Multi-phonon resonant and nonresonant scattering of electrons in doped semiconductors with diamond-type-lattices

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Abstract: Time-resolved spectroscopies acquired rates of electron-phonon scattering of helium-like states of Mg double donor centers in silicon. The fastest rates of intracenter dephasing and depopulation correlate spectrally to the two-phonon branches of silicon.

Electronic scattering in solids accompanied by emission and absorption of lattice phonons belong to the class of ultrafast processes in solids due to inherent short lifetimes of lattice phonons, as a rule between hundreds femtosecond and tens picosecond at ambient temperatures. Electron-phonon (e-ph) interactions [1] dominate electron scattering rates in bulk semiconducting crystals (Fig. 1) when other fast mechanisms, such as electron-electron (e-e) and electron-defect interactions remain at a moderate level, that is a common case for low to moderate densities of defects in crystalline lattices.

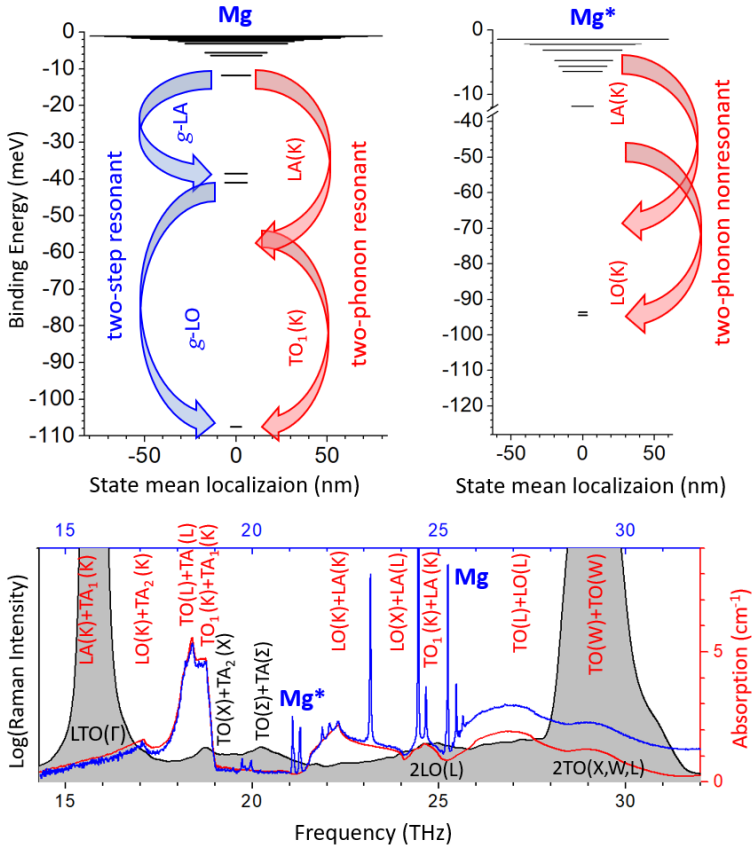

Fig. 1. Multi-phonon scattering of electrons bound to excited states of atomic like impurity spectra of a semiconductor: case of Si doped by a He-like Mg donor. Upper plot shows different types of impurity-phonon resonances on the energy spectra diagram of neutral Mg center and Mg* molecular complex. Low plot shows: (filled grey) Raman spectrum of Si, representing phononic density of states for the spectral range of sum (one to three) phonons; (red) infrared absorption spectrum representing combination of infrared-active phonons; (blue) absorption spectrum of Si:Mg sample. Note different correlation of Mg and Mg* transitions (discrete lines) to the Raman-active [2] and Infrared-active [3] combinations of principal phonons.

The one-phonon (e-1ph) interactions (such as in Fig. 1, two-step) are the strongest process and usually sufficient for quantitative description of electrical transport or nonequilibrium dynamics of free electrons [1]. The higher order, two-phonon (e-2ph) and higher (e-Nph), interactions are generally several orders of magnitude weaker processes for both bulk (3D) and 2D-solids (graphene, etc.) [2-4]. As a rule, a typical e-1ph process implies a short-range interaction that fails already for polar semiconductors. Semiconductors with a diamond-type (nonpolar) lattice exhibit no e-1ph interactions with optical lattice phonons; instead, e-Nph scattering with high-energy phonons at the critical points of the Brillouin zone dominate. An e-2ph scattering considers a two-step process via an intermediate state, with the probability inversely proportional to its off-shell energy extents, expressed as resonances of the e-energy steps to the ph-energies [1], and becomes non-vanishing only at that resonances. Such resonances are generally missing in discrete, atomic-like spectra of optically active impurity centers in semiconductors (Fig. 1, red arrows) and this e-ph scattering is considered to be strongly damped. For silicon (Si), doped by hydrogen-like donors with interstate energies in the one-phonon spectrum (< 15 THz), acoustic phonons serve the low-energy e-1ph process: the e-1ph rates correlate with the Si phonon density spectrum [5] while intervalley phonons enhance additionally the e-1ph rates for the large-energy processes for donors [6]. Deeper centers, such as helium-like donors, possessing interstate gaps of the energies in 2ph- and 3ph- ranges (15-30 THz, Fig. 1), can exhibit solely multi-phonon / combination scattering.

Several time-resolved techniques have been used for determination of e-ph scattering rates in Si doped by H-like and He-like centers at lattice temperatures below 5K. Decay rates of excited atomic-like states (T_1^{-1}) were accessed by a pump-probe and transient grating techniques, while dephasing time (T_2) was acquired by a photon echo measurement [7].

We observed clear, systematic dependences of scattering rates to the impurity-intervalley-phonon resonances in the e-1ph schemes, with typical T_1 values for different e-transitions < 100 ps. For nonresonant e-transitions, where large energy atomic gaps correlate only to the difference-phonon spectrum, T_1 is lower, commonly an order of magnitude. Once the spectrum of the e-2ph scattering process correlates to the two-phonon absorption spectrum of a semiconductor lattice (that means a combination-phonon spectrum of infrared-active phonons, see the Mg center transitions in Fig. 1), the e-2ph rates are enhanced significantly, the corresponding T_1 values for different e-transitions of Mg < 30 ps. The only to date elaborated theoretical formalism for impurity-2ph scattering, based on adiabatic approximation [8], returns the T_1^{-1} rates up to 10^{12} s^{-1}, in the same order of the observed experimentally in boron doped diamond [9]. This work also points on the resonant enhancement (factor > 10^4) of scattering probability for the overtones of principle phonons at the boundaries of the Brillouin zone. The T_1 and T_2 times for different e-transitions of a Mg* donor complex, lying off the two-phonon absorption in Si (Fig. 1), have been determined to be about twice longer that those for a neutral Mg center.

Our experimental results indicate overall high probability of two-phonon scattering processes accompanying intracenter transitions in semiconductors with diamond-type lattices.

References

[1] N. E. Lee, J. J. Zhou, H. Y. Chen, et al., “Multiphonon diffuse scattering in solids from first principles: Application to layered crystals and two-dimensional materials,” Nature Commun. **11,** 1607 (2020).

[2] C. S. Wang, J. M. Chen, R. Becker, A. Zdetsis, “Second order Raman spectrum and phonon density of states of silicon,” Phys. Lett. A **44,** 517 (1973).

[3] D. Franta, D. Nečas, L. Zajíčková, and I. Ohlídal, “Dispersion model of two-phonon absorption: application to c-Si,” Opt. Mater. Express **8,** 1641 (2014).

[4] M. Zacharias, H. Seiler, F. Caruso et al., “Ab initio electron-two-phonon scattering in GaAs from next-to-leading order perturbation theory,” Phys. Rev. B **104,** 205109 (2021).

[5] N. Q. Vinh, B. Redlich, A. F. G. van der Meer, et al., “Time-Resolved Dynamics of Shallow Acceptor Transitions in Silicon,” Phys. Rev. X **3,** 011019 (2013).

[6] V. V. Tsyplenkov, E. V. Demidov, K. A. Kovalevsky, and V. N. Shastin, "Relaxation of Excited Donor States in Silicon with Emission of Intervalley Phonons," Semicond. **42**, 1016-1022 (2008).

[7] N. Dessmann, S. G. Pavlov, A. Pohl et al., “Combination of ultrafast time-resolved spectroscopy techniques for the analysis of electron dynamics of heliumlike impurity centers in silicon,” Phys. Rev. B **106,** 195205 (2022).

[8] N. A: Bekin, “On the Two-Phonon Relaxation of Excited States of Boron Acceptors in Diamond,” Semicond. **53,** 1340-1347 (2019).

[9] S. G. Pavlov, S. A. Tarelkin, V. S. Bormashov et al., “Dynamics of infrared excitations in boron doped diamond,” Diamond & Related Materials **92**, 259–265 (2019).