Formally linear response theory of pump-probe experiments

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By linearizing the density of both the pump- and probe-excited states and neglecting the overlap between femtosecond laser pulses, the Kubo response theory is 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 band structure method, e.g., based on a Green's function approach. In particular, even the time-dependent zeroth order dynamic conductivity as obtained by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker method for fcc Ni(100) predicts a relatively slow demagnetization process over 100 fs after the impact of the probe pulse, which is in reasonably good agreement with available experimental data.

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I. INTRODUCTION

Many fundamental processes in matter like electron-electron or electron-phonon scattering in solids, occur on a time scale ranging from a few tenths of a femtosecond ($1\ \mathrm{fs}=10^{-15}\ \mathrm{s}$) to a picosecond ($1\ \mathrm{ps}=10^{-12}\ \mathrm{s}$). 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.

The effect of a pump pulse on the sample can be analyzed either by measuring the probe pulse characteristics or by monitoring the effects created by the probe pulse in the presence and absence of the pump pulse. In both techniques, the excited states created by the pump pulse can be directly inspected, e.g., via a spectral analysis. In addition, by varying the delay time of the probe pulse, a time-resolved spectrum of the excited sample is obtained, which in turn tracks the specific ultrafast process that characterizes the relaxation of the system. Viewed oppositely, this implies that any ultrafast process (at least in principle) can be studied, if the pump-probe experimental setup is known. Besides the complexity of the investigated system (sample preparation), characteristics of the pulses (i.e., intensity, carrier frequency, and polarization) and the delay time, the measured dynamics also depends on the relative orientation of the pump and the probe polarization planes.

Inspired by the pioneering work of Freeman and co-workers[1] ultrafast magnetism in dilute magnetic semiconductors rapidly gained attention because of its huge technological potential.[2] Presently, it is believed that on the basis of "spin dynamics" in magnetic semiconductor quantum structures, such as "spin superlattices," "digital magnetic structures,"[3] etc., a much faster generation of computers will soon become reality, for an overview see Ref. 4. It should be noted that femtosecond spectroscopy methods are already in technological or academic use, e.g., as optical data storage,[5] in lithography[6] or in near-field spin microscopy techniques.[7]

Despite all the experimental work performed so far, however, except a few attempts,[8–10] *ab initio* like theoretical investigations in realistic systems—at least according to our knowledge—are still lacking completely. The present paper provides therefore a scheme that describes pump-probe experiments within the response theory of Kubo, which in turn can be implemented using, e.g., the spin-polarized relativistic screened Korringa-Kohn-Rostoker method[11–14] via a contour integration technique.[15,16]

The paper is organized as follows. Section II summarizes briefly the slowly varying envelope approximation, which provides the most frequently used models of laser pulses. After introducing the basic concepts of the Kubo theory in Sec. III A, the linear response of an arbitrary system to a femtosecond pulse is deduced and found to yield a compact expression, which can be viewed as a generalization of the well-known Kubo formula for the ac conductivity. By neglecting the overlap of pulses in time and linearizing the densities in both the probe- and pump-excited states, a formally linear response theory is developed for pump-probe experiments in Secs. III B and III C. Applied for laser pulses, the scheme straightforwardly provides in Sec. III D the dynamic conductivity as a sum of a zeroth and first order contribution, which in turn are shown to depend besides the optical properties of the investigated system, also on the characteristics of both pulses. Explicit and hence directly implementable expressions for the involved current-current and three-current correlation functions are given in Sec. IV. In Sec. V, the proposed theory of pump-probe experiments is illustrated in terms of the time-dependent zeroth order dynamic conductivity of fcc Ni(100) as obtained by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker method. The main results thereof are finally summarized in Sec. VI.

II. FEMTOSECOND LASER PULSES

The slowly varying envelope approximation (SVEA) states that the change in the complex amplitude of an

optical pulse is slow with respect to the average oscillation period. This approximation seems to be valid for pulse durations that are approximately 10 times bigger than the average oscillation period. Therefore, the SVEA is a reasonable approach to be considered for most of the effects occurring due to the interaction of femtosecond pulses with matter.[17]

If the z axis is taken along the propagation direction then within the SVEA a femtosecond pulse can be viewed as an amplitude modulated quasimonochromatic plane wave,[18]

$$\vec{E}(z,t) = \vec{e}\tilde{E}(t)\exp(i\tilde{q}z) = \vec{\mathcal{E}}_0\tilde{\mathcal{E}}(t)\exp[i(\tilde{q}z - \omega_p t)], \quad (1)$$

where $\vec{e}$ is the unit polarization vector, $\mathcal{E}_0$ the real amplitude ($\vec{\mathcal{E}}_0 = \vec{e}\mathcal{E}_0$), ω_p the carrier frequency, $\tilde{q}$ the corresponding complex wave vector component along the z axis, and $\tilde{E}(t) = \mathcal{E}_0\tilde{\mathcal{E}}(t)\exp(-i\omega_p t)$. In the case of a linear medium $\tilde{q} = q_0\tilde{n}(\omega_p)$, with $\tilde{n}(\omega_p)$ being the complex refractive index and $q_0 = \omega_p/c$ the propagation constant in vacuum.

Equation (1) assumes that the complex amplitude varies faster along the propagation direction than across it and therefore the complex, dimensionless envelope of the electric field $\tilde{\mathcal{E}}(t)$ does not depend on z, i.e.,[17]

$$\tilde{\mathcal{E}}(t) = \mathcal{E}(t)\exp[-i\varphi(t)],$$

where $\mathcal{E}(t)$ is the real, dimensionless envelope and $\varphi(t)$ the time-dependent phase. It must be pointed out that in femtosecond laser optics the validity of Eq. (1) is restricted by the bandwidth $\Delta\omega$ of the pulse and by the time dependence of its envelope $\tilde{\mathcal{E}}(t)$, i.e., $\Delta\omega$ must be much smaller than the carrier frequency ω_p, and, see Ref. 19,

$$\left|\frac{d\tilde{\mathcal{E}}(t)}{dt}\right| \ll \omega_p|\tilde{\mathcal{E}}(t)|.$$

Alternatively to Eq. (1), one can use the following form:

$$\vec{E}(z,t) = \vec{\mathcal{E}}_0\mathcal{E}(t)\exp[-i\phi(t)]\exp(i\tilde{q}z),$$

with the time-dependent exponential part defining the phase factor of the pulse $\phi(t) = \omega_p t + \varphi(t)$, of which the first derivative with respect to time yields the time-dependent (instantaneous) carrier frequency

$$\omega(t) = \omega_p + \frac{d\varphi(t)}{dt}.$$

A constant phase pulse is a pulse that shows no frequency variation in time. A linear variation of $\varphi(t)$ with respect to t, i.e., $d\varphi(t)/dt = \text{const} \neq 0$, simply shifts the carrier frequency.[20] If $d\varphi(t)/dt$ is time dependent, the pulse is said to be frequency modulated or chirped. According to a positive or negative sign of $d^2\varphi(t)/dt^2$, the instantaneous carrier frequency increases (decreases) along the pulse and the pulse is called up (down) chirped.[21] If the phase distortion is quadratic, one has a linear chirp and higher order terms lead to a nonlinear chirp.[22]

Nowadays, ultrashort light pulses can be completely characterized in time as well as in frequency by measuring $\tilde{E}(t)$ or its Fourier transform $\tilde{E}(\omega)$ via frequency-resolved optical gating.[23,24] The decomposition of $\phi(t)$ into a carrier frequency ω_p and a time-dependent phase $\varphi(t)$, however, is not unique. Quite commonly ω_p is chosen at the pulse peak; a better choice is to identify ω_p by the intensity weighted average frequency, i.e.,[19]

$$\omega_p = \frac{\int_{-\infty}^{\infty} dt\ \omega(t)|\tilde{\mathcal{E}}(t)|^2}{\int_{-\infty}^{\infty} dt|\tilde{\mathcal{E}}(t)|^2}.$$

Although for the pulse duration Δt and the spectral width $\Delta\omega$ several formulations are known and used in the literature,[17,25] the bandwidth theorem,[26]

$$\Delta t\ \Delta\omega \geqslant 2\pi \quad \text{or } \Delta t\ \Delta\omega \geqslant \tfrac{1}{2} \quad (2)$$

ensures that the duration and spectral width of a pulse cannot be simultaneously arbitrarily small. For example, a 10 fs laser pulse covers almost the whole visible spectrum. Viewed oppositely, this means that for a given spectrum, only one pulse envelope exists, which has the shortest possible duration.[25] In fact, Eq. (2) can be viewed as a kind of counterpart of the Heisenberg uncertainty principle in pulsed optics and therefore one often refers to these equations (2) as the uncertainty principles of Fourier analysis, or simply calls them time-frequency uncertainty relations.[27]

III. GENERALIZED KUBO THEORY

A. Linear response to a laser pulse

The dynamic response of a physical quantity $\vec{B}(t)$ to an external force $\vec{X}(t)$ conjugated to the displacement $\vec{A}(t)$,[28]

$$\delta\langle B_\mu\rangle_t = \langle B_\mu(t)\rangle_X - \langle B_\mu(t)\rangle_0 = \sum_\nu \tilde{\chi}_{\mu\nu} X_\nu(t) \quad (\mu,\nu = x,y,z) \quad (3)$$

defines the dynamic linear admittance (susceptibility) $\tilde{\chi}_{\mu\nu}$. Here $\langle B_\mu(t)\rangle_X$ and $\langle B_\mu(t)\rangle_0$ are the mean values of $B_\mu(t)$ with respect to the (probability) densities $\rho_X(t)$ in the presence of $\vec{X}(t)$ and ρ_0 in the absence of $\vec{X}(t)$,

$$\langle B_\mu(t)\rangle_X = \mathrm{Tr}[\rho_X(t)B_\mu(t)] \quad \text{and } \langle B_\mu(t)\rangle_0 = \mathrm{Tr}[\rho_0 B_\mu(t)]. \quad (4)$$

The Hamiltonian of the system,

$$H_X(t) = H_0 + H_{\text{ext}}(t),$$

consists of an unperturbed part H_0 and an external perturbation $H_{\text{ext}}(t)$, which in the dipole approximation is given by

$$H_{\text{ext}}(t) = -\sum_\mu A_\mu(t)X_\mu(t).$$

In principle, within the interaction picture, $\rho_X(t)$ can be determined by solving iteratively the below equation,

$$\rho_X(t) \simeq \rho_0(t) - \frac{i}{\hbar}\int_{t_0}^{t} dt'[H_{\text{ext}}(t'),\rho_X(t')]. \tag{5}$$

However, in dealing with near-equilibrium states, for which the external perturbation starts in the infinite past ($t_0 \to -\infty$), when the density matrix can be assumed to be of canonical form,[29] namely

$$\rho_0(-\infty) = \rho_0 = \frac{\exp(-\beta H_0)}{\text{Tr}[\exp(-\beta H_0)]}, \tag{6}$$

a linearization (first order iteration) of Eq. (5) then simply leads to

$$\rho_X(t) \simeq \rho_0 - \frac{i}{\hbar}\int_{-\infty}^{t} dt'[H_{\text{ext}}(t'),\rho_0]. \tag{7}$$

In Eq. (6) $\beta=(k_B T)^{-1}$ is the so-called inverse temperature with k_B being the Boltzmann constant and T the temperature. By using this form of $\rho_X(t)$ in Eq. (4), the dynamic response in Eq. (3) can be written as

$$\delta\langle B_\mu\rangle_t = \frac{i}{\hbar}\sum_\nu \int_{-\infty}^{t} dt'\ \text{Tr}\{[A_\nu,\rho_0]B_\mu(t-t')\}X_\nu(t'), \tag{8}$$

where the Heisenberg operator $B_\mu(t-t')$ is defined as

$$B_\mu(t-t') = \exp\left(+\frac{i}{\hbar}H_0(t-t')\right)B_\mu \exp\left(-\frac{i}{\hbar}H_0(t-t')\right),$$

$$B_\mu = B_\mu(0).$$

Introducing[30] now the response function $\Phi_{\mu\nu}(t) \equiv \Phi_{B_\mu A_\nu}(t)$ as a response to a δ-type force,

$$\Phi_{\mu\nu}(t) = \frac{i}{\hbar}\langle[B_\mu(t),A_\nu]\rangle_0, \tag{9}$$

$\delta\langle B_\mu\rangle_t$ in Eq. (8) is of the form

$$\delta\langle B_\mu\rangle_t = \sum_\nu \int_{-\infty}^{t} dt'\ \Phi_{\mu\nu}(t-t')X_\nu(t'). \tag{10}$$

Recalling that the density ρ_0 for the equilibrium state was assumed to be canonical, see Eq. (6), in terms of the so-called canonical correlation,[28,31]

$$\langle a;b\rangle = \frac{1}{\beta}\int_0^\beta d\lambda\langle a(-i\hbar\lambda)b\rangle_0 = \langle b;a\rangle, \tag{11}$$

with

$$\langle a(-i\hbar\lambda)b\rangle_0 = \langle \exp(+\lambda H_0)a\ \exp(-\lambda H_0)b\rangle_0$$
$$= \text{Tr}[\rho_0 \exp(+\lambda H_0)a\ \exp(-\lambda H_0)b],$$

the response function $\Phi_{\mu\nu}(t)$ in Eq. (9) is finally given by

$$\Phi_{\mu\nu}(t) = \beta\langle B_\mu(t);\dot{A}_\nu\rangle = -\beta\langle\dot{B}_\mu(t);A_\nu\rangle, \tag{12}$$

where dotted symbols denote first derivatives with respect to t.

In the case of a perturbing electric field, B_μ picks up the meaning of a current density J_μ corresponding to the displacement A_μ,

$$J_\mu = \dot{A}_\mu = \frac{dA_\mu}{dt} = \frac{i}{\hbar}[H_0,A_\mu], \tag{13}$$

and the response function $\Phi_{\mu\nu}(t)$ in Eq. (12) can be written as

$$\Phi_{\mu\nu}(t) = \beta\langle J_\mu(t);\dot{A}_\nu\rangle = \beta\langle J_\mu(t);J_\nu\rangle.$$

Furthermore, if the electric field is pulsed as given by Eq. (1), suppressing its spatial dependence, Eq. (10) leads to

$$\delta\langle J_\mu\rangle_t = \sum_\nu \tilde{\sigma}_{\mu\nu}(\omega_p,t)\mathcal{E}_{0\nu}\exp(-i\omega_p t), \tag{14}$$

where the carrier frequency- and time-dependent conductivity is given by

$$\tilde{\sigma}_{\mu\nu}(\omega_p,t) = \beta\int_0^\infty d\xi\langle J_\mu(\xi);J_\nu\rangle\tilde{\mathcal{E}}(t-\xi)\exp(+i\omega_p\xi). \tag{15}$$

Because in Eq. (1) the pulse is of the form of an amplitude modulated quasimonochromatic plane wave, for the identity envelope, i.e., $\tilde{\mathcal{E}}(t-\xi)=1$, Eq. (15) recovers the well-known Kubo formula of the ac conductivity,

$$\tilde{\sigma}_{\mu\nu}(\omega) = \beta\int_0^\infty d\xi\langle J_\mu(\xi);J_\nu\rangle\exp(+i\omega\xi) = \beta\ \mathcal{L}[\langle J_\mu(t);J_\nu\rangle]_{-i\omega}, \tag{16}$$

with $\omega\equiv\omega_p$ now denoting the frequency of the monochromatic plane wave, and where

$$\mathcal{L}[f(y)]_x = \int_0^\infty dy\ f(y)\exp(-xy), \quad x\in\mathbb{C}, \tag{17}$$

is the Laplace transform of a function $f(y)$ satisfying the Dirichlet conditions.[32] Note that as long as $\tilde{\sigma}_{\mu\nu}(\omega)$ only depends on the properties of the system investigated, $\tilde{\sigma}_{\mu\nu}(\omega_p,t)$ does depend on the pulse characteristics via the pulse envelope and the carrier frequency.

Since $\tilde{\sigma}_{\mu\nu}(\omega_p,t)$ is directly related to the experimentally observed quantity $\delta\langle J_\mu\rangle_t$, for numerical reasons, it is more convenient to take the Fourier transform of Eq. (14),

$$\delta\langle J_\mu\rangle_\omega = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\ \delta\langle J_\mu\rangle_t\exp(+i\omega t) = \sum_\nu \tilde{\sigma}_{\mu\nu}(\omega_p,\omega)\mathcal{E}_{0\nu},$$

in which according to Eq. (15) the dynamic conductivity $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$,

$$\tilde{\sigma}_{\mu\nu}(\omega_p,\omega) = \frac{\beta}{2\pi}\int_{-\infty}^{\infty} dt\, \exp[+i(\omega-\omega_p)t] \times \int_0^{\infty} d\xi\, \langle J_\mu(\xi);J_\nu\rangle \tilde{\mathcal{E}}(t-\xi)\exp(+i\omega_p\xi),$$

is now resolved with respect to the spectral components of the pulse, e.g., for $\omega \in [\bar{\omega}-\Delta\omega, \bar{\omega}+\Delta\omega]$, where $\bar{\omega}$ denotes the mean frequency. By interchanging the order of integration and making use of the well-known properties of Fourier transforms,[26]

$$\mathcal{F}[f(y)]_x = \int_{-\infty}^{\infty} dy\ f(y)\exp(-ixy), \quad x \in \mathbb{R}, \tag{18}$$

where $f(y)$ is an absolutely integrable function, the frequency-frequency representation of the linear dynamic conductivity is finally given in the following compact form:

$$\tilde{\sigma}_{\mu\nu}(\omega_p,\omega) = \frac{\beta}{2\pi}\mathcal{F}[\tilde{\mathcal{E}}(t)]_{\omega_p-\omega}\mathcal{L}[\langle J_\mu(t);J_\nu\rangle]_{-i\omega} = \frac{1}{2\pi}\mathcal{F}[\tilde{\mathcal{E}}(t)]_{\omega_p-\omega}\tilde{\sigma}_{\mu\nu}(\omega), \tag{19}$$

with $\tilde{\sigma}_{\mu\nu}(\omega)$ being obtained from Eq. (16) for all spectral components of the pulse probing the investigated system.

Because in the case of ultrashort laser pulses nonlinear effects occur at arbitrary low intensities,[25] in addition to $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$ in Eq. (19), second order response functions also must be determined. Although these latter quantities are straightforwardly obtained by evaluating the dynamic response $\delta\langle B_\mu\rangle_t = \langle B_\mu(t)\rangle_X - \langle B_\mu(t)\rangle_0$ for $\rho_X(t)$ as given by Eq. (5) [after the substitution of Eq. (7) on the right-hand side], the corresponding expressions are not listed here for reasons which will become clear in the following section.

B. A formally linear response to pump and probe pulses

Based on the superposition principle of electromagnetic fields, the total external electric field in a pump-probe experiment is given by[33–36]

$$\vec{E}(\vec{r},t) = \vec{E}_{\rm pu}(\vec{r},t) + \vec{E}_{\rm pr}(\vec{r},t) = \vec{e}_{\rm pu}\mathcal{E}_{{\rm pu},0}\tilde{\mathcal{E}}_{\rm pu}(t)\exp[i(\vec{q}_{\rm pu}\vec{r}-\omega_{\rm pu}t)] + \vec{e}_{\rm pr}\mathcal{E}_{{\rm pr},0}\tilde{\mathcal{E}}_{\rm pr}(t-\tau)\exp[i(\vec{q}_{\rm pr}\vec{r}-\omega_{\rm pr}t)], \tag{20}$$

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 $\vec{e}_{\rm pr}$. The polarizations are important if orientational effects must be investigated.[33] In all other cases a parallel configuration can be assumed, i.e., $\vec{e}_{\rm pu}=\vec{e}_{\rm pr}$,[37] and almost always linear polarizations.[20]

In Eq. (20) the probe pulse has a peak at $t=\tau$, the pump pulse at $t=0$. For a positive delay time τ of the probe pulse ($\tau>0$), the probe pulse interacts with the system after the pump pulse, whereas in the case of a negative delay time ($\tau<0$) the interaction of the probe pulse with the system happens before the pump field acts. The only constraint applied to the delay time τ is that it must be of the same order of magnitude as the pulse duration,[20] see also Sec. II.

In considering an external electric field as defined in Eq. (20), 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.[37] This is reasonable to assume having, for example, "spin dynamics" in mind, namely incoherent ultrafast processes that occur after the phase of the pump-excited state is lost.[38] Furthermore, as shown immediately below, this separation in time of the pump and probe pulses allows one to account also for nonlinear effects, which in the case of ultrashort laser pulses are quite important, because—due to the wide spectral bandwidth of pulses—they occur even for arbitrary low intensities.[25] Although nonlinear effects can be introduced right from the beginning into the quadratic response theory outlined at the end of Sec. III A by using there Eq. (20), in this manner the formal linearity of the proposed scheme cannot be preserved and therefore must be abandoned. Note also, that not only nonlinear effects are missing, when the time-dependent part of Eq. (20) is substituted into Eq. (10), but also that the resulting $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr},\omega)$ cannot be related to any relaxation process within a pump-probe experiment, because the pump-excited state is not probed, if one uses Eq. (19). In contrast to this, $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pu},\omega)$ as obtained from Eq. (19) represents a linear response of the system to the pump pulse and therefore is a physically meaningful quantity, which can be used as a reference to be extracted from the response of the system to the delayed probe pulse.

In pump-probe experiments a canonical density ρ_0 as given by Eq. (6) for the equilibrium state can be always assumed and also that the pump-pulse perturbation is switched on in the infinite past ($t_0\to-\infty$). Furthermore, because in Eq. (20) 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, see Fig. 1. 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 $t>\tau$, i.e.,

$$\delta\langle B_\mu\rangle_t = \langle B_\mu(t)\rangle_{\rm pr} - \langle B_\mu(\tau)\rangle_{\rm pu} = \sum_\nu \tilde{\chi}_{\mu\nu}X_\nu, \tag{21}$$

where

$$\langle B_\mu(t)\rangle_{\rm pr} = {\rm Tr}[\rho_{\rm pr}(t)B_\mu(t)] \quad \text{and} \quad \langle B_\mu(\tau)\rangle_{\rm pu} = {\rm Tr}(\rho_{\rm pu}B_\mu),$$

with $B_\mu=B_\mu(\tau)$ and $\rho_{\rm pu}=\rho_{\rm pu}(\tau)$. Here and in the following the subscripts "pu" and "pr" specify with which of the pulses, pump or probe, the system is actually interacting. Compared to Eq. (3), one notes that quite obviously in Eq. (21) the dynamic response refers to the change of $\vec{B}(t)$ with respect to its mean value for the pump-excited state and not for that in the initial equilibrium state.

If within the interaction picture the only source of perturbation is the probe pulse then from the linearization of the density,

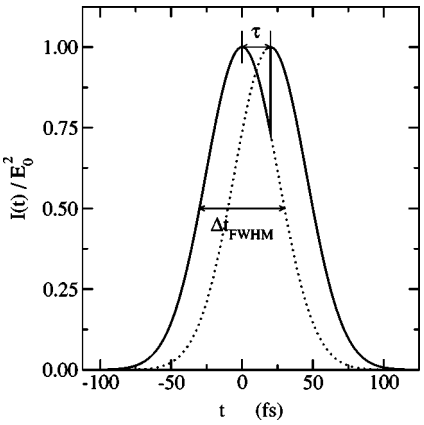

FIG. 1. Time-dependent normalized intensity resulting from two time-delayed (τ=20 fs) identical, linearly polarized Gaussian femtosecond (t_{FWHM}=60 fs) laser pulses (parallel configuration) neglecting the time overlap of the pulses.

$$\rho_{\mathrm{pr}}(t) \simeq \rho_{\mathrm{pu}} - \frac{i}{\hbar}\int_{\tau}^{t} dt'[H_{\mathrm{pr}}(t'),\rho_{\mathrm{pu}}], \tag{22}$$

where according to the dipole approximation,

$$H_{\mathrm{pr}}(t) = -\sum_{\nu} A_{\nu}(t)X_{\mathrm{pr},\nu}(t),$$

one directly obtains[39]

$$\delta\langle B_{\mu}\rangle_{t} \simeq \sum_{\nu}\int_{\tau}^{t} dt' \frac{i}{\hbar}\langle[B_{\mu}(t),A_{\nu}(t')]\rangle_{\mathrm{pu}} X_{\mathrm{pr},\nu}(t'),$$

by assuming

$$\mathrm{Tr}[\rho_{\mathrm{pu}}B_{\mu}(t)] - \mathrm{Tr}(\rho_{\mathrm{pu}}B_{\mu}) \simeq 0.$$

Here $A_{\nu}(t)$ and $B_{\mu}(t)$ are Heisenberg operators and

$$\langle[B_{\mu}(t),A_{\nu}(t')]\rangle_{\mathrm{pu}} = \mathrm{Tr}\{\rho_{\mathrm{pu}}[B_{\mu}(t),A_{\nu}(t')]\},$$

is the mean value of their commutator with respect to the pump-excited state. Introducing now in analogy with Eq. (9) the response function $\Phi_{\mu\nu}(t,t')$,

$$\Phi_{\mu\nu}(t,t') = \frac{i}{\hbar}\langle[B_{\mu}(t),A_{\nu}(t')]\rangle_{\mathrm{pu}}, \tag{23}$$

the dynamic response can be written as

$$\delta\langle B_{\mu}\rangle_{t} = \sum_{\nu}\int_{\tau}^{t} dt'\ \Phi_{\mu\nu}(t,t')X_{\mathrm{pr},\nu}(t'). \tag{24}$$

Because in accordance with the SVEA, see Sec. II, the external force $\vec{X}_{\mathrm{pr}}(t)$ is given by

$$\vec{X}_{\mathrm{pr}}(t) = \vec{\mathcal{X}}_{\mathrm{pr},0}\tilde{\mathcal{X}}_{\mathrm{pr}}(t-\tau)\exp(-i\omega_{\mathrm{pr}}t),$$

where $\vec{\mathcal{X}}_{\mathrm{pr},0}$ is the real amplitude and $\tilde{\mathcal{X}}_{\mathrm{pr}}(t)$ the complex dimensionless envelope of the probe pulse, Eq. (24) directly yields the generalized, frequency- and time-dependent admittance $\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)$,[39]

$$\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau) = \int_{0}^{t-\tau} d\xi\ \Phi_{\mu\nu}(t,t-\xi)\tilde{\mathcal{X}}_{\mathrm{pr}}(t-\tau-\xi) \times\exp(+i\omega_{\mathrm{pr}}\xi), \tag{25}$$

such that

$$\delta\langle B_{\mu}\rangle_{t} = \sum_{\nu}\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)\mathcal{X}_{\mathrm{pr},0\nu}\exp(-i\omega_{\mathrm{pr}}t).$$

By taking the Fourier transform of this equation,

$$\delta\langle B_{\mu}\rangle_{\omega} = \frac{1}{2\pi}\int_{\tau}^{\infty} dt\ \delta\langle B_{\mu}\rangle_{t}\exp(+i\omega t) = \sum_{\nu}\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)\mathcal{X}_{\mathrm{pr},0\nu},$$

the frequency-frequency representation of the admittance $\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)$,

$$\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau) = \frac{1}{2\pi}\int_{\tau}^{\infty} dt\ \tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)\exp[+i(\omega-\omega_{\mathrm{pr}})t],$$

is defined according to Eq. (25) by

$$\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau) = \frac{1}{2\pi}\int_{\tau}^{\infty} dt\exp[+i(\omega-\omega_{\mathrm{pr}})t] \times\int_{0}^{t-\tau} d\xi\ \Phi_{\mu\nu}(t,t-\xi)\tilde{\mathcal{X}}_{\mathrm{pr}}(t-\tau-\xi) \times\exp(+i\omega_{\mathrm{pr}}\xi). \tag{26}$$

It should be noted that, e.g., in Ref. 39 the frequency-time representation of the admittance $\tilde{\chi}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)$ in Eq. (25) at $t=\Delta t_{\mathrm{pr}}$ (probe-pulse duration) is considered to be directly measurable in a pump-probe experiment.

C. Density linearization for the pump-excited state

Similar to $\rho_{\mathrm{pr}}(t)$ in Eq. (22), the density for the pump-excited state is written

$$\rho_{\mathrm{pu}} \simeq \rho_0 - \frac{i}{\hbar}\int_{-\infty}^{\tau} dt[H_{\mathrm{pu}}(t),\rho_0] \equiv \rho_{\mathrm{pu}}^{(0)} + \rho_{\mathrm{pu}}^{(1)}, \tag{27}$$

with a zeroth order density

$$\rho_{\mathrm{pu}}^{(0)} = \rho_0 \tag{28}$$

and a first order density $\rho_{\mathrm{pu}}^{(1)}$ given within the dipole approximation by

$$\rho_{\mathrm{pu}}^{(1)} = \frac{i}{\hbar}\sum_{\kappa}\int_{-\infty}^{\tau} dt[A_{\kappa}(t),\rho_0]X_{\mathrm{pu},\kappa}(t). \tag{29}$$

By substituting the expression in Eq. (27) for ρ_{pu} into Eq. (23), two contributions to the response function $\Phi_{\mu\nu}(t,t')$ arise, namely $\Phi_{\mu\nu}^{(0)}(t,t')$ and $\Phi_{\mu\nu}^{(1)}(t,t')$, such that

$$\Phi_{\mu\nu}(t,t') = \Phi_{\mu\nu}^{(0)}(t,t') + \Phi_{\mu\nu}^{(1)}(t,t'), \tag{30}$$

where

$$\Phi^{(0)}_{\mu\nu}(t,t') = \frac{i}{\hbar}\langle[B_\mu(t),A_\nu(t')]\rangle_0 \tag{31}$$

corresponds to the zeroth order density in Eq. (28). By making use of the invariance properties of the trace, $\Phi^{(0)}_{\mu\nu}(t,t')$ turns out to be a time-time representation of the response function $\Phi_{\mu\nu}(t)$ known from the linear response theory of Kubo,[28] see Eq. (9). The other response function $\phi^{(1)}_{\mu\nu}(t,t')$ is defined by the first order density $\rho^{(1)}_{\rm pu}$ introduced in Eq. (29) as

$$\Phi^{(1)}_{\mu\nu}(t,t') = \frac{i}{\hbar}\{\mathrm{Tr}[B_\mu(t),A_\nu(t')]\rho^{(1)}_{\rm pu}\}.$$

It is a matter of simple algebra to show that

$$\Phi^{(1)}_{\mu\nu}(t,t') = \sum_\kappa \int_{-\infty}^{\tau} dt''\, \Phi_{\mu\nu\kappa}(t,t',t'')X_{{\rm pu},\kappa}(t'') \tag{32}$$

$$(t \geqslant t' \geqslant \tau \geqslant t''),$$

where the second order response function is given by

$$\begin{aligned}\Phi_{\mu\nu\kappa}(t,t',t'') &= -\frac{1}{\hbar^2}\langle[[B_\mu(t),A_\nu(t')],A_\kappa(t'')]\rangle_0 \\ &= -\frac{1}{\hbar^2}\langle[A_\kappa(t''),[A_\nu(t'),B_\mu(t)]]\rangle_0. \end{aligned}\tag{33}$$

Strictly speaking, Eq. (33) provides a three-time representation of the second order response function known from nonlinear response theory.[40] In contrast to the quadratic response theory, however, in the present scheme $\Phi_{\mu\nu\kappa}(t,t',t'')$ couples the κth and νth Cartesian component of the pump and probe pulses, respectively, at $t'' \leqslant \tau$ and $t' \geqslant \tau$, to the μth Cartesian component of the dynamic response at $t \geqslant t'$. Furthermore, in spite of the presence of $\Phi_{\mu\nu\kappa}(t,t',t'')$ in the dynamic response $\delta\langle B_\mu\rangle_t$, the formal linearity of the response promoted by a linearization of both $\rho_{\rm pr}(t)$ and $\rho_{\rm pu}=\rho_{\rm pu}(\tau)$ densities, see Eqs. (22) and (27), is preserved, because Eq. (21) still applies. Quite obviously, a nonlinear response theory like that in Refs. 8 and 41, which needs at least a second iteration to be considered in Eq. (5) cannot show these features, when dealing with pump-probe experiments.

In the case of a canonical density ρ_0, see Eq. (11), Eq. (31) directly leads to

$$\Phi^{(0)}_{\mu\nu}(t,t') = \beta\langle B_\mu(t);\dot{A}_\nu(t')\rangle = -\beta\langle\dot{B}_\mu(t);A_\nu(t')\rangle, \tag{34}$$

which is nothing but the time-time representation of the canonical correlation in Eq. (12). Therefore, substituting the kernel in Eqs. (25) and (26) by this zeroth order response function $\Phi^{(0)}_{\mu\nu}(t,t'=t-\xi)$, immediately yields a strictly linear complex admittance $\tilde{\chi}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)$ and its Fourier transform $\tilde{\chi}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)$. The only deficiency of these strictly linear or zeroth order admittances is that they do not account for orientational effects, which are caused by the difference in the polarization of the pump and probe pulse,[33] i.e., $\vec{e}_{\rm pu} \neq \vec{e}_{\rm pr}$ in Eq. (20).

Alternatively Eq. (33) can also be written as

$$\begin{aligned}\Phi_{\mu\nu\kappa}(t,t',t'') &= -\frac{1}{\hbar^2}\mathrm{Tr}\{B_\mu(t)[A_\nu(t'),[A_\kappa(t''),\rho_0]]\} \\ &\simeq \beta\langle B_\mu(t);\dot{A}_\nu(t');\dot{A}_\kappa(t'')\rangle, \end{aligned}\tag{35}$$

where, by extending the conventional canonical correlation introduced in Eq. (11),

$$\begin{aligned}\langle a(t);b(t');c(t'')\rangle = &\frac{1}{\beta}\int_0^\beta d\lambda' \int_0^{\lambda'} d\lambda''\langle a(t-i\hbar\beta)b(t'-i\hbar\lambda') \\ &\times c(t''-i\hbar\lambda'')\rangle_0 \\ &+\frac{1}{\beta}\int_0^\beta d\lambda' \int_0^{\lambda'} d\lambda''\langle a(t-i\hbar\beta) \\ &\times c(t''-i\hbar\lambda')b(t'-i\hbar\lambda'')\rangle_0. \end{aligned}$$

With this expression used in Eq. (32), in the case of a pump pulse,

$$\vec{X}_{\rm pu}(t) = \vec{\xi}_{{\rm pu},0}\tilde{\xi}_{\rm pu}(t)\exp(-i\omega_{\rm pu}t),$$

Eq. (25) immediately provides a first order complex admittance,

$$\begin{aligned}\tilde{\chi}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},t;\tau) = &\sum_\kappa \xi_{{\rm pu},0\kappa}\exp(-i\omega_{\rm pu}t) \\ &\times\int_0^{t-\tau} d\xi\, \tilde{\xi}_{\rm pr}(t-\tau-\xi)\exp(+i\omega_{\rm pr}\xi) \\ &\times\int_{t-\tau}^{\infty} d\xi'\Phi_{\mu\nu\kappa}(t,t-\xi,t-\xi')\tilde{\xi}_{\rm pu}(t-\xi') \\ &\times\exp(+i\omega_{\rm pu}\xi'). \end{aligned}$$

The Fourier transform of $\tilde{\chi}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},t;\tau)$ then leads to

$$\begin{aligned}\tilde{\chi}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau) = &\frac{1}{2\pi}\sum_\kappa \xi_{{\rm pu},0\kappa}\int_\tau^\infty dt\exp[+i(\omega-\omega_{\rm pu})t] \\ &\times\int_0^{t-\tau} d\xi\, \tilde{\xi}_{\rm pr}(t-\tau-\xi)\exp(+i\omega_{\rm pr}\xi) \\ &\times\int_{t-\tau}^{\infty} d\xi'\,\Phi_{\mu\nu\kappa}(t,t-\xi,t-\xi')\tilde{\xi}_{\rm pu}(t-\xi') \\ &\times\exp(+i\omega_{\rm pu}\xi'). \end{aligned}\tag{36}$$

In analogy with Eq. (25), here the integration over ξ' introduces a second order complex admittance,

$$\begin{aligned}\tilde{\chi}_{\mu\nu\kappa}(\omega_{\rm pu},t,\xi;\tau) = &\int_{t-\tau}^{\infty} d\xi'\,\Phi_{\mu\nu\kappa}(t,t-\xi,t-\xi')\tilde{\xi}_{\rm pu}(t-\xi') \\ &\times\exp(+i\omega_{\rm pu}\xi'), \end{aligned}$$

that depends on the pump-pulse characteristics (envelope and carrier frequency) and material properties. In addition, $\tilde{\chi}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ depends also on the probe-pulse characteristics and the amplitude of the pump pulse.

D. Zeroth and first order dynamic conductivities

Because according to Eq. (13) in the case of laser pulses the zeroth order canonical response function as introduced in Eq. (34) is given by

$$\Phi^{(0)}_{\mu\nu}(t,t') = \beta\langle J_\mu(t); J_\nu(t')\rangle,$$

Eq. (25) reduces to

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau) = \frac{\beta}{2\pi}\exp[+i(\omega-\omega_{\rm pr})\tau] \times\int_0^\infty dt\,\exp[+i(\omega-\omega_{\rm pr})t] \times\int_0^t d\xi\langle J_\mu(\xi);J_\nu\rangle\tilde{\mathcal{E}}_{\rm pr}(t-\xi)\exp(+i\omega_{\rm pr}\xi)$$

and hence the frequency-frequency representation of the generalized, strictly linear conductivity for the pump-probe experiments can be written as

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau) = \frac{1}{2\pi}\exp[+i(\omega-\omega_{\rm pr})\tau]\mathcal{L}[\tilde{\mathcal{E}}_{\rm pr}(t)]_{i(\omega_{\rm pr}-\omega)}\tilde{\sigma}_{\mu\nu}(\omega), \tag{37}$$

where in terms of ω this quantity is now resolved with respect to the spectral components of the probe pulse, e.g., for $\omega\in[\overline{\omega}_{\rm pr}-\Delta\omega_{\rm pr},\overline{\omega}_{\rm pr}+\Delta\omega_{\rm pr}]$, $\overline{\omega}_{\rm pr}$ being the mean frequency of the probe pulse.

As can be seen from Eq. (37), independent of the envelope of the probe pulse, $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)$ oscillates in τ with an amplitude that depends on ω but not on τ. The quantity of interest, however, namely the time-resolved response $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)$ of the system, due to the rather complicated dependence of $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)$ on ω, oscillates in t with a time-dependent amplitude, a fact that immediately follows from a Fourier transform of Eq. (37):

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau) = \mathcal{F}[\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)]_t. \tag{38}$$

One also observes that by comparing the zeroth order conductivity $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau\to0)$ with the strictly linear conductivity $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr,}\omega)$—as follows from Eq. (19) when the probe pulse would interact alone with the system—that the only difference occurring is the type of the transformation performed for the probe pulse envelope.

By observing that in the case of laser pulses, Eq. (35) directly yields

$$\Phi_{\mu\nu\kappa}(t,t',t'') = \beta\langle J_\mu(t);J_\nu(t');J_\kappa(t'')\rangle,$$

the first order conductivity resolved according to the spectral components of the pump pulse, e.g., for $\omega\in[\overline{\omega}_{\rm pu}-\Delta\omega_{\rm pu},\overline{\omega}_{\rm pu}+\Delta\omega_{\rm pu}]$ with $\overline{\omega}_{\rm pu}$ being the mean frequency of the pump pulse, immediately follows from Eq. (36) as

$$\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau) = \frac{\beta}{2\pi}\exp[+i(\omega-\omega_{\rm pu})\tau]\sum_\kappa\mathcal{E}_{{\rm pu},0\kappa} \times\int_0^\infty dt\,\exp(+i\omega t) \times\int_0^t d\xi\,\tilde{\mathcal{E}}_{\rm pr}(t-\xi)\exp(+i\omega_{\rm pr}\xi) \times\int_0^\infty d\xi'\langle J_\mu(t);J_\nu(t-\xi);J_\kappa(-\xi')\rangle \times\tilde{\mathcal{E}}_{\rm pu}(\tau-\xi')\exp(+i\omega_{\rm pu}\xi'). \tag{39}$$

Although for this form one can also take advantage on the properties of Laplace transforms, see Eq. (17), for $\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ a similar expression to that in Eq. (37) cannot be deduced.

In accordance with Eq. (30), the total and formally linear dynamic conductivity is finally obtained by combining the zeroth and first order conductivities as given by Eqs. (37) and (39),

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

Because $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pu},\omega)$ can be determined on the same footing as $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$, pump-probe experiments such as in Refs. 19 and 20 can also be described numerically within the present response scheme. Unlike Refs. 8, 41, 42, and 43, where the time-dependent dipole interaction between the electronic subsystem and external pulse fields is directly introduced in the Hamiltonian and the linear and nonlinear susceptibilities are calculated within the response theory of Kubo, here, in order to evaluate $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ no time-dependent Schrödinger or Dirac equation has to be solved, see the following section. Another major difference is that here both contributions $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)$ and $\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ to the dynamic conductivity explicitly depend on the pulse characteristics, see Eqs. (37) and (39), whereas in Refs. 8, 41, 42, and 43, these dependencies enter via time-dependent current matrix elements.

IV. NUMERICAL FRAMEWORK

A. Current-current correlations and the Luttinger formula

By using the contour deformation method,[44] the current-current correlation in Eq. (16) is given by

$$\langle J_\mu(t);J_\nu\rangle = \frac{i}{\hbar\beta}\int_t^\infty dt'\langle[J_\mu(t'),J_\nu]\rangle_{\rm eq}, \tag{40}$$

where the suffix "eq" refers to the unperturbed equilibrium density $\rho_{\rm eq}$,[29] i.e.,

$$\rho_{\rm eq} = \frac{1}{V}f(H_0) = \frac{1}{V}\frac{1}{\exp[\beta(H_0-\varepsilon_F)]+1}, \tag{41}$$

with $f(H_0)$ being the Fermi-Dirac distribution function, H_0 the unperturbed Hamiltonian of the system, ε_F the Fermi level, and V the reference volume.

In a representation, which diagonalizes the Hamiltonian H_0, the mean value of the current-current commutator in Eq. (40) can be written as

$$\langle [J_\mu(t'), J_\nu] \rangle_{\mathrm{eq}} = \frac{1}{V} \sum_{m,n} [f(\varepsilon_m) - f(\varepsilon_n)] \times \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t' \right) J^\mu_{mn} J^\nu_{nm},$$

with $J^\mu_{mn} = \langle m | J_\mu | n \rangle$, $|m\rangle$ and $|n\rangle$ being eigenstates of H_0. Therefore, according to Eq. (40), the current-current correlation function is given by

$$\langle J_\mu(t); J_\nu \rangle = -\frac{1}{\beta V} \sum_{m,n} \frac{f(\varepsilon_m) - f(\varepsilon_n)}{\varepsilon_m - \varepsilon_n} J^\mu_{mn} J^\nu_{nm} \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t \right).$$

The indefinite integral in the Laplace transform of this expression,

$$\mathcal{L}[\langle J_\mu(t); J_\nu \rangle]_{-i\omega} = -\frac{1}{\beta V} \sum_{m,n} \frac{f(\varepsilon_m) - f(\varepsilon_n)}{\varepsilon_m - \varepsilon_n} J^\mu_{mn} J^\nu_{nm} \times \int_0^\infty dt \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n + \hbar\omega) t \right), \tag{42}$$

converges if and only if the real part of the exponent is positive.[45] As this is not the case in Eq. (42), a small real parameter $\delta > 0$ must be introduced in order to make the integral convergent,

$$\int_0^\infty dt \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n + \hbar\omega) t \right) = -\frac{1}{\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n + \hbar\omega + i\delta)},$$

which in turn directly leads to

$$\mathcal{L}[\langle J_\mu(t); J_\nu \rangle]_{-i\omega} = -\frac{i\hbar}{\beta V} \sum_{m,n} \frac{f(\varepsilon_m) - f(\varepsilon_n)}{\varepsilon_m - \varepsilon_n} \frac{J^\mu_{mn} J^\nu_{nm}}{\varepsilon_m - \varepsilon_n + \zeta}, \tag{43}$$

with ζ being a complex frequency,

$$\zeta = \hbar\omega + i\delta.$$

In defining the below quantity,[16]

$$\tilde{\Sigma}_{\mu\nu}(\zeta) = \frac{i\hbar}{V} \sum_{m,n} \frac{f(\varepsilon_m) - f(\varepsilon_n)}{\varepsilon_m - \varepsilon_n + \zeta} J^\mu_{mn} J^\nu_{nm},$$

Eq. (43) can be written as

$$\mathcal{L}[\langle J_\mu(t); J_\nu \rangle]_{-i\omega} = \frac{\tilde{\sigma}_{\mu\nu}(\zeta)}{\beta},$$

where $\tilde{\sigma}_{\mu\nu}(\zeta)$ is then given by the well-known Luttinger formula,[46]

$$\tilde{\sigma}_{\mu\nu}(\zeta) = \frac{\tilde{\Sigma}_{\mu\nu}(\zeta) - \tilde{\Sigma}_{\mu\nu}(0)}{\zeta}. \tag{44}$$

Just like the widely used Wang-Callaway formula for the optical conductivity,[47] for details see Ref. 48, the Luttinger formula (44) and consequently Eqs. (19) and (37), have the advantage that they also simultaneously provide the absorptive and the dispersive parts on the same footing without using Kramers-Kronig relations.[16] Furthermore, it was shown[16] that for $\delta > 0$ not only the field is turned on at $t = -\infty$,[49] but that the interaction of the system with its surroundings and all scattering processes at $T \neq 0$ are also described. It has already been demonstrated[50] that the Green's function and the current matrices needed to evaluate the optical conductivity tensor $\tilde{\sigma}_{\mu\nu}(\zeta)$ can be computed by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker (SKKR) method for layered systems[11–14] with an arbitrary high precision.

B. Three-current correlations

For the three-current correlations entering Eq. (39), a carefully revised contour deformation method,[44] provides after some algebra

$$\langle J_\mu(t); J_\nu(t'); J_\kappa(t'') \rangle = -\frac{1}{\hbar^2 \beta} \int_{t'}^\infty d\tau' \int_{t''}^\infty d\tau'' \langle [J_\mu(t), J_\kappa(\tau'')] J_\nu(\tau') - [J_\mu(t), J_\nu(\tau')] J_\kappa(\tau'') \rangle_0,$$

which in the case of the equilibrium density $\rho_0 = \rho_{\mathrm{eq}}$ as given in Eq. (41) and within the representation diagonalizing the unperturbed Hamiltonian H_0 can be written as

$$\begin{aligned}
\langle J_\mu(t); J_\nu(t'); J_\kappa(t'') \rangle = {} & \frac{1}{\beta V} \sum_{m,n,p} \frac{f(\varepsilon_m) J^\mu_{mn} J^\kappa_{np} J^\nu_{pm}}{(\varepsilon_p - \varepsilon_m)(\varepsilon_n - \varepsilon_p)} \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t \right) \exp\left(\frac{i}{\hbar} (\varepsilon_p - \varepsilon_m) t' \right) \exp\left(\frac{i}{\hbar} (\varepsilon_n - \varepsilon_p) t'' \right) \\
& - \frac{1}{\beta V} \sum_{m,n,p} \frac{f(\varepsilon_m) J^\kappa_{mn} J^\mu_{np} J^\nu_{pm}}{(\varepsilon_p - \varepsilon_m)(\varepsilon_m - \varepsilon_n)} \exp\left(\frac{i}{\hbar} (\varepsilon_n - \varepsilon_p) t \right) \exp\left(\frac{i}{\hbar} (\varepsilon_p - \varepsilon_m) t' \right) \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t'' \right) \\
& - \frac{1}{\beta V} \sum_{m,n,p} \frac{f(\varepsilon_m) J^\mu_{mn} J^\nu_{np} J^\kappa_{pm}}{(\varepsilon_p - \varepsilon_m)(\varepsilon_n - \varepsilon_p)} \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t \right) \exp\left(\frac{i}{\hbar} (\varepsilon_n - \varepsilon_p) t' \right) \exp\left(\frac{i}{\hbar} (\varepsilon_p - \varepsilon_m) t'' \right) \\
& + \frac{1}{\beta V} \sum_{m,n,p} \frac{f(\varepsilon_m) J^\nu_{mn} J^\mu_{np} J^\kappa_{pm}}{(\varepsilon_p - \varepsilon_m)(\varepsilon_m - \varepsilon_n)} \exp\left(\frac{i}{\hbar} (\varepsilon_n - \varepsilon_p) t \right) \exp\left(\frac{i}{\hbar} (\varepsilon_m - \varepsilon_n) t' \right) \exp\left(\frac{i}{\hbar} (\varepsilon_p - \varepsilon_m) t'' \right).
\end{aligned} \tag{45}$$

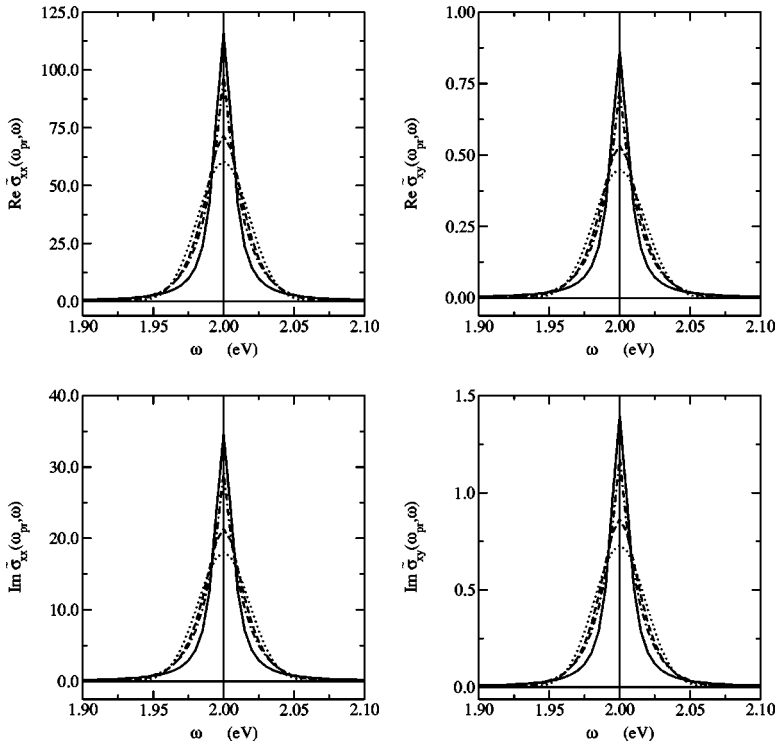

FIG. 2. Linear response to a femtosecond laser probe pulse as given by the dimensionless dynamic conductivity $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$ for $\mu,\nu=x,y$ in the case of fcc Ni(100), when the pulse has a carrier frequency $\omega_p=2$ eV (marked by a thin vertical line), a duration $\Delta t=60$ fs (taken as full width at half-maximum, i.e., FWHM) and an envelope of a double exponential (full line), a Gaussian (dotted line), a hyperbolic secant (dashed line), and a Lorentzian shape (dotted-dashed line), respectively.

Finally, by introducing Eq. (45) into Eq. (39), one obtains the first order dynamic conductivity. Although the expression for $\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ can again be related to the Laplace transform of the envelopes $\tilde{\mathcal{E}}_{\rm pu}(t)$ and $\tilde{\mathcal{E}}_{\rm pr}(t)$, for pulses that are not of double exponential form, in the evaluation of $\tilde{\sigma}^{(1)}_{\mu\nu}(\omega_{\rm pr},\omega_{\rm pu},\omega;\tau)$ additional approximation must be made in order to calculate the corresponding integrals.

V. RESULTS AND DISCUSSIONS

As an application for the scheme presented in here the linear dynamic conductivity for fcc Ni(100) is evaluated by using the zero order contribution only and the characteristics of pulses taken from Ref. 51. The optical conductivity tensor $\tilde{\sigma}_{\mu\nu}(\omega)$ in Eqs. (19) and (37) has been calculated for photon energies ω within the visible regime (from 0 to 6 eV with an increment of 0.27 eV) by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker (SKKR) method[16] for the layered system $Ni/Ni_{12}/Ni(100)$ with the well-known LDA equilibrium lattice constant $a_{\rm theo}=3.466$ Å $=6.55$ a.u.

As can be seen from Fig. 2, the linear response to a femtosecond laser pulse strongly depends on the pulse characteristics. The Fourier transform of the pulse envelope, for example, acts as a frequency filter on the optical conductivity tensor $\tilde{\sigma}_{\mu\nu}(\omega)$ around the carrier frequency ω_p of the pulse. Although the Fourier transform $\mathcal{F}[\mathcal{E}(t)]_{\omega_p-\omega}$ of the real and even envelopes $\mathcal{E}(t)=\mathcal{E}(-t)\in\mathbb{R}$ of a double exponential, Gaussian, hyperbolic secant or Lorentzian pulse assumed in Fig. 2, is symmetric with respect to its mean frequency, here $\overline{\omega}=\omega_p$, a similar feature not necessarily is valid for $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$. The reason is quite simple, in order to have $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$ as a function of $\omega\in[\omega_p-\Delta\omega,\omega_p+\Delta\omega]$ symmetric with respect to ω_p, besides of $\mathcal{F}[\mathcal{E}(t)]_{\omega_p-\omega}$ also $\tilde{\sigma}_{\mu\nu}(\omega)$ must be symmetric around the carrier frequency within the bandwidth $\Delta\omega$ of the pulse. Because this is only approximatively the case for $\omega_p=2$ eV as considered in Fig. 2, $\tilde{\sigma}_{\mu\nu}(\omega_p,\omega)$ is slightly asymmetric as a function of the spectral component ω of the pulse.

This small asymmetry of the optical conductivity in the vicinity of 2 eV becomes even more pronounced, when—in accordance with Eq. (37)—$\tilde{\sigma}_{\mu\nu}(\omega)$ is multiplied by the Laplace transform $\mathcal{L}[\mathcal{E}_{\rm pr}(t)]_{i(\omega_{\rm pr}-\omega)}$ of the real and even enveloped probe pulse of carrier frequency $\omega_{\rm pr}=2$ eV (not shown). Therefore it is not surprising at all that $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)$ is independent of the delay time τ of the probe pulse and asymmetrically oscillates around $\omega_{\rm pr}=2$ eV, see Fig. 3. Furthermore, as can be seen in this figure, although the real and imaginary part of both the diagonal and the off-diagonal conductivity tensor elements oscillate quite differently with respect to ω, the nodes of Re $\tilde{\sigma}^{(0)}_{x\nu}(\omega_{\rm pr},\omega;\tau)$ and Im $\tilde{\sigma}^{(0)}_{x\nu}(\omega_{\rm pr},\omega;\tau)$ for $\nu=x,y$ not only are similarly positioned, but their number is directly proportional to the delay time τ.

In practice, instead of the Fourier transform introduced in Eq. (38), one performs

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)=\int_{\omega_{\rm min}}^{\omega_{\rm max}}\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},\omega;\tau)\times\exp[-i(\omega-\omega_{\rm pr})t]d\omega\quad\text{for } t\geqslant\tau>0,$$

which introduces so-called leakage, namely, noticeable, unphysical oscillations in $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)$ as a function of t. This

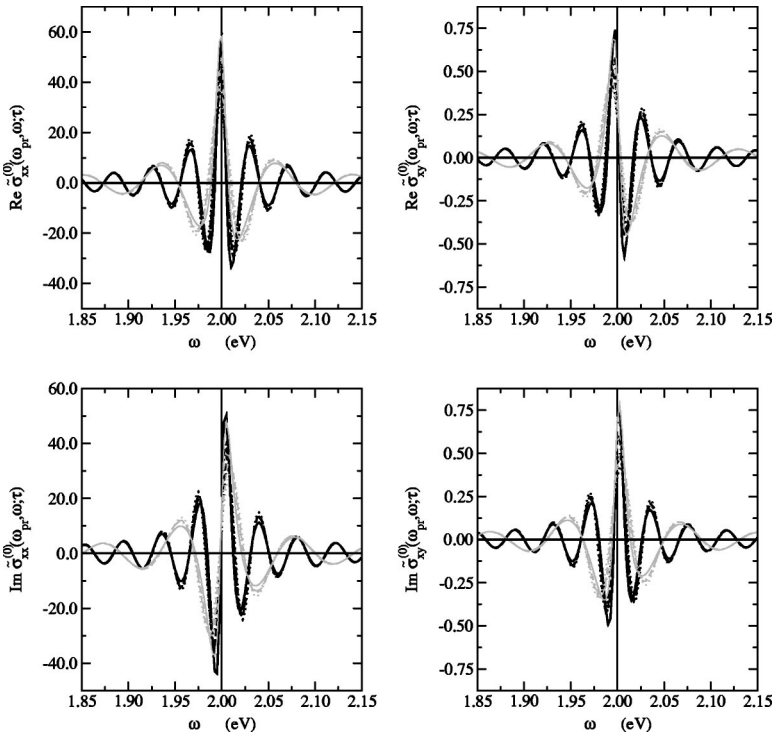

FIG. 3. Strictly linear, frequency- and delay time-dependent (in grey for $\tau=50$ fs and in black for $\tau=100$ fs) dimensionless zeroth order dynamic conductivity $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)$ in the case of fcc Ni(100), when the probe pulse has a carrier frequency $\omega_{\mathrm{pr}}=2$ eV (marked by a thin vertical line), a duration $\Delta t=60$ fs (assumed as FWHM) and an envelope of a double exponential (full line), a Gaussian (dotted line), a hyperbolic secant (dashed line), and a Lorentzian shape (dotted-dashed line), respectively.

leakage can be eliminated multiplying $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)$ with a window function $w(\omega-\omega_{\mathrm{pr}};\omega_c)$ centered at ω_{pr}, for which one assumes that all $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)$ values necessary for the Fourier transform are in a symmetric interval $[\omega_{\mathrm{pr}}-\omega_c,\omega_{\mathrm{pr}}+\omega_c]$ around ω_{pr}, where $\omega_c=\min(\omega_{\mathrm{pr}}-\omega_{\min},\omega_{\max}-\omega_{\mathrm{pr}})>0$. Hence by considering

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)=\int_{\omega_{\min}}^{\omega_{\max}} w(\omega-\omega_{\mathrm{pr}};\omega_c)\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)\times\exp[-i(\omega-\omega_{\mathrm{pr}})t]d\omega,$$

the time-dependent zeroth order dynamic conductivity is obtained as the convolution of $\mathcal{F}[\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)]_t$ with the Fourier transform of the window function $\mathcal{F}[w(\omega-\omega_{\mathrm{pr}};\omega_c)]_t$. There are several window functions and windowing methods known in the literature.[26] For illustrative purposes in Fig. 4 a Gauss window with a damping $\alpha\in\mathbb{R}$ has been used

$$w(\omega-\omega_{\mathrm{pr}};\omega_c)=\exp\left[-\alpha\left(\pi\frac{\omega-\omega_{\mathrm{pr}}}{\omega_c}\right)^2\right]\quad \text{if } |\omega-\omega_{\mathrm{pr}}|<\omega_c.$$

By inspecting Fig. 4 one immediately observes that the strictly linear time-dependent dynamic conductivity, independent of the delay time τ, is a function of the time interval $t-\tau$ (measured immediately after the probe pulse hits the sample) rather than a function of t and τ, i.e.,

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},t;\tau)=\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},t-\tau). \tag{46}$$

The fact that Eq. (46) applies in general for an arbitrary carrier frequency and duration of the probe pulse, is a direct consequence of the Fourier transform of $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},\omega;\tau)$ as given by Eq. (37). This means that independent of whether one starts the probe pulse to interact with the pump-excited system or not, all elements of the strictly linear time-dependent dynamic conductivity tensor vanish at the very moment the probe pulse hits the sample. The other feature of the zeroth order dynamic conductivity shown in Fig. 4, namely

$$\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},t-\tau)\simeq\tilde{\sigma}_{\mu\nu}(\omega_{\mathrm{pr}})\tilde{\mathcal{E}}_{\mathrm{pr}}(t-\tau), \tag{47}$$

is valid if and only if

$$\tilde{\sigma}_{\mu\nu}(\omega)\simeq\tilde{\sigma}_{\mu\nu}(\omega_{\mathrm{pr}}),\quad \forall\ \omega\in[\omega_{\mathrm{pr}}-\Delta\omega,\omega_{\mathrm{pr}}-\Delta\omega],$$

namely when the changes in the optical conductivity of the system in the vicinity of the carrier frequency ω_{pr} can be neglected, see also Eq. (37). Because in case of fcc Ni(100) the optical conductivity $\tilde{\sigma}_{\mu\nu}(\omega)$ is almost a constant function of ω in the close vicinity of 2 eV, $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\mathrm{pr}},t-\tau)$ can be approximated sufficiently well by Eq. (47), if the carrier frequency of the probe pulse $\omega_{\mathrm{pr}}=2$ eV, see Fig. 4. As a direct consequence of Eq. (47), one observes that independent of the pulse envelope the ratio of the strictly linear dynamic conductivity tensor elements

$$\frac{\tilde{\sigma}^{(0)}_{xy}(\omega_{\mathrm{pr}},t-\tau)}{\tilde{\sigma}^{(0)}_{xx}(\omega_{\mathrm{pr}},t-\tau)}\simeq\frac{\tilde{\sigma}_{xy}(\omega_{\mathrm{pr}})}{\tilde{\sigma}_{xx}(\omega_{\mathrm{pr}})},\quad \forall\ t>\tau, \tag{48}$$

is not a time-dependent quantity. For fcc Ni(100), e.g., and $\omega_{\mathrm{pr}}=2$ eV, $\tilde{\sigma}_{xy}(\omega_{\mathrm{pr}})/\tilde{\sigma}_{xx}(\omega_{\mathrm{pr}})\simeq(1+i)/100$. If Eq. (47) applies, due to the difference in magnitude between the off-diagonal and diagonal tensor elements, the off-diagonal elements fall off faster below a given threshold than the diagonal ones. For example, by considering the lowest detectable limit of the conductivity, namely 0.001 (10^{15} Hz), one can conclude that fcc Ni(100) is completely demagnetized in approximately 100 fs after a Gaussian probe pulse

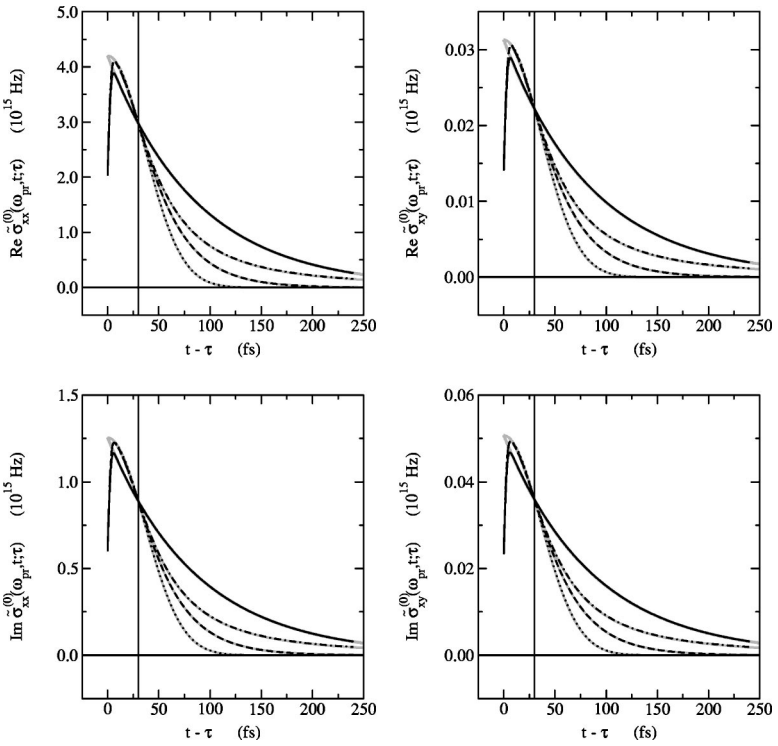

FIG. 4. Gauss-windowed (damping factor $\alpha=3$) strictly linear, time- and delay time-dependent optical conductivity $\tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)$ as compared with $\tilde{\mathcal{E}}_{\rm pr}(t-\tau)\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr})$ shown in grey, where $\tilde{\mathcal{E}}_{\rm pr}(t-\tau)$ is the envelope of the probe pulse (double exponential, full line; Gaussian, dotted line; hyperbolic secant, dashed line; and Lorentzian, dotted-dashed line) with a duration of $\Delta t_{\rm FWHM}=60$ fs (thin vertical lines mark $t-\tau=\Delta t_{\rm FWHM}/2$) and a carrier frequency of $\omega_{\rm pr}=2$ eV. Here $\tilde{\sigma}_{xx}(\omega_{\rm pr})=4.2036+1.2551i(10^{15}$ Hz) and $\tilde{\sigma}_{xy}(\omega_{\rm pr})=0.031\,305+0.050\,733\,i(10^{15}$ Hz), respectively.

($\omega_{\rm pr}=2$ eV and $\Delta t_{\rm FWHM}=60$ fs) hits the sample (see Fig. 4), because for $t-\tau=100$ fs, $\tilde{\sigma}^{(0)}_{xx}(\omega_{\rm pr},t-\tau)=(0.769\,30+0.209\,20\,i)\times10^{-1}$ and $\tilde{\sigma}^{(0)}_{xy}(\omega_{\rm pr},t-\tau)=(0.619\,80+0.994\,29\,i)\times10^{-3}\simeq0$, both in units of 10^{15} Hz. In comparison with the experimentally found demagnetization time for Ni of 80 fs,[52] our result of 100 fs seems to be in reasonably good agreement considering that the first order dynamic conductivity has not been included. Note that even when Eq. (48) holds, the Kerr rotation and ellipticity angles are still time-dependent quantities, because Eq. (48) provides only the prefactor entering the expression for the complex Kerr angle within the two-media approach,[53] while the term multiplied by this prefactor is function of $\tilde{\sigma}^{(0)}_{xx}(\omega_{\rm pr},t-\tau)$.

Another interesting aspect is that the time-integrated strictly linear (zeroth order) dynamic conductivity as obtained from Eq. (38) by using Eqs. (18) and (37),

$$\int_{\tau}^{\infty} dt\ \tilde{\sigma}^{(0)}_{\mu\nu}(\omega_{\rm pr},t;\tau)=\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr})\int_{0}^{\infty}\tilde{\mathcal{E}}_{\rm pr}(t)dt,$$

is independent of the delay time τ and a constant quantity proportional to the optical conductivity $\tilde{\sigma}_{\mu\nu}(\omega_{\rm pr})$ and to the duration Δt of the probe pulse.

VI. SUMMARY

A response theory of pump-probe experiments has been developed in Sec. III B by using the slowly varying envelope approximation for the pulses and neglecting their overlap in time. Although the latter assumption restricts the applicability of the theory to so-called incoherent ultrafast processes like "spin dynamics," its advantages are obvious, (i) it allows one to treat the effect of each of the pulses separately, (ii) linearization of the densities in both pump- and probe-excited states, see Eqs. (22) and (27), directly leads to a formally linear response theory, which also includes second order response functions of the investigated system, see Eqs. (32) and (35). In addition, the current-current and three-current correlations needed to evaluate the dynamic conductivity must be known only for the initial equilibrium state of the system, see Sec. IV. In spite of this feature of the proposed scheme, but unlike the Kubo response theory, in here the zeroth order (strictly linear) and first order conductivities, as can be seen from Eqs. (37) and (39), do depend both on the characteristics of the pulses.

The proposed formally linear response theory for pump-probe experiments was illustrated by calculating the time-dependent strictly linear (zeroth order) conductivity for fcc Ni(100) in terms of the spin-polarized relativistic screened Korringa-Kohn-Rostoker (SKKR) method. The obtained demagnetization time of about 100 fs (depending on the probe-pulse envelope) fits rather well to the experimentally found value of 80 fs.

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