Molecules in condensed phase lose their coherences due to the interaction with the surrounding. Simple systems like a diatomic molecule in a solid rare gas allow us to investigate vibrational and electronic decoherences. Here, we will present the coherent dynamics of a Cl2 in solid Ar investigated by using a fs-pump-probe technique.

The wavepacket focusing can be achieved with a negatively chirped pump pulse to compensate the dispersion of the wavepacket in anharmonic potentials. By applying the different negatively chirped pulses, the real decoherence by the matrix can be distinguished from the dispersion. Due to the anharmonicity of potentials, the wavepacket is dispersed and then fully or fractionally revives if the vibrational coherence is retained during the rephasing time. We observed the quarter and half revivals with a nonchirped pump pulse and determined the decoherence time for the vibrational wavepacket. In addition to the vibrational coherence of molecules, the coherence of the lattice vibration of solid Ar will be discussed.

The electronic coherence between the electronic ground state and the excited B state of Cl2 in solid Ar was studied by using a phase locked pulse pair (PLPP) as a pump pulse. Depending on the relative phase of the PLPP, more or less population can be transferred to the B state after every vibrational period if molecules keep the phase information on the PLPP. We observed vibrational recurrence around 220 fs in the LIF interferogram using a PLPP at 520 nm. The contribution of zero phonon line and phonon side band to the recurrence will be discussed.