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New Scales: Properties of Nanostructures in the Femtosecond Regime

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INTRODUCTION

Many fundamental processes in matter like electron–electron or electron–phonon scattering in solids, occur on a time scale ranging from a few tenth of a femtosecond (1 fs = 10^{-15} s) to a picosecond (1 ps = 10^{-12} s). These ultrafast phenomena are experimentally accessed by monitoring the interaction of ultrashort light pulses with a given sample. Among the experimental techniques available nowadays, the so–called pump–probe methods are most frequently used. In such a pump–probe experiment one needs two ultrafast laser pulses: a pump pulse, which excites the investigated system and a probe pulse, delayed in time, which explores the relaxation of the excited system.

Based on the superposition principle of electromagnetic fields, within the Slowly Varying Envelope Approximation (SVEA) the total external electric field in a pump–probe experiment is given by

$$\begin{aligned}\vec{E}(\vec{r},t) &= \vec{E}_{\rm pu}(\vec{r},t) + \vec{E}_{\rm pr}(\vec{r},t) \qquad (1)\\ &= \vec{e}_{\rm pu}\mathcal{E}_{\rm pu,0}\,\tilde{\mathcal{E}}_{\rm pu}(t)\exp\left[i\left(\vec{q}_{\rm pu}\,\vec{r}-\omega_{\rm pu}t\right)\right]\\ &+ \vec{e}_{\rm pr}\,\mathcal{E}_{\rm pr,0}\tilde{\mathcal{E}}_{\rm pr}(t-\tau)\exp\left[i\left(\vec{q}_{\rm pr}\vec{r}-\omega_{\rm pr}t\right)\right] \ ,\end{aligned}$$

where $\tilde{\mathcal{E}}_{\rm pu}(t)$ and $\tilde{\mathcal{E}}_{\rm pr}(t)$ are the time–dependent envelopes of the pump and the probe pulse propagating in the directions $\vec{q}_{\rm pu}$ and $\vec{q}_{\rm pr}$ with the carrier frequencies $\omega_{\rm pu}$ and $\omega_{\rm pr}$ and having the polarizations $\vec{e}_{\rm pu}$ and $\vec{e}_{\rm pr}$. In considering an external electric field as defined in Eq. (1), it is assumed that no significant overlap in time exists between the pump and probe pulse, i.e., all coherence coupling effects can be neglected.

GENERALIZED KUBO THEORY

Because in Eq. (1) a possible overlap between the pump and probe pulse is neglected, it can be considered that for $t > \tau$ the probe pulse interacts only with the pump–excited system. The problem to be solved reduces therefore in finding the dynamic linear response of the pump–excited system with respect to the probe pulse at times $t > \tau$. If within the interaction picture the only source of perturbation is the probe pulse then from the linearization of the density,

$$\rho_{\rm pr}(t) \simeq \rho_{\rm pu} + \frac{i}{\hbar}\int_{\tau}^{t} dt' \,\left[\vec{r}(t'),\rho_{\rm pu}\right]\vec{E}_{\rm pr}(t') \ , \quad (2)$$

where the density for the pump–excited state is written

$$\begin{aligned}\rho_{\rm pu} &\simeq \rho_0 + \frac{i}{\hbar}\sum_{\kappa}\int_{-\infty}^{\tau} dt\,\left[\vec{r}(t),\rho_0\right]\vec{E}_{\rm pu}(t)\\ &\equiv \rho_{\rm pu}^{(0)} + \rho_{\rm pu}^{(1)} \ .\end{aligned}$$

Accordingly the frequency–frequency representation of the generalized, strictly linear conductivity for the pump–probe experiments is given by [1]

$$\begin{aligned}\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau) &= \frac{1}{2\pi}\exp\left[+i\left(\omega-\omega_{\rm pr}\right)\tau\right]\\ &\times\mathcal{L}\left[\tilde{\mathcal{E}}_{\rm pr}(t)\right]\Big|_{i(\omega_{\rm pr}-\omega)}\,\tilde{\sigma}_{\mu\nu}(\omega) \ , \qquad (3)\end{aligned}$$

where in terms of ω this quantity is now resolved with respect to the spectral components of the probe pulse. The first order conductivity, on the other hand, is resolved with respect to the spectral components of the pump pulse and hence is written

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as [1]

$$\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau) = \frac{\beta}{2\pi}\exp\left[+i\left(\omega-\omega_{\rm pu}\right)\tau\right] \times \sum_{\kappa}\mathcal{E}_{{\rm pu},0\kappa}\int_0^\infty dt\ \exp\left(+i\omega t\right) \int_0^t d\xi\,\tilde{\mathcal{E}}_{\rm pr}(t-\xi)\exp\left(+i\omega_{\rm pr}\xi\right) \int_0^\infty d\xi'\ \left\langle J_\mu(t);\ J_\nu(t-\xi);\ J_\kappa(-\xi')\right\rangle \times\tilde{\mathcal{E}}_{\rm pu}(\tau-\xi')\exp\left(+i\omega_{\rm pu}\xi'\right)\ . \tag{4}$$

Although for this form one can also take advantage on the properties of Laplace transforms, for $\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ a similar expression to that in Eq. (3) cannot be deduced. The total and formally linear dynamic conductivity is finally obtained by combining the zeroth and first order conductivities as given by Eqs. (3) and (4),

$$\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau) = \tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau) + \tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)\ .$$

SUMMARY

By linearizing the density of both the pump– and probe–excited states and neglecting the overlap between femtosecond laser pulses, the Kubo response theory has been extended to describe pump–probe experiments. The main advantages of this response scheme is that although second order responses are included, it formally remains a linear theory and therefore all obtained expressions can be implemented straightforwardly within any standard bandstructure method.

REFERENCES

[1] A. Vernes and P. Weinberger, Phys. Rev. B **71**, 165108 (2005).