Simple theory of hot electron dynamics observed by femtosecond ellipsometry

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The dynamics of the linear and angular momenta of hot electrons in metals are of key importance for the design and operation of hot electron devices such as spin and tunnel valve transistors. The corresponding relaxation times are expected to lie in the subpicosecond range and must be studied with experimental techniques of adequate (femtosecond) temporal resolution. Here we report a simple theory of the ultrafast ellipsometric response of metals after excitation with femtosecond optical pulses. Although developed in the relaxation time approximation, the theory allows electron linear and angular momentum relaxation times to be extracted. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2172576]

The time scales for the linear and angular momentum dynamics of electrons and holes are important for transport phenomena, where the carrier energy, direction of motion, and spin polarization play a significant role.[1,2] The corresponding spin, momentum, and energy scattering times are expected to be of the order of a few femtoseconds.[3] Using femtosecond optical pump-probe measurements, we observed the combined contributions of the specular inverse Faraday effect (SIFE) and specular optical Kerr effect (SOKE) to the ellipsometric response of metals to optical excitation.[4,5] After separation of the two effects, a small time delay was observed to occur between the SIFE and SOKE signals, while the SIFE and SOKE signals were themselves found to contain additional unexpected structure. This was qualitatively interpreted in terms of the finite relaxation time values for the linear (SOKE) and circular (SIFE) polarizations of the sample, and hence, the electronic linear and angular momenta of hot electrons. In this paper, we develop a theory in the relaxation time approximation that allows the signals from Refs. 4 and 5 to be fitted and certain relaxation times to be determined.

The magnitudes of the SIFE and SOKE depend upon the nonvanishing components of the third-order optical susceptibility tensor $\chi^{(3)}$ and the pump intensity and polarization.[6] The dependence upon the Stokes parameters of the pump was calculated by Popov *et al.* in the continuous wave excitation limit and specialized to the case of circular and linear pump polarizations.[7] Results for the elliptically polarized pump were later given in order to analyze the data of Wilks and Hicken.[8] To apply the theories to a femtosecond pump-probe experiment, an additional assumption had to be invoked that the signal should have the shape of the pump-probe intensity cross correlation and the magnitude derived from the continuous wave model.[6,8] Here we explicitly include the electron momentum relaxation within the description of the shape of time-resolved SIFE and SOKE signals obtained from metallic samples.[9]

The probability of optical absorption of a photon with energy $E=\hbar\omega$ can be described as $\int_{-\infty}^{+\infty} N(\varepsilon)|M(\varepsilon,\varepsilon+E)|H(\varepsilon+E)d\varepsilon$, where $N(\varepsilon)$ and $H(\varepsilon)$ are the total numbers of electrons and holes per unit energy at the energy level ε. The function $M(\varepsilon,\varepsilon+E)$ is proportional to the probability of an electron transition from an individual state at energy ε to an individual state at energy $\varepsilon+E$ upon absorption of a photon of energy E. The probe signal in optical pump-probe experiments is proportional to pump-induced changes in the absorption probability $\Delta S(E,t)$. By retaining only the terms in $\Delta S(E)$ that are of first order in the numbers of nonequilibrium electrons $n(\varepsilon,t)$ and holes $h(\varepsilon,t)$ at energy level ε, we obtain

$$\begin{aligned}\Delta S(E,t) \propto & \int_{\varepsilon_F-2E}^{\varepsilon_F-E} N_0(\varepsilon)|M(\varepsilon,\varepsilon+E)|h(\varepsilon+E,t)d\varepsilon \\ & - \int_{\varepsilon_F-E}^{\varepsilon_F} N_0(\varepsilon)|M(\varepsilon,\varepsilon+E)|n(\varepsilon+E,t)d\varepsilon \\ & - \int_{\varepsilon_F-E}^{\varepsilon_F} h(\varepsilon,t)|M(\varepsilon,\varepsilon+E)|H_0(\varepsilon+E)d\varepsilon \\ & + \int_{\varepsilon_F}^{\varepsilon_F+E} n(\varepsilon,t)|M(\varepsilon,\varepsilon+E)|H_0(\varepsilon+E)d\varepsilon, \quad (1)\end{aligned}$$

where N_0 and H_0 are the unperturbed numbers of electrons and holes, respectively. We neglect the modification of $M(\varepsilon,\varepsilon+E)$ by the pump and consider only the effects of the pump-induced electron redistribution within the phase space.

In the SIFE and SOKE, the sample acquires a linear and (or) circular polarization upon absorption of an intense elliptically polarized pump pulse that results from the transfer of linear and (or) angular momenta to the optically excited hot electrons. The linearly polarized probe pulse can be represented as a superposition of either two circularly polarized pulses with opposite helicities or two linearly polarized pulses with polarization axes that are parallel and perpendicular to that of the pump. The probe polarization is then modified due to the difference in the refraction and absorption of the two circularly polarized components for the SIFE

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and (or) the linearly polarized components for the SOKE. We describe the SIFE and SOKE signals $\Delta S_P(E,t)$ by Eq. (1) in which $n(\varepsilon,t)$ and $h(\varepsilon,t)$ are replaced by $\delta n_P(\varepsilon,t)$ and $\delta h_P(\varepsilon,t)$, and P denotes either the SIFE or the SOKE. $\delta n_P(\varepsilon,t)$ is the difference in the number of electrons with the angular momentum parallel and antiparallel to the pump linear momentum in the case of the SIFE, and the difference in the number of electrons with the linear momentum parallel and perpendicular to the axis of linear birefringence in the case of the SOKE. Similarly, $\delta h_P(\varepsilon,t)$ represents the modified momentum distribution of the holes. The orientation of the electron linear momentum vector varies continuously, and hence, our assignment of an electron to one of the two populations with orthogonal linear momentum is an assumption. Likewise, the angular momentum may occupy a range of values depending on the coupling scheme within the metal. To account for the transient rotation and ellipticity of the probe pulse, $\Delta S_P(E,t)$ and $M(\varepsilon,\varepsilon+E)$ are now considered to be complex numbers, so that the transient rotation and ellipticity are given by the real and imaginary parts of $\Delta S_P(E,t)$. We also assume that the sample is isotropic and nonmagnetic, and so $M(\varepsilon,\varepsilon+E)$ does not depend on the electronic angular and linear momenta.

Let us consider the number of electrons or holes at an energy level ε. We assume that every electron with this energy is in one of the four states defined by the two possible values of the linear momentum and the angular momentum. The number of electrons or holes changes either via elastic scattering between these four states or via inelastic scattering from the energy level. In the relaxation time approximation, both these processes and the excitation by the pump pulse can be described by the following system of rate equations:

$$\dot{\rho}^{\uparrow Y} = -\alpha^{\uparrow Y}\rho^{\uparrow Y} + \alpha^{\uparrow\uparrow YZ}\rho^{\uparrow Z} + \alpha^{\uparrow\downarrow YY}\rho^{\downarrow Y} + \alpha^{\uparrow\downarrow YZ}\rho^{\downarrow Z} + sqI(t),$$

$$\dot{\rho}^{\uparrow Z} = \alpha^{\uparrow\uparrow ZY}\rho^{\uparrow Y} - \alpha^{\uparrow Z}\rho^{\uparrow Z} + \alpha^{\uparrow\downarrow ZY}\rho^{\downarrow Y} + \alpha^{\uparrow\downarrow ZZ}\rho^{\downarrow Z} + rqI(t),$$

$$\dot{\rho}^{\downarrow Y} = \alpha^{\downarrow\uparrow YY}\rho^{\uparrow Y} + \alpha^{\downarrow\uparrow YZ}\rho^{\uparrow Z} - \alpha^{\downarrow Y}\rho^{\downarrow Y} + \alpha^{\downarrow\downarrow YZ}\rho^{\downarrow Z} + spI(t),$$

$$\dot{\rho}^{\downarrow Z} = \alpha^{\downarrow\uparrow ZY}\rho^{\uparrow Y} + \alpha^{\downarrow\uparrow ZZ}\rho^{\uparrow Z} + \alpha^{\downarrow\downarrow ZY}\rho^{\downarrow Y} - \alpha^{\downarrow Z}\rho^{\downarrow Z} + rpI(t), \tag{2}$$

where the source terms in the right-hand side are equal to $I(E,\varepsilon,x,t)=[2A_0\zeta_{e(h)}(E,\varepsilon)/\sigma E]\sqrt{\ln 2/\pi}\exp\{-(x/l_0)-[4\ln(2)t^2/\sigma^2]\}$, where $\zeta_e(E,\varepsilon)$ and $\zeta_h(E,\varepsilon)$ are electron and hole absorption coefficients, respectively. $\rho_j^{\sigma 1\sigma 2}$ is the number of electrons $[n_P(\varepsilon,t)]$ or holes $[h_P(\varepsilon,t)]$ in state $\sigma 1\sigma 2$, where $\sigma 1$ represents either $\uparrow$ ("angular momentum up") or $\downarrow$ ("angular momentum down") and $\sigma 2$ represents either Y (linear momentum parallel to the y axis) or Z (linear momentum parallel to the z axis). $\alpha^{\sigma 1\sigma 2}$ is the total scattering rate (inelastic and elastic processes) for state $\sigma 1\sigma 2$. $\alpha^{\sigma 1\sigma 1'\sigma 2\sigma 2'}$ is the elastic scattering rate from state $\sigma 1'\sigma 2'$ to state $\sigma 1\sigma 2$. A_0 is the pump pulse energy per unit excited volume, which is equal to Sl_0, where S is the pump spot area and l_0 is the optical skin depth of the material. σ is the full width at half maximum (FWHM) of the pump pulse; t is the time variable; and p and q describe the efficiency of the angular momentum orientation so that for a linearly polarized pump $p=q=0.5$, and for left and right circular polarizations $p=1$, $q=0$, and $p=0$, $q=1$, respectively. Let us choose the coordinate system so that the z axis is at 45° from the probe plane of polarization. Then, r and s describe the efficiency of the linear momentum orientation parallel to the z and y axes, respectively. With the pump either circularly polarized or linearly polarized parallel or perpendicular to the probe, $r=s=0.5$, while for linear polarization parallel to the z and y axis, $r=1$, $s=0$, and $r=0$, $s=1$, respectively.

Let us define $\rho_0=\rho^{\uparrow Y}+\rho^{\uparrow Z}+\rho^{\downarrow Y}+\rho^{\downarrow Z}$, $\rho_{\updownarrow}=(\rho^{\uparrow Y}+\rho^{\uparrow Z})-(\rho^{\downarrow Y}+\rho^{\downarrow Z})$, and $\rho_{YZ}=(\rho^{\uparrow Z}+\rho^{\downarrow Z})-(\rho^{\uparrow Y}+\rho^{\downarrow Y})$, where ρ_0 is the total number of hot electrons (holes), and $\rho_{\updownarrow}$ and ρ_{YZ} are the angular and linear momentum polarizations of the hot electron (hole) population $\delta n_{\mathrm{SIFE}}(\varepsilon,t)$ and $\delta n_{\mathrm{SOKE}}(\varepsilon,t)$ $[\delta h_{\mathrm{SIFE}}(\varepsilon,t)$ and $\delta h_{\mathrm{SOKE}}(\varepsilon,t)]$.

If we assume that the total scattering rates $\alpha^{\sigma 1\sigma 2}$ are all equal to the same constant α, and that the scattering rates $\alpha^{\sigma 1\sigma 1'\sigma 2\sigma 2'}$ are equal to $\alpha^{\updownarrow}$ if $\sigma 1'\neq\sigma 1$ and $\sigma 2'=\sigma 2$, α^{YZ} if $\alpha 1'=\sigma 1$ and $\alpha 2'\neq\sigma 2$, and α^{X} if $\alpha 1'\neq\sigma 1$ and $\sigma 2'\neq\sigma 2$, then we obtain

$$\rho^* = \frac{A_0\zeta_{n(h)}B^*\exp[-(x/l_0)+(\sigma\alpha^*/4\sqrt{\ln 2})^2-\alpha^* t]}{2E} \times\left[1+\mathrm{erf}\left(\frac{2\sqrt{\ln 2}t}{\sigma}-\frac{\sigma\alpha^*}{4\sqrt{\ln 2}}\right)\right], \tag{3}$$

where * refers to either 0, $\updownarrow$, or YZ, and $\alpha_0=\alpha-\alpha^{YZ}-\alpha^{\updownarrow}-\alpha^{X}$, $\alpha_{\updownarrow}=\alpha-\alpha^{YZ}+\alpha^{\updownarrow}+\alpha^{X}$, $\alpha_{YZ}=\alpha+\alpha^{YZ}-\alpha^{\updownarrow}+\alpha^{X}$, $B_0=1$, $B_{\updownarrow}=(r+s)(q-p)$, and $B_{YZ}=(r-s)(q+p)$. Note that the relaxation rates depend on the energy. For example, the Fermi liquid theory predicts that the inelastic scattering rate of electrons is proportional to the square of the excess energy relative to the Fermi level. Hence, one needs to evaluate the integrals in (1). However, in our simplified approach we avoid this in the following way. We divide the integration interval in every integral in (1) into several parts so that in each interval we can neglect the dependence of the various relaxation rates on energy. Then we can write $\Delta S_P(E,t)$ as

$$\Delta S_P(E,t) \propto \sum_{j=1}^{\xi_h} k_{h,j}^P \delta h_P(\alpha_{h,j}^P,t) + \sum_{j=1}^{\xi_n} k_{n,j}^P \delta n_P(\alpha_{n,j}^P,t), \tag{4}$$

where the complex coefficients $k_{h,j}^P$ and $k_{n,j}^P$ are given by

$$k_{h,j}^P = \int_{\varepsilon_{h,j}}^{\varepsilon_{h,j+1}} A_0\zeta_h(E,\varepsilon)B^*[N_0(\varepsilon-E)M(\varepsilon-E,\varepsilon) - M(\varepsilon,\varepsilon+E)H_0(\varepsilon+E)]d\varepsilon,$$

$$k_{n,j}^P = \int_{\varepsilon_{n,j}}^{\varepsilon_{n,j+1}} A_0\zeta_n(E,\varepsilon)B^*[M(\varepsilon,\varepsilon+E)H_0(\varepsilon+E) - N_0(\varepsilon-E)M(\varepsilon-E,\varepsilon)]d\varepsilon,$$

$$\varepsilon_{h,1}=\varepsilon_F-E,\quad \varepsilon_{h,\xi_h+1}=\varepsilon_F,\quad \varepsilon_{n,1}=\varepsilon_F,\quad \varepsilon_{n,\xi_n+1}=\varepsilon_F+E,$$

and the functions $\delta h_P(\alpha_{h,j}^P,t)$ and $\delta n_P(\alpha_{n,j}^P,t)$ are defined by Eq. (3), but without the prefactor $A_0\zeta_{e(h)}B^*$. In the simplest case when one neglects the energy dependence of the scat-

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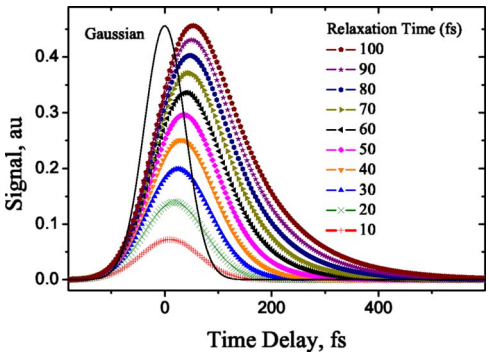

FIG. 1. (Color online) The dependence of the signal shape defined in Eq. (3) on the relaxation time $\tau^* = 1/\alpha^*$ is illustrated. The FWHM was set to 90 fs. The signal was convolved with the temporal form of the probe intensity.

tering rate, there are only two terms in the right-hand side of (4): one for electrons and one for holes.

Let us now assume that parameters r, s, q, and p must depend on the orientation of the quarter wave plate φ in the experiment with an elliptically polarized pump as $r=\frac{1}{2}(1+\sin 4\varphi)$, $s=\frac{1}{2}(1-\sin 4\varphi)$, $q=\frac{1}{2}(1+\sin 2\varphi)$, and $p=\frac{1}{2}(1-\sin 2\varphi)$; and on the orientation of the polarizer ψ in an experiment with a linearly polarized pump as $r=\frac{1}{2}(1+\sin 2\psi)$, $s=\frac{1}{2}(1-\sin 2\psi)$, $q=p=\frac{1}{2}$. Assuming that the rotation and ellipticity signals are equal to the real and imaginary parts of (4), respectively, then the SIFE and SOKE contributions to the time-resolved signals described by Eq. (4) will vary as predicted by the equations from Ref. 8.

The main result of this calculation is that a transient SIFE or SOKE signal may, in general, contain several contributions of the form (3), convolved with the spatio-temporal form of the probe intensity. The experimental data from Ref. 5 required two such contributions. Let us consider a signal that is described by a single term of the type given in (3). The dependence of the signal shape on the relaxation time $\tau^* = 1/\alpha^*$ is illustrated in Fig. 1. Increasing the relaxation time gives rise to four effects: the time-resolved signal becomes larger and more asymmetric, it broadens, and the position of the maximum shifts towards positive time delay.

So far, we have neglected any transport effects. However, ballistic electron transport takes place on femtosecond time scales and may reduce the signal if hot electrons are able to leave the excited volume of the sample.[10] This may be accounted for crudely by replacing the skin depth l_0 in the equations above with $l_0 + l_{\mathrm{bal}}$, where the ballistic range l_{bal} is understood to be the average distance a hot electron can travel from the surface before it is first scattered. This distance depends on the average collision frequency and the electron group velocity and has a value of about 100 nm for noble metals.

In summary, we have developed a simple analytical model that allows time-resolved signals from ellipsometric pump-probe measurements, such as those in Refs. 4, 5, and 8, to be fitted. The fitting provides values for electronic relaxation times that are important for device operation, while the model also represents a useful starting point for a more rigorous analysis that takes into account the realistic electronic band structure of the sample.

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