# Ultrafast pump-probe detection of X-ray induced transient optical reflectivity changes in GaAs

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Abstract: The optical index modulation was theoretically estimated and demonstrated under short X-ray excitation in low-temperature-grown GaAs (LT\_GaAs). Hot-electron thermalization time <1 ps, carrier recombination time <2 ps and the duration of the index perturbation was determined by the carrier recombination time which was of order -2 ps in LT\_GaAs with a high density of recombination defects. Predictions of radiation –induced changed in the optical refractive index were in reasonably good agreement with the limited experimental data available, suggesting that LT\_GaAs was a highly promising material for high speed single transient ionizing radiation detector.

Key words: low-temperature-grown GaAs; X-ray detectors; index perturbation; picoseconds temporal resolution

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# 超快泵探针对 GaAs 中 X 射线诱导的 瞬态光学反射率变化的探测

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摘 要:理论估算并实验验证了在 X 射线脉冲激发下低温砷化镓的光学折射率调制特性。泵浦-探 针实验表明,低温砷化镓中存在的高密度复合缺陷大大减小了载流子寿命,使超热电子的弛豫时间小 于 1×10<sup>-12</sup> s,载流子的复合时间小于 2×10<sup>-12</sup> s,折射率的扰动时间约为 2×10<sup>-12</sup> s。通过理论分析,给出 了自由载流子和俄歇效应对该弛豫过程的定量估算,与实验结果吻合较好。该研究表明低温生长砷化 镓是一种有效的可用于单次瞬态皮秒时间分辨 X 射线探测的材料。

关键词:低温生长砷化镓; X射线探测器; 折射率扰动; 皮秒时间分辨

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## **0** Introduction

Measuring X –ray pulse temporal history, including pulse shape and relative timing with optical lasers with tens of picoseconds temporal resolution is a challenging problem, particularly when from a single event <sup>[1–3]</sup>. Conventional X– ray detectors collect the charge generated by the interaction of the incident radiation with the sensor medium and generate voltages or currents that are transported, usually via signal cables, to remote recorders. The degradation of the high frequency signals components during signal generation and transmission limits the temporal response of state –of–the– artelectronic detection systems<sup>[4–5]</sup>.

A new class of sensor, called rad optic sensors, has previously reported over coming these limitations by using light to detect X-ray radiation ionization in place <sup>[6]</sup>. K. L. Baker et al. made use of a change in 805 nm optical probe beam in CdSe ionized by an X-ray radiation <sup>[7]</sup>, while Gahl et al. measured the change in optical reflectivity from GaAs caused by 40 eV photons from an X-ray free– electron laser <sup>[8]</sup>, but these early approaches were lacking a well –understood physics framework, and especially lacking the material with more faster recombination time.

It's interested in low –temperature –grown GaAs (LT\_GaAs), and in particular how the X –ray photo – absorption process stimulates a range of many –body responses that can be probed by optical transmission near the band gap. It was successfully demonstrated in this work that a significant negative index shift was associated with the absorption of X–rays, and the work was began to develop detectors. This will provide a new approach for X –ray ultrafast detectors for synchrotron and inertial confinement fusion research.

#### 1 Method

The experiments conducted to test this X –ray detector were carried out on the ultra–short pulse laser facility using the arrangement illustrated in Fig.1. An 800 nm Ti:Sapph oscillator was regenerative amplified at 10 Hz to yield pulses with -2 mJ energy and -200 fs duration. Of this beam, through

multi amplify; -200 mJ was focused on a 20  $\mu$ m thick pure Cu foil located at the center of a vacuum chamber. This produced an X –ray source dominated by the Copper K –alpha. The remaining oscillator energy pumped an optical parametric amplifier (OPA) to generate a 1900 nm idler that was then sent through a second harmonic generation (SHG) crystal, yielding a 950 nm probe beam. The initial pulse width was < 100 fs, corresponding to a spectral width of roughly 25 nm FWHM. This was filtered to obtain a 6 ps transform–limited impulse and then relay into the GaAs(or LT\_GaAs) surface.



Fig.1 Transient X –ray induced optical reflectivity change  $(\Delta R/R)$  measurement schematic

The sample was positioned 20 mm away from focus at  $45^{\circ}$  from target normal. The details of experiment are shown in the Fig.1. The GaAs sample were cut from a  $350 \,\mu$ m semi –insulating GaAs wafer and mounted on sapphire substrates using a thin crystal bond adhesive. The LT – GaAs semiconductor samples were grown by MBE at a temperature of 200 °C and annealed at 600 °C. X –ray pulses were synchronized with probe beams from an optical parametric amplifier system.

#### 2 Results

The reflectivity changes of  $\Delta R/R$  which is induced by the X –ray pulse was successfully observed, occurring on time scales from femtoseconds to many hundreds of picoseconds. Fig.2 shows this evolution for different delay ranges.

As shown in Fig.2(a) the carrier curve cannot return to the same status as it is before excited. The reason is as follows: the defect states of the intrinsic GaAs is very little, thus, the carrier cannot recombine immediately through the defect. As a result, the carrier is trapped in the higher energy level. As a result of these problems with the GaAs,



Fig.2 Temporal characteristics of the transient X-ray induced optical reflectivity change ( $\Delta R/R$ ) on different delay time

the bulk LT–GaAs is considered as an alternative material system. In this case, the time response is greatly improved over the intrinsic GaAs sample and occurs in < 1 ps as given by the rise time of the bleaching signal (positive feature). Carrier trapping is also faster and occurs in 2-3 ps, as given by the decay of the bleaching signal. These operating parameters for the LT–GaAs system are very promising and may be further improved through additional materials engineering and processing, if needed.

The results clearly demonstrate there are two principal manifestations of the carrier dynamics: bleaching and trap absorption (mediated by Arsenic precipitates). The relaxation of the X-ray excited carriers to the bottom of the conduction band is given by the rise time of the bleaching signaland occurs in  $\sim$ 1 ps. The trapping is given by the decay of the bleaching signal and occurs in 2–3 ps. This is different from intrinsic GaAs. The sample grown at low temperature has defects. Thus, the capture effect of the defect state plays an important role in the recombination of the carriers, which helps the recovery time is shortened to 2–3 ps.

## **3** Discussions

In this section, the underlying physical mechanisms behind these results is discussed detailedly. The time evolution of the induced absorption effect, namely the index change,  $\triangle R$ , can be divided into two stages: bleaching and trapping <sup>[9]</sup>. Bleaching results from the radiation excitation of carriers into the excited states of the conduction band and decays through the relaxation of excited carriers to the bottom of the conduction band.

Before X -rays arrive, the ideal semiconductor density of states at the direct band gap is assumed to have an empty conduction band and a fully occupied valence band. In the X-ray excitation case, for energies below a few MeV, the dominant X-ray absorption mechanism is photoelectron generation, where a single very hot photoelectron is produced and left a deep core hole. This single hot carrier interacts with the material on relatively short distance scales, ultimately producing a cascade of lower energy secondary carriers along the track of the primary photoelectron. The deep core hole is filled by a higher-level core electron, mostly accompanied by the emission of an Auger electron. This replaces a deep core hole with two higher ones, and the Auger electron in elastically scatters many more carriers to the conduction and valence bands. This is repeated with higher core levels, creating multiple localized holes that eventually cascade up to become mobile valence band holes [10-11]. A simplified model for describing this result is shown in Fig.3. Transit X -ray excitation fills the states at the bottom of the conduction band, blocking absorption of photons with energies just above the band gap energy. Induced absorption for photons with energy just below the band gap could arise from transient localized states at the band edges, as seen with doped semiconductors.

Since the number of pairs large and the e -h pair volume is very small, the concentration of free carriers, N, produced by a single X-ray photon can be large enough to locally cause complete bleaching of the excision absorption in a very small volume along the track of the primary phot oelectron. Trap absorption results from the optical excitation of these traps (As precipitates) into the conduction band. Both processes contribute to the change in the index of refraction (nonlinear index).

It supposed most of the incident X-rays were absorbed in the LT\_GaAs, the density of X -ray absorption sites created by each X -ray pump pulse is  $\sim 10^{17}$  cm<sup>-3</sup>. The



Fig.3 Radiation absorption creates electron-hole pairs that modulate the absorption spectrum

multiple shallower holes from subsequent Auger processes may increase this by another order of magnitude. It suppose the cumulative carriers density created by each X-ray pump pulse is  $\sim 10^{20}$  cm<sup>-3</sup>. In transport studies, carrier concentration levels of  $\sim 10^{18}$  cm<sup>-3</sup> and above have caused observable optical refractive changes as large as  $10^{-2}$  near the band gap <sup>[12]</sup>.

#### 4 Conclusions

In conclusion, X-ray pulse induced transient optical reflectivity changes was established in GaAs as a powerful tool for high –speed X –ray detection. These properties make low–temperature–grown GaAs a highly promising material for radiation detections devices. Future work will focus on developing faster devices, characterizing both the rise and fall times, the development of imaging array technology. It also note that this same effect can be used to detect any particle that produces electron –hole pairs in semiconductors, thus opens a new field of time resolved studies of radiation detectors for various radiation particles.

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