Nonlinear photomagnetism of metals: Theory of nonlinear photoinduced dc current

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Photoinduced magnetic flux has recently been observed in normal metals exposed to light. This effect is partly due to the fact that the light reflected from a metal surface transfers to the conduction electrons some of its quasimomentum. This creates a dc surface current which, for an appropriate geometry, brings about the photomagnetic effect. There is another contribution to the current that is due to anisotropy of the probabilities of electron transitions induced by the light, in combination with diffuse reflection of the electrons at the surface. We present here a theory of the dependence of the photoinduced current on the intensity of light Q. We assume that the light intensity is either constant or the time scale of its variation is much larger than the inverse Rabi frequency corresponding to the interband electron transition. At comparatively low intensities the current is proportional to Q. At higher intensities it varies as $Q^{1/2}$. The physical origin of such behavior is analyzed. Various factors that allow a lowering of the critical intensity for the onset of the nonlinear behavior are discussed.

I. INTRODUCTION

In a recent paper[1] observation of the photomagnetism of metals was reported. In a sample of metal of such a form that a circular dc current can exist, a buildup of magnetization under illumination was observed. In Ref. 2 a microscopic theory of the photoinduced surface current bringing about the photomagnetism was worked out. A case where the current is due to the light-excited interband transitions of the electrons was considered. It was pointed out that there is a transfer of quasimomentum from the light partly absorbed at the metal surface to the conduction electrons which is one of the sources of the dc surface current. It was indicated that there is another contribution to the current due to anisotropy of the probabilities of electron transitions induced by the light, in combination with diffuse reflection of the electrons at the surface.

In Ref. 3 yet another possible source of photomagnetism was demonstrated, i.e., photoinduced bulk current. It can also be excited by light absorbed at a metal surface, in particular, by light propagating perpendicularly to the surface. The magnetic flux associated with this current is expected to be much larger than the magnetic flux associated with the surface current.

In all these papers only linear dependence of the photoinduced magnetic flux was considered theoretically and observed in experiment. The present paper is devoted to the case where the magnetic flux is a nonlinear function of the light intensity Q. We work out a theory of such dependence. We have some experimental evidence of a nonlinear dependence of the observed magnetic flux which could be hopefully increased along with the increase of the pumping power. Therefore, in our opinion, a theoretical understanding of the nonlinear behavior of the photocurrent is called for.

The physics of the nonlinear behavior of the light-induced magnetic flux may be described as follows. The quasimomentum conservation in the interaction of light with the Bloch electrons of a metal establishes correspondence between pairs of states. One of the states of such a pair belongs to the lower band (band 1) while another one belongs to the upper band (band 2). As an example, we will consider the case where before the illumination has started the state in the lower band is occupied while the state in the upper band is empty. Under the influence of an ac electric field of coherent light the occupation probabilities of the states belonging to such a pair begin to oscillate with time. The frequency of oscillation is the Rabi frequency Ω_R which is given by

$$\hbar\Omega_R = |\mathbf{E}\cdot\mathbf{T}_{21}| ,$$

where $\mathbf{E}$ is the amplitude of electric field inside the metal while $\mathbf{T}_{21}$ is the corresponding transition matrix element [see Eq. (6)]. Ω_R is proportional to the amplitude of electric field or, in other words, to $Q^{1/2}$. If the intensity of light, Q, is so low that this frequency is smaller than the average collision rate $1/\tau$ given below by Eq. (12) then the Rabi oscillation does not actually happen. If, on the other hand, $\Omega_R\tau \gg 1$ the Rabi oscillation takes place and the dc current should be a nonlinear function of Q. We wish to emphasize that in our paper by dc current we actually mean such current whose variation is slow on the time scale of Ω_R^{-1}. This means, in particular, that the laser pulse duration should be larger than the smaller of the two quantities Ω_R^{-1} and τ.

Reasoning of this sort was applied for investigation of saturation of intervalence band transitions in semiconductors by James and Smith in Refs. 4 and 5 (see also

Ref. 6). It was also used for investigation of linear-circular dichroism of drag current due to nonlinear intersubband absorption of light in Ref. 7. The most important difference between the cases considered in the quoted papers and the present one is the already mentioned essential spatial inhomogeneity of the problem.

This reasoning is good for semiconductors provided the electric-field amplitude can be considered as coordinate independent. In metals, however, the light intensity falls off within the layer of the width δ of the order of 10^{-5}–10^{-6} cm while the current excited by light exists over the length about mean free path l. Under these circumstances the onset of nonlinearity demands much higher intensities as the interband transitions take place in a comparatively narrow layer near the metal surface. It turns out that for the case

$$l \gg \delta \tag{1}$$

we are actually interested in the big dimensionless parameter determining the onset of the nonlinear intensity dependence is

$$\gamma = \frac{4(\Omega_R \tau)^2 \overline{m} \delta}{\bar{p} \tau} , \tag{2}$$

where $\overline{m}$ is the average (over both bands) value of the electron effective mass and $\bar{p}$ is the average value of the quasimomentum of the electrons excited by light. The appearance of parameter $\Omega_R \tau$ seems natural for nonlinear problems of this sort (see Refs. 4 and 5) while δ/l is, in fact, the ratio of the volume where the light penetrates to the volume where the photomagnetic current flows [see derivation of Eq. (39)]. After integration over all the electrons taking part in these transitions we obtain the $Q^{1/2}$ dependence for the photomagnetic response. We wish to emphasize that to develop this dependence the parameter γ need not be particularly large; it should just be somewhat bigger than unity.

One should discuss separately the case of small electron groups that can move almost parallel to the sample's surface. These are, for instance, Fermi surface "necks" in copper and gold or flat parts of electron surfaces of constant energy. In these groups the component of electron velocity perpendicular to the sample surface can be so small that the electrons would remain within the surface layer δ during the time τ. Then the nonhomogeneity of the electric field is of no importance and the nonlinear parameter is the same as in semiconductors, namely,

$$\gamma_s = 2(\Omega_R \tau)^2 . \tag{3}$$

It does not contain the small factor δ/l. As a result, the critical intensities for such groups can be much smaller. The phase volumes occupied by such groups are usually also small. They, however, can bring about nonlinear intensity dependence at comparatively low intensities in metals where the ratio l/δ is not too big and the orientation of the sample's surface is chosen in the appropriate way.

We do not consider in the present paper contribution of the surface bands into the surface current (cf. with Ref. 8). We think, however, that it might be very interesting to investigate their role both theoretically and experimentally, choosing for this purpose the crystallographic orientation of a monocrystalline sample's surface as well as the frequency of the light.

By measurement of nonlinear photomagnetism one can obtain important information concerning the values of the interband transition amplitudes induced by light. This way one can also get the data about the relaxation rates of the Bloch electrons in metals. One can also verify the assumptions concerning the distribution of the conduction electrons perturbed by light and to check under which circumstances its deviation from the equilibrium may still be considered as small. It seems that, unlike the semiconductors where the electron distribution function may be strongly nonequilibrium,[6] in metals one may assume that the deviation is small up to rather high intensities of light.

It is seen from Eq. (2) that the critical intensity for the onset of nonlinearity, Q_c, goes down when the average collision rate τ goes up. Q_c is also very sensitive to the value of the matrix element $\mathbf{T}_{21}$. In regard to the values of the transition matrix elements that can speak *for a given frequency of light* of two types of metals. In the metals of the first type transitions are possible between a band of well-localized states (such as d band) and the conduction band. For the frequency interval used in Ref. 1 such metals as copper belong to the first type. As in the case of atomic photoeffect, the values of transition matrix elements for these metals may be rather big. One can come to the same conclusion by analysis of the experiment[9,10] and theory[11,12] of light absorption in copper as well as the linear photomagnetic response observed in Ref. 1 (see above and Sec. V). For the metals of the second type only transitions where the localized states are not involved are possible. If an approximation of almost free electrons is valid for calculation of the transition amplitudes they should be small, the smallness being determined by the ratio of the pseudopotential constant to some characteristic energy of the order of Fermi energy. Such a case was considered in Ref. 13 (where the part of the current due to the quasimomentum transfer was calculated in the almost-free-electron approximation and experimental evidence for this effect was presented in the form of spatially asymmetric photoemission from rough silver films). One should expect that in this case the critical intensity for the onset of the nonlinear behavior should be (for the same values of τ) much bigger than in the first case. As so far most of the experimental data have been obtained on copper we will make most numerical estimates for copper.

II. PROBABILITY OF INTERBAND ELECTRON TRANSITIONS INDUCED BY LIGHT

The Hamiltonian of interaction of conduction electrons with light has the form

$$\mathcal{H}_{\text{int}} = \tfrac{1}{2}(\mathcal{K} + \mathcal{K}^{\dagger}) , \tag{4}$$

$$\mathcal{K} = (ie/2\omega m_0) e^{-i\omega t} [e^{ik_x x}(-i\hbar \mathbf{E}\cdot\boldsymbol{\nabla}) + (-i\hbar \mathbf{E}\cdot\boldsymbol{\nabla}) e^{ik_x x}] . \tag{5}$$

Here ω is the frequency of light and m_0 is the free electron mass; $\mathbf{E}(z)$ denotes the amplitude of the electric field that depends on coordinate z along the direction perpendicular to the metal surface. We will consider the time-averaged action of the field introducing a term describing the interband transitions into the Boltzmann equation for the electrons. To allow for the z dependence of the electron distribution function we can imagine that the sample is divided into slabs, the thickness of each of them being so small that the field in a slab can be considered as independent of z. This approach permits us to take into account the z dependence of the intensity of light.

The matrix element of the interband transition between the Bloch states with quasimomentum $\mathbf{p}$ in the lower band 1 and with quasimomentum $\mathbf{p}'$ in the upper band 2 is

$$\tfrac{1}{2}\langle 2\mathbf{p}'|\mathcal{H}|1\mathbf{p}\rangle=\frac{ie}{2m_0\omega}\mathbf{E}\cdot\mathbf{P}_{21}e^{-i\omega t}\delta_{p_x',p_x+\hbar k_x}\times\delta_{p_y',p_y}\delta_{p_z',p_z}\ , \tag{6}$$

where $\delta_{p'p}$ is the Kronecker symbol. Here $\mathbf{P}_{21}=(2m_0\omega/e)\mathbf{T}_{21}$ is given by

$$\mathbf{P}_{21}(\mathbf{p}',\mathbf{p})=\frac{1}{\mathcal{V}_0}\int d^3r\, u_{\mathbf{p}'}^{(2)*}(\mathbf{r})\left[-i\hbar\frac{\partial}{\partial\mathbf{r}}\right]u_{\mathbf{p}}^{(1)}(\mathbf{r})\ , \tag{7}$$

$\mathcal{V}_0$ being the volume of the primitive cell and $u_{\mathbf{p}}^{(1,2)}(\mathbf{r})$ being the Bloch amplitudes.

In what follows we will use the notation

$$\hbar\Omega_R=\frac{e}{2m_0\omega}|\mathbf{E}\cdot\mathbf{P}_{21}(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})|\ , \tag{8}$$

where Ω_R is the Rabi frequency.

In the linear theory the transition probability from Bloch state 1, $\mathbf{p}$, to state 2, $\mathbf{p}'=\mathbf{p}+\hbar\mathbf{k}$, is given by

$$G\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)})\ ,$$

$$G(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})=\frac{\pi}{2\hbar}\left[\frac{e}{m_0\omega}\right]^2|\mathbf{E}(z)\cdot\mathbf{P}_{21}(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})|^2\ . \tag{9}$$

We will see that in nonlinear theory the δ function in this equation will be replaced by a more complicated expression. This will appear to be the main modification of the Boltzmann equation we used in the linear theory to calculate the nonequilibrium parts of the distribution functions.

The x component of the current density is given by the usual equation

$$j_x=2e\int\frac{d^3p}{(2\pi\hbar)^3}[v_x^{(2)}(\mathbf{p})f_{\mathbf{p}}^{(2)}+v_x^{(1)}(\mathbf{p})f_{\mathbf{p}}^{(1)}]\ . \tag{10}$$

Here $f_{\mathbf{p}}^{(1,2)}$ are the nonequilibrium parts of the electron distribution functions in the lower and upper bands, respectively; $v_x^{(1,2)}(\mathbf{p})$ are the x components of the electron velocities.

III. BOLTZMANN EQUATION

To work out the nonlinear equation for the variation of the electron distribution function due to interaction with light we will use an "intermediate" equation for the density matrix rather than a perturbation theory equation of the type (9). Let us denote by $\psi e^{-i\omega t}$ the density matrix between the states $2,\mathbf{p}+\hbar\mathbf{k}$ and $1,\mathbf{p}$. We have (see Refs. 4 and 5)

$$\frac{\partial\psi}{\partial t}=i(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\psi+\frac{ie}{m_0\omega}\mathbf{E}(z)\cdot\mathbf{P}_{21}(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})(\mathcal{F}_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\mathcal{F}_{\mathbf{p}}^{(1)})-\frac{\psi}{\tau}\ , \tag{11}$$

where

$$\frac{1}{\tau}=\frac{1}{2}\left[\frac{1}{\tau^{(1)}}+\frac{1}{\tau^2}\right]\ . \tag{12}$$

Here $\mathcal{F}^{(1,2)}$ are the full distribution functions. They satisfy the following equations:

$$\frac{\partial\mathcal{F}_{\mathbf{p}}^{(1,2)}}{\partial t}+v_z^{(1,2)}\frac{\partial\mathcal{F}_{\mathbf{p}}^{(1,2)}}{\partial z}=\mp2\Omega_R\,\mathrm{Re}\psi-\left[\frac{\partial\mathcal{F}^{(1,2)}}{\partial t}\right]_{\mathrm{coll}}\ . \tag{13}$$

The first term on the right-hand side originates from the commutator between the off-diagonal part of the density matrix and the Hamiltonian of interaction of the Bloch electrons with the electromagnetic field [Eq. (5)]. The last term on the right-hand side represents the intraband transitions on conserving the electron quasimomentum which may be due to collisions of the electrons with defects (or phonons). Performing analysis of the equation for the density matrix in the resonant approximation (i.e., taking into account only two bands) one can see that on the one hand $\mathrm{Im}\psi$ can be expressed through $\mathrm{Re}\mathbf{P}_{21}$. The term proportional to $\mathrm{Im}\mathbf{P}_{21}$ is not resonant and therefore is beyond the approximation accepted throughout the paper. On the other hand, far away from the threshold where

$$\left[\frac{\omega-\omega_{\mathrm{threshold}}}{2m}\right]^{1/2}\gg k$$

one can always find such a gauge transformation that $\mathbf{P}_{21}$ would be real (up to the small terms proportional to k). It is in this gauge that we have written Eq. (13). Naturally, the final expressions for the observables, such as $f_{\mathbf{p}}^{(1,2)}$, do not depend of the gauge.

Here we consider quantum transitions of the electrons while the electric field is treated classically. Frequency ω is assumed to be big enough so that the conditions for the energy and quasimomentum conservation allow transitions between the occupied states in the lower band and empty states in the upper band (we do not consider here the threshold effects due to the transitions to the immediate vicinity of the Fermi level). The amplitude of the electric field, $\mathbf{E}(z)$, can be complex. It means that these equations are valid for any polarization of the light, including circular (or, in general, elliptical).

We assume that wave vector $\mathbf{k}$ has an x component k_x along the metal surface. Strictly speaking, the spatial

variation of function $\mathbf{E}(z)$ can describe not only its damping but also an oscillation along the z direction. This means that the z component of the quasimomentum can also be transferred to the electrons at the surface, thus creating a dc current or voltage along the direction. However, we shall disregard this effect as we do not consider here in detail the z component of the current.

The distribution function of the electrons excited by a laser light should have a part that is very sharp in the quasimomentum space. This means that one can present the full distribution functions as the sums of the smooth parts $F_{\mathbf{p}}^{(1,2)}$ and resonant (sharp) parts $f_{\mathbf{p}}^{(1,2)}$. For the latter the "out" term of the collision operator should be much bigger than the "in" term. This means that to describe the collisional variation of a sharp part of the distribution function $f_{\mathbf{p}}^{(1,2)}$ one may use the relaxation-time approximation:

$$\left[\frac{\partial \mathcal{F}_{\mathbf{p}}^{(1,2)}}{\partial t}\right]_{\text{coll}} = \frac{\mathcal{F}_{\mathbf{p}}^{(1,2)} - F_{\mathbf{p}}^{(1,2)}}{\tau^{(1,2)}} \ . \tag{14}$$

As for $F_{\mathbf{p}}^{(1,2)}$, below we consider these as the equilibrium Fermi functions.

For a stationary case (or, to be more exact, in the case where the time scale for the amplitude of light variation is larger than Ω_R^{-1}) where the time derivatives vanish we have from Eq. (11)

$$\mathrm{Re}\psi = -\frac{\hbar^2 \Omega_R \tau^{-1}(\mathcal{F}_{\mathbf{p}+\hbar\mathbf{k}}^{(2)} - \mathcal{F}_{\mathbf{p}}^{(1)})}{(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)} - \hbar\omega - \varepsilon_{\mathbf{p}}^{(1)})^2 + (\hbar/\tau)^2} \ , \tag{15}$$

whereas for the nonequilibrium parts of the distribution functions

$$v_z^{(1,2)} \frac{\partial f_{\mathbf{p}}^{(1,2)}}{\partial z} + \frac{f_{\mathbf{p}}^{(1,2)}}{\tau^{(1,2)}} = \mp 2\Omega_R \mathrm{Re}\psi(z) \ . \tag{16}$$

IV. ROLE OF SPATIAL NONHOMOGENEITY

As the amplitude of the electric field falls off as a function of the distance z from the metal surface the Rabi frequency is coordinate dependent. We assume that the real part of the dielectric susceptibility of metal for the frequency ω is negative and the electric-field amplitude falls off as $e^{-\kappa z/2}$. Accordingly,

$$\Omega_R(z) = \Omega_0 e^{-\kappa z/2} \ , \tag{17}$$

where Ω_0 is given by Eq. (8) with $\mathbf{E} = \mathbf{E}_{(0)}$. We have to solve Eq. (16) allowing for the z dependence of $\Omega_R(z)$.

We will use the boundary conditions of elastic and completely diffuse electron scattering at the metal's surface. They were discussed in detail in Refs. 2 and 3. For the current parallel to the metal's surface (surface current) we are going to calculate they can be used in the following simplified form:

$$f_{\mathbf{p}}^{(1,2)}(0) = 0 \quad \text{for} \quad v_z^{(1,2)} > 0 \ . \tag{18}$$

We will be interested in the case where the electron mean free path l is much bigger than the light penetration depth, $\delta = 1/\kappa$. Then the main contribution to the current comes from the electrons with $v_z > 0$. For these electrons Eqs. (16) and (18) give

$$f_{\mathbf{p}}(z)^{(1,2)} = \frac{1}{v_z^{(1,2)}} \exp\left[-\frac{z}{v_z^{(1,2)}\tau}\right] \times \int_0^z dz' \exp\left[\frac{z'}{v_z^{(1,2)}\tau}\right] [\mp 2\Omega_R(z')\mathrm{Re}\psi(z')] \ , \tag{19}$$

while the contribution of the electrons with $v_z < 0$ is of higher order in the small parameter δ/l and therefore we will neglect it. The electrons with the positive z component of velocity give the principal contribution to the surface current (which falls off over the distances z of the order of l). For such distances taking into account that $\psi(z)$ falls off over the distances of the order of δ one can rewrite Eq. (19) as

$$f_{\mathbf{p}}(z)^{(1,2)} = \mp \frac{2\Omega_0}{v_z^{(1,2)}} \int_0^z dz' e^{-\kappa z'/2} \mathrm{Re}\psi(z') \ . \tag{20}$$

In order to obtain a self-consistent equation for $\mathrm{Re}\psi(z)$ we write solutions of Eqs. (16) for $z \ll l$. For this purpose it is convenient to introduce a new function

$$\varphi(z) = e^{\kappa z/2} \mathrm{Re}\psi(z)$$

and

$$N_0 = \frac{\hbar^2 \Omega_0 \tau^{-1}}{(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}/2}^{(2)} - \epsilon_{\mathbf{p}-\hbar\mathbf{k}/2}^{(1)} - \hbar\omega)^2 + (\hbar/\tau)^2} \ .$$

Then one can present Eq. (15) in the form

$$\varphi(z) = -N_0(F^{(2)} - F^{(1)}) - N_0[f_{\mathbf{p}}^{(2)}(z) - f_{\mathbf{p}}^{(1)}(z)] \ .$$

Inserting in this definition Eq. (20) for the resonant distribution functions we get the following integral equation:

$$\varphi(z) + \frac{4N_0\Omega_0}{\bar{v}_z} \int_0^z dz' \exp(-\kappa z')\varphi(z') = -N_0(F_{\mathbf{p}}^{(2)} - F_{\mathbf{p}}^{(1)}) \ , \tag{21}$$

where

$$\frac{2}{\bar{v}_z} = \frac{1}{v_z^{(1)}} + \frac{1}{v_z^{(2)}} \ .$$

It is equivalent to the differential equation

$$\frac{d\varphi}{dz} + \frac{4N_0\Omega_0}{\bar{v}_z} \exp(-\kappa z)\varphi(z) = 0 \ , \qquad \varphi(0) = -N_0(F_{\mathbf{p}}^{(2)} - F_{\mathbf{p}}^{(1)}) \ . \tag{22}$$

Its solution is given by

$$\varphi(z) = N_0(F_{\mathbf{p}}^{(1)} - F_{\mathbf{p}}^{(2)}) \exp\left\{\frac{4N_0\Omega_0}{\kappa\bar{v}_z}[\exp(-\kappa z) - 1]\right\} \ . \tag{23}$$

To obtain the singular part of the distribution function let us make use of the fact that $\mathrm{Re}\psi(z)$ has essentially nonvanishing values only within the region $z < \kappa^{-1} \ll l$.

It is only this region that contributes to the integral over z' in Eq. (19) after we have inserted into it $\varphi(z')$ from Eq. (23). As a result, the integral over z' is readily calculated and we get

$$f^{(1,2)}=\mp\frac{\bar{v}_z}{2v_z^{(1,2)}}(F_{\mathbf{p}}^{(1)}-F_{\mathbf{p}+\hbar\mathbf{k}}^{(2)})\exp\left[-\frac{z}{v_z^{(1,2)}\tau^{(1,2)}}\right]\times\left[1-\exp\left[-\frac{4N_0\Omega_0}{\kappa\bar{v}_z}\right]\right]. \quad (24)$$

V. CALCULATION OF CURRENT

Now we embark on a discussion of the equation for the surface current density that one can get from Eq. (10) by integration over z:

$$\mathbf{g}=\int_0^\infty dz\ \mathbf{j}(z)\ . \quad (25)$$

For calculations and order-of-magnitude estimates of the expected effects we will exploit here a simple model of the electron spectrum. We assume the electron spectrum in both bands to be isotropic and quadratic:

$$\varepsilon^{(1)}(p)=-p^2/2m_1,\quad \varepsilon^{(2)}(p)=\varepsilon_g+p^2/2m_2\ . \quad (26)$$

As for the probabilities of the interband transitions, we make one of the simplest assumptions compatible with the isotropic model, namely,

$$\mathbf{P}_{21}(\mathbf{p}',\mathbf{p})=\alpha(\mathbf{p}'+\mathbf{p})\ , \quad (27)$$

where α is a real dimensionless constant. In reality the angular dependence of the matrix element may be much more complicated. This, however, is of little consequence as we are going to use this equation only for rough order-of-magnitude estimates. Actually, for such estimates *any* form of the matrix element is suitable (so far as all the quasimomenta involved are of the order of p_F) so we take the simplest one. What is of importance, though, are the numerical values of the coefficient α. They are discussed at the end of this section.

Making use of Eqs. (24) and (10) and integrating over z [see Eq. (25)] one calculates the current density which is a sum of two terms:

$$g_x^{(1,2)}=e\int\frac{d^3p}{(2\pi\hbar)^3}\bar{v}_z\frac{p_z\mp\hbar k_x/2}{m^{(1,2)}}\tau^{(1,2)}\times\left[1-\exp\left[-\frac{4N_0\Omega_0}{\kappa\bar{v}_z}\right]\right]. \quad (28)$$

In what follows it will be convenient to assume that the resonant transitions connect the state $\mathbf{p}-\hbar\mathbf{k}/2$ in the lower band and the state $\mathbf{p}+\hbar\mathbf{k}/2$ in the upper one. In such a case the interband matrix element [Eq. (27)] depends only on quasimomentum $\mathbf{p}$ which essentially simplifies the calculations.

Let us begin with a calculation of the term proportional to $\mathbf{k}$ in the parentheses in Eq. (28). The term proportional to 1 in the square brackets cancels the first term in the expansion of the exponent and does not contribute to the current. Therefore further on we will discuss only the term having the resonant factor in the exponent.

The energy difference in the equation for N_0 up to the first order in $\mathbf{k}$ is given by

$$\varepsilon^{(2)}_{\mathbf{p}+\hbar\mathbf{k}/2}-\varepsilon^{(1)}_{\mathbf{p}-\hbar\mathbf{k}/2}-\hbar\omega=\frac{1}{2\bar{m}}(\mathbf{p}^2+\eta\mathbf{k}\cdot\mathbf{p})+\varepsilon_g-\hbar\omega\ , \quad (29)$$

where

$$\frac{1}{\bar{m}}=\frac{1}{m_1}+\frac{1}{m_2}\ ,$$

$$\frac{1}{m^-}=\frac{1}{m_2}-\frac{1}{m_1}\ ,$$

$$\eta=\bar{m}/m^-\ .$$

Calculating the contribution to the surface current we are interested in we can neglect the term proportional to η in Eq. (29). For further calculation it will be convenient to present the exponent as

$$\frac{4\Omega_0N_0}{\kappa\bar{v}_z}=\frac{U(p)}{(p-p_0-i\hbar/l)(p-p_0+i\hbar/l)}\ , \quad (30)$$

where

$$p_0=\sqrt{2\bar{m}(\hbar\omega-\varepsilon_g)}\ ,$$

$$l=\tau(p+p_0)/2\bar{m}\ ,$$

and

$$U(p)=\frac{16(\hbar\Omega_0)^2\bar{m}^2}{\tau\kappa\bar{v}_z(p+p_0)^2}\ . \quad (31)$$

Apart from p_0, U depends also on the direction of quasimomentum $\mathbf{p}$. The inequality

$$p_0l\gg\hbar$$

permits us to do the p integration in the pole approximation. The same condition permits us to take all the slowly varying quantities [such as $l(p)$] at $p=p_0$. Thus we are left with the integral

$$J=\int_{-\infty}^{\infty}\frac{dp}{2\pi i}\exp\left[-\frac{U(p_0)}{(p-p_0-i\hbar/l_0)(p-p_0+i\hbar/l_0)}\right], \quad (32)$$

where $l_0=l(p_0)$. Now we expand the exponent and make use of the identity

$$\frac{1}{n!}=\oint_\Gamma\frac{dx}{2\pi i}\frac{e^x}{x^{n+1}}\ , \quad (33)$$

where Γ is a loop encircling the point $x=0$. As a result, we get

$$J=\sum_{n=0}^{\infty}\oint_\Gamma\frac{dx}{2\pi i}\frac{e^x}{x^{n+1}}\oint_C\frac{dp}{2\pi i}\frac{1}{(p-p_0-i\hbar/l_0)^n}\times\left[-\frac{U(p_0)}{(p-p_0+i\hbar/l_0)}\right]^n. \quad (34)$$

Here C is a loop around the pole $p=p_0+i\hbar/l_0$.

One can easily sum the geometric progression in Eq. (34). As a result, we get

$$J=\oint_\Gamma \frac{dx}{2\pi i x} e^x \oint_C \frac{dp}{2\pi i} \frac{(p-p_0)^2+(\hbar/l_0)^2}{(p-p_0)^2+(\hbar/l_0)^2+U(p_0)/x} . \tag{35}$$

Let us deform the contour of integration Γ into Δ. The latter we define in such a way that the pole

$$p_1=p_0+i\sqrt{(\hbar/l_0)^2+U(p_0)/x}$$

would remain within C and

$$|p_1-p_0| \ll p_0 .$$

Then all the approximations formulated above will be valid while on the contour Δ we have

$$|x| \gg U(p_0)/p_0^2 .$$

Thus

$$J=\oint_\Delta \frac{dx}{2\pi i x} e^x \frac{U(p_0) l_0/\hbar}{\sqrt{x^2+xU(p_0)(l_0/\hbar)^2}} . \tag{36}$$

One can see that there are no regions on contour Δ whose contribution would be anomalously big. Inserting Eq. (36) into Eq. (28) one can check that the skipping electron trajectories (corresponding to small values of $\bar{v}_z$) give virtually no contribution to the current. Therefore

$$g=g^{(\mathrm{lin})} \quad \text{if} \quad U(p_0)(l_0/\hbar)^2 \ll 1 , \tag{37}$$

$$g=\beta g^{(\mathrm{lin})}/\sqrt{Q/Q_c} \quad \text{if} \quad U(p_0)(l_0/\hbar)^2 \gg 1 , \tag{38}$$

where β is a numerical factor of the order of unity. The critical intensity Q_c is determined by the condition

$$U(p_0)(l_0/\hbar)^2=1 . \tag{39}$$

Making use of Eq. (31) at $v_z \approx v$ one can get Eq. (2). Here

$$f_{\mathrm{p}}^{(2)}=\frac{\hbar^2\Omega_0^2(1+\tau_2\tau_1^{-1})\exp(-\kappa z)(F_{\mathrm{p}}^{(1)}-F_{\mathrm{p}}^{(2)})}{[\varepsilon_{\mathrm{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathrm{p}}^{(1)}]^2+(\hbar^2/4)(\tau_1^{-1}+\tau_2^{-1})^2+\hbar^2\Omega_0^2(1+\tau_2\tau_1^{-1})\exp(-\kappa z)} . \tag{40}$$

In the same approximation $f_{\mathrm{p}}^{(1)}=0$. Equation (40) shows that the condition for the onset of nonlinear behavior is

$$4\Omega_0^2\tau_2^2\tau_1(\tau_1+\tau_2)^{-1} \approx 1 . \tag{41}$$

For $\tau_1 \approx \tau_2$ this condition coincides with Eq. (3). Inserting Eq. (40) into equations for the currents shows that an electron belonging to a small group makes roughly the same contribution to the current as an electron with $v_z \approx v$. Hence the contribution of the whole small group into the full current has a small factor of the order of the phase volume of the group. There are no other small factors. Thus this contribution is sensitive to much smaller electric fields.

For ordinary electron groups the critical intensity of light, Q_c, is defined by Eq. (2). For $Q \ll Q_c$ one can use $g^{(\mathrm{lin})}$ is a value of a current for sufficiently small intensities where the linear approximation is valid.

Calculating the term proportional to p_x in the parentheses in Eq. (28) we follow the same procedure and, as a result, get the same condition for the onset of the nonlinear dependence. However, the value of the numerical coefficient β should be different in this case.

One should also consider a contribution due to expansion of a resonant factor up to the terms linear in $\eta\mathbf{k}\cdot\mathbf{p}$. If one takes this into account an extra factor

$$U(p_0)[(p-p_0)^2+(\hbar/l_0)^2]^{-2}$$

in Eq. (35) will appear. As a result, there will be two simple poles at $p=p_0+i\hbar/l_0$ and at $p=p_1$ within contour C. The sum of the residues in these poles will result in extra factor in the integrand of Eq. (36),

$$[1+2\sqrt{1+U(p_0)(l_0/\hbar)^2/x}\,]^{-1} .$$

Thus a nonlinear current due to this term will be of the order of

$$g^{(\mathrm{lin})}Q_c/Q .$$

It will be intensity independent. We could not think of a geometry of the experiment where such a contribution would be predominant. In other words, it could be difficult to discern it as compared to the contribution given by Eq. (38).

Let us now discuss a contribution of a small electron group moving almost parallel to the surface. For them the approximations made at the derivation of Eq. (24) are not valid. To be specific, we assume that the electron effective mass in the conduction band is anisotropic (like for the case of the "neck" of the Fermi surface of noble metals) and its z component is very big. Then one can neglect the term with the space derivative in Eq. (16). As a result, one of the equations becomes algebraic and in the main order in δ/l one gets

the results of the linear theory—see Refs. 1–3. For $Q \gg Q_c$ we have

$$g \approx \beta \frac{g^{(\mathrm{lin})}}{\sqrt{Q/Q_c}} \propto \sqrt{Q} , \tag{42}$$

where β is of the order of 1. Here we wish to emphasize the point already mentioned in the Introduction. Because of the essential spatial nonhomogeneity of the electromagnetic field distribution within the metal ($\delta \ll l$) there is an important difference between the nonlinear theories of photomagnetic current in metals and semiconductors (see Refs. 4–6). It manifests itself in the first place in the nonlinear parameter γ, Eq. (2). The factor δ/l is, in fact, a ratio of the volume where the light is absorbed within the metal to the volume where the pho-

tomagnetic current flows. Let us mention that for the onset of nonlinear behavior one usually does not need a very strong inequality $\gamma \gg 1$. Indeed, the exponential function in Eq. (24) rapidly reaches its asymptotical value. One obtains the nonlinear parameter by equating the exponent in Eq. (24) to 1 for some typical value of $\bar{v}_z$ corresponding to the resonance condition

$$\varepsilon^{(2)}_{\mathbf{p}+\hbar\mathbf{k}} - \hbar\omega - \varepsilon^{(1)}_{\mathbf{p}} = 0 \ . \tag{43}$$

Let us now make estimates of the nonlinearity parameter given by Eq. (2). To do this we should express Ω_R^2 through the ac field within the metal, with regard of the electrodynamical boundary conditions. As a result, we get

$$|E|^2 \approx \frac{8\pi Q}{|\epsilon| c} \ . \tag{44}$$

Again having in mind the case of copper, we have taken the value of $|\varepsilon|$ for $\hbar\omega = 2.4$ eV equal to 5 (see Ref. 9). The main difficulty for an estimate of γ is in the value of the matrix element $\mathbf{P}_{21}$. In copper within the frequency interval we are interested in it is determined by the transitions between the well localized d states and conduction band. We assume it to be about $2p_F$; this value is in a reasonable correspondence with the experimental data[9,11] on copper. Then one can present the parameter of nonlinearity in the form

$$\gamma = 4|E|^2 \left(\frac{e\tau}{2m_0\hbar\omega} \right)^2 |\mathbf{P}_{21}|^2 \frac{\delta}{l} \ . \tag{45}$$

For $\tau \approx 3 \times 10^{-10}$ sec γ becomes of the order of 1 at the intensity of the order of 50 kW/cm^2 which, incidentally is much smaller than the intensities used for the second harmonic generation (see, for instance, Ref. 14).

The estimates look entirely different for small groups of electrons. For this case we assume $\tau \approx 10^{-11}$ sec. Then γ_s is of the order of unity for $Q \approx 10$ W/cm^2.

Let us give an estimate for the surface current density g for the light intensities of the order of Q_c for normal electron groups. The simplest way to do this is to use the fact that at the edge of applicability of the linear theory where $Q \approx Q_c$ the linear theory should still give a correct order-of-magnitude estimate. Then we arrive at a conclusion that for the data given above for copper the surface current density, g, should be of the order of 100 A/cm.

Finally, let us make estimates of the local heating of the metal surface by the adsorbed light. To do this, let us assume that all the intensity of the absorbed light turns into the heat which is removed from the surface by an ordinary heat conduction. We also assume that the heat conductivity is related to the conductivity σ by the Wiedemann-Franz law. As a result, we get

$$\frac{|\nabla T|}{T} \approx \frac{3e^2 Q}{\pi^2 |\varepsilon| T^2 \sigma} \ . \tag{46}$$

For $\sigma \approx 10^{10}\ \Omega^{-1}\,\mathrm{cm}^{-1}$, $T = 4$ K we have $|\nabla T|/T \approx 1$ cm^{-1}. This estimate seems to be acceptable for any reasonable rate of laser pulse repetition.

VI. CONCLUSION

In the Introduction we mentioned two main sources of photomagnetism, i.e., the photoinduced surface current and the photoinduced bulk current. In the present paper we investigated the nonlinear behavior of the current of the first type. According to theoretical considerations, the bulk current should behave basically in the same way. However, the photomagnetism associated with it, as well as its nonlinear part, should be much bigger—see Ref. 3. The linear photomagnetism associated with the bulk current has been recently investigated by one of the authors.[15] Investigation of the nonlinear photomagnetism in various metals and under various conditions (crystal surface orientation, light polarization direction, temperature interval, impurity contents, etc.) can be of considerable interest for several reasons.

First, this is a way to study properties of a system of electrons highly excited within conduction bands as well as modes of relaxation of these electrons. Some processes that are not typical for a more usual situation of electrons slightly displaced from the equilibrium can be of much more importance for the electrons highly above the Fermi level and the holes well below it. Among these one can name in the first place the electron-electron collisions. Their investigation may provide a lot of useful information in the future.

Second, this is a powerful way to study interaction of the electrons with the light. It should prove very interesting to investigate this phenomenon together with the intensity dependence of light reflection and absorption.

Third, this could provide a way to study various aspects of interaction of conduction electrons with the surface of a metal. Again this effect can provide a unique possibility to investigate this for high energy electrons.

In summary, we have worked out a theory of the nonlinear photomagnetic response. The theory predicts the dependence for the threshold intensity for the onset of the nonlinear behavior Q_c on the matrix element of the interband transition induced by light, the quasimomenta of the electrons taking part in these transitions, electron effective masses in both bands, electron mean free paths, and the penetration depth of the light. For $Q \gg Q_c$ the theory predicts a $Q^{1/2}$ dependence of the photomagnetic response.

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