Nonlinear optical properties of metals in a strong electromagnetic field

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The change produced in the nonlinear optical properties of the surface of a metal by high-power laser radiation is analyzed. The dispersion relations for the nonlinear current are calculated on the basis of perturbation theory using the density-matrix formalism. The solutions of the kinetic equation which describes the evolution of the distribution function of the electron gas of a metal in the field of an intense electromagnetic wave are chosen as the zeroth approximation for the diagonal elements of the density matrix. It is shown that under the action of an intense heating laser pulse on the surface of a metal, an electron distribution function that is different from the equilibrium function is formed in the irradiation zone, and this substantially changes the dispersion relations for the imaginary part of the nonlinear current j_n'' and the associated nonlinear optical response of the surface. It is shown that the scale of the change in j_n'' is determined by, besides the parameters of the heating laser pulse, the relaxation times of the electron gas in the metal, so that these times can be determined experimentally.

I. INTRODUCTION

There are a large number of theoretical and experimental works on the nonlinear optical response of a surface and the determination of the connection of this response with the microscopic electronic characteristics of the medium (see, for example, Ref. 1). Experimental techniques are trending toward shorter and correspondingly more powerful laser pulses for probing a surface. There naturally arises the question of how this radiation affects the electronic characteristics of a surface, specifically, the electron distribution function (EDF). It is well known that for plasma and gaseous media, a deviation of the EDF from the equilibrium form results in a large change in the nonlinear optical properties of such media, specifically, a higher yield of optical harmonics.[2,3] Such a possibility has not been considered for metals, probably because it is believed that since the electron-electron relaxation time is short ($\tau_{ee}\sim 10^{-12}$–10^{-15} sec), the deviations of the EDF from the equilibrium form are very small and cannot influence the macrophysical characteristics of media. These assertions are poorly substantiated, however, especially for nonlinear optical phenomena, where transitions between excited states can play a decisive role and the nonlinear response times of the medium are at least comparable to τ_{ee}.

A number of experiments[4,5] which indicate directly or indirectly that the electron distribution function of a metal in a strong electromagnetic field is of a nonequilibrium nature have now been performed. It has been proved experimentally that the form of this function affects the reflection coefficient of a metal surface irradiated with high-power laser radiation,[6] and this leads naturally to the idea that a similar effect should occur for the nonlinear optical characteristics of the surface. The methodological principles for analyzing this problem were formulated in Ref. 7, but we know of no attempts to assess the effect of the nonequilibrium nature of the EDF on the nonlinear optical response of a surface. This problem is also of interest because nonlinear optical methods for probing a surface provide a great deal of information.[8] Specifically, they directly yield data on the evolution of the electronic subsystem of a metal in a strong electromagnetic field.

In the present paper we analyze the processes which could change the nonlinear optical properties of a surface in a strong electromagnetic field.

2. FORMULATION OF THE PROBLEM

In its classical formulation, the problem of finding the nonlinear response of a metal surface irradiated with high-power electromagnetic radiation reduces to finding the nonlinear current. The solution of this problem in the perturbation theory approximation is well known and has been analyzed in detail by Bloembergen.[9] In this method, however, the equilibrium thermodynamic Fermi distribution is used as the zeroth-order approximation for the density matrix (see, for example, Ref. 10), and on the basis of the considerations stated above, this approximation may not be adequate for pico- and subpicosecond laser pulses. The alternative to this approximation is to choose a different initial approximation which more accurately reflects the real physical situation.

Proceeding from what we have said above, we reformulate the problem as follows: let two beams of different radiation be incident on the surface of a metal—a heating beam with frequency ω_h and a probing beam with frequencies ω_1 and ω_2. We require that the following conditions be satisfied: 1) the probing field must not affect, to a first approximation, the electron distribution in the conduction band; 2) the heating field alters the electron distribution only as a result of intraband transitions; and, 3) the coupling of the heating field with the electron gas in the metal is local. These conditions

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impose certain restrictions on the parameters of the model chosen. First, the heating radiation must be much more intense than the probing radiation: $I_h \gg I_{1,2}$. In certain ranges of the frequencies and intensities of the heating radiation, the populations can change mainly as a result of intraband transitions. The criterion for the external field to influence the change in the populations and off-diagonal elements of the density matrix as a result of interband transitions can be determined by analyzing the exact solution for a general two-level system (see, for example, Ref. 11). In the case when the frequencies of the heating field $\omega_h \ll \omega_{mn}$ (ω_{mn} is the frequency of an interband transition), the effect of the field on the change in the slowly varying part of the density matrix, resulting, specifically, from two-photon interband transitions, will be small if its intensity

$$I_h \ll [1+(\omega_{mn}T_{mn})^2]/(\hbar^{-1}Q\omega_{mn}T_{mn}^2),$$

where T_{mn} is the relaxation time of the off-diagonal element of the density matrix (transverse relaxation time) and Q is the joint matrix element of the two-photon transition. According to calculations, the transition is saturated in the optical range for heating radiation power densities of the order of hundreds of TW/cm^2, which is much higher than the threshold for surface damage, even for femptosecond pulses. The satisfaction of the third condition requires a normal skin effect, which for most metals occurs in the near-IR range of the spectrum[12,13] and results in the relation $\hbar\omega_h \leqslant 1$ eV.

In the case when the conditions indicated above are satisfied, the effect of the heating field on the electron system of the metal can be analyzed on the basis of the kinetic equation for the electron distribution function, taking the electron-electron and electron-phonon interactions into account. The slowly varying solution obtained can be used as the initial state of the metal surface to describe the nonlinear optical response of the medium to the probe radiation.

3. ELECTRON DISTRIBUTION FUNCTION OF A METAL IN A STRONG ELECTROMAGNETIC FIELD

To analyze the effect of a strong high-frequency electromagnetic field on the electron distribution in a metal, we employ an equation for the one-particle density matrix:

$$\frac{\partial f(\mathbf{p},t)}{\partial t} = S_{ee} + S_{ep}, \tag{1}$$

where $f(\mathbf{p},t)$ is the one-particle density matrix (which, in what follows, we refer to as the electron distribution function), $\mathbf{p}$ is the canonical momentum, and S_{ee} and S_{ep} are the electron-electron and electron-phonon collision integrals, respectively.

The equation (1) was derived in the standard manner from the Heisenberg equations under the assumption that the distribution function is spatially uniform.[14] This assumption is justified in the case of the normal skin effect. In the case at hand it is applicable if[12,13]

$$v/\omega \ll \delta,$$

where v is the characteristic electron velocity, δ is the depth of the skin layer, and ω is the frequency of the field. At optical frequencies ($\omega \sim 10^{14}$–10^{15} sec^{-1}) for metals $\delta \sim 10^{-6}$ cm and $v/\omega \sim 10^{-7}$ cm. Therefore, the dependence of the distribution function on the spatial coordinates and the transport processes associated with this dependence can be neglected.

We obtain an equation for the slowly varying component of the distribution function by averaging Eq. (1) over one period of the field:

$$\frac{\partial f(\mathbf{p})}{\partial t} = -\frac{1}{\tau_{ee}}[f(\mathbf{p})-f_0(\mathbf{p})] + \frac{2\pi}{\hbar}\sum_{n=-\infty}^{\infty}\sum_{\mathbf{k}}|C_{\mathbf{k}}|^2[2N(\mathbf{k},t)+1]J_n^2(\mathbf{ak}) \times [f(\mathbf{p}+\mathbf{k},t)-f(\mathbf{p},t)]\delta(\varepsilon_{\mathbf{p}+\mathbf{k}}-\varepsilon_{\mathbf{p}}-n\hbar\omega), \tag{2}$$

where $f_0(\mathbf{p}) = [\exp(\varepsilon_{\mathbf{p}} - \varepsilon_F/k_BT) + 1]^{-1}$ is the equilibrium distribution function; $N(\mathbf{k},t)$ is the distribution function of phonons in a state with quasimomentum $\mathbf{k}$. Since the electron-phonon coupling constants are small, we assume below that this distribution is the equilibrium distribution with a time-dependent temperature $T_i(\mathbf{r},t)$; J_n are Bessel functions; $\mathbf{a} = e\mathbf{E}_0/m\hbar\omega^2$ is the amplitude of the electron oscillations in a high-frequency field $\mathbf{E} = \mathbf{E}_0\sin(\omega t)$; $\varepsilon_{\mathbf{p}} = p^2/2m$ is the electron energy; and, ε_F is the Fermi energy. The quantity C_k is related to the matrix element for scattering of an electron by an acoustic phonon from the state $\mathbf{p}$ into the state $\mathbf{p}'$:

$$|\mathbf{M}^k_{\mathbf{p},\mathbf{p}'}|^2 = \delta_{\mathbf{p}',\mathbf{p}-\mathbf{k}}\frac{1}{L^3}|C_{\mathbf{k}}|^2.$$

Here, L^3 is the normalization volume.

The first term on the right-hand side of Eq. (2) is the electron-electron collision integral in the τ approximation. We note that the electron-electron collision integral does not contain any terms which correspond to induced absorption (emission) of photons, since the field may be regarded as uniform over distances of the order of the screening length.

The equation (2) shows that the induced absorption (emission) of photons is possible in electron-phonon collisions [the second term on the right-hand side of Eq. (2)].

In studying the kinetic processes in the electron gas of the metal, the fact that, as a rule, the distribution function over the direction of the momentum relaxes much more quickly ($\tau \sim 10^{-14}$ sec) than the energy distribution function ($\tau \sim 10^{-13}$ sec) must be taken into account.[15] This fact makes it possible to simplify the kinetic equation even more. Averaging Eq. (2) over the directions of the momentum and neglecting the contribution of electron-phonon scattering (the term with $n=0$ in the electron-phonon collision integral) to energy relaxation, we obtain

$$\frac{\partial f(\varepsilon)}{\partial t} = -\frac{1}{\tau}[f(\varepsilon)-f_0(\varepsilon)] + \sum_{n=1}^{\infty}\left\{\nu_n^{(+)}[f(\varepsilon+n\hbar\omega) - f(\varepsilon)] + \nu_n^{(-)}[f(\varepsilon-n\hbar\omega)-f(\varepsilon)]\right\}, \tag{3}$$

where $\nu_n^{(\pm)}$ are the electron-phonon collision frequencies, given by

$$\nu_n^{(\pm)}=\frac{2\pi}{\hbar}\sum_{\mathbf{k}}|C_{\mathbf{k}}|^2[2N(\mathbf{k},t)+1]J_n^2(\mathbf{ak})^2\delta(\varepsilon_{\mathbf{p}+\mathbf{k}}-\varepsilon_{\mathbf{p}}\mp n\hbar\omega). \quad (4)$$

The argument of the Bessel function ($\mathbf{ak}\sim X=eE_0v_F/\hbar\omega^2$) is a parameter which determines the multiphonon nature of the process.[16] Since in our problem the field strength is limited by the plasma-formation threshold ($E_0<10^7$ V/cm for femptosecond pulses), $X<1$ always holds, and the contribution of multiphoton processes can be neglected compared to one-photon processes.

A further simplification, which makes possible a simple estimate of the contribution of electron-photon collisions, consists of the following. Let $\nu_1^{(+)}$ and $\nu_1^{(-)}$ be energy-independent and given by

$$\nu_1^{(+)}=\nu, \quad \nu_1^{(-)}=\nu\theta(\varepsilon-\hbar\omega), \quad (5)$$

where ν is an effective electron-photon collision frequency. The physical reason for the appearance of the Heaviside step function $\theta(\varepsilon)$ in Eq. (5) is that the electron energy cannot be negative [see Eq. (4)]. The parameter ν equals the number of photons absorbed by one electron per unit time, and it can be estimated as

$$\nu=W/(n_1\hbar\omega\delta),$$

where W is the power density of the absorbed radiation and n_1 is the effective electron density. Finally, we write the kinetic equation in the form

$$\frac{\partial f(\varepsilon)}{\partial t}=-\frac{1}{\tau_{ee}}[f(\varepsilon)-f_0(\varepsilon)]+\nu[f(\varepsilon+\hbar\omega)-f(\varepsilon)]+\nu\theta(\varepsilon-\hbar\omega)[f(\varepsilon-\hbar\omega)-f(\varepsilon)]. \quad (6)$$

The equation (6) was previously derived in Ref. 17 phenomenologically. The solution of this equation was obtained in Ref. 18 in the form of a series in the small parameter $\kappa=\nu\tau_{ee}$,

$$f(\varepsilon,t)=\sum_{n=0}^{\infty}\kappa_0^n f_n(\varepsilon,t), \quad (7)$$

where κ_0 is a small dimensionless parameter equal to the number of incident field photons per electron over the time τ_{ee}:

$$\kappa_0=W_0\tau_{ee}/(n_1\hbar\omega\delta), \quad (8)$$

where W_0 is the peak power density of the absorbed radiation.

Substituting this expansion into the kinetic equation, we obtain a system of recurrence relations whose solution has the form

$$f_n(\varepsilon,x)=\int_{-\infty}^{x}dy_1\beta(y_1)\int_{-\infty}^{y_1}dy_2\beta(y_2)\ldots\int_{-\infty}^{y_n}dy_n\beta(y_n)\times\Lambda^n[f_0(\varepsilon,T_e(x))]\exp(y_n-x), \quad (9)$$

where $x=t/\tau_{ee}$; $\beta(t)=W(t)/W_0$; $T_e=T_e(\mathbf{r},t)$ is the temperature of the electron gas, as determined from the heat-conduction equation,[19] which makes it possible to explicitly take into account the diffusion spreading of heat; Λ^n is the result of the displacement operator Λ operating n times on the function $f(\varepsilon,t)$:

$$\Lambda[f(\varepsilon)]=f(\varepsilon,\hbar\omega)-f(\varepsilon)+\theta(\varepsilon-\hbar\omega)[f(\varepsilon-\hbar\omega)-f(\varepsilon)]. \quad (10)$$

The expression for $f_n(\varepsilon,x)$ can be substantially simplified if

$$\left|\frac{\partial\Lambda^n[f_0(\varepsilon,x)]}{\partial x}\right|\ll\left|\frac{\partial\beta(x)}{\partial x}\right|.$$

Estimating the derivative on the left-hand side of this inequality, we find that it differs from zero over narrow energy ranges of width $\cong T_e$ adjoining points on the energy axis where $\varepsilon_F-\varepsilon\pm\hbar\omega=0$. Since most observed physical quantities are determined by the higher-order moments of the distribution function, for $T_e\ll\hbar\omega$ the contribution of the time dependence $T_e(t)$ to the kinetics of these quantities does not exceed $T_e/\hbar\omega$. In this case

$$f(\varepsilon,x)=\sum_{n=0}^{\infty}\kappa_0^n\Lambda^n[f_0(\varepsilon,T_e)]\Theta_n(x), \quad (11)$$

where the function $\Theta_n(x)$ describing the temporal behavior of $f(\varepsilon,x)$ is given by

$$\Theta_n(x)=\int_{-\infty}^{x}dy_1\beta(y_1)\int_{-\infty}^{y_1}dy_2\beta(y_2)\ldots\int_{-\infty}^{y_n}dy_n\beta(y_n)\times\exp(y_n-x).$$

It can be shown that for laser pulse durations $\tau_1\gg\tau_{ee}$, the form of the function $\Theta_n(t)$ simplifies, and $\Theta_n(t)=\exp(-nt^2/\tau_i^2)$ in the case of a laser pulse with a Gaussian temporal profile.

The main assumption leading to the form (11) of the solution is that it must be possible to write the collision integrals in the τ-approximation. In so doing, it was assumed that $f(\varepsilon,t)$ relaxes to the equilibrium function $f_0(\varepsilon,T_e)$ with a time-dependent electron temperature $T_e(t)$.[19]

It is well known that the effect of ultrashort laser pulses on a metal surface is to produce a dynamically nonequilibrium situation in which the electron temperature T_e decouples form the lattice temperature T_i, but both the electron and phonon distribution functions individually retain their equilibrium form.[5]

Analysis of the solution (11) gives several different results. Figure 1 displays the electron distribution function, calculated from Eq. (11), for sodium with different values of the laser radiation characteristics. The figure also displays the equilibrium electron distribution function with temperature T_e, as calculated from the data of Ref. 19 (dashed lines). From Fig. 1 and the expression (11) it is evident that in the region of interaction of the radiation with the metal surface, there is formed predominantly an electron distribution function with a periodic stepped structure with step $\hbar\omega$ the amplitude of the steps being proportional to κ^n, and the temporal behavior of each step being determined by the form of $\Theta_n(t)$. The curves *1* and *2* were obtained for a 1.17 eV heating photon and power densities $W=200$ and 600

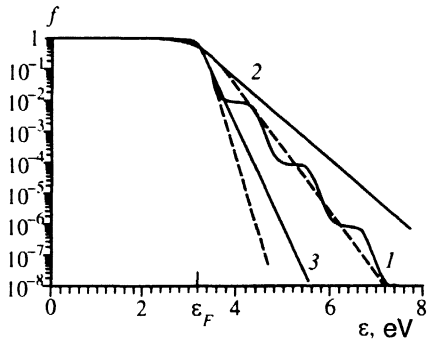

FIG. 1. Electron distribution function computed for sodium irradiated with high-power laser radiation.

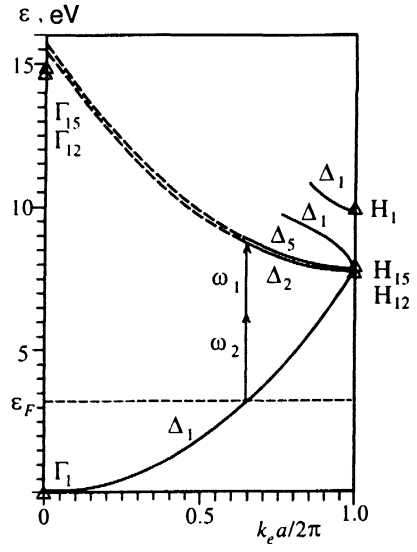

FIG. 2. Na[100] dispersion curves.[20]

MW/cm^2, respectively. The curve 3 was obtained for a 0.5 eV heating photon and a power density of 200 MW/cm^2. As the intensity of the heating radiation increases or the energy of the heating photons decreases ($\kappa \to 1$), the stepped structure is smoothed out, and in the limit, as can be seen clearly in the figure, the solution (11) approaches the solutions obtained in Ref. 19.

4. ANALYSIS OF THE CHANGES IN THE NONLINEAR RESPONSE OF A METAL

It follows from what we have said above that the actual number of electrons occupying nonequilibrium states is extremely small, and most electrons occupy equilibrium states. It is very important, however, that a small number of electrons occupy states above the Fermi level that are essentially empty in the case of equilibrium heating.

In the long-wavelength approximation (assuming that the size of the "main region" of the crystal is much smaller than the wavelength of the radiation and the wave vector of the radiation is small compared to that of an electron in the crystal) the following expression can be derived for the nonlinear component of the current density:[7,10]

$$\begin{aligned}\mathbf{j}(\omega_1+\omega_2)=&\frac{e}{8\pi mc^2}[(\omega_1+\omega_2)(\mathbf{a}_1\mathbf{a}_2)[\hat{\varepsilon}(\omega_1+\omega_2)-1]\\&\times(\mathbf{q}_1+\mathbf{q}_2)+2\omega_1\mathbf{a}_2(\mathbf{q}_1[\hat{\varepsilon}(\omega_1)-1]\mathbf{a}_1)\\&+2\omega_2\mathbf{a}_1(\mathbf{q}_2[\hat{\varepsilon}(\omega_2)-1]\mathbf{a}_2)]\\&+\frac{e^3}{\hbar^2m^3c^2}\sum_{lmnk_e}(\mathbf{a}_1\mathbf{p}_{lm})(\mathbf{a}_2\\&\times\mathbf{p}_{mn})\,\mathbf{p}_{nl}B(l,m,n,\omega_1,\omega_2)+P_{1\leftrightarrow 2},\end{aligned}\qquad(12)$$

where

$$\begin{aligned}&B(l,n,m,\omega_1,\omega_2)\\&=(\omega_1+\omega_2-\omega_{ln}+iT_{ln}^{-1})^{-1}\\&\times\left[\frac{f_l-f_m}{\omega_1-\omega_{lm}^{+}+iT_{lm}^{-1}}-\frac{f_m-f_n}{\omega_1-\omega_{mn}+iT_{mn}^{-1}}\right],\end{aligned}$$

$\mathbf{a}_1$ and $\mathbf{q}_1$ are, respectively, the components of the vector potential and the wave vector; f_m are the diagonal elements of the density matrix; $\mathbf{p}_{mn}=-i\hbar\int\psi_{ml}^{*}\nabla\psi_{nl}dV$ are the matrix elements of the momentum operator; and, ψ_{nl} is the electron wave function. The summation extends over the band indices and the electron wave vector k_e; it is assumed that the matrix elements depend on k_e. The symbol $P_{1\leftrightarrow 2}$ represents a term which is the same as the last term except that the frequencies of the field are interchanged. The bracketed terms are presented in Ref. 7, where an expression can also be found for the permittivity tensor $\hat{\varepsilon}(\omega_1)$.

It follows from Eq. (12) that the nonequilibrium structure arising in the electron distribution function (11) in a strong field can affect the imaginary part of the current density, i.e., the so-called nk-dependences in the linear case.

In the case of a transverse electromagnetic field in a crystal, the second and third terms in brackets in Eq. (12) vanish. The contribution of the remaining terms to the imaginary part of the nonlinear current as a function of the transverse relaxation time and the frequencies of the probe field can be different. Without loss of generality, we shall consider the change produced in the nonlinear current by the imaginary part of the nonlinear polarizability. We note that the imaginary part of the linear permittivity will have a similar effect. In analyzing the experimental data, this contribution can also be taken into account on the basis of measurements of the linear response at the corresponding frequency.

Let the model system be a sodium surface. We shall analyze the change in the contribution to the nonlinear response of the electrons in states with symmetry Δ. Figure 2 displays the dispersion curves[20] for Na[100] (here $a=8.11$ a.u. is the lattice constant; the triangles correspond to the energies at the symmetry points Γ and **H**). The dashed line represents our extrapolated curve, obtained on the basis of the parabolic-band model. It is evident from the figure that for optical frequencies, interband transitions can actually occur between the lowest bands. In this connection, we focus on the two-band model. In this case, the last term in expression (12) will have the following form:

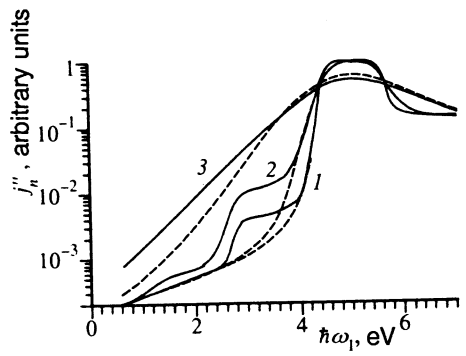

FIG. 3. Dispersion curves for the imaginary part of the nonlinear current for different values of the heating-radiation power density: $W=50$ (*1*), 200 (*2*), and 800 MW/cm^2 (*3*).

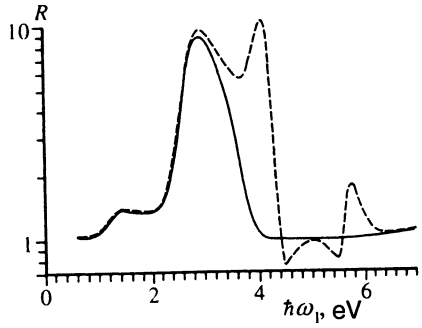

FIG. 4. Dispersion curves of the ratio of the imaginary parts of the nonlinear current in the nonequilibrium and equilibrium cases (see text).

$$\mathbf{j}(\omega_1+\omega_2)=\sum_{k_e}\frac{(\mathbf{a}_1\mathbf{G})(\mathbf{a}_2\mathbf{p}_{12})\mathbf{p}_{21}(f_1-f_2)}{(\omega_1+\omega_2-\omega_{21}+iT_{21}^{-1})(\omega_1-\omega_{21}+iT_{21}^{-1})}+P_{1\leftrightarrow 2}, \tag{13}$$

where $\mathbf{G}$ is the reciprocal lattice vector of the crystal. In the calculations it was assumed that the matrix elements and the relaxation time are independent of the electron wave vector $\mathbf{k}_e$. We note that six terms in the sum (11) were included in the computational results presented below.

In the case of the nonequilibrium electron distribution function (11), the dispersion curves for the imaginary part of the nonlinear current must have a stepped form (Fig. 3). Figure 3 displays the curve for nonlinear summation of the frequencies $\omega_1+\omega_2$ with $\hbar\omega_2=1.18$ eV, $\hbar\omega_h=1.17$ eV, $\tau_{ee}=10^{-13}$ sec, $T_{12}=6\cdot10^{-14}$ sec, and $W=50$ (curve *1*), 200 (*2*), and 800 MW/cm^2 (*3*). The step width is determined by the energy of the heating photons, the reciprocal lattice vector, the Fermi energy ε_F, the number of the step, and the character of the dispersion law, and can be found from the expression

$$\Delta S_n \approx m^{-1/2}\hbar G(\sqrt{\varepsilon_F+n\hbar\omega_h}-\sqrt{\varepsilon_F+(n-1)\hbar\omega_h}). \tag{14}$$

Since the amplitude of a step is proportional to κ, i.e., the power of the heating radiation and the electron-electron relaxation time, the quantities mentioned above can be judged according to their behavior. As the power density of the heating radiation increases, the dispersion curves become smoother and approach the curves obtained when the heating of the electron gas is an equilibrium process. The dashed lines represent the analogous dependences obtained under the assumption of equilibrium heating of the surface.

Figure 4 displays the ratio R of the imaginary part of the nonlinear current j''_n, calculated for the nonequilibrium case, to j''_n calculated for equilibrium heating (solid line) and to j''_n calculated for a "cold" metal at an electron gas temperature of 300 K (the process $\omega_1+\omega_2$ with $\hbar\omega_2=1.18$ eV, $\hbar\omega_h=1.17$ eV, $\tau_{ee}=10^{-13}$ sec, $T_{12}=6\cdot10^{-14}$ sec, and $W=200$ MW/cm^2). It is clear from the figure that the difference between the curves is greater in the spectral range corresponding to transitions whose initial states lie $\hbar\omega_h$ above the Fermi level (in this case the frequency of the direct interband transition from the Fermi level is 5.54 eV). As the frequency of the probe radiation increases further, the contributions of states lying above and below the state corresponding to a direct two-photon transition cancel one another. As a result of this cancellation, the effect of the nonequilibrium electrons remains even in the high-frequency region of the spectrum; this is especially clearly seen in comparing the nonequilibrium response to the response of a "cold" lattice (the features on the dashed curve correspond to the peaks in the population difference $f(\varepsilon)-f_0(\varepsilon)$ [compare Fig. 1]. In the case when the probe radiation frequency ω_2 is comparable to ω_1, however, the increase in the effect of nonequilibrium electrons in the high-frequency region of the spectrum will not be so large because of the effect of the transposed term. In certain spectral regions, R depends on the ratio of τ_{ee} and T_{12} and on the parameters of the heating and probe radiation, and it can reach several orders of magnitude.

The stepped character of the dispersion curves is more pronounced for $\omega_2\leqslant\omega_h$ (Fig. 5). displays j''_n as a function of the frequency ω_1 of the probing radiation; the curves were obtained for different frequency conversion processes with heating photon energies of 1.17 (a) and 0.5 eV (b) ($\tau_{ee}=10^{-12}$ sec, $T_{12}=6\cdot10^{-14}$ sec, and $W=80$ NW/cm^2). Curve *1* corresponds to frequency summation $\omega_1+\omega_2$ ($\omega_2=1.01\omega_h$) and curve *2* corresponds to second harmonic generation at the frequency $2\omega_1$; the dashed line corresponds to the dispersion curve of the imaginary part of the linear current. One can see from the figure that as the frequency of the heating radiation increases, the stepped character of the curves becomes more pronounced. Numerical calculations show that the change in the response in the nonlinear case is much greater than for the linear case.

We now consider the effect of the relaxation times T_{12} and τ_{ee} on the magnitude of the nonlinear current. We note that the characteristic transverse relaxation times, determined by both nonadiabatic and adiabatic interactions, are, as a rule, less than the characteristic electron-electron times, determined by inelastic interactions of the electronic subsystem in the bottom band. As expected, when the relaxation time τ_{ee} decreases, the electron distribution function thermalizes more rapidly and, correspondingly, the heating field becomes less influential (Fig. 6). Here, the coefficient R is the ratio of

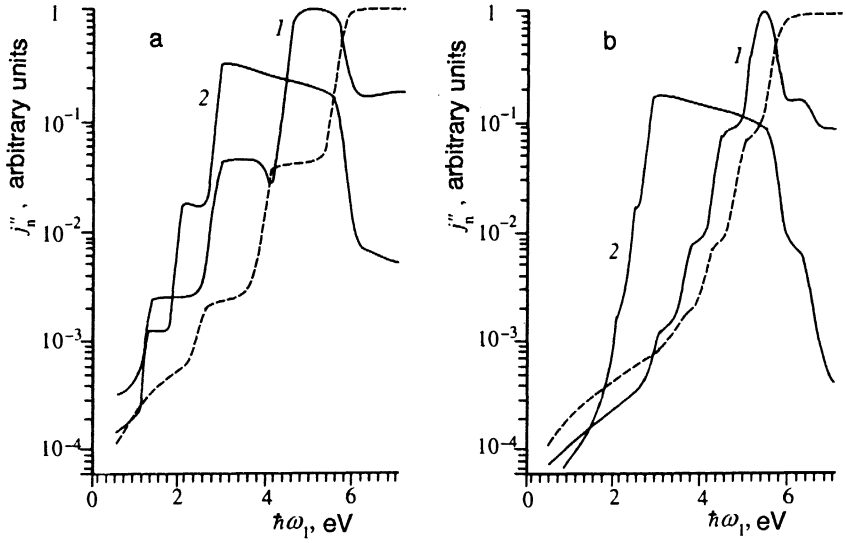

FIG. 5. Manifestation of nonequilibrium in different wave interaction processes (see text).

the imaginary part of the nonlinear current in the presence of the heating field to the imaginary part of the nonlinear current on the "cold" surface, Fig. 6 displays the curves obtained for different ratios of the relaxation times $T_{12}=0.1\tau_{ee}$ (curve *1*) and $T_{12}=0.5\tau_{ee}$ (curves *2* and *3*). Here, $\hbar\omega_1=3.51$ eV, $\hbar\omega_2=1.18$ eV, $\hbar\omega_h=1.17$ eV, and $W=100$ (curves *1* and *2*) and 400 MW/cm^2 (curve *3*). As the transverse relaxation time increases, the contribution of the equilibrium electrons to the imaginary part of the current density (13) decreases, the consequently the difference between the equilibrium and nonequilibrium cases increases. It is evident from the figure that the nonlinear processes make it possible to observe this effect for shorter relaxation times than in the analysis of the linear response (the dashed lines correspond to the same curves for the linear case). A similar result also obtains as the heating radiation power density increases.

The results show that the nonequilibrium structure produced by powerful laser radiation in the electron gas of a metal results in large changes in the linear and nonlinear responses of the surface. Analysis of the latter will definitely improve surface diagnostics. It is obvious that the complicated band structure of different metals can mask the effect of the nonequilibrium electrons. Nonetheless, these difficulties can be overcome by choosing suitable probe and heating radiation, as well as the geometry of the experiment. We wish to underscore once again that because the number of nonequilibrium electrons is small, the absolute magnitude of the nonlinear response can be small, but its relative change, as shown above, can be very large, making this effect a "threshold" effect and creating the potential for direct observation of the effect. At the same time, the direct functional dependence of the observed response on fundamental characteristics of an electron gas, such as the energy relaxation time τ_{ee} and the transverse relaxation time T_{mn}, makes it very important to perform such measurements and this, together with the progress made in modern femtosecond laser technology, can be easily realizable experimentally.

5. CONCLUSIONS

A possible mechanism by which strong laser radiation can influence the nonlinear response of the surface of a metal was demonstrated for the example of a quite simple model. In the analysis a number of assumptions were made, which, in our opinion, do not distort the physical picture of the phenomenon. Specifically, there is no doubt that in the real case, all interband transitions, and not just those in the symmetry direction, must be taken into account. Although for simple metals (such as sodium) this is not so important because of the high degree of symmetry of the Fermi surface, for metals with a more complicated band structure the latter can change substantially the dispersion relations for the nonlinear current. In spite of the obvious limitation of the model considered, however, the main conclusion that the frequency dependence of the nonlinear current changes in a strong field is also valid for more complicated systems. This must be taken into account when performing experiments on nonlinear optical probing of a surface, since for a number of rea-

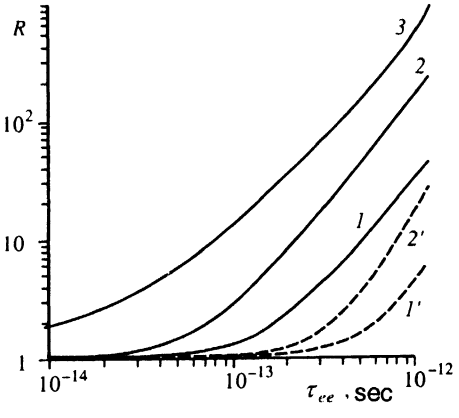

FIG. 6. Effect of the relaxation time on the magnitude of the nonlinear current (see text).

sons, specifically, intensification on nonuniformities, the field at the surface cannot be assumed *a priori* to be weak, even for moderate laser radiation intensities.

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