Electrodynamics of the propagation of ultrashort light pulses in metals

É. M. Belenov, L. G. Grechko, and A. P. Kanavin
P. N. Lebedev Physics Institute, Russian Academy of Sciences, 117924 Moscow, Russia

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The penetration of intense femtosecond light pulses into metals is analyzed. Short light pulses can penetrate into metals and propagate in them even if the field has spectral components at frequencies below the plasma frequency.

1. Laser physics has now succeeded in generating ultrashort light pulses with a length τ_p as short as one oscillation period and with fields $\mathscr{E}$ on the order of atomic fields. For pulses of this sort, the field which penetrates into a metal interacts with the electrons of the metal in a very nonlinear way. This circumstance has implications for the dynamics of the field in the metal. It leads to qualitatively new results in the reflection, penetration, and propagation of ultrashort light pulses as they interact with metals.

2. We consider a linearly polarized ultrashort light pulse which is incident normally from vacuum on a plane metal surface. In the constitutive equations we incorporate the circumstance that the electrons of the metal, which determine the dynamics of the field in the medium, interact not only with the external electromagnetic wave but also with the periodic lattice of the solid. This interaction with the lattice determines the dependence of the electron energy $\epsilon(k)$ on the wave number k and leads through Bragg reflection to umklapp effects when the electrons reach the boundary of the Brillouin zone.

The effect of umklapp processes on the response of a metal to an ultrashort light pulse is taken into the account under the assumption that the field is spatially uniform. We denote by $\Delta\epsilon$ the width of the conduction band. Adopting the approximation of strong coupling along one of the principal crystallographic axes, with which the polarization direction of the external field coincides, we write the electron energy as $\epsilon(k)=\epsilon(k_\perp)+\Delta\epsilon[1-\cos(k_\parallel a)]$, where $k_\parallel$ and $k_\perp$ are the components of the wave vector along and across this axis, and a is the lattice constant.

In the semiclassical approximation, the electron distribution function in the field $\mathscr{E}(t)$ obeys the kinetic equation

$$\frac{\partial f(k,t)}{\partial t}+\frac{e\mathscr{E},(t)}{\hbar}\frac{\partial f(k,t)}{\partial k}=J_{st}(f), \tag{1}$$

where $J_{st}(f)$ is a collision integral. For simplicity we restrict the discussion to the case in which a dominant role is played by elastic collisions of electrons with impurities. We write the collision integral in the form

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$$j_{st}(f)=\int W(k,k')(f(k)-f(k'))dk', \tag{2}$$

where the transition probability $W(k, k')$ is assumed to be a slowly varying quantity. In the reduced-band scheme, umklapp processes at the boundary of the Brillouin zone can be dealt with by means of the boundary conditions

$$f(k_\perp, g/2)=f(k_\perp, -g/2), \tag{3}$$

where g is a reciprocal-lattice vector. Expression (2) effectively reflects the equivalence of the states electrons in momentum space which are shifted with respect to each other by a reciprocal-lattice vector.

From (1) and (2) we find a closed system of equations for the current density in the metal,

$$j=\frac{e}{\hbar}\int\frac{\partial\epsilon}{\partial k}f(k,t)dk,$$

and for the energy of the electron gas in the conduction band, $E=\int\epsilon(k)f(k, t)dk$:

$$\frac{dj}{dt}+\nu j+\left(\frac{2\pi e}{\hbar g}\right)^2\mathscr{E}(E-E_0)=0,\quad \frac{dE}{dt}-\mathscr{E}j=0. \tag{4}$$

Here E_0 is the value of the energy in the case of a uniform energy distribution of the electrons in the conduction band, and ν is the rate of elastic collisions of electrons.

System of equations (4) is equivalent to the system of "shortened" Bloch equations for the level population difference $n\sim E-E_0$, the polarization amplitude $P\sim j$, and the amplitude of the pulse field $\mathscr{E}$ when the applied field is resonant with the transition frequency. Here we are allowing for the circumstance that the pulse length τ_p is shorter than the relaxation time τ_1 of the medium of two-level particles. The time τ_2 is determined by the rate ν, and the transition dipole moment μ is determined by the magnitude of the reciprocal-lattice vector: $\mu=ea=2\pi e/g$.

In metals, the conduction electrons undergo collisions of a variety of types: with each other, with phonons, with impurity atoms, and with other lattice defects. For electron–electron collisions, the electrons, separated from each other by a distance $\sim$1–3 Å, typically traverse comparatively large distances without colliding with each other. Effects of this sort are a consequence of the Pauli principle and the screening of the Coulomb interaction in the medium. The mean free path l of an electron at $T=300$ K is $\sim 10-4$ cm. The time between electron–electron collisions corresponding to this mean free path is $\tau_{ee}\sim l/v_F\sim 10^{-12}$ s, where v_F is the electron velocity at the Fermi surface.[1] With decreasing temperature, τ_{ee} increases as $1/T^2$. The temperature dependence of the electron–phonon interaction time is[2] $\tau_{ep}\sim 1/T$. At sufficiently low temperatures, a governing role is played in kinetic effects by the scattering of electrons by impurities and lattice defects. This scattering is characterized by a time τ_{ed}. If the crystal is sufficiently pure, the time τ_{ed} can be extremely long. For copper at low temperatures, for example, τ_{ed} can be $\sim 10^{-8}$ s.[1]

3. For a sufficiently short field pulse, in the femtosecond length range, $\nu\tau_p \ll 1$, we can ignore the collisional term in the first equation in (4) in comparison with $\partial j/\partial t$. In this case we find from (4) a Josephson time dependence for the current density[1)]

$$j = j_0 \sin\left(\frac{2\pi e}{\hbar g}\int_{-\infty}^{t} \mathscr{E}(t)dt\right), \tag{5}$$

where j_0 is the maximum value of the current, which is determined by the extent to which the conduction band is filled with electrons. Expression (5) corresponds physically to motion of the electron subsystem of the metal as a whole in the conduction band. We also note that in the absence of a two-level system, whose dynamics is described in the approximation of slowly varying amplitudes and phases, the Rabi frequency is governed not by the amplitude of the field but by its value at a given instant.[3,4]

4. The penetration of an ultrashort pulse into a metal and its propagation in the metal are governed by the wave equation

$$\frac{\partial^2 \mathscr{E}}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2 \mathscr{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial j}{\partial t}, \tag{6}$$

along with constitutive equation (5) and the conditions that the electric and magnetic components of the field must be continuous at the boundary of the medium. In a weak field, $\mu \int \mathscr{E}(t)dt \ll \hbar$, for a pulse of frequency ω, we find a linear wave equation from (6) for a medium with a dielectric constant $\epsilon(\omega) = 1 - \omega_{pl}^2/\omega^2$, where ω_{pl} is the plasma frequency of the metal. In the case of a strong field, the dependence of the current on the field becomes very nonlinear, and Eq. (6) reduces to the sine-Gordon equation

$$\frac{\partial^2 \varphi}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2 \varphi}{\partial t^2} = \frac{1}{\lambda^2}\sin(\varphi), \tag{7}$$

where

$$\varphi = \frac{\mu}{\hbar}\int_{-\infty}^{t} \mathscr{E}(t)dt, \quad \lambda^2 = \hbar c^2/4\pi j_0 \mu.$$

We know that among the solutions of (7) there are soliton solutions, in particular, 2π and 0π pulses. In the simplest case, the soliton is a bell-shaped 2π pulse:

$$\mathscr{E}(x,t) = \mathscr{E}_p \operatorname{sech}\left\{\frac{t - x/v}{\tau_p}\right\}, \tag{8}$$

where $\mathscr{E}_p = \hbar/\mu\tau_p$ is the amplitude of the pulse in the medium, τ_p is the length of the pulse, and v is the velocity of the pulse, which is related to τ_p by

$$\frac{1}{v^2} = \frac{1}{c^2} + \frac{\tau^2}{\lambda^2}. \tag{9}$$

5. If we wish to form pulse (8) in a metal by means of an external source,[2)] the incident pulse must have the same temporal shape. In this case the reflection coefficient

at $\omega_{pl}\tau_p \gg 1$ is approximately one, $R \simeq 1-2/(\omega_{pl}\tau_p)$, and the "amplitude" $\mathscr{E}_0$ of the field in the incident wave does not depend on the pulse length. It is determined exclusively by the properties of the medium:

$$\mathscr{E}_0=\sqrt{\frac{\pi\hbar j_0}{\mu}}. \tag{10}$$

Let us plug in some numbers. Setting $\tau_p \sim 10^{-14}$ s, $\mu \sim 1.5 \times 10^{-17}$ esu, $j_0=\zeta e n_e v_g$ (the constant ζ is determined by the initial filling of the Brillouin zone by electrons; for metals such as copper, $\zeta \sim 1-2p_F/g$ has a value $\leqslant 0.1$), $n_e \sim 10^{23}$ cm^{-3}, and $v_g \sim 10^8$ cm/s, we find $j_0 \sim 5 \times 10^{20}$ esu and $\mathscr{E}_p \sim 2 \times 10^6$ V/cm. The total energy of the incident pulse corresponding to this field is $\epsilon_0 \sim 10^2$ mJ/cm^2.

6. Unipolar pulse (8) (a pulse half a wavelength long) can be generated only under conditions of waveguide propagation of the field. The extremely short pulse incident from vacuum must be bipolar and must contain two half-waves; i.e., it must be a one-wavelength pulse. This is a consequence of the equation div$\mathscr{E}=0$ for the field in vacuum. This pulse is obviously a 0π pulse with an area

$$\int_{-\infty}^{+\infty} \mathscr{E}(x,t)dt=0.$$

A bipolar pulse of this sort can be thought of as, for example, a superposition of two waves of the type in (8) which are separated in time (or space): $\mathscr{E}_0(x, t)-\mathscr{E}_0(x, t+\tau)$. In the metal the pulse excites a field $\mathscr{E}_2(x, t)-\mathscr{E}_2(x, t+\tau)$, which is generally not a soliton solution of the sine-Gordon equation. Nevertheless, at sufficiently large values of τ the solution is approximately a soliton solution, and the estimates above are valid for finding its parameters. The pulse in (8), as it propagates further in the medium, converts into a soliton solution of the sine-Gordon equation.[7] The same can be said of an incident pulse which consists of more than one wavelength. In this case the solution of the sine-Gordon equations describes not one but a series of separating 0π pulses. While in the case of a weak field, in which the Fourier components of the pulse will (depending on the frequency ω) either propagate into the metal ($\omega > \omega_{pl}$) or undergo a complete plasma reflection ($\omega < \omega_{pl}$), in strong fields all frequency components of the pulse will propagate into the medium, will interact in a nonlinear way with each other, and will generate solitons which propagate in the metal.

[1)]For the case of a collisionless electron gas in a crystal lattice, expression (5) for the current density can be derived from the equations for the work performed by the field on an electron, $\partial\epsilon/\partial t=eE(t)v_g$, along with the equation describing the time evolution of the group velocity of the electrons, $\partial v_g/\partial t=(eE/\hbar^2)\partial^2\epsilon(k)/\partial k^2$ [see Ref. 1] for the case with an explicit functional dependence $\epsilon(k)$].

[2)]Similar "half-wavelength" pulses have been examined theoretically in connection with the scattering of a strong field in a Raman-active medium[5]. They were also demonstrated experimentally in Ref. 6.

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