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Resonance Raman scattering caused by non-adiabatic transitions

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Excited states in molecules and impurity centers in crystals with high symmetry are usually degenerate. After optical excitation, due to Jahn-Teller effect the symmetry is usually lowered. The initial stage of this process which occurs in the vicinity of the symmetric configuration is essentially determined by the nonadiabaticity of the electronic and nuclear motion. To study this effect, the optical probing, such as measurements of absorption and resonance Raman scattering (RRS) spectra may be used. Raman scattering can give us much more information about vibrational modes (by changing the polarization of the incident light) than absorption or at least it can be used as a complementary method for studing these systems.

The purpose of this communication is to explore the effects of strong non-adiabaticity of the JT dynamic in the RRS excitation profiles of impurity centers of solids. The cases of two-fold degenerate or quasi-degenerate excited electronic states are considered. Our study is based on the recently proposed method [1] of calculation of optical transitions in centers with the Jahn-Teller and pseudo-Jahn-Teller effect in the final state. The calculations of Raman excitation profiles (REPs) are fully quantum mechanical. The results are compared with that of the semiclassical theory of REPs of systems with JT effect in the excited state. Notable differences in the results of the purely quantum mechanical theory presented here and the semiclassical theory are demonstrated. The most notable result is the manifestations of different life times of states in the shapes of the spectral lines that arise from different interaction with phonons. Such a difference is observed also in the region of vibronic parameters, usually believed to belong to the area of applicability of the semiclassical theory.

1. Pae, K.; Vaikjärv, T.; Hizhnyakov, V. (2010). Vibronic transitions to a state with Jahn-Teller effect: contribution of phonon continuum. *In: Program & Abstracts: 20th International Symposium on the Jahn-Teller Effect, University of Fribourg, 16-20 Aug 2010. (Toim.) Atanasov, M.*. Fribourg: University of Fribourg, 2010, 77 - 77.