frac{\sqrt{M+2}}{2 \sqrt{\pi}}\binom{M}{j}(-1)^{j} \int_{0}^{t} \nabla^{k} e^{-j t L_{i}} e^{-(M+2) s L_{i}} f \frac{d s}{\sqrt{s}} \equiv \mathrm{I}_{0}+\sum_{j=1}^{M} \mathrm{I}_{j} .
\end{aligned}
$$

For $\mathrm{I}_{0}$, it follows from Minkowski's inequality, Propositions 3.1 and 3.2, Lemma 3.2 and the assumption $t<[\operatorname{dist}(E, F)]^{2 k}$ that

$$
\begin{aligned}
\left\|\mathrm{I}_{0}\right\|_{L^{2}(F)} & \lesssim \int_{0}^{t}\left\|\sqrt{s} \nabla^{k} e^{-s L_{i}} e^{-(M+1) s L_{i}} f\right\|_{L^{2}(F)} \frac{d s}{s} \\
& \lesssim \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \frac{d s}{s} \\
& \lesssim \exp \left\{-\frac{\widetilde{C}_{1}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\} \\
& \times \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}_{2}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\} \frac{d s}{s}\|f\|_{L^{2}(E)} \\
& \lesssim \exp \left\{-\frac{\widetilde{C}_{1}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\} \frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\|f\|_{L^{2}(E)} \\
& \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)},
\end{aligned}
$$

where $\widetilde{C}, \widetilde{C}_{1}, \widetilde{C}_{2}$ are positive constants such that $\widetilde{C}_{1}+\widetilde{C}_{2}=\widetilde{C}$.
For each $\mathrm{I}_{j}, j \geq 1$, by Lemma 3.2 and Propositions 3.1 and 3.2 , we have

$$
\begin{aligned}
\left\|\mathrm{I}_{j}\right\|_{L^{2}(F)} & \lesssim \frac{1}{\sqrt{t}} \int_{0}^{t}\left\|\left(\sqrt{j t} \nabla^{k} e^{-j t L_{i}}\right) \circ\left(e^{-(M+2) s L_{i}}\right) f\right\|_{L^{2}(F)} \frac{d s}{\sqrt{s}} \\
& \lesssim \frac{1}{\sqrt{t}}\|f\|_{L^{2}(E)} \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\} \frac{d s}{\sqrt{s}} \\
& \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)},
\end{aligned}
$$

which together with the estimate of $\mathrm{I}_{0}$ implies that

$$
\begin{equation*}
\|\mathrm{I}\|_{L^{2}(F)} \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)} \tag{6.4}
\end{equation*}
$$

where and in what follows, $\widetilde{C}$ always denotes a positive constant. We now estimate O by writing

$$
\begin{aligned}
\mathrm{O} & \sim \int_{t}^{\infty} \sqrt{s} \nabla^{k} e^{-s L_{i}} e^{-M s L_{i}}\left(I-e^{-t L_{i}}\right)^{M} e^{-s L_{i}} f \frac{d s}{s} \\
& \sim \int_{t}^{\infty}\left(\sqrt{s} \nabla^{k} e^{-s L_{i}}\right) \circ\left(e^{-s L_{i}}-e^{-(s+t) L_{i}}\right)^{M} \circ\left(e^{-s L_{i}}\right) f \frac{d s}{s} .
\end{aligned}
$$

Using the analytic property of semigroups and Lemma 3.1, we have that for all $g \in L^{2}\left(\mathbb{R}^{n}\right)$ supported in the closed set $E$ and $t<s$,

$$
\begin{aligned}
\left\|\left[e^{-s L_{i}}-e^{-(s+t) L_{i}}\right] g\right\|_{L^{2}(F)} & =\left\|-\int_{0}^{t} \frac{\partial}{\partial r}\left(e^{-(s+r) L_{i}}\right) g d r\right\|_{L^{2}(F)} \\
& \lesssim \int_{0}^{t}\left\|(s+r) L_{i} e^{-(s+r) L_{i}} g\right\|_{L^{2}(F)} \frac{d r}{s+r} \\
& \lesssim \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\} \frac{d r}{s+r}\|g\|_{L^{2}(E)} \\
& \lesssim \frac{t}{s} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\}\|g\|_{L^{2}(E)}
\end{aligned}
$$

Thus,

$$
\begin{equation*}
\left\|\frac{s}{t}\left[e^{-s L_{i}}-e^{-(s+t) L_{i}}\right] g\right\|_{L^{2}(F)} \lesssim \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\}\|g\|_{L^{2}(E)} \tag{6.5}
\end{equation*}
$$

Therefore, by Minkowski's inequality, (6.5), Lemma 3.2, Propositions 3.1 and 3.2, and the change of variables, we obtain

$$
\begin{aligned}
\|\mathrm{O}\|_{L^{2}(F)} \lesssim & \int_{t}^{\infty}\left\|\left(\sqrt{s} \nabla^{k} e^{-s L_{i}}\right) \circ\left(\frac{s}{t}\left[e^{-s L_{i}}-e^{-(s+t) L_{i}}\right]\right)^{M} \circ\left(e^{-s L_{i}}\right) f\right\|_{L^{2}(F)} \\
& \times\left(\frac{t}{s}\right)^{M} \frac{d s}{s} \\
\lesssim & \|f\|_{L^{2}(E)} \int_{t}^{\infty}\left(\frac{t}{s}\right)^{M} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\} \frac{d s}{s} \\
\lesssim & \left(\frac{[\operatorname{dist}(E, F)]^{2 k}}{t}\right)^{-M}\|f\|_{L^{2}(E)}
\end{aligned}
$$

Combining this estimate with (6.4), we have

$$
\left\|\nabla^{k} L_{i}^{-1 / 2}\left(I-e^{-t L_{i}}\right)^{M} f\right\|_{L^{2}(F)} \lesssim\left(\frac{[\operatorname{dist}(E, F)]^{2 k}}{t}\right)^{-M}\|f\|_{L^{2}(E)}
$$

that is, (6.1) holds.
Now, we prove (6.2). Using (6.3) and the change of variables, we have that

$$
\begin{aligned}
\nabla^{k} L_{i}^{-1 / 2}\left(t L_{i} e^{-t L_{i}}\right)^{M} f & =\frac{1}{2 \sqrt{\pi}} \int_{0}^{\infty} \nabla^{k} e^{-s L_{i}}\left(t L_{i} e^{-t L_{i}}\right)^{M} f \frac{d s}{\sqrt{s}} \\
& \sim \int_{0}^{\infty} \nabla^{k} e^{-(M+1) s L_{i}}\left(t L_{i} e^{-t L_{i}}\right)^{M} f \frac{d s}{\sqrt{s}} \\
& \sim \int_{0}^{t} \nabla^{k} e^{-(M+1) s L_{i}}\left(t L_{i} e^{-t L_{i}}\right)^{M} f \frac{d s}{\sqrt{s}}+\int_{t}^{\infty} \cdots \equiv \mathrm{B}+\mathrm{D}
\end{aligned}
$$

An application of the analytic property of semigroups, Propositions 3.1 and 3.2, and Lemmas 3.1 and 3.2 yields that

$$
\begin{aligned}
& \|\mathrm{B}\|_{L^{2}(F)} \quad \lesssim \frac{1}{\sqrt{t}} \int_{0}^{t}\left\|\left(\sqrt{\frac{t}{2}} \nabla^{k} e^{-\frac{t}{2} L_{i}}\right) \circ\left(e^{-(M+1) s L_{i}}\right) \circ\left(\frac{t}{2} L_{i} e^{-\frac{t}{2} L_{i}}\right) \circ\left(t L_{i} e^{-t L_{i}}\right)^{M-1} f\right\|_{L^{2}(F)} \frac{d s}{\sqrt{s}} \\
& \\
& \lesssim \frac{1}{\sqrt{t}} \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \frac{d s}{\sqrt{s}} \\
& \\
& \lesssim \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)},
\end{aligned}
$$

where $C$ is a positive constant.
For the estimate of D , similarly to the estimate for B , we write

$$
\mathrm{D}=\int_{t}^{\infty}\left(\sqrt{s} \nabla^{k} e^{-s L_{i}}\right) \circ\left(\frac{t}{s}\right)^{M} \circ\left[s L_{i} e^{-(s+t) L_{i}}\right]^{M} f \frac{d s}{s}
$$

and we estimate $s L_{i} e^{-(s+t) L_{i}} f$ by

$$
\begin{aligned}
\left\|s L_{i} e^{-(s+t) L_{i}} f\right\|_{L^{2}(F)}= & \left\|\frac{s}{t} e^{-s L_{i}} \int_{0}^{t} \frac{\partial}{\partial r}\left(r L_{i} e^{-r L_{i}}\right) f d r\right\|_{L^{2}(F)} \\
\lesssim & \left\|\frac{s}{t} e^{-s L_{i}} \int_{0}^{t}\left[L_{i} e^{-r L_{i}} f-r L_{i}^{2} e^{-r L_{i}} f\right] d r\right\|_{L^{2}(F)} \\
\lesssim & \left\|\frac{s}{t} e^{-s L_{i}} \int_{0}^{t} L_{i} e^{-r L_{i}} f d r\right\|_{L^{2}(F)} \\
& +\left\|\frac{s}{t} e^{-s L_{i}} \int_{0}^{t} r L_{i}{ }^{2} e^{-r L_{i}} f d r\right\|_{L^{2}(F)} \equiv \mathrm{V}_{1}+\mathrm{V}_{2} .
\end{aligned}
$$

By Minkowski's inequality, Lemma 3.1 and $r<t<s$, we obtain

$$
\begin{aligned}
\mathrm{V}_{1} & \lesssim \frac{s}{t} \int_{0}^{t}\left\|L_{i} e^{-(s+r) L_{i}}(f)\right\|_{L^{2}(F)} d r \lesssim \frac{s}{t} \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{(s+r)^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} \frac{d r}{s} \\
& \lesssim \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)} .
\end{aligned}
$$

Similarly, we have that

$$
\begin{aligned}
\mathrm{V}_{2} & \lesssim \frac{s}{t} \int_{0}^{t}\left\|\left[(r+s) L_{i}\right]^{2} e^{-(r+s) L_{i}} f\right\|_{L^{2}(F)} \frac{d r}{r+s} \\
& \lesssim \frac{s}{t}\|f\|_{L^{2}(E)} \int_{0}^{t} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{(r+s)^{1 /(2 k-1)}}\right\} \frac{d r}{r+s} \\
& \lesssim \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\}\|f\|_{L^{2}(E)},
\end{aligned}
$$

which together with the estimate of $\mathrm{V}_{1}$ shows that the family $\left\{s L_{i} e^{-(s+t) L_{i}}\right\}_{t>0}$ of operators satisfies the $k$-Davies-Gaffney estimate in $s$. Thus, using Minkowski's inequality, Lemmas 3.1 and 3.2, Propositions 3.1 and 3.2 , and the change of variables, we obtain

$$
\begin{aligned}
\|\mathrm{D}\|_{L^{2}(F)} & \lesssim \int_{t}^{\infty}\left\|\left(\sqrt{s} \nabla^{k} e^{-s L_{i}}\right) \circ\left(\frac{t}{s}\right)^{M} \circ\left(s L_{i} e^{-(s+t) L_{i}}\right)^{M} f\right\|_{L^{2}(F)} \frac{d s}{s} \\
& \lesssim\|f\|_{L^{2}(E)} \int_{t}^{\infty}\left(\frac{t}{s}\right)^{M} \exp \left\{-\frac{\widetilde{C}[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{s^{1 /(2 k-1)}}\right\} \frac{d s}{s} \\
& \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)} .
\end{aligned}
$$

Combining the estimates for B and D , we obtain that

$$
\left\|\nabla^{k} L_{i}^{-1 / 2}\left(t L_{i} e^{-t L_{i}}\right)^{M} f\right\|_{L^{2}(F)} \lesssim\left(\frac{t}{[\operatorname{dist}(E, F)]^{2 k}}\right)^{M}\|f\|_{L^{2}(E)},
$$

which shows that (6.2) also holds. This finishes the proof of Lemma 6.1.
With the help of Lemma 6.1, we show that the Riesz transform $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ is bounded from $H_{L}^{p}\left(\mathbb{R}^{n}\right)$ to the classical Hardy space $H^{p}\left(\mathbb{R}^{n}\right)$, which when $p=1, i=2$ and $k=1$ was first obtained in [28].

Theorem 6.1. Let $k \in \mathbb{N}, p \in(n /(n+k), 1]$, $L_{1}$ be the $2 k$-order divergence form homogeneous elliptic operator with complex bounded measurable coefficients and $L_{2}$ the $2 k$-order Schrödinger type operator. Then, for all $i \in\{1,2\}$, the Riesz transform $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ is bounded from $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ to the classical Hardy space $H^{p}\left(\mathbb{R}^{n}\right)$.

Proof. Let $i \in\{1,2\}$. We first claim that to prove Theorem 6.1, it suffices to show that $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ maps each $\left(H_{L_{i}}^{p}, \epsilon, M\right)$-molecule $m$ as in Definition 4.2 with $\epsilon>0$ and $M>n(1 / p-1 / 2) /(2 k)$ into a classical $H^{p}\left(\mathbb{R}^{n}\right)$-molecule in [43] up to a harmless constant multiple.

Indeed, assume this claim for the moment. For any $f \in \mathbb{H}_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$, by Theorem 4.1, there exist $\left\{\lambda_{j}\right\}_{j=0}^{\infty} \in l^{p}$ and a sequence $\left\{m_{j}\right\}_{j=0}^{\infty}$ of $\left(H_{L i}^{p}, \epsilon, M\right)$-molecules such that $f=\sum_{j=0}^{\infty} \lambda_{j} m_{j}$ is a molecular $\left(H_{L}^{p}, 2, \epsilon, M\right)$-representation of $f$ and

$$
\|f\|_{H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)} \sim\left(\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right)^{1 / p}
$$

Moreover, from the $L^{2}\left(\mathbb{R}^{n}\right)$-boundedness of $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ and the fact that $f=\sum_{j=0}^{\infty} \lambda_{j} m_{j}$ holds in $L^{2}\left(\mathbb{R}^{n}\right)$, it follows that

$$
\begin{equation*}
\nabla^{k}\left(L_{i}^{-1 / 2}\right) f=\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left(\sum_{j=0}^{\infty} \lambda_{j} m_{j}\right)=\sum_{j=0}^{\infty} \lambda_{j} \nabla^{k}\left(L_{i}^{-1 / 2}\right) m_{j} \tag{6.6}
\end{equation*}
$$

in $L^{2}\left(\mathbb{R}^{n}\right)$ and hence in the space $\mathcal{S}^{\prime}\left(\mathbb{R}^{n}\right)$ of Schwartz distributions, which, together with the above claim, implies that (6.6) is a classical molecular decomposition of $\nabla^{k}\left(L_{i}^{-1 / 2}\right) f$ in $H^{p}\left(\mathbb{R}^{n}\right)$. Thus, by the molecular characterization of $H^{p}\left(\mathbb{R}^{n}\right)$ in [43], we further obtain that

$$
\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right) f\right\|_{H^{p}\left(\mathbb{R}^{n}\right)} \lesssim\left(\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right)^{1 / p} \sim\|f\|_{H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)}
$$

which, combined with a density argument, then shows that $\nabla^{k}\left(L_{i}^{-1 / 2}\right)$ is bounded from $H_{L_{i}}^{p}\left(\mathbb{R}^{n}\right)$ to $H^{p}\left(\mathbb{R}^{n}\right)$.

Let $m$ be an $\left(H_{L_{i}}^{p}, \epsilon, M\right)$-molecule associated with the cube $Q$ as in Definition 4.2 with $\epsilon>0$ and $M>n(1 / p-1 / 2) /(2 k)$. To prove the above claim, we need prove that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ is a classical $H^{p}\left(\mathbb{R}^{n}\right)$-molecule in [43] up to a harmless constant multiple. To this end, we only need show that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ is a following defined $H^{p}\left(\mathbb{R}^{n}\right)$-molecule in [33, 32], from which it follows that it is also a classical molecule in [43]. In what follows, for any $\gamma \in \mathbb{R}$, we denote by $\lfloor\gamma\rfloor$ the maximal integer not more than $\gamma$. Let $p \in(0,1]$ and $Q$ be a cube in $\mathbb{R}^{n}$. A function $\widetilde{m} \in L^{2}\left(\mathbb{R}^{n}\right)$ is called an $H^{p}\left(\mathbb{R}^{n}\right)$-molecule associated to $Q$ if there exists a positive constant $\epsilon \in(0, \infty)$ such that
(i) for all $j \in \mathbb{Z}_{+}$,

$$
\begin{equation*}
\|\widetilde{m}\|_{L^{2}\left(S_{j}(Q)\right)} \lesssim\left[2^{j} l(Q)\right]^{n(1 / p-1 / 2)} 2^{-j \epsilon} \tag{6.7}
\end{equation*}
$$

(ii) there exists a non-negative integer $M \in \mathbb{Z}_{+}$with $M \geq\lfloor n(1 / p-1)\rfloor$ such that for all multi-indices $\alpha$ with $0 \leq|\alpha| \leq M$,

$$
\begin{equation*}
\int_{\mathbb{R}^{n}} x^{\alpha} \widetilde{m}(x) d x=0 \tag{6.8}
\end{equation*}
$$

We first prove that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ satisfies (6.7). For all $j \in\{0,1,2\}$, by the $L^{2}\left(\mathbb{R}^{n}\right)$ boundedness of $\nabla^{k}\left(L_{i}{ }^{-1 / 2}\right)$ and (4.7), we have

$$
\begin{equation*}
\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right) m\right\|_{L^{2}\left(S_{j}(Q)\right)} \lesssim\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right) m\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\|m\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim|Q|^{1 / 2-1 / p} \tag{6.9}
\end{equation*}
$$

When $j>2$, we write

$$
\begin{aligned}
\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right) m\right\|_{L^{2}\left(S_{j}(Q)\right)} \leq & \left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left(I-e^{-[l(Q)]^{2 k} L_{i}}\right)^{M} m\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& +\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left[I-\left(I-e^{-[l(Q)]^{2 k} L_{i}}\right)^{M}\right] m\right\|_{L^{2}\left(S_{j}(Q)\right)} \equiv \mathrm{I}+\mathrm{O} .
\end{aligned}
$$

An application of Lemma 6.1 and (4.7) gives that

$$
\begin{aligned}
\mathrm{I} & \lesssim\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left(I-e^{-[l(Q)]^{2 k} L_{i}}\right)^{M}\left(m \chi_{2^{j-2} Q}\right)\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& +\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left(I-e^{-[l(Q)]^{2 k} L_{i}}\right)^{M}\left(m \chi_{\mathbb{R}^{n} \backslash\left(2^{j+1} Q\right)}\right)\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& +\left\|\nabla^{k}\left(L_{i}^{-1 / 2}\right)\left(I-e^{-[l(Q)]^{2 k} L_{i}}\right)^{M}\left(m \chi_{2^{j+1} Q \backslash\left(2^{j-2} Q\right)}\right)\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \lesssim\left[\frac{\operatorname{dist}\left(S_{j}(Q), 2^{j-2} Q\right)}{l(Q)}\right]^{2 k M}\left\|m \chi_{2^{j-2} Q}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \\
& +\left[\frac{\operatorname{dist}\left(S_{j}(Q), \mathbb{R}^{n} \backslash\left(2^{j+1} Q\right)\right)}{l(Q)}\right]^{2 k M}\left\|m \chi_{\mathbb{R}^{n} \backslash\left(2^{j+1} Q\right)}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}+\left\|m \chi_{2^{j+1} Q \backslash\left(2^{j-1} Q\right)}\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \\
\lesssim & 2^{-2 j k M}[l(Q)]^{n(1 / 2-1 / p)}+\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \epsilon} .
\end{aligned}
$$

Let $\tilde{\epsilon} \equiv \min \{\epsilon, 2 k M-n(1 / p-1 / 2)\}>0$. We then have

$$
\begin{equation*}
\mathrm{I} \lesssim\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \tilde{\epsilon}} \tag{6.10}
\end{equation*}
$$

To estimate O, from Lemma 6.1 and (4.3), we deduce that

$$
\begin{aligned}
\mathrm{O} & \lesssim \sup _{1 \leq \ell \leq M}\left\|\nabla^{k} L_{i}^{-1 / 2} e^{-\ell[l(Q)]^{2 k} L_{i}} m\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \sim \sup _{1 \leq \ell \leq M}\left\|\nabla^{k} L_{i}^{-1 / 2}\left(\frac{\ell}{M}[l(Q)]^{2 k} L_{i} e^{-\frac{\ell}{M}[l(Q)]^{2 k} L_{i}}\right)^{M}\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M} m\right\|_{L^{2}\left(S_{j}(Q)\right)} \\
& \sim \sup _{1 \leq \ell \leq M} \| \nabla^{k} L_{i}{ }^{-1 / 2}\left(\frac{\ell}{M}[l(Q)]^{2 k} L_{i} e^{-\frac{\ell}{M}[l(Q)]^{2 k} L_{i}}\right)^{M} \\
& \times\left[\chi_{2^{j-2} Q}\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M}\right] m \|_{L^{2}\left(S_{j}(Q)\right)}
\end{aligned}
$$

$$
\begin{aligned}
& +\sup _{1 \leq \ell \leq M} \| \nabla^{k} L_{i}^{-1 / 2}\left(\frac{\ell}{M}[l(Q)]^{2 k} L_{i} e^{-\frac{\ell}{M}[l(Q)]^{2 k} L_{i}}\right)^{M} \\
& \times\left[\chi_{\mathbb{R}^{n} \backslash 2^{j+1} Q}\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M}\right] m\left\|_{L^{2}\left(S_{j}(Q)\right)}+\sup _{1 \leq \ell \leq M}\right\| \nabla^{k} L_{i}^{-1 / 2}\left(\frac{\ell}{M}[l(Q)]^{2 k}\right. \\
& \left.\times L_{i} e^{-\frac{\ell}{M}}[l(Q)]^{2 k} L_{i}\right)^{M}\left[\chi_{2^{j+1} Q \backslash 2^{j-2} Q}\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M}\right] m \|_{L^{2}\left(S_{j}(Q)\right)} \\
\lesssim & 2^{-2 j k M}\left\|\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M} m\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}+\left\|\chi_{2^{j+1} Q \backslash 2^{j-1} Q}\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M} m\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \\
\lesssim & 2^{-2 j k M}\left\{\sum_{\widetilde{k}=0}^{\infty}\left\|\left([l(Q)]^{-2 k} L_{i}^{-1}\right)^{M} m\right\|_{L^{2}\left(S_{\tilde{k}}(Q)\right)}^{2}\right\}^{1 / 2}+\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \epsilon} \\
\lesssim & 2^{-2 j k M}\left\{\sum_{\widetilde{k}=0}^{\infty} 2^{-2 \widetilde{k}[\epsilon+n(1 / p-1 / 2)]}\right\}^{1 / 2}[l(Q)]^{n(1 / 2-1 / p)}+\left[2^{j} l(Q)\right]^{n(1 / 2-1 / p)} 2^{-j \epsilon} \\
\lesssim & 2^{-j \tilde{\epsilon}}\left[2^{j} l(Q)\right]^{n / p-2 / p},
\end{aligned}
$$

which, together with (6.10), implies that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ satisfies (6.7) with $\epsilon$ therein replaced by $\tilde{\epsilon}$.

Now, we prove that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ satisfies (6.8) by borrowing some idea from the proof of Theorem 7.4 in [36]. Let $D\left(\sqrt{L_{i}}\right)$ be the domain of $\sqrt{L_{i}}$ and $R\left(L_{i}^{-1 / 2}\right)$ the range of $L_{i}^{-1 / 2}$. From $[6,40]$, it follows that $D\left(\sqrt{L_{i}}\right)=D\left(\mathfrak{a}_{i}\right)$, where $D\left(\mathfrak{a}_{i}\right) \subset W^{k, 2}\left(\mathbb{R}^{n}\right)$ is the domain of the sesquilinear form associated to $L_{i}$, which implies that $R\left(L_{i}^{-1 / 2}\right) \subset W^{k, 2}\left(\mathbb{R}^{n}\right)$. Let $\left\{\varphi_{j}\right\}_{j=1}^{\infty} \subset C_{\mathrm{c}}^{\infty}\left(\mathbb{R}^{n}\right)$ such that
(i) $\sum_{j=1}^{\infty} \varphi_{j}(x)=1$ for almost every $x \in \mathbb{R}^{n}$;
(ii) for each $j \in \mathbb{N}$, there exists a ball $B_{j} \subset \mathbb{R}^{n}$ such that $\operatorname{supp} \varphi_{j} \subset 2 B_{j}, \varphi_{j} \equiv 1$ on $B_{j}$ and $0 \leq \varphi_{j} \leq 1$;
(iii) there exists a positive constant $C_{\varphi}$ such that for all $j \in \mathbb{N}$ and $x \in \mathbb{R}^{n}$,

$$
\sum_{\ell=1}^{k}\left|\nabla^{\ell} \varphi_{j}(x)\right| \leq C_{\varphi}
$$

(iv) there exists $N_{\varphi} \in \mathbb{N}$ such that $\sum_{j=1}^{\infty} \chi_{2 B_{j}} \leq N_{\varphi}$.

For all $j \in \mathbb{N}$ and multi-indices $\alpha$, let $\eta_{j} \in C_{\mathrm{c}}^{\infty}\left(\mathbb{R}^{n}\right)$ such that $\eta_{j} \equiv 1$ on $2 B_{j}$ and $\operatorname{supp} \eta_{j} \subset$ $4 B_{j}$. Since $R\left(L_{i}^{-1 / 2}\right) \subset W^{k, 2}\left(\mathbb{R}^{n}\right)$ and $\eta_{j} x^{\alpha} \in C_{\mathrm{c}}^{\infty}\left(\mathbb{R}^{n}\right)$, we have

$$
\int_{\mathbb{R}^{n}} x^{\alpha} \nabla^{k} L_{i}^{-1 / 2} m(x) d x=\int_{\mathbb{R}^{n}} x^{\alpha} \nabla^{k-1}\left(\sum_{j=1}^{\infty} \varphi_{j} \nabla L_{i}^{-1 / 2}\right) m(x) d x
$$

$$
\begin{aligned}
& =\sum_{j=1}^{\infty} \int_{\mathbb{R}^{n}} x^{\alpha} \nabla^{k-1}\left(\varphi_{j} \nabla L_{i}^{-1 / 2}\right) m(x) d x \\
& =\sum_{j=1}^{\infty} \int_{\mathbb{R}^{n}} \eta_{j} x^{\alpha} \nabla^{k-1}\left(\nabla L_{i}^{-1 / 2}\right) m(x) d x \\
& =\sum_{j=1}^{\infty}(-1)^{k-1} \int_{\mathbb{R}^{n}}\left(\nabla^{k-1}\left(\eta_{j} x^{\alpha}\right)\right) \nabla\left(L_{i}^{-1 / 2}\right) m(x) d x .
\end{aligned}
$$

Thus, for all $|\alpha| \leq k-1=n(1 /[n /(n+k)]-1)$, we obtain

$$
\begin{aligned}
\left|\int_{\mathbb{R}^{n}} x^{\alpha} \nabla^{k} L_{i}^{-1 / 2} m(x) d x\right| & \leq \sum_{j=1}^{\infty} \mid \int_{\mathbb{R}^{n}}\left(\nabla^{k-1}\left(\eta_{j} x^{\alpha}\right)\right) \nabla\left(L_{i}^{-1 / 2} m(x) d x \mid\right. \\
& \leq \sum_{j=1}^{\infty}\left|\int_{\mathbb{R}^{n}} \eta_{j} \nabla\left(L_{i}-1 / 2\right) m(x) d x\right| \\
& =\sum_{j=1}^{\infty}\left|\int_{\mathbb{R}^{n}} \eta_{j} \nabla\left(\varphi_{i} L_{i}^{-1 / 2}\right) m(x) d x\right| \\
& =\sum_{j=1}^{\infty}\left|\int_{\mathbb{R}^{n}} \nabla\left(\eta_{j}\right) \varphi_{i} L_{i}^{-1 / 2} m(x) d x\right|=0
\end{aligned}
$$

which implies that $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ satisfies (6.8) with $p$ and $M$ respectively replaced by $n /(n+k)$ and $n(1 /[n /(n+k)]-1)$. Thus, $\nabla^{k}\left(L_{i}^{-1 / 2}\right) m$ is a classical $H^{p}\left(\mathbb{R}^{n}\right)$ molecule in [43], which completes the proof of Theorem 6.1.

On the Hardy space $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$, we further obtain its characterization by the Riesz transforms $\nabla^{k}\left(L_{1}^{-1 / 2}\right)$. To this end, we first introduce some notions.

Definition 6.1. Let $p \in(0,1]$ and $L_{1}$ be the $2 k$-order divergence form homogenous elliptic operator with complex bounded measurable coefficients. The Riesz transform Hardy space $H_{L_{1}, \text { Riesz }}^{p}\left(\mathbb{R}^{n}\right)$ is defined to be the completion of the set

$$
\mathbb{H}_{L_{1}, \operatorname{Riesz}}^{p}\left(\mathbb{R}^{n}\right) \equiv\left\{f \in L^{2}\left(\mathbb{R}^{n}\right): \nabla^{k}\left(L_{1}^{-1 / 2}\right) f \in H^{p}\left(\mathbb{R}^{n}\right)\right\}
$$

with respect to the quasi-norm

$$
\|f\|_{H_{L_{1}, \text { Riesz }}^{p}\left(\mathbb{R}^{n}\right)} \equiv\left\|\nabla^{k}\left(L_{1}^{-1 / 2}\right) f\right\|_{H^{p}\left(\mathbb{R}^{n}\right)}
$$

for all $f \in \mathbb{H}_{L_{1}, \text { Riesz }}^{p}\left(\mathbb{R}^{n}\right)$.
We also need the following notion of $L^{p}-L^{q} k$-off-diagonal estimates, which when $k=1$ previously appeared in [3] (see also [32]).

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Definition 6.2. Let $k \in \mathbb{N}, r, q \in(1, \infty)$ and $r \leq q$. A family $\left\{S_{t}\right\}_{t>0}$ of operators is said to satisfy the $L^{r}-L^{q} k$-off-diagonal estimate, if there exist positive constant $C$ and $\widetilde{C}$ such that for all closed sets $E, F \subset \mathbb{R}^{n}$ and $f \in L^{r}\left(\mathbb{R}^{n}\right) \cap L^{2}\left(\mathbb{R}^{n}\right)$ supported in $E$,

$$
\left\|S_{t} f\right\|_{L^{q}(F)} \leq C t^{\frac{n}{2 k}\left(\frac{1}{q}-\frac{1}{r}\right)} \exp \left\{-\widetilde{C} \frac{[\operatorname{dist}(E, F)]^{2 k /(2 k-1)}}{t^{1 /(2 k-1)}}\right\}\|f\|_{L^{r}(E)}
$$

On the $L^{r}-L^{q} k$-off-diagonal estimate of the $2 k$-order divergence form homogeneous elliptic operator $L_{1}$ with complex bounded measurable coefficients, we have the following useful lemma.

Lemma 6.2. Let $L_{1}$ be the $2 k$-order divergence form homogeneous elliptic operator with complex bounded measurable coefficients and $r \in(1,2]$ such that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimate. Then the family $\left\{t L_{1} e^{-t L_{1}}\right\}_{t>0}$ of operators also satisfies the $L^{r}-L^{2} k$-off-diagonal estimate.

Proof. By the analytical property of the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$, we have $\left\{t L_{1} e^{-t L_{1}}\right\}_{t>0}=$ $\left\{2\left(\frac{t}{2} L_{1} e^{-\frac{t}{2} L_{1}}\right)\left(e^{-\frac{t}{2} L_{1}}\right)\right\}_{t>0}$. Since the $k$-Davies-Gaffney estimate is just the $L^{2}-L^{2} k$ -off-diagonal estimate, it follows from Proposition 3.1 and Lemma 3.1 that $\left\{\frac{t}{2} L_{1} e^{-\frac{t}{2} L_{1}}\right\}_{t>0}$ satisfies the $L^{2}-L^{2} k$-off-diagonal estimate. Moreover, by the fact that $\left\{e^{-\frac{t}{2} L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimate and an argument similar to the proof of Lemma 3.2 with $\left\{A_{t}\right\}_{t>0}$ and $\left\{B_{s}\right\}_{s>0}$, respectively, replaced by $\left\{\frac{t}{2} L_{1} e^{-\frac{t}{2} L_{1}}\right\}_{t>0}$ and $\left\{e^{-\frac{t}{2} L_{1}}\right\}_{t>0}$, we obtain that $\left\{t L_{1} e^{-t L_{1}}\right\}_{t>0}$ also satisfies the $L^{r}-L^{2} k$-off-diagonal estimate, which completes the proof of Lemma 6.2.

Proposition 6.1. Let $L_{1}$ be the $2 k$-order divergence form homogeneous elliptic operator with complex bounded measurable coefficients and $r \in(1,2]$ such that the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimate. Then for all $p \in(0,1]$ such that $p>r n /(n+k r)$ and $h \in \mathbb{H}_{L_{1}, \operatorname{Riesz}}^{p}\left(\mathbb{R}^{n}\right)$,

$$
\|h\|_{H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)} \leq C\left\|\nabla^{k} L_{1}^{-1 / 2} h\right\|_{H^{p}\left(\mathbb{R}^{n}\right)}
$$

To prove Proposition 6.1, we need recall some results concerning the homogenous HardySobolev space $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$; see, for example, [13, 27, 44, 45].

Definition 6.3. Let $k \in \mathbb{N}$ and $p \in(0,1]$. The homogeneous Hardy-Sobolev space $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ is defined to be the space

$$
\dot{H}^{k, p}\left(\mathbb{R}^{n}\right) \equiv\left\{f \in \mathcal{S}^{\prime}\left(\mathbb{R}^{n}\right) / \mathscr{P}_{k-1}\left(\mathbb{R}^{n}\right):\|f\|_{\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)} \equiv \sum_{|\sigma|=k}\left\|\partial^{\sigma} f\right\|_{H^{p}\left(\mathbb{R}^{n}\right)}<\infty\right\}
$$

where $\mathcal{S}^{\prime}\left(\mathbb{R}^{n}\right)$ denotes the space of all Schwartz distributions on $\mathbb{R}^{n}$ and $\mathscr{P}_{k-1}\left(\mathbb{R}^{n}\right)$ the class of all polynomials of order strictly less than $k$ on $\mathbb{R}^{n}$.

Let $\ell \in \mathbb{N}$ be fixed. Let $\mathcal{S}\left(\mathbb{R}^{n}\right)$ denote the space of all Schwartz functions on $\mathbb{R}^{n}$ and $\phi \in \mathcal{S}\left(\mathbb{R}^{n}\right)$ such that
(i) $\phi$ is radial, $\operatorname{supp} \phi \subset\left\{x \in \mathbb{R}^{n}:|x|<1\right\}$ and for all $\xi \neq 0, \int_{0}^{\infty}|\hat{\phi}(t \xi)|^{2} \frac{d t}{t}=1$, where $\hat{\phi}$ denotes the Fourier transform of $\phi$,
(ii) for all $|\gamma| \leq \ell, \int_{\mathbb{R}^{n}} x^{\gamma} \phi(x) d x=0$.

For any given $\phi \in \mathcal{S}\left(\mathbb{R}^{n}\right)$ as above and all $f \in \mathcal{S}^{\prime}\left(\mathbb{R}^{n}\right)$, let $Q_{t} f \equiv \phi_{t} * f$, where $\phi_{t} \equiv$ $t^{-n} \phi(x / t)$ for all $t \in(0, \infty)$ and $x \in \mathbb{R}^{n}$. Let $p, q \in(0, \infty)$ and $\alpha \in \mathbb{R}$ such that $|\alpha|<\ell+1$. The homogenous Triebel-Lizorkin space $\dot{F}_{p, q}^{\alpha}\left(\mathbb{R}^{n}\right)$ is defined to be the space

$$
\begin{aligned}
\dot{F}_{p, q}^{\alpha}\left(\mathbb{R}^{n}\right) \equiv\left\{f \in \mathcal{S}^{\prime}\left(\mathbb{R}^{n}\right) / \mathscr{P}\left(\mathbb{R}^{n}\right):\right. & \|f\|_{\dot{F}_{p, q}^{\alpha}\left(\mathbb{R}^{n}\right)} \\
& \left.\equiv\left\|\left\{\int_{0}^{\infty}\left(t^{-\alpha}\left|Q_{t} f\right|\right)^{q} \frac{d t}{t}\right\}^{1 / q}\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}<\infty\right\}
\end{aligned}
$$

where $\mathscr{P}\left(\mathbb{R}^{n}\right)$ denotes the class of all polynomials on $\mathbb{R}^{n}$; see, for example, $[27,44,45]$.
Let $\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)$ for $k \in \mathbb{N}$ denote the homogenous Sobolev space of order $k$ endowed with the norm $\|\cdot\|_{\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)} \equiv\left\|\nabla^{k}(\cdot)\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}$. It is known that the homogeneous Sobolev space $\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)$ and Hardy-Sobolev space $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ coincide, respectively, with the TriebelLizorkin space $\dot{F}_{2,2}^{k}\left(\mathbb{R}^{n}\right)$ and $\dot{F}_{p, 2}^{k}\left(\mathbb{R}^{n}\right)$ with equivalent norms (see, for example, [44, p. 242]).
Definition 6.4. Let $k \in \mathbb{N}, \ell \geq k$ be any fixed positive integer and $p \in(0,1]$. A function $b$ is called an $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$-atom if it satisfies that
(i) there exists a ball $B \subset \mathbb{R}^{n}$ such that $\operatorname{supp} b \subset B$,
(ii) for any $|\gamma| \leq \ell, \int_{\mathbb{R}^{n}} x^{\gamma} b(x) d x=0$,

$$
\begin{equation*}
\|b\|_{\dot{F}_{2,2}^{k}\left(\mathbb{R}^{n}\right)} \leq|B|^{1 / 2-1 / p} \tag{iii}
\end{equation*}
$$

Lemma 6.3. Let $p \in(0,1], k \in \mathbb{N}$ and $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$. Then there exist $\left\{\lambda_{j}\right\}_{j=0}^{\infty} \in l^{p}$ and a sequence $\left\{b_{j}\right\}_{j=0}^{\infty}$ of $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$-atoms such that $f=\sum_{j=0}^{\infty} \lambda_{j} b_{j}$ in $\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$, and $\|f\|_{\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)} \sim\left\{\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right\}^{1 / p}$.
Proof. For any $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$, by the coincidence of Sobolev spaces and HardySobolev spaces with Triebel-Lizorkin spaces, we know that $f \in \dot{F}_{2,2}^{k}\left(\mathbb{R}^{n}\right) \cap \dot{F}_{p, 2}^{k}\left(\mathbb{R}^{n}\right)$. From this and a slight modification on the proof of [45, Proposition 4.3] together with the same observation as in Theorem 4.2 on the convergence of the atomic decomposition for elements in the tent spaces, we deduce all the desired conclusions of Lemma 6.3, which completes the proof of Lemma 6.3.

Proof of Proposition 6.1. For all $g \in L^{2}\left(\mathbb{R}^{n}\right)$, define the operator $S_{1}$ by setting, for all $x \in \mathbb{R}^{n}$,

$$
S_{1} g(x) \equiv\left\{\iint_{\Gamma(x)}\left|t^{k} \sqrt{L_{1}} e^{-t^{2 k} L_{1}} g(y)\right|^{2} \frac{d y d t}{t^{n+1}}\right\}^{1 / 2}
$$

For all $h \in \mathbb{H}_{\text {Riesz, } L_{1}}^{p}\left(\mathbb{R}^{n}\right)$, let $f \equiv L_{1}^{-1 / 2} h$. Then $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ and, by Lemma 6.3, there exist $\left\{\lambda_{j}\right\}_{j=0}^{\infty} \in l^{p}$ and a sequence $\left\{b_{j}\right\}_{j=0}^{\infty}$ of $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$-atoms such that $f=\sum_{j=0}^{\infty} \lambda_{j} b_{j}$ in $\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ and, moreover, $\left(\sum_{j=0}^{\infty}\left|\lambda_{j}\right|^{p}\right)^{1 / p} \sim\|f\|_{\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)}$. By Theorem 5.1 with $L$ replaced by $L_{1}$, to show Proposition 6.1, we only need prove that for all $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right) \cap \dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$ with $p \in(n r /(n+k r), 1]$,

$$
\begin{equation*}
\left\|S_{1} \sqrt{L_{1}} f\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} \lesssim\|f\|_{\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)} \tag{6.12}
\end{equation*}
$$

To prove (6.12), it suffices to prove that for all $\dot{H}^{k, p}\left(\mathbb{R}^{n}\right)$-atoms $b$,

$$
\begin{equation*}
\left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{p}\left(\mathbb{R}^{n}\right)} \lesssim 1 \tag{6.13}
\end{equation*}
$$

Indeed, if (6.13) holds, by the $L^{2}\left(\mathbb{R}^{n}\right)$-boundedness of $S_{1}$ which is deduced from (4.5), and [5, Theorem 1.1], we obtain

$$
\left\|S_{1} \sqrt{L_{1}} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\sqrt{L_{1}} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \sim\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \sim\|f\|_{\dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)}
$$

which together with an argument similar to the proof of (5.11) yields that for almost every $x \in \mathbb{R}^{n},\left|S_{1} \sqrt{L_{1}} f(x)\right| \leq \sum_{j=0}^{\infty}\left|\lambda_{j} S_{1} \sqrt{L_{1}} b_{j}(x)\right|$. This combined with (6.13) shows that (6.12) is valid.

We now prove (6.13). For $j \in \mathbb{N}$, let $\mathcal{R}\left(S_{j}(Q)\right) \equiv \cup_{x \in S_{j}(Q)} \Gamma(x)$ be the saw-tooth region based on $S_{j}(Q) \subset \mathbb{R}^{n}$. By Minkowski's inequality, Hölder's inequality and Fubini's theorem, we obtain

$$
\begin{aligned}
\left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{p}\left(\mathbb{R}^{n}\right)}^{p} & \lesssim \sum_{j=0}^{\infty}\left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{p}\left(S_{j}(Q)\right)}^{p} \\
\lesssim & \left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{2}(4 Q)}^{p}|Q|^{n\left(\frac{1}{p}-\frac{1}{2}\right) p}+\sum_{j=3}^{\infty}\left\|S_{1} \sqrt{L_{1} b}\right\|_{L^{2}\left(S_{j}(Q)\right)}^{p}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right) p} \\
\lesssim & \left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{2}(4 Q)}^{p}|Q|^{\left(\frac{1}{p}-\frac{1}{2}\right) p} \\
& +\sum_{j=3}^{\infty}\left\{\iint_{\mathcal{R}\left(S_{j}(Q)\right)}\left|t^{2 k} L_{1} e^{-t^{2 k} L_{1}} b(y)\right|^{2} \frac{d y d t}{t^{2 k+1}}\right\}^{p / 2}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right) p} \\
\lesssim & \left\|S_{1} \sqrt{L_{1}} b\right\|_{L^{2}(4 Q)}^{p}|Q|^{\left(\frac{1}{p}-\frac{1}{2}\right) p} \\
& +\sum_{j=3}^{\infty}\left\{\int_{2^{j-2} Q} \int_{2^{j-3} l(Q)}^{\infty}\left|t^{2 k} L_{1} e^{-t^{2 k} L_{1}} b(y)\right|^{2} \frac{d y d t}{t^{2 k+1}}\right\}^{p / 2}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right) p} \\
& +\sum_{j=3}^{\infty}\left\{\int_{\mathbb{R}^{n} \backslash 2^{j-2} Q} \int_{0}^{\infty} \cdots\right\}^{p / 2}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right) p}
\end{aligned}
$$

$$
\equiv \mathrm{I}+\sum_{j=3}^{\infty}\left(\mathrm{J}_{j}\right)^{p}+\sum_{j=3}^{\infty}\left(\mathrm{V}_{j}\right)^{p} .
$$

For I, by the $L^{2}\left(\mathbb{R}^{n}\right)$-boundedness of $S_{1},(6.11)$ and $[6$, Theorem 1.1] we have

$$
\begin{equation*}
\mathrm{I} \lesssim\left\|\sqrt{L_{1}} b\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}^{p}|Q|^{\left(\frac{1}{p}-\frac{1}{2}\right) p} \lesssim\|b\|_{\dot{F}_{2,2}^{k}\left(\mathbb{R}^{n}\right)}^{p}|Q|^{\left(\frac{1}{p}-\frac{1}{2}\right) p} \lesssim 1 \tag{6.14}
\end{equation*}
$$

To estimate $\mathrm{J}_{j}$, recall the following embedding theorem (see, for example, [44]) that for all $f \in \dot{F}_{\frac{n r}{} k_{n+k r}, 2}\left(\mathbb{R}^{n}\right)$,

$$
\begin{equation*}
\|f\|_{L^{r}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\nabla^{k} f\right\|_{L^{\frac{n r}{n+k r}\left(\mathbb{R}^{n}\right)}} \tag{6.15}
\end{equation*}
$$

For each $\mathrm{O}_{j}$, using Minkowski's inequality, Lemma 6.2, (6.15), Lemma 6.2, Hölder's inequality and (6.11), we obtain

$$
\begin{aligned}
\mathrm{J}_{j} & \lesssim\left\{\int_{2^{j-3} l(Q)}^{\infty}\left\|t^{2 k} L_{1} e^{-t^{2 k} L_{1}} b\right\|_{L^{2}\left(2^{j-2} Q\right)}^{2} \frac{d t}{t^{1+2 k}}\right\}^{1 / 2}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right)} \\
& \lesssim\left\{\int_{2^{j-3} l(Q)}^{\infty} t^{2 n\left(\frac{1}{2}-\frac{1}{r}\right)} \frac{d t}{t^{1+2 k}}\right\}^{1 / 2}\left|2^{j} l(Q)\right|^{n\left(\frac{1}{p}-\frac{1}{2}\right)}\|b\|_{L^{r}(Q)} \\
& \lesssim\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{r}\right)-k}\|b\|_{L^{r}(Q)} \lesssim\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{r}\right)-k}\left\|\nabla^{k} b\right\|_{L^{\frac{r n}{n+k r}}(Q)} \\
& \lesssim\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{r}\right)-k}\left\|\nabla^{k} b\right\|_{L^{2}(Q)}|l(Q)|^{\frac{n+k r}{r}-\frac{n}{2}} \lesssim 2^{\left[n\left(\frac{1}{p}-\frac{1}{r}\right)-k\right] j} .
\end{aligned}
$$

Let $\alpha \equiv \frac{n}{r}+k-\frac{n}{p}$. Since $p \in\left(\frac{n r}{n+k r}, 1\right]$, we then have $\alpha>0$ and

$$
\begin{equation*}
\sum_{j=3}^{\infty}\left(\mathrm{J}_{j}\right)^{p} \lesssim \sum_{j=3}^{\infty} 2^{-\alpha j p} \lesssim 1 \tag{6.16}
\end{equation*}
$$

To estimate $\mathrm{V}_{j}$, we write

$$
\begin{aligned}
\mathrm{V}_{j} \lesssim & {\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{2}\right)}\left\{\int_{\mathbb{R}^{n} \backslash 2^{j-2} Q} \int_{0}^{2^{j-3} l(Q)}\left|t^{2 k} L_{1} e^{-t^{2 k} L_{1}} b(y)\right|^{2} \frac{d y d t}{t^{2 k+1}}\right\}^{1 / 2} } \\
& +\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{2}\right)}\left\{\int_{\mathbb{R}^{n} \backslash 2^{j-2} Q} \int_{2^{j-3} l(Q)}^{\infty} \cdots\right\}^{1 / 2} \equiv \mathrm{~V}_{j, 1}+\mathrm{V}_{j, 2}
\end{aligned}
$$

Similarly to the estimate of $\mathrm{J}_{j}$, we have

$$
\begin{equation*}
\mathrm{V}_{j, 2} \lesssim 2^{-\alpha j} \tag{6.17}
\end{equation*}
$$

To estimate $\mathrm{V}_{j, 1}$, let $\beta \in(2 k+2 n(1 / r-1 / 2), \infty)$. By Lemma 6.2, (6.11) and (6.15), there exists a positive constant $\widetilde{C}$ such that

$$
\mathrm{V}_{j, 1} \lesssim\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{2}\right)}
$$

$$
\begin{aligned}
& \times\left[\int_{0}^{2^{j-3} l(Q)} t^{2 n\left(\frac{1}{2}-\frac{1}{r}\right)} \exp \left\{-\widetilde{C} \frac{\left[2^{j} l(Q)\right]^{2 k /(2 k-1)}}{t^{2 k /(2 k-1)}}\right\} \frac{d t}{t^{2 k+1}}\right]^{1 / 2}\|b\|_{L^{r}(Q)}, \\
\lesssim & {\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{2}\right)}\left[\int_{0}^{2^{j-3} l(Q)} t^{2 n\left(\frac{1}{2}-\frac{1}{r}\right)}\left[\frac{t}{2^{j} l(Q)}\right]^{\beta} \frac{d t}{t^{2 k+1}}\right]^{1 / 2}\left\|\nabla^{k} b\right\|_{L^{\frac{r n}{n+k r}(Q)}} } \\
\lesssim & {\left[2^{j} l(Q)\right]^{n\left(\frac{1}{p}-\frac{1}{2}\right)}\left[\frac{1}{\left[2^{j} l(Q)\right]^{\beta}} \int_{0}^{2^{j-3} l(Q)} t^{2 n\left(\frac{1}{2}-\frac{1}{r}\right)+\beta-2 k-1} d t\right]^{1 / 2} } \\
& \times\left\|\nabla^{k} b\right\|_{L^{2}(Q)}|l(Q)|^{n+k r} r-\frac{n}{2} \\
& 2^{j\left[n\left(\frac{1}{p}-\frac{1}{r}\right)-k\right]}
\end{aligned}
$$

which together with (6.17) shows that

$$
\sum_{j=3}^{\infty}\left(\mathrm{V}_{j}\right)^{p}=\sum_{j=3}^{\infty} 2^{-\alpha j p} \lesssim 1
$$

This, combined (6.14) and (6.16), implies (6.13), which completes the proof of Proposition 6.1.

Combining Theorem 6.1 and Proposition 6.1, we obtain the following Riesz transform characterization of $H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)$. We point out that Theorem 6.2 when $k=1$ is just the Riesz transform characterization of $H_{-\operatorname{div}(\mathrm{AD})}^{p}\left(\mathbb{R}^{n}\right)$ for $p \in(0,1]$, which is exactly [32, Theorem $5.2]$ in the case that $p \in(0,1]$.

Theorem 6.2. Let $k \in \mathbb{N}, L_{1}$ be the $2 k$ order divergence form homogeneous elliptic operator and $r \in(1,2]$ such that $r n /(n+k r) \leq 1$ and the semigroup $\left\{e^{-t L_{1}}\right\}_{t>0}$ satisfies the $L^{r}-L^{2} k$-off-diagonal estimates. Then for all $p \in(r n /(n+k r), 1]$,

$$
H_{L_{1}}^{p}\left(\mathbb{R}^{n}\right)=H_{\mathrm{Riesz}, L_{1}}^{p}\left(\mathbb{R}^{n}\right)
$$

with equivalent norms.
Remark 6.1. We point out that a key fact used in the proof of Proposition 6.1 (and hence Theorem 6.2) is $\left\|\sqrt{L_{1}} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)}$, which comes from [6, Theorem 1.1]. This inequality for $L_{2}$ is equivalent to the following inequality that for all $f \in \dot{W}^{k, 2}\left(\mathbb{R}^{n}\right)$,

$$
\left\|V^{k / 2} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)} \lesssim\left\|\nabla^{k} f\right\|_{L^{2}\left(\mathbb{R}^{n}\right)},
$$

which seems impossible even when $V \equiv 1$. Thus, the method used in the proof of Proposition 6.1 seems unsuitable for obtaining a counterpart of Proposition 6.1 for $L_{2}$.

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Jun Cao and Dachun Yang (Corresponding author):
School of Mathematical Sciences, Beijing Normal University, Laboratory of Mathematics and Complex Systems, Ministry of Education, Beijing 100875, People's Republic of China E-mails: caojun1860@mail.bnu.edu.cn (J. Cao) and dcyang@bnu.edu.cn (D. Yang)


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    *Corresponding author

