Photomagnetism of metals: Microscopic theory of photoinduced bulk current

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It is shown that visible light incident perpendicular to the surface of a normal metal and partly reflected at the surface can excite in the metal a dc *bulk* current that extends over a distance of the electron mean free path. For the case where the current is short circuited by another conductor (for instance, a superconductor) the magnetic flux built up in such a loop is estimated. It is expected to be much larger than the photoinduced magnetic flux associated with the dc *surface* current that was recently observed in normal metals illuminated by light falling obliquely on a metal surface. A microscopic theory of such a photomagnetic dc current is worked out for the case where the current is due to light-excited interband transitions of the electrons and their interaction with the metal surface.

I. INTRODUCTION AND PHENOMENOLOGICAL CONSIDERATIONS

In a recent paper,[1] observation of the photomagnetism of metals was reported. In a double-connected metal sample illuminated in such a way that a circular dc *surface* current could be excited a build up of magnetic flux was observed. In a subsequent paper[2] dealing with a microscopic theory of the effect, two contributions to the surface current were discussed. One of them is due to the quasimomentum transfer to the conduction electrons from the light partially reflected at the metal surface. Another is due to the anisotropy of the electron transitions with regard to the light polarization direction, in combination with diffuse reflection of the electrons at the surface. Both contributions exist for the light falling obliquely on the metal surface.

The purpose of the present paper is to point out that, along with the surface current, a dc *bulk* current can be excited by light[3] and to work out a microscopic theory of that effect. The surface current is rather sensitive to the geometry. It vanishes, if, for instance, the light falls normally on a plane that is perpendicular to an axis of symmetry of the metallic crystal. To the contrary, the bulk current is present for any angle of incidence. We shall calculate the excited current for the simplest case of light incident normally on a metal surface. The light is partly reflected from the surface and partly absorbed due to the interaction with the conduction electrons.

In this section we repeat the phenomenological arguments given in Ref. 3. The microscopic theory will be presented in the following sections, starting by establishing the Boltzmann equation for the excited electrons in Sec. II and giving its solution in Sec. III. After defining a simple model for the electron spectrum in Sec. IV, the excited current is explicitly calculated in Sec. V and compared in Sec. VI with the phenomenological estimate of Sec. I. In Sec. VII the calculated effect is compared with the photoinduced surface current (and the magnetic flux associated with it) as observed and calculated in Refs. 1, 2, and 4.

There are two ways in which bulk current can be produced in a metal by incident electromagnetic radiation: interband and intraband transitions of the electrons. For visible light the first effect often plays the dominant role, and in view of current experiments with visible light (see Refs. 1 and 4) only interband transitions in a metal are considered in this paper. We would like to mention here that in semiconductors a surface current due to the interband transitions and diffuse scattering of the electrons from the semiconductor surface was observed and discussed theoretically in Ref. 5 whereas intraband transitions were considered to calculate the voltage (in semiconductors) (Refs. 6 and 7) and the bulk current (in metals) (Ref. 7) produced by the radioelectric effect.

For the sake of definiteness, let us consider transitions from a full valence band into the empty states of the conduction band above the Fermi level. Let us assume that the average effective mass of the electrons in the conduction band is much smaller than that of the electrons in the valence band, so that most of the current is carried by the former. If the z axis is perpendicular to the illuminated surface of the metal, all the excited electrons fall into two groups: those with $v_z>0$ and $v_z<0$, where v_z is the z component of the electron velocity. Both groups contribute to the electron current density j_z, the first group directly, the second after a reflection from the surface. Taking these processes into account one can calculate the light-induced current density $j_z^{(l)}(z)$.

There are two characteristic lengths that determine the spatial variation of this quantity, i.e., the light penetration depth δ and the electron mean free path l. We will be particularly interested in the case $l \gg \delta$, where the effect under consideration is rather big. Then l is the characteristic length over which the current density $j_z^{(l)}(z)$ falls off exponentially. The total current density is

$$j_z = j_z^{(l)}(z) + \sigma \mathscr{E}_z(z) = \text{const} , \tag{1}$$

where $\mathcal{E}_z$ is the dc electric field along the z direction which builds up in a stationary situation in order to guarantee charge conservation, and σ is the static electric conductivity. The actual value of the electric field is determined by the boundary conditions at the surfaces of the illuminated metal.

The simplest and most effective arrangement is achieved if the normal current excited by the light is short-circuited by a superconducting loop. Then we have no voltage,[8]

$$\int_0^L \mathcal{E}_z(z)dz=0 \ , \tag{2}$$

and the total current density, determining the magnetic flux, is obtained from Eq. (1) as

$$j_z=\frac{1}{L}\int_0^L j_z^{(l)}(z)dz \ . \tag{3}$$

Here we assume that the light falls perpendicularly on the metal surface, $z=0$; $L>z>0$ corresponds to the interior of the metal.

Let us give an order-of-magnitude estimate of the expected photomagnetic effect. The intensity of light will be measured by the time-averaged Poynting vector in the ingoing wave Q_z. For the Poynting vector of the reflected wave Q_z' we have

$$Q_z'=-(1-r)Q_z \ . \tag{4}$$

Then the absorbed energy flux is rQ_z, or the absorbed photon flux is

$$rQ_z/\hbar\omega \ , \tag{5}$$

where ω is the frequency of light. Assuming that interband transitions of the electrons are mainly responsible for the absorption of the photons, the last ratio also gives the number of electrons excited per second within the volume that equals unit area times the penetration depth δ.

Now, the excited electrons move away from the penetration layer, producing the current $j_z^{(l)}(z)$ which falls off over the distance l, so that

$$j_z^{(l)}(z)=j_0e^{-z/l} \ . \tag{6}$$

For j_0 one writes

$$\frac{j_0}{e}=\eta\frac{rQ_z}{\hbar\omega} \ , \tag{7}$$

where η is a numerical coefficient of the order of (but somewhat smaller than) unity. Its value can be calculated on the basis of a microscopic theory (that will be done in Sec. V).

Making use of Eqs. (3), (6), and (7) we arrive at

$$j_z=\eta e\frac{rQ_z}{\hbar\omega}\frac{l}{L} \ . \tag{8}$$

The total current J is

$$J=\eta e\frac{rQ_z}{\hbar\omega}\frac{l}{L}S^{(l)} \ , \tag{9}$$

where $S^{(l)}$ is the area of the illuminated part of the metal.

Now we are able to give an order-of-magnitude estimate of J. Assuming $\eta=0.5$, $r=0.1$, $l/L=0.1$, $Q_z=1$ W/cm^2, $S^{(l)}=0.1$ cm^2, and $\omega=3\times10^{15}$ s^{-1} we obtain $J\approx0.3$ mA.

The magnetic flux through the closed loop formed by a small normal-metal part and the superconducting short circuit is given by

$$\Phi=c^{-1}\mathcal{L}J \ , \tag{10}$$

where $\mathcal{L}$ is the self-inductance of the system. As a result, we get for the magnetic flux measured in units of the magnetic-flux quantum, $\Phi_0=\pi c\hbar/e$,

$$\frac{\Phi}{\Phi_0}=\eta\frac{re^2}{\pi c^2\hbar^2}\frac{l}{L}\frac{Q_zS^{(l)}\mathcal{L}}{\omega} \ . \tag{11}$$

The order of magnitude of this ratio, assuming $\mathcal{L}=5$ cm$=5\times10^{-9}$ H, is $\Phi/\Phi_0\approx10^3$. This means that the expected effect is very large. In Sec. VII we will show that this effect is, for instance, much bigger than the one due to the photoinduced surface current observed in metals in Refs. 1 and 4 and discussed theoretically in Refs. 1 and 2.

II. BOLTZMANN EQUATION

To calculate the dc current we shall use the Boltzmann equation for the stationary distribution function of the electrons in the upper band $f_{\mathbf{p}}$. Omitting (where no confusion arises) the band index (2) we can write the Boltzmann equation in the form

$$v_z\frac{\partial f_{\mathbf{p}}}{\partial z}=\left[\frac{\partial f_{\mathbf{p}}}{\partial t}\right]_{\mathrm{E}}+\left[\frac{\partial f_{\mathbf{p}}}{\partial t}\right]_{\mathrm{coll}} \ . \tag{12}$$

Here the term on the left-hand side allows for the z dependence of the distribution function. The terms on the right-hand side represent the interband transitions due to the light and the collisions with, for instance, the impurities. In analogy with Ref. 2 we write the latter as

$$\left[\frac{\partial f_{\mathbf{p}}}{\partial t}\right]_{\mathrm{coll}}=-\frac{f_{\mathbf{p}}-f_{\mathbf{p}0}}{\tau_{\mathbf{p}}} \ , \tag{13}$$

where $f_{\mathbf{p}}^{(0)}$ is the equilibrium distribution function and $\tau_{\mathbf{p}}$ is the relaxation time.

The first term on the right-hand side of Eq. (12) is calculated by means of the Fermi golden rule applied to the interaction Hamiltonian between the electron and the field,

$$\mathcal{H}_{\mathrm{int}}=\tfrac{1}{2}(\mathcal{K}+\mathcal{K}^{\dagger}) \ , \tag{14}$$

$$\mathcal{K}=\frac{i\hbar e}{2cm_0}(\nabla\cdot\mathbf{A}+\mathbf{A}\cdot\nabla) \ , \tag{15}$$

where m_0 is the free electron mass and $\mathbf{A}$ is the vector potential. We assume the light wave to be of the form

$$\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}=\mathbf{E}_0\exp[-i\omega t+ikz-\tfrac{1}{2}\kappa z] \ , \tag{16}$$

where $\delta=1/\kappa$ gives the penetration depth. The commutator $[\nabla,\mathbf{A}]$ vanishes and we have

$$\mathcal{H}=\frac{\hbar e}{\omega m_0}\exp(-i\omega t)\exp[ikz-\tfrac{1}{2}\kappa z](\mathbf{E}_0\cdot\nabla) \ . \qquad (17)$$

To allow for the z dependence of the electron distribution function $f_{\mathbf{p}}$ in Eq. (12) we can imagine that the sample is divided into slabs of thickness Δz, Δz being so small that the field in a slab can be considered as independent of z. On the other hand, Δz is still large compared to the lattice constant and to the distance traveled by an electron within a time equal to the period of the light wave. This approach permits to take into account the z dependence of the light intensity.

We assume the frequency of light ω, to be large enough, so that the conditions for the energy and quasimomentum conservation allow transitions between the lower band 1 (that is full) and the upper band 2 (that is filled up to the Fermi level). Not considering the threshold effect, we use here the zero-temperature approximation. Then we have $f_{\mathbf{p}}^{(0)}=0$ in the upper band and $f_{\mathbf{p}}^{(0)}=1$ in the lower band. This gives in the first order of the light intensity

$$\left[\frac{\partial f_{\mathbf{p}}}{\partial t}\right]_{\mathbf{E}}=\sum_{p'} G(\mathbf{p},\mathbf{p}')\delta(\varepsilon_{\mathbf{p}'}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)}) \ , \qquad (18)$$

where

$$G(\mathbf{p},\mathbf{p}')=\frac{\pi}{2\hbar}\left[\frac{e}{m_0\omega}\right]^2|\mathbf{E}(z)\cdot\mathbf{P}_{21}(\mathbf{p},\mathbf{p}')|^2 \ . \qquad (19)$$

Here $|\mathbf{E}(z)|^2=|\mathbf{E}_0|^2\exp(-\kappa z)$ [the factor e^{ikz} drops out when the absolute value is taken in Eq. (19)] and $\mathbf{P}_{21}(\mathbf{p},\mathbf{p}')$ is the interband transition matrix element given by

$$\mathbf{P}_{21}(\mathbf{p},\mathbf{p}')=-i\hbar\int d^3r\,\psi_{\mathbf{p}}^{(2)*}(\mathbf{r})\nabla\psi_{\mathbf{p}'}^{(1)}(\mathbf{r}) \ . \qquad (20)$$

Here $\psi_{\mathbf{p}}^{(i)}(\mathbf{r})$ are the normalized Bloch functions, $\mathbf{p}$ being the quasimomentum and $i=1,2$ referring to the lower (1) and upper (2) bands, respectively.

Since the z dependence of $\mathbf{E}$ has been taken out of the integral, Eq. (20) vanishes unless $\mathbf{p}'=\mathbf{p}$. Writing the Bloch functions in the form

$$\psi_{\mathbf{p}}^{(i)}(\mathbf{r})=\exp(i\mathbf{p}\cdot\mathbf{r}/\hbar)u_{\mathbf{p}}^{(i)}(\mathbf{r}) \ ,$$

we can write Eq. (20) as

$$\mathbf{P}_{21}(\mathbf{p},\mathbf{p})=-i\hbar\int d^3r\,u_{\mathbf{p}}^{(2)*}(\mathbf{r})\nabla u_{\mathbf{p}}^{(1)}(\mathbf{r}) \ . \qquad (21)$$

Going back to the Boltzmann equation (12) we obtain

$$v_z\frac{\partial f_{\mathbf{p}}}{\partial z}+\frac{f_{\mathbf{p}}}{\tau_{\mathbf{p}}}=D_0e^{-\kappa z} \ , \qquad (22)$$

where we have made use of Eq. (13). D_0, taking into account Eqs. (18) and (19) with $\mathbf{p}=\mathbf{p}'$, is given by

$$D_0=\frac{\pi}{2\hbar}\left[\frac{e}{m_0\omega}\right]^2|\mathbf{E}_0\cdot\mathbf{P}_{21}(\mathbf{p},\mathbf{p})|^2\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)}) \ . \qquad (23)$$

III. SOLUTION OF THE BOLTZMANN EQUATION

The general solution of the Boltzmann equation (22) reads

$$f_{\mathbf{p}}(z)=\frac{1}{v_z}e^{-z/v_z\tau}\int_a^z dz'e^{z'/v_z\tau}D_0e^{-\kappa z'}+Ce^{-z/v_z\tau} \ , \qquad (24)$$

where C is independent of z, but, like D_0, may depend on all other variables. C, as well as the lower integration limit a, will be determined by the boundary conditions. To do this, we have to distinguish two cases, $v_z<0$ and $v_z>0$.

A. Case $v_z<0$

For this case $f_{\mathbf{p}}=f_{\mathbf{p}}^{(-)}$ is due to interband transitions taking place within a layer of width $\delta=1/\kappa$ near the surface $z=0$ of the metal. For $z\gg\delta$, $f_{\mathbf{p}}=0$. Therefore, we have to put $C=0$ and $a\to\infty$. With

$$\beta=1/|v_z|\tau \qquad (25)$$

we have

$$f_{\mathbf{p}}^{(-)}(z)=\frac{1}{|v_z|}e^{\beta z}\int_z^\infty dz'e^{-\beta z'}D_0e^{-\kappa z'} \ . \qquad (26)$$

The z' integration yields

$$f_{\mathbf{p}}^{(-)}(z)=\frac{D_0}{|v_z|}\frac{e^{-\kappa z}}{\beta+\kappa}=D_0\frac{e^{-\kappa z}}{1/\tau+\kappa|v_z|} \quad (v_z<0) \ . \qquad (27)$$

B. Case $v_z>0$

Here $f_{\mathbf{p}}=f_{\mathbf{p}}^{(+)}$ consists of two parts. One is due to interband transitions with $v_z>0$ and vanishes for $z=0$; the other stems from the electrons that were created by interband transitions with $v_z<0$ and have been reflected by the surface $z=0$. The latter part is just represented by the last term in Eq. (24). Thus

$$f_{\mathbf{p}}^{(+)}(z)=\frac{1}{v_z}e^{-\beta z}\int_0^z dz'e^{\beta z'}D_0e^{-\kappa z'}+Ce^{-\beta z} \ . \qquad (28)$$

The z' integration is again trivial and yields

$$f_{\mathbf{p}}^{(+)}(z)=D_0\frac{e^{-\kappa z}-e^{-\beta z}}{1/\tau-\kappa v_z}+Ce^{-\beta z} \quad (v_z>0) \ . \qquad (29)$$

C has to be determined in such a way that at the surface $z=0$ there is no net current in the z direction, i.e.,

$$\int_{v_z>0}d^3p\,f^{(+)}(0)v_z+\int_{v_z<0}d^3p\,f^{(-)}(0)v_z=0 \ . \qquad (30)$$

Taking the values of $f^{(+)}(0)$ and $f^{(-)}(0)$ from Eqs. (29) and (27), we obtain

$$\int_{v_z>0}d^3p\left[Cv_z-\frac{D_0v_z}{1/\tau+\kappa v_z}\right]=0 \ . \qquad (31)$$

In order to determine the relation between C and D_0 we are going to use a specific model. This will be defined in Sec. IV.

IV. ISOTROPIC ELECTRON SPECTRUM MODEL

For the further calculations we exploit a simple model of an isotropic electron spectrum. It will enable us to make order-of-magnitude estimates of the expected effects.

We assume the electron spectrum in both bands to be isotropic and quadratic:

$$\varepsilon^{(1)}(p)=p^2/2m_1,\quad \varepsilon^{(2)}(p)=\varepsilon_g+p^2/2m_2 \ . \tag{32}$$

Here we assume the effective mass to be positive if the curvature of the band is positive, or, in other words, $\partial^2\varepsilon/\partial p_i^2>0$. As for the probabilities of the interband transitions, we make one of the simplest assumptions compatible with the isotropic model, namely

$$\mathbf{P}_{21}(\mathbf{p},\mathbf{p})=2\alpha\mathbf{p} \ , \tag{33}$$

where α is a 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 estimates. What is of importance, though, are the numerical values of the coefficient α. In the cases where the almost free-electron model is applicable α should be considered as much smaller than one. The smallness is determined by the parameter proportional to the ratio of the pseudopotential constant(s) to some characteristic energy of the order of the Fermi energy.

V. CALCULATION OF THE CURRENT

For simplicity, we again start by assuming that the effective mass of the electrons in the upper band is much smaller than the absolute value of the effective mass of the electrons in the lower band. Then it is sufficient to calculate only the contribution to the current stemming from the upper band. At the end of this section we drop this assumption and give a formula for the contribution of both bands to the current.

We have now to find the relation between C and D_0 in Eq. (31). According to Eq. (33), D_0 [Eq. (23)] takes the form

$$D_0=B|\mathbf{E}_0\cdot\mathbf{p}|^2\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)}) \ , \tag{34}$$

$$B=\frac{2\pi}{\hbar}\left[\frac{e\alpha}{m_0\omega}\right]^2 . \tag{35}$$

The simplest way of satisfying condition (31) would be to put

$$C=\frac{D_0}{1/\tau+\kappa v_z} \ . \tag{36}$$

However, we have to keep in mind that C represents the excited electrons that have been reflected by the surface. If the reflection were specular Eq. (36) would be justified. However, it is more realistic to assume that the electrons are elastically, but diffusely reflected by the surface. Therefore, C should have the energy dependence, but not the angular dependence (in particular, favoring the electrons with $\mathbf{p}$ in the direction of $\mathbf{E}_0$) of D_0 [Eq. (34)]. Consequently, we take

$$C=\mathcal{A}\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)}) \ . \tag{37}$$

With the help of Eqs. (34) and (37) we are now able to evaluate the integral in Eq. (31). It takes the form

$$\int_{v_z>0}d^3p\left[\mathcal{A}v_z-B\frac{(\mathbf{E}_0\cdot\mathbf{p})^2v_z}{1/\tau+\kappa v_z}\right]\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)})=0 \ . \tag{38}$$

According to Eq. (32)

$$v_z=\partial\varepsilon_{\mathbf{p}}^{(2)}/\partial p_z=p_z/m_2 \ ;$$

the argument of the δ function is

$$\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}}^{(2)}=-p^2/2m+\hbar\omega' \ , \tag{39}$$

where

$$\frac{1}{m}=\frac{1}{m_2}-\frac{1}{m_1},\quad \hbar\omega'=\hbar\omega-\varepsilon_g \ . \tag{40}$$

The integrals in Eq. (38) are straightforward and lead to the result

$$\mathcal{A}=B|E_0|^2\frac{m_2p_{\omega'}}{\kappa}F_1\left[\frac{m_2}{\tau\kappa p_{\omega'}}\right] , \tag{41}$$

where

$$p_{\omega'}=\sqrt{2m\hbar\omega'}$$

and

$$F_1(\gamma)=\int_0^1 d\zeta\frac{\zeta(1-\zeta^2)}{\zeta+\gamma}=\frac{2}{3}+\frac{1}{2}\gamma-\gamma^2-\gamma(1-\gamma^2)\ln\frac{1+\gamma}{\gamma} \ . \tag{42}$$

The density of the current excited by the light is given by

$$j_z^{(l)}(z)=2e\int_{v_z<0}\frac{d^3p}{(2\pi\hbar)^3}f_{\mathbf{p}}^{(-)}(z)v_z+2e\int_{v_z>0}\frac{d^3p}{(2\pi\hbar)^3}f_{\mathbf{p}}^{(+)}(z)v_z \ . \tag{43}$$

What we actually need is the total current density j_z, Eq. (1), which is given by Eq. (3). Using Eqs. (27) and (29), we obtain

$$j_z=\frac{1}{L}\int_0^L dz\, j_z^{(l)}(z)=\frac{2e}{L}\int_0^L dz\int_{v_z>0}\frac{d^3p}{(2\pi\hbar)^3}v_z\left[-D_0\frac{e^{-\kappa z}}{1/\tau+\kappa v_z}+D_0\frac{e^{-\kappa z}-e^{-\beta z}}{1/\tau-\kappa v_z}+Ce^{-\beta z}\right] . \tag{44}$$

Assuming that $\kappa L\gg1$, $\beta L\gg1$ and remembering that $\beta=1/\tau|v_z|$, the z integration yields

$$j_z=\frac{2e}{L}\int_{v_z>0}\frac{d^3p}{(2\pi\hbar)^3}v_z\left[-D_0\frac{1}{\kappa(1/\tau+\kappa v_z)}+D_0\frac{\tau}{\kappa}+C\tau v_z\right] . \tag{45}$$

Introduction of the expressions (34), (37), and (41) leads to integrals similar to the ones in Eq. (38). The result is

$$j_z = \frac{2\pi e}{Lh^3} B |E_0|^2 \frac{m}{m_2} p_{\omega'}^4 \frac{\tau}{\kappa} F(\gamma) , \tag{46}$$

where [compare with Eq. (42)]

$$F(\gamma) = (\tfrac{2}{3} - \gamma) F_1(\gamma) + \tfrac{1}{4}$$
$$= \tfrac{25}{36} - \tfrac{1}{3}\gamma - \tfrac{7}{6}\gamma^2 + \gamma^3 + \gamma(\gamma - \tfrac{2}{3})(1-\gamma^2) \ln \frac{1+\gamma}{\gamma} . \tag{47}$$

Here

$$\gamma = \frac{m_2}{\tau \kappa p_{\omega'}} = \frac{1}{\kappa l} = \frac{\delta}{l} \tag{48}$$

gives the ratio of the penetration depth of the electromagnetic wave to the mean free path

$$l = \tau \frac{p_{\omega'}}{m_2} \tag{49}$$

of the high-energy electrons in the conduction band with velocity $p_{\omega'}/m_2$. Introducing l and δ into Eq. (46) and substituting for B Eq. (35) we obtain the final result

$$j_z = \frac{em}{2\pi L \hbar^4} \left[\frac{ea}{m_0 \omega} \right]^2 |E_0|^2 p_{\omega'}^3 l \delta F \left[\frac{\delta}{l} \right] . \tag{50}$$

Let us look at the limiting cases $\gamma = \delta / l \ll 1$ and $\gamma \gg 1$. We have

$$\gamma \ll 1, \quad F(\gamma) \to \tfrac{25}{36} , \tag{51}$$

$$\gamma \gg 1, \quad F(\gamma) \to \frac{3}{10\gamma} . \tag{52}$$

The physics of these results is rather transparent. If the mean free path l of the electrons is large compared to the penetration depth δ, then $\gamma \ll 1$, and the average current density j_z [see Eqs. (1) and (3)] is proportional to $l\delta$ because δ is the thickness of the layer in which interband transitions take place and l is the length over which the current $j_z^{(l)}$ [see Eq. (6)] decays due to the collisions in the bulk of the metal.

In the opposite case, if the mean free path l of the electrons is much smaller than the penetration depth δ (or, in other words, $\gamma \gg 1$), of all the electrons excited in the layer of thickness δ only those within a thin layer of the width l near the surface are sensitive to the presence of the surface scattering. Elsewhere, because of the symmetry of the transition probability in regard of exchange of $\mathbf{p}$ by $-\mathbf{p}$, there is a cancellation of the contributions with $v_z > 0$ and $v_z < 0$. Then the previous result is reduced by a factor $\frac{54}{125}(l/\delta)$ and j_z is proportional to l^2.

We can add to this that, whereas the actual behavior of the current in the intermediate case $\delta / l \approx 1$ and the actual numerical coefficients in the extreme cases are sensitive to the model for the electron spectrum and the transition probabilities, the dependencies on the electron mean free path and the penetration depth given by Eq. (50), in combination with (51) or (52) for the limiting cases are quite general.

At the end of this section we wish to give an equation for the two-band contribution to the current for the case where for both bands the conditions $l^{(1,2)} \gg \delta$ are fulfilled. We have

$$j_z = \frac{25}{36} \frac{em}{2\pi L \hbar^4} \left[\frac{ea}{m_0 \omega} \right]^2 |E_0|^2 p_{\omega'}^4 \delta \left[\frac{\tau^{(2)}}{m_2} - \frac{\tau^{(1)}}{m_1} \right] , \tag{53}$$

where $\tau^{(1,2)}$ are the times of relaxation and $m_{1,2}$ are the effective masses (with their appropriate signs) in the first and second bands, respectively.

VI. COMPARISON OF PHENOMENOLOGICAL AND MICROSCOPIC APPROACHES

Now we are going to calculate the rate of the interband electron transitions, per square centimeter, induced by the light. On the one hand, this should be equal to the rate of photon absorption by the electrons, $rQ_z/\hbar\omega$. On the other, the same quantity in the limit $l \gg \delta$ should determine the current density, Eq. (8). Comparing both expressions we shall be able to determine the coefficient η entering this equation.

One obtains the rate of electron generation in the upper band, which is equal to the rate of photon absorption within the layer of the width δ near the surface, by integration of Eq. (18) over $2d^3p$ and dz. One gets

$$\frac{rQ_z}{\hbar\omega} = \delta \frac{1}{3\pi^2 \hbar^3} m p_{\omega'}^3 B |E_0|^2 , \tag{54}$$

where B is defined by Eq. (35). Equation (54) can be used in principle to establish the relation between the coefficient r (defining the light absorption) and the matrix element $\mathbf{P}_{21}$ as well as the electron dispersion laws in the two bands. However, we use it here for comparison with Eq. (46), which, in view of Eq. (49), for the most interesting case $l \gg \delta$ ($\delta = 1/\kappa$) gives

$$j_z = \frac{25}{36} \delta \frac{e}{4\pi^2 \hbar^3} B |E_0|^2 m p_{\omega'}^3 \frac{l}{L} . \tag{55}$$

By comparison of Eqs. (8), (54), and (55) we get

$$\eta = \tfrac{25}{48} . \tag{56}$$

The value of this coefficient is model sensitive. However, we wish to emphasize that for any type of interaction of the electrons with light (i.e., for any realistic dependence on $\mathbf{p}$ of the matrix element $\mathbf{P}_{21}$) our microscopic calculation confirms the estimate given by Eq. (8) with the numerical coefficient η of the order of 1.

VII. CONCLUSION

On the basis of the calculation presented one can see that the expected effect is rather large. It is interesting to compare it to the photoinduced surface current (and to the magnetic flux associated with it). Such a current in metals was observed in Refs. 1 and 4 and discussed theoretically in Refs. 1 and 2. It exists for an oblique incidence of light on the metal surface and has, in general, two contributions. One of them is associated with the transfer of the x or y component of the quasimomentum from the light to the conduction electrons. These are the components parallel to the surface and are conserved at the metal surface. Another contribution, the so-called photogalvanic current, is due to the asymmetry in the electron distribution brought about by the surface scattering of the conduction electrons.[5,2]

In the geometry considered in the present paper the z component of the quasimomentum (i.e., the one perpendicular to the surface) is not conserved at the metal surface. The contribution of the quasimomentum transfer to the bulk current associated with the interband transitions is of little significance and we do not consider it here. Therefore, it is worthwhile to compare the photomagnetism in the present geometry with that associated with the photogalvanic surface current.

We begin with pointing out that the current investigated in the present paper is proportional to the value of $|E_{0x}|^2$ at the metal surface, while the photogalvanic surface current is proportional to $\mathrm{Re}(E_{0x}^* E_{0z})$. To compare these two values, one should keep in mind some facts concerning the free-electron contribution to the dielectric susceptibility of a typical metal. In the zeroth approximation in the electron scattering it is given by

$$\epsilon=\epsilon_0-\frac{\omega_p^2}{\omega^2}\ , \tag{57}$$

where ϵ_0 is a contribution due to atomic core polarization (usually comprising several units within the frequency range we are interested in), whereas the plasma frequency ω_p is given by

$$\omega_p^2=\frac{4\pi n e^2}{m}\ . \tag{58}$$

Here n is the free-electron concentration, whereas m is their average effective mass. To give an estimate for typical metals, we take n of the order of 10^{23} cm^{-3} and m of the order of the free-electron mass. We get

$$\omega_p^2 \approx 3\times 10^{32}\ \mathrm{s}^{-2}\ ,$$

which means that for visible light the absolute value of the second term in Eq. (57) is at least bigger than ten so that $\epsilon(\omega)$ is negative. It means, in its turn, that visible light is strongly reflected at the metal surface.

Now, we wish to indicate the following points. First, if the absolute value of the metal dielectric susceptibility $|\epsilon|$ is much bigger than 1, $|E_{0z}|$ is smaller than $|E_{0x}|$ by a factor $|\epsilon|^{-1/2}$. Second, in the zeroth approximation in the electron scattering ϵ appears to be negative. As a result, in the same approximation E_{0z} is completely out of phase relative to E_{0x}, so that the time average of $E_x E_z$, $\mathrm{Re}(E_{0x}^* E_{0z})=0$. This quantity differs from zero only in the next approximation, and there it is proportional either to the intensity of the interband scattering, i.e., to α^2, or to the intensity of intraband scattering, i.e., to $1/\omega\tau$.[9] These are the main reasons as to why the current considered in the present paper may be big compared to that considered in Refs. 1, 2, and 4.

To this statement we can add the following consideration. The photogalvanic surface current is very sensitive to the elastic electron reflection at the surface (see Ref. 5). It vanishes for pure specular reflection. The bulk current calculated in the present paper is not particularly sensitive to the character of reflection. We made our calculation for the diffuse reflection as we consider this case to be more realistic. However, the calculation could have been easily done for the specular reflection with a slight alteration of the result (coefficient η).

We may also mention that the photogalvanic contribution is very sensitive to the polarization of the light and, according to Refs. 2 and 5 can even vanish for the polarization in a particular plane. On the other hand, the effect can be enhanced for circular polarization. To the contrary, the current considered in the present paper is not very sensitive to the polarization of the light. All this means that investigation of surface and bulk currents reveal different properties of the conduction electrons. We would like to remark once again that the case of normal incidence of light was considered here only for simplicity. For oblique incidence, the excitation of a dc bulk current should also take place.

Generally, the experimental investigation of this effect may be of considerable interest. First, under the illumination the electrons are highly excited above the Fermi level and in such a way one has a unique possibility to investigate the properties of these electrons and their relaxation. Second, together with the light absorption, this is a tool to investigate the interaction of the electrons with light. Third, in this way one can learn a lot about the interaction of highly energetic conduction electrons with the metal surface.

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[8] The implication is that the superconductor is in good contact with the normal conductor. Such a contact can be easily achieved at the far (unilluminated) surface of the normal conductor. One way to make a good contact at the illuminated

surface of the normal metal is to cover it with a superconducting gauze, the dimensions of a mesh being smaller than the electron mean free path. If the gauze is covered by a nontransparent substance only the normal metal will be illuminated.

[9]The theory developed in the present paper, with slight modifications, may prove to be also applicable to semiconductors and semimetals outside the region of their transparency. However, because of the small electron concentration in these materials, it may be easier to measure not the current but the voltage built up across an insulated sample.