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COHERENCE DECAY TIMES OF PHONONS IN MOLECULAR-IONIC CRYSTALS MEASURED BY TIME-RESOLVED CARS.

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A CARS spectrometer for the study of the vibrational dephasing processes in molecular crystals has been set up.

Decay times as short as few picoseconds can be obtained by deconvolution with an instrumental function whose width is less than 10 ps.

The coherence decay times of the internal and external phonons of a K_2SO_4 oriented single crystal have been measured from 10 K to room temperature. The phonons measured so far all belong to the totally symmetric irreducible representation of the crystal space group. The 1145 and 1087 cm^{-1} modes do not show any appreciable asymmetry of the decay profile: even at 10 K the decay time is shorter than the instrumental resolution. A decay of the 617 cm^{-1} mode can be detected only at low temperature, with a lifetime of about 15 ps at 10 K. The vibration at 983 cm^{-1} instead does show a remarkable lengthening of the coherence decay time at low temperature. The measured T_2 ranges from ~300 ps at 10 K to ~20 ps at 100 K.

The different behaviour of the modes considered is consistent with an energy decay mechanism, involving three- and multi-phonon processes, and can be put into relation with the density of phonon states. The 983 cm^{-1} vibrational level is in fact rather isolated, and only decay processes involving a large number of phonons can contribute to its lifetime at very low temperature; in contrast, the phonons in the ~600 and ~1100 cm^{-1} regions have several close by levels accessible by three-phonon scattering events involving internal and external phonons.