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Vibrational Relaxation in Solid SO₂

L. Angeloni, E. Castellucci, R. Righini, P. R. Salvi
Dipartimento di Chimica, Università di Firenze, FIRENZE, Italy

Vibrational relaxation and depnasing of crystal phonons and vibrons have been the object of considerable interest in recent years [1]. The temperature dependence of vibrational bandwidths can be regarded as a critical step for testing current theories on anharmonicity in molecular crystals.

Recently we have reported on picosecond time resolved experiments in simple molecular ionic crystals and given novel informations as to the vibrational decay processes in these systems [2,3].

In most of the cases so far studied the most efficient decay route was found to be through depopulation of the vibrational excitons into the lattice phonon manifold involving a third order anharmonic coupling.

The study of the phonon relaxation dynamics in simple molecular crystals such as SO₂ may, on the other hand, offer further arguments of discussion as to the strength of the anharmonic coupling coefficients and the most probable relaxation pathways of the vibrational excitations.

In fact, being the molecule characterized by three internal coordinates and by intermolecular interactions weaker than in the previous cases, it is easier to assess the role of intramolecular coupling and depnasing in the decay processes.

We have measured the vibrational decay time of the ν_1 vibrational exciton of solid SO₂ by means of picosecond CARS in the temperature range of 10 K to the melting point (196 K). Preliminary results point out the importance of depnasing processes involving the low frequency ν_2 vibration in determining the ν_1 linewidth dependence on temperature. Further time domain experiments on ν_2 as well as high resolution Raman measurements which are in due course will help clarifying better the relaxation mechanisms in solid SO₂.

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