Real-time propagations for correlated systems in Fock space

Johannes Flick\(^1\)(flick.fhbo-berlin.mp.d)\(^1\), Heiko Appel\(^1\), Angel Rubio\(^1,2\)

\(^1\) Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany
\(^2\) NanoBio Spectroscopy group and ETSF, Universidad del País Vasco, San Sebastián, Spain

(i) Second-order coupling Hamiltonians and time-resolved photoemission spectroscopy

The Born-Oppenheimer approximation is ubiquitous in most studies of electronically excited systems. However, a few systematic studies indicate that the electromagnetic coupling to the solvent plays a significant role, which can e.g. influence the transition dynamics. For these cases, matrix elements of the correlation Hamiltonians have been developed, which can be used to calculate the effect of the solvent on the transition dynamics. In this work, we perform exact diagonalizations and real-time propagations for the model in Fock space. We calculate the effect of the nonlinear coupling on the ground-state photoemission and time-resolved photoemission spectroscopy.

Time-resolved photoemission spectroscopy

To include second-order virtual displacements effects in the electron-phonon coupling, we add the following term to the Hamiltonian:

\[
H_{ph} = \sum_{\alpha, \beta} \frac{1}{2} \sum_{\lambda, \lambda'} \left( \langle \alpha | \phi \rangle \langle \beta | \phi \rangle \right) \cdot \hat{X}_{\lambda} \cdot \hat{X}_{\lambda'}
\]

where \(\hat{X}_{\lambda}\) are the phonon operators.

Time-resolved photoemission spectroscopy

To follow the photoemission spectroscopy dynamics, we propagate the initial state as an initial state and calculate the PES spectra at different times.

(ii) Spontaneous emission in cavity QED

In this work, we study the dynamics of correlated photoelectron-emissions. Our model system consists of an atom placed in an optical cavity. Axial and quantized electromagnetic modes are coupled to the atom-coupling pair.

Exact dynamics in cavity QED for one-photon and two-photon processes

With quantized electric field:

\[
\hat{E}(\omega) = \sum_{\omega, \lambda, \lambda'} \left( \langle \phi_{\alpha} | \phi_{\beta} \rangle \right) \cdot \hat{X}_{\lambda} \cdot \hat{X}_{\lambda'}
\]

where \(\hat{X}_{\lambda}\) are the electric field operators.

Projected density matrix:

\[
\rho_{\alpha, \beta} = \langle \phi_{\alpha} | \phi_{\beta} \rangle
\]

Photon energy density:

\[
\epsilon(\omega) = \langle \phi_{\alpha} | \hat{H}_0 | \phi_{\beta} \rangle
\]

Conclusions

The model shows that the spontaneous emission rate is real-time dependent. This is important for understanding the role of the cavity in the dynamics of the transition matrix element involving the correlated emission of two photons.

References