Nonlinear absorption of ultrashort laser pulses in thin metal films

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Self-consistent simulations of the ultrafast electron dynamics in thin metal films were performed. A regime of nonlinear oscillations was observed that corresponds to ballistic electrons bouncing back and forth against the films' surfaces. When an oscillatory laser field is applied to the film, the field energy is partially absorbed by the electron gas. Maximum absorption occurs when the period of the external field matches the period of the nonlinear oscillations, which, for sodium films, lies in the infrared range. Possible experimental implementations are discussed. © 2005 Optical Society of America

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The recent progress in the study of metallic nanostructures is due mainly to the development of ultrafast spectroscopy techniques that allow the experimentalist to probe the electron dynamics on a femtosecond (and, more recently, attosecond) time scale. Typical pump–probe experiments involve perturbing the system with a first stronger pulse followed by a second weaker pulse that acts as a diagnostic tool. By modulating the relative amplitude of the signals as well as the delay between the pump and the probe, it is possible to assess with great precision the dynamic relaxation of the electron gas.[1–4]

In this Letter we focus on the ultrafast electron dynamics in thin metallic films. Several experiments have shown[1,2] that electron transport in thin metal films occurs on a femtosecond time scale and involves ballistic electrons traveling at the Fermi velocity of the metal, v_F. These findings were corroborated by accurate numerical simulations[5] that highlighted a regime of slow nonlinear oscillations corresponding to ballistic electrons bouncing back and forth on the films' surfaces. These oscillations were recently measured in transient reflection experiments on thin gold films.[6] The existence of this regime prompted us to analyze the possibility of boosting energy absorption in the film by optically exciting the electron gas at the frequency of the nonlinear oscillations.

In the rest of this Letter, time is normalized in units of the inverse plasmon frequency $\omega_{\mathrm{pe}}^{-1}$, velocity in units of Fermi speed v_F, and length in units of $L_F=v_F/\omega_{\mathrm{pe}}$. For alkali metals we have $L_F=0.59(r_s/a_0)^{1/2}$ Å (1 Å=0.1 nm), $\omega_{\mathrm{pe}}^{-1}=1.33\times10^{-2}(r_s/a_0)^{3/2}$ fs, $E_F=50.11(r_s/a_0)^{-2}$ eV, and $T_F=5.82\times10^5(r_s/a_0)^{-2}$ K, where r_s is the Wigner–Seitz radius. We concentrate primarily on sodium films, for which $r_s=4a_0$ ($a_0=0.529$ Å is the Bohr radius).

We consider a system of electrons interacting via a Coulomb potential and confined within a slab of thickness L. The ion background is represented by a fixed density with soft edges, $n_i(x)=\bar{n}_i\{1+\exp[(|x|-L/2)/\sigma_i]\}^{-1}$, where $\bar{n}_i=3/(4\pi r_s^3)$ is the ion density of the bulk metal and $\sigma_i \ll L$ is a diffuseness parameter.[7] In this jellium model, the self-consistent electrostatic potential depends only on the coordinate normal to the surface (here denoted x). Thus the motion of an electron parallel to the surface of the film is completely decoupled from the motion normal to the surface, and a one-dimensional model can be adopted.

The electrons are initially prepared in a Fermi–Dirac equilibrium at a finite (but small) temperature. They are subsequently excited by imposition of a constant velocity shift $\Delta v_x=0.08v_F$ on the initial distribution.[7] This procedure is appropriate when no linear momentum is transferred parallel to the plane of the surface (i.e., when $q_\parallel=0$) and is relevant to the excitation of the film with optical pulses.[8] For $q_\parallel=0$, only longitudinal modes (volume plasmon with $\omega=\omega_{\mathrm{pe}}$) can be excited.

After the excitation is applied, the electron distribution function $f_e(x,v_x,t)$ starts evolving in time according to the semiclassical Vlasov equation

$$\frac{\partial f_e}{\partial t}+v_x\frac{\partial f_e}{\partial x}+\frac{e}{m_e}\frac{\partial \phi}{\partial x}\frac{\partial f_e}{\partial v_x}=0, \tag{1}$$

where m_e is the electron mass and e denotes the absolute electron charge. The electrostatic potential is obtained self-consistently, at each instant, from Poisson's equation

$$\frac{\mathrm{d}^2\phi}{\mathrm{d}x^2}=\frac{e}{\varepsilon_0}[n_e(x,t)-n_i(x)], \tag{2}$$

with $n_e=\int f_e \mathrm{d}v_x$. As a reference case, we studied a sodium film with initial temperature $T_e=0.008T_F\simeq300$ K, diffuseness parameter $\sigma_i=0.3L_F$, and thickness $L=50L_F\simeq59$ Å.[8]

The time evolution of the thermal E_{th} and center-of-mass E_{cm} energies was analyzed[5] (Fig. 1). During an initial rapidly oscillating phase, E_{cm} is almost entirely converted into thermal energy (Landau damping). After saturation, a slowly oscillating regime appears, with a period equal to $50\,\omega_{\mathrm{pe}}^{-1}\approx5.3$ fs. This period is close to the time of flight of electrons traveling at the Fermi velocity and bouncing back and forth on the film's surfaces (further details are provided in our previous paper[5]).

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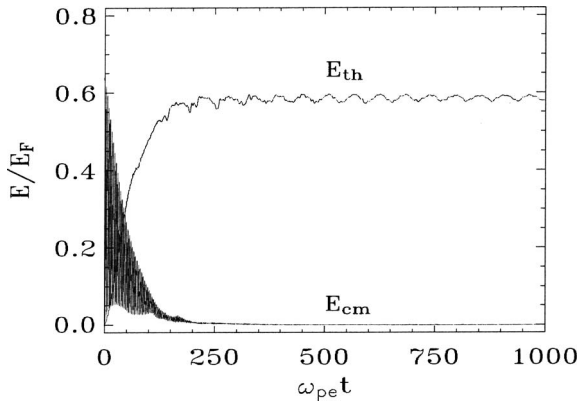

Fig. 1. Initial time evolution of the thermal and center-of-mass energies for film thickness $L=50L_F$.

The nonlinear oscillations above appear for all reasonable values of the physical parameters. Preliminary studies suggest that electron–electron collisions do not destroy this regime either, at least for relatively low excitation energies and short times. It is tempting, therefore, to investigate whether some kind of resonant absorption can be achieved when the system is externally excited at the same frequency of the nonlinear oscillations.

A similar situation was investigated by Taguchi *et al.*[9] (building on an idea that is due to Brunel[10]) to simulate the interaction of an argon cluster with a strong laser field ($\approx 10^{15}$ to $\approx 10^{16}$ W/cm^2). In their simulations, the neutral cluster is quickly ionized by the laser field, which heats the electrons to 10 eV. At these temperatures the electrons behave classically and are initially described by a Maxwell–Boltzmann distribution. In that case, the electrons' transit velocity through the cluster is not clearly defined and depends on the intensity of the laser (indeed, in the classical case, there is no natural oscillatory regime like the one shown in Fig. 1). For a degenerate electron gas, the transit velocity is unambiguously given by the Fermi velocity of the metal, and thus we expect an even neater resonance to occur.

Our conjecture can be tested in the following way: At time $\omega_{pe}t=1000$ ($\approx$106 fs for a sodium film), when the oscillatory regime is well established, we switch on a small external electric field, uniform in space and sinusoidal in time with period T: $E_{ext}=E_0\sin(2\pi t/T)$, where E_0 is the (constant) field amplitude. The simulation is then continued with the external field on for another 4000 $\omega_{pe}^{-1}\approx 425$ fs. This situation corresponds to a laser pulse that is switched on quickly and lasts for a duration longer than 425 fs.

For an electron transit velocity exactly equal to v_F we would expect resonance for a laser period $T=2L/v_F$ (=100 in units of ω_{pe}^{-1}). The factor 2 comes from the fact that the electric field must keep the same sign during a transit from one surface to the other and must reverse sign during the return transit. We note that the resonance is expected to fall into the IR domain. Indeed, for a laser period $T=100$ $\omega_{pe}^{-1}=10.6$ fs, the corresponding wavelength is $\lambda=3.2$ μm.

We can estimate the amplitude E_0 of the laser field by noting that the total energy of the laser pulse is $U=(\frac{1}{2}\varepsilon_0 E_0^{\,2})c\tau S$, where c is the speed of light in vacuum, τ is the pulse duration, and S is the surface of the laser spot. Typical values for IR lasers[11] are $S=0.01$ mm^2 and $U=1$ μJ, and, by taking a pulse duration $\tau=400$ fs (similar to the duration used in the simulations), we obtain an electric field $E_0=4.3\times 10^8$ V/m. In the numerical simulations, the electric field is normalized to $\bar{E}\equiv m_e v_F\omega_{pe}/e=1.70\times 10^{12}(r_s/a_0)^{-5/2}$ V/m, yielding $\bar{E}=5.31\times 10^{10}$ V/m for sodium films. Therefore, by taking field amplitude $E_0=0.01\bar{E}$, we get a dimensional value that is realistic for an IR laser pulse. This external field amplitude is an order of magnitude smaller than the self-consistent electric field that is present at the films' surfaces.

The results for the reference case ($L=50L_F$) are shown in Fig. 2, where the electron thermal energy is plotted against time. We observed that the absorption is clearly enhanced for $\omega_{pe}T=106$ and $\omega_{pe}T=150$, whereas for larger or smaller values virtually no energy is absorbed. We also verified that the resonance does not depend on the phase of the external oscillating field.

The resonant period is close, but not exactly equal, to the predicted value $\omega_{pe}T=100$, and the resonance displays a certain broadness. The latter property can be explained by noting that a certain dispersion exists in the electron velocities near v_F that generates a dispersion in the resonant period. If the period is $T=2L/v$, the resonance broadness should be $|\delta T|=(2L/v^2)\delta v$. To estimate the broadness, we plot, in Fig. 3, the variation of the velocity distribution at the center of the film: $\delta f(v_x)=|f_e(x=0,v_x,\omega_{pe}t=1000)-f_e(x=0,v_x,t=0)|$. The distribution is indeed modified near the Fermi velocity, as expected (see also Fig. 3 of Ref. 5). Note that δf is not symmetric about $v_x=0$ because the initial excitation was not symmetric either. On closer inspection, the peaks occur at a velocity slightly smaller (in absolute value) than v_F, roughly

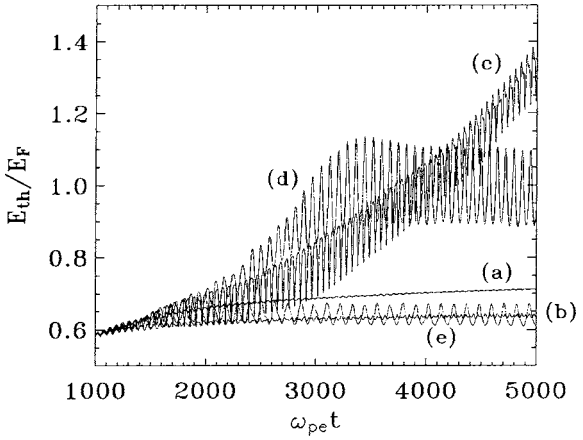

Fig. 2. Time evolution of the thermal energy in the presence of an external electric field. The external field is switched on at $\omega_{pe}t=1000$. $\omega_{pe}T=$(a) 27, (b) 73, (c) 106, (d) 150, (e) 230. The results are for a film of thickness $L=50L_F$.

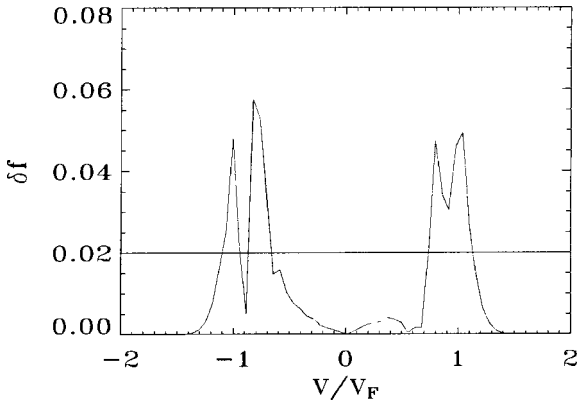

Fig. 3. Variation of the electron velocity distribution with respect to the initial Fermi–Dirac equilibrium, at the center of the film, at time $\omega_{pe}t=1000$.

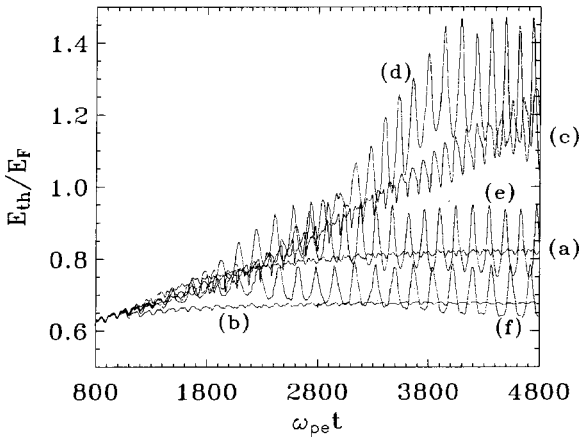

Fig. 4. Same as Fig. 2 for a film of thickness $L=100L_F$. $\omega_{pe}T=$(a) 90, (b) 150, (c) 212, (d) 250, (e) 290, (f) 350.

$|v_x|\simeq 0.9v_F$. Their broadness can be estimated by assuming that a deviation of 2% (relative to the maximum $f_e=1$) is significant. Then v varies in the interval $0.7v_F<v<1.1v_F$ (and equivalently for negative velocities), so $\delta v\simeq 0.4v_F$. This yields a broadness $\omega_{pe}\delta T\simeq 50$, with $90<\omega_{pe}T<140$. This estimate is compatible with the simulation results of Fig. 2, where the resonance has clearly disappeared at $\omega_{pe}T=73,230$.

To test the robustness of this nonlinear resonance effect, we repeated the same numerical experiment with a thicker film, $L=100L_F$ (the initial evolution for this case was shown in our previous paper[5]). The resonant period is expected to scale linearly with the film's thickness, and indeed we observed enhanced absorption for $\omega_{pe}T=212,250$ (Fig. 4).

In contrast, we observed that the resonance virtually disappears for thicker films, $L=200L_F$ or larger. We interpret this result by noting that the existence of the resonance depends on nonequilibrium electrons traveling coherently through the film. The phase space portraits of the electron distribution function (Fig. 3 in our previous work[5]) show a complex structure of traveling vortices. It is probable that, for thicknesses larger than a certain threshold, the necessary coherence is lost, so the resonance cannot manifest itself.

The resonance also disappears for small amplitudes of the external field. For the reference case $L=50L_F$, the resonance is still observed for $E_0/\bar{E}=0.005$ but no longer for $E_0/\bar{E}=0.001$. This result may be related to the fact that the absorbed energy at resonance U_{abs} scales quadratically with the field amplitude: $U_{abs}\propto e^2{E_0}^2T^2/m_e$ (this formula becomes exact for the harmonic oscillator). For small fields the resonance is thus weak, and other factors (e.g., Landau damping) can easily erase it completely.

In summary, we have shown the existence of a nonlinear absorption regime in the electron dynamics of thin metal films. This effect is generic and should not, in principle, depend on the nature of the metal. The resonance occurs in the IR domain and should be accessible via experiments employing ultrafast laser sources with standard specifications. This absorption mechanism could be used as an optical diagnostic technique to determine, for instance, the thickness of the film or to obtain information on the electronic distribution.

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