拉曼散射在共振及近共振时的条件：广义短时间近似

Raman Scattering at Resonant or Near-Resonant Conditions: A Generalized Short-Time Approximation

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中文关键词 共振拉曼 散射持续时间 已三烯 短时间近似

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中文摘要

研究了频率失谐时共振拉曼散射的动力学过程．当入射光子能量远离共振吸收能量时，时域内的失相使散射过程变快．这使得频率失谐如同照相机的快门功能，具有规律的散射持续时间，为普通的稳态测量提供了控制散射时间的有效工具．基于这个理论对两个多模式模型系统以及反式-1,3,5-己三烯和鸟嘌呤-胞嘧啶Watson-Crick碱基对分子的共振拉曼光谱进行了研究．除了这些特殊的物理效应，快散射机制可以简化光谱，同时使散射理论得到简化．当入射光子频率在共振区域时，拉曼光谱中会出现较强的多倍频成分；当入射光子频率与第一共振吸收频率之间的失谐量为振动能量时，在快散射过程中，这些多倍频成分逐渐消失．因此，利用入射光子与共振频域的失谐可以明显地简化拉曼光谱，从复杂光谱中去除多倍频和软模的影响，并且可以避免共振态的解离和荧光衰减引起的干扰．

英文摘要

We investigate the dynamics of resonant Raman scattering in the course of the frequency de-tuning. The dephasing in the time domain makes the scattering fast when the photon energy is tuned from the absorption resonance. This makes frequency detuning to act as a cam-era shutter with a regulated scattering duration and provides a practical tool of controlling the scattering time in ordinary stationary measurements. The theory is applied to resonant Raman spectra of a couple of few-mode model systems and to trans-1,3,5-hexatriene and guanine-cytosine (G-C) Watson-Crick base pairs (DNA) molecules. Besides some particular physical effects, the regime of fast scattering leads to a simplification of the spectrum as well as to the scattering theory itself. Strong overtones appear in the Raman spectra when the photon frequency is tuned in the resonant region, while in the mode of fast scattering, the overtones are gradually quenched when the photon frequency is tuned more than one vibra-tional quantum below the first absorption resonance. The detuning from the resonant region thus leads to a strong purification of the Raman spectrum from the contamination by higher overtones and soft modes and purifies the spectrum also in terms of avoidance of dissociation and interfering fluorescence decay of the resonant state. This makes frequency detuning a very useful practical tool in the analysis of the resonant Raman spectra of complex systems and considerably improves the prospects for using the Raman effect for detection of foreign substances at ultra-low concentrations.

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