Optical Response in Molecular Systems With Fast and Slow Delayed Dissipation

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A theoretical and computational treatment of systems including delayed dissipation has been done in terms of a reduced density matrix satisfying a generalized Liouville-von Neumann equation.[1] The delayed dissipation leads to a time integral with a memory supermatrix term derived from the time-correlation of atomic displacements in the medium.

Thermal equilibrium in this case is only reached when the system has attained a Boltzmann distribution, and has been there long enough for the correlations to subside, according to a memory time which depends on the decay of the correlation function. When thermal equilibrium is a starting condition, these previous correlations must still be considered.

We consider cases where the time correlation function of the medium decays exponentially (fast) or as an inverse power (slow). The slow case is solved with a general procedure by calculating the difference between solutions with and without excitation by light pulses.

We present examples for the slow delayed relaxation of CO/Cu(001) and the fast delayed relaxation of Ag_3/Si(111)

1. A. S. Leathers and D. A. Micha, J. Phys. Chem. **110**, 749 (2006)

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