ULTRAFAST WAVE PACKET DYNAMICS IN JAHN-TELLER SURFACES: Ag ATOMS IN Xe CRYSTALS

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The dynamics on Jahn-Teller surfaces involving pseudorotation, radial oscillations and the symmetric breathing mode has been already explored in isolated triatomics like Na_3. This contribution deals with the additional effects occuring in the condensed phase and originating from the energy dissipation to the lattice by phonon emission. A study with femtosecond pump-probe spectroscopy is carried out for substitutional Ag atoms in a rare gas lattice (Xe) of cubic (fcc) symmetry. Excitation from the electronic ground state with s-symmetry to the p-orbitals of the Ag atom leads to a dynamic Jahn-Teller splitting which removes the degeneracy of the p-orbitals by a coupling to nontotally symmetric phonon modes. Energy dissipation of the initially pseudorotating wave packets leads to structural relaxation and to a thermalized population in a strongly statically deformed geometry according to the large Stokes shift of about 2 eV. The initial population in the Jahn-Teller state is probed by a transient bleaching of the ground state resulting in a life time of several ps. The observed subpicosecond dynamics is related to the cubic and noncubic modes derived from MCD experiments. The subsequent energy dissipation of about 0.4 eV corresponding to 80 phonons is detected by fluorescence dip spectroscopy. The static deformation and the thermalization in it is completed in 5 - 7 ps. This very fast energy relaxation cannot be attributed to single phonon emission events for a phonon period of about 1 ps and is discussed with respect to higher order terms in Jahn-Teller surfaces.