Photomagnetism of metals: Microscopic theory of the photoinduced surface current

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Photoinduced magnetic flux has been observed recently in normal metals exposed to unpolarized visible 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. Both contributions are compared. The relation between them may be particularly sensitive to the polarization of the light.

I. INTRODUCTION

In a recent paper[1] the observation of the photomagnetism of metals was reported. In a sample of metal illuminated in such a way that a circular dc current could exist (see Fig. 1) a buildup of magnetization was observed. Estimates of the current made in the same paper were based on the conservation of the quasimomentum of light and conduction electrons.

The quasimomentum of light and its conservation in relation to the homogeneity of the medium where light propagates have been discussed in detail by Peierls.[2] In Ref. 3 (cf. also with Ref. 4) equations for the quasimomentum flux of light are given and the quasimomentum conservation in the interaction of light with the conduction electrons is discussed.

These considerations, however, permit one to make only a rough estimate of the observed effect. The purpose of the present paper is to work out a microscopic theory of a surface current excited by light. This should permit one to make a more detailed comparison with future experimental data and to obtain important information concerning the high-energy electrons excited by light.

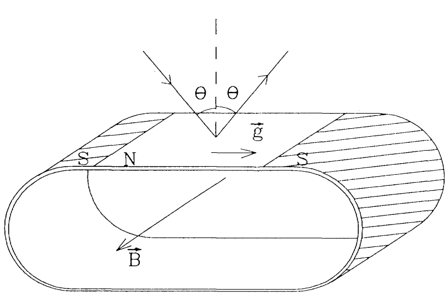

FIG. 1. An example of an arrangement where a dc circular current is excited under the action of light. Light falls obliquely on a surface of uniformly illuminated normal metal at an angle θ to the perpendicular and is partly reflected. The dc surface current is excited due to the quasimomentum transfer and photogalvanic effect. The current is short circuited by other metal, preferably a superconductor, so that the whole current encircles the orifice. The current creates magnetic flux within the loop.

The effect we are going to discuss has much in common with a phenomenon well known for semiconductors, i.e., drag of electrons by a traveling electromagnetic wave. It was first investigated by Barlow in Ref. 5 where it was treated classically as a Hall current in ac crossed electric and magnetic fields. Later a voltage produced by a traveling wave was calculated in much detail for semiconductors in Refs. 6 and 7 and for plasmas in Ref. 8 (in view of a vast amount of the literature we are giving here only references to some of the earliest papers). It was observed by Danishevskii *et al.*,[9] Gibson, Kimmit, and Walker,[10] and Khaikin and Yakubovskii.[11] There is, however, an important difference between the manifestation of the effect in semiconductors and in metals. In semiconductors, where the conductivity is rather small, the current excited by a traveling wave is also small. It may be difficult to measure it directly. Therefore one usually measures the voltage built up across the sample. In metals, because of the high conductivity, the voltage is too small. To the contrary, the sensitivity of the existing devices is so high that the current usually can be measured with a rather high accuracy.

The arrangement investigated in Ref. 1 differs from those used in most cases for semiconductors also in that instead of a traveling wave within the bulk we have in metals a wave reflected from a conductor's surface. However, in both cases the effect is due (at least partly) to the fact that the quasimomentum of an electromagnetic wave is fed into the electron system. A surface current due to the transfer of the *momentum* of light was discussed by Ivchenko and Pikus[12] and Normantas and Pikus.[13] They treated in detail the normal incidence of light on a semiconductor surface and were particularly interested in the

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case of anisotropic energy surfaces. They indicated that a surface current may appear also in an isotropic case for oblique incidence of light.

In semiconductors a surface current of a different nature, which may be called photogalvanic, was also discussed. It was predicted by Margarill and Entin[14] and observed as well as discussed in more detail by Al'perovich *et al.*;[15] they were particularly interested in the case where light excited interband transitions in a semiconductor. This effect is due to a diffuse scattering of the electrons from the surface (see Fig. 2). In general the transition probability is an anisotropic function of the electron quasimomentum. The anisotropy exists even in the case of an isotropic electron spectrum due to the directional asymmetry brought about by the electric ac field vector **E**. Let us assume that **E** lies in the plane xz where z is perpendicular and x is parallel to the surface. In the simplest (isotropic) case one can consider the situation where the transition probability has an item proportional to $(\mathbf{Ep})^2$, **p** being the electron quasimomentum. As a result of the invariance of the transition probability to the change of the sign of the quasimomentum, the number of electrons generated by light and moving, say, along the negative direction of the x axis and, at the same time, towards the surface would be equal to the number of electrons moving along $+x$ and from the surface. The electrons of the first group would be scattered from the surface and if the scattering were diffuse, they would not give appreciable contribution to the current. The electrons of the second group would be scattered only in the bulk of the sample and therefore their contribution to the current parallel to the surface would be larger.

This effect should disappear for specular scattering of the electrons at the surface.[15] It should not exist for polarization of ac electric field **E** in the y direction, i.e., perpendicular to the plane of light propagation. This last statement is valid for an isotropic case which can be seen directly from the form of the angular dependence of the transition probability indicated above. It should also be

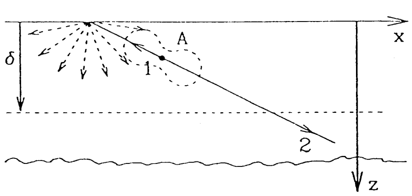

FIG. 2. A section of the normal metal (depicted in Fig. 1) near the illuminated surface. The vector of the ac electric field within the surface layer is parallel to the arrows. Arrows 1 and 2 indicate two opposite directions where the transition probability has maxima. Angular distribution of the transition probability (having a central symmetry) is indicated by the dashed line. Electron 1 moves from point A along negative x and, at the same time, along negative z towards the surface where it experiences diffuse scattering. Its contribution to g_x is small. Electron 2 passes a much longer distance moving from the surface and being scattered within the bulk. Its contribution to the surface current is much larger. As a result, there is a surplus of excited electrons moving in the x direction.

true if, for instance, the plane of light propagation coincides with a plane of symmetry of the crystal or the direction of the surface current coincides with an axis of symmetry of the crystal (provided it has a center of symmetry). This effect should also be considerably reduced in metal films of thickness smaller than the light penetration depth (provided, of course, that the conditions for the electron scattering are the same at both surfaces of the film). One should also mention that the conditions for penetration of an ac electric field differ substantially in metals and semiconductors. In the range of optical frequencies well below the so-called plasma edge the dielectric susceptibility of a metal is negative and its modulus may be rather large (see Sec. X). It means that in the first approximation the z component of the ac field is entirely out of phase in regard to its x component (i.e., the phase difference is $\pi/2$). In this approximation one gets no contribution to the photogalvanic current. In the next approximation the dielectric susceptibility is modified due to the absorption of light. The phase difference between E_z and E_x is not exactly equal to $\pi/2$. As a result, there is some dc surface photogalvanic current, though it should be usually reduced in comparison to semiconductors.

However, in general, in bulk metal specimens two contributions to the dc surface current excited by illumination should exist. In what follows we shall compare both contributions.

II. PHENOMENOLOGY

We start with phenomenological expressions for both contributions to the current writing these for an isotropic conductor. However, generalization for an anistropic case is rather simple.

In the lowest approximation in the light intensity, an equation for the dc bulk current density **j**, due to the drag of the electrons by an electromagnetic wave, can be written as (here and henceforth we use the Einstein summation convention)

$$j_i = i\frac{\xi}{2}\left[E_l\frac{\partial E_l^*}{\partial x_i} - E_l^*\frac{\partial E_l}{\partial x_i}\right] + i\frac{\eta}{2}\left[E_l\frac{\partial E_i^*}{\partial x_l} - E_l^*\frac{\partial E_i}{\partial x_l}\right] \tag{1}$$

where $\xi(\omega)$ and $\eta(\omega)$ are transport coefficients which in general depend on the frequency of light. Here all the terms are bilinear in ac electric field components E_i and E_i^* and their first derivatives over the coordinates. Indeed, these components are proportional to $e^{-i\omega t}$ and $e^{i\omega t}$, respectively. These terms change their sign for a traveling wave if it changes the direction of its propagation. The symmetry allows also the term proportional to $i(\mathbf{E}^*\mathrm{div}\mathbf{E} - \mathrm{c.c.})$; we have not included it as $\mathrm{div}\mathbf{E}=0$. In principle, two more terms containing the first derivatives could enter the equation for the current density. One of them gives a dc current perpendicular to the surface, which we do not consider here. Another one has components parallel to the surface. However, it will not be present in our microscopic calculations for it could appear only in higher approximations of the perturbation theory; henceforth we shall omit it.

The first term of Eq. (1) can be transformed with the help of the identity

$$[\mathbf{E},\mathrm{curl}\mathbf{E}^*]-[\mathbf{E}^*,\mathrm{curl}\mathbf{E}]=E_l(\nabla E_l^*)-E_l^*(\nabla E_l)+(\mathbf{E}^*\nabla)\mathbf{E}-(\mathbf{E}\nabla)\mathbf{E}^* , \quad (2)$$

which gives for the current density

$$\mathbf{j}=i\frac{\xi}{2}([\mathbf{E},\mathrm{curl}\mathbf{E}^*]-[\mathbf{E}^*,\mathrm{curl}\mathbf{E}])+i\frac{\eta'}{2}[(\mathbf{E}\nabla)\mathbf{E}^*-(\mathbf{E}^*\nabla)\mathbf{E}] \quad (3)$$

where $\eta'=\xi+\eta$. The first term of Eq. (3) can be transformed making use of the Maxwell equation $\mathrm{curl}\mathbf{E}=(i\omega/c)\mathbf{H}$ and we get

$$\mathbf{j}=\frac{4\pi\xi\omega}{c^2}\mathbf{Q}+i\frac{\eta'}{2}[(\mathbf{E}\nabla)\mathbf{E}^*-(\mathbf{E}^*\nabla)E] \quad (4)$$

where $\mathbf{Q}=(c/4\pi)\mathrm{Re}[\mathbf{E},\mathbf{H}^*]$ is the time-averaged Poynting vector inside the metal.

This is an equation for the bulk current whereas we are interested in the density of a *surface* one $\mathbf{g}$. Indeed, one observes in the experiment a jump of a stationary magnetic field $\Delta\mathbf{B}$ across a surface where a dc current flows. The jump is given by $[\mathbf{n},\Delta\mathbf{B}]=(4\pi/c)\mathbf{g}$ where $\mathbf{n}$ is a unit vector along the normal to the metal surface. This is the manifestation of the photomagnetism (see Fig. 1). If the electron mean free path l is much smaller than the electric field penetration depth δ, one can obtain the surface current density $\mathbf{g}$ by integrating Eq. (4) over the distance to the metal surface z. If the intensity of the ac electric field varies as $|\mathbf{E}(z)|^2\propto e^{-z/\delta}$, then the integration is equivalent to multiplication by δ.

In the case where the electron mean free path l is larger than or of the order of the penetration depth δ, the collisions with the surface are important within the whole layer of the width δ and one cannot obtain a surface current by the integration of Eq. (4). Rather one should start with an equation for the surface current

$$g_\mu=\frac{4\pi\zeta_1\omega}{c^2}Q_\mu+\frac{i\zeta_2}{2}\left[E_\nu\frac{\partial E_\mu^*}{\partial x_\nu}-E_\nu^*\frac{\partial E_\mu}{\partial x_\nu}\right]+\frac{i\zeta_3}{2}\left[E_\mu\frac{\partial E_\nu^*}{\partial x_\nu}-E_\mu^*\frac{\partial E_\nu}{\partial x_\nu}\right] , \quad (5)$$

where μ,ν run through values x,y; E_μ are the components of the electric field at the surface.

An important feature of Eqs. (4) and (5) is that they in general give a current not only along the x axis but also along the y axis. The physical origin of this current may be visualized as follows.

The primary source of the x component of the current is associated with the fact that, due to the quasimomentum transfer, the electrons with, say, $p_x>0$ outnumber those with $p_x<0$. As a result, there is a net electron current along the x axis. In regard to the transitions excited by light this means that the number of transitions where the electrons with, say, $p_x>0$ are involved is larger than that of the electrons with $p_x<0$.

On the other hand, the transition probabilities are in general even functions of the quasimomentum $\mathbf{p}$ (for more details see Sec. V). It means that a probability can have a part asymmetric in p_x, then it should be also asymmetric in p_y (or p_z). For instance, it can have an item $2p_xp_y\mathrm{Re}(E_x^*E_y)$ (this is just the simplest possibility that takes into consideration the proper symmetry and permits us to see the physics. On the basis of this example one can see the essential features characteristic of a general case). The presence of such a term means that if the electrons with $p_x>0$ are in a majority in the upper band the electrons with $p_y>0$ should also have a majority there [provided $\mathrm{Re}(E_xE_y^*)>0$]. Since there is no quasimomentum transfer in the y direction that would have meant nothing for the y component of the current if both bands were exactly alike. As, however, the velocities as well as the relaxation times in both bands differ, an asymmetric redistribution of the electrons between the bands should, in general, result in a current in the y direction. One can also add that such a redistribution should make a contribution to j_x in addition to the one that is directly associated with the quasimomentum transfer. This is reflected by the fact that the second term in Eq. (1) can contribute both to j_x and j_y.

The phenomenological equation for the photogalvanic surface current according to Ref. 15 has the form

$$\mathbf{g}=\frac{\lambda}{2}\{[\mathbf{E}-\mathbf{n}(n\mathbf{E})](\mathbf{nE}^*)+\mathrm{c.c.}\} . \quad (6)$$

There could be another contribution,[15] namely $i\lambda'[\mathbf{n},[\mathbf{E},\mathbf{E}^*]]$. It can appear in higher approximations of the perturbation theory, and we are not going to consider it. It is important to note that Eq. (6) also gives in general a current not only along the x axis but also along the y axis.

III. PROBABILITY OF INTERBAND ELECTRON TRANSITIONS INDUCED BY LIGHT

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

$$\mathcal{H}_{\mathrm{int}}=\tfrac{1}{2}(\mathcal{K}+\mathcal{K}^\dagger) , \quad (7)$$

$$\mathcal{K}=(ie/2\omega m_0)e^{-i\omega t}[e^{ik_xx}(-i\hbar\mathbf{E}\nabla)+(-i\hbar\mathbf{E}\nabla)e^{ik_xx}] . \quad (8)$$

Here ω is the frequency of light, m_0 is the free-electron mass; here and henceforth $\mathbf{E}(z)$ will denote 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

$$\langle 2\mathbf{p}'|\mathcal{H}|1\mathbf{p}\rangle=\frac{ie}{m_0\omega}\mathbf{E}\mathbf{P}_{21}e^{-i\omega t}\delta_{p_x',p_x+\hbar k_x}\delta_{p_y',p_y}\delta_{p_z',p_z} \tag{9}$$

where $\delta_{\mathbf{p}'\mathbf{p}}$ is the Kronecker symbol. Here

$$\mathbf{P}_{21}(\mathbf{p}',\mathbf{p})=\frac{1}{\Omega_0}\int d^3r\, u_{\mathbf{p}'}^{(2)*}(\mathbf{r})\left[-i\hbar\frac{\partial}{\partial\mathbf{r}}+\frac{\mathbf{p}+\mathbf{p}'}{2}\right]u_{\mathbf{p}}^{(1)}(\mathbf{r})\ , \tag{10}$$

Ω_0 being the volume of the primitive cell and $u_{\mathbf{p}}^{(1,2)}(\mathbf{r})$ being the Bloch amplitudes.

The actual calculation of transition amplitudes for a particular metal may be rather complicated. For an important example of almost-free electrons this has been done in Refs. 16–18. In the present paper we shall not use the results of such calculations. However, we shall exploit extensively the symmetry properties of the transition amplitudes derived below. The transition probability from Bloch state $1,\mathbf{p}$ to state $2,\mathbf{p}'=\mathbf{p}+\hbar\mathbf{k}$ is given by $G\delta(\varepsilon_{\mathbf{p}}^{(1)}+\hbar\omega-\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)})$ where

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

The variation of the electron distribution function due to interaction with light is

$$\left[\frac{\partial f_{\mathbf{p}}^{(1)}}{\partial t}\right]_{\mathbf{E}}=G\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})(f_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-f_{\mathbf{p}}^{(1)})\ , \tag{11}$$

$$\left[\frac{\partial f_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}}{\partial t}\right]_{\mathbf{E}}=-G\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})(f_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-f_{\mathbf{p}}^{(1)})\ . \tag{12}$$

Here we consider quantum transitions of the electrons while the electric field is treated classically. Frequency ω is assumed to be large 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). 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 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. It means that the z component of the quasimomentum can also be transferred to the electrons at the surface, thus creating a current or voltage along this direction. However, as has already been mentioned, we shall disregard this effect.

IV. BOLTZMANN EQUATION

The system of Boltzmann equations for the time-averaged electron distribution functions $f^{(1,2)}$ has the form

$$v_z^{(1,2)}\frac{\partial f^{(1,2)}}{\partial z}=\left[\frac{\partial f^{(1,2)}}{\partial t}\right]_{\mathbf{E}}+\left[\frac{\partial f^{(1,2)}}{\partial t}\right]_{\text{coll}}\ . \tag{13}$$

Here the term on the left-hand side allows for the coordinate dependence of the electron distribution function. The terms on the right-hand side represent interband transitions brought about by the illumination and intraband transitions not conserving the quasimomentum which may be due, for instance, to collisions of the electrons with defects.

As the distribution of the electrons excited by a laser light is usually very sharp in the quasimomentum space the "out" term of the collision operator should be much larger than the "in" term. This means that to describe the collisional variation of the sharp part of the distribution function (we are interested in) one may use the relaxation-time approximation

$$\left[\frac{\partial f_{\mathbf{p}}^{(1,2)}}{\partial t}\right]_{\text{coll}}=\frac{f_{\mathbf{p}}^{(1,2)}-f_{\mathbf{p}0}^{(1,2)}}{\tau_{\mathbf{p}}^{(1,2)}} \tag{14}$$

where $f_{\mathbf{p}0}^{(1,2)}$ are the equilibrium distribution functions whereas $\tau_{\mathbf{p}}^{(1,2)}$ are the relaxation times for the electrons in bands 1 and 2, respectively.

To calculate the current in the lowest approximation in the light intensity we should insert into Eqs. (11) and (12) the electron distribution functions in the zeroth-order approximation. As the energy $\hbar\omega$ is much larger than the thermal energy we may, for all practical purposes (except for the analysis of the threshold phenomena), consider the case of zero temperature. Then for the transitions that are allowed by the Pauli principle we have $f_{p0}^{(1)}=1$ and $f_{p0}^{(2)}=0$. In the lowest approximation in the light intensity the Boltzmann equations for the variations $\Delta f_{\mathbf{p}}=f_{\mathbf{p}}-f_{\mathbf{p}0}$ of the electron distribution functions have the form

$$\begin{aligned}v_z^{(1)}(\mathbf{p})\frac{\partial\Delta f_{\mathbf{p}}^{(1)}}{\partial z}+\frac{\Delta f_{\mathbf{p}}^{(1)}}{\tau_{\mathbf{p}}^{(1)}}&=-G(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ ,\\ v_z^{(2)}(\mathbf{p}+\hbar\mathbf{k})\frac{\partial\Delta f_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}}{\partial z}+\frac{\Delta f_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}}{\tau_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}}&=G(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ .\end{aligned} \tag{15}$$

Let us start with calculation of the current initiated by the quasimomentum transfer for the simplest situation where one can neglect the terms with the spatial derivatives in Eqs. (15). This is the case where the electron mean free path l is much smaller than the electromagnetic wave penetration depth δ, so that the electron scattering is comparatively strong. As a result, we get

$$\Delta f^{(1,2)}=\mp\tau^{(1,2)}G\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ . \tag{16}$$

For the x component of the current density $j_x(z)$ we have

$$j_x=2e\int\frac{d^3p}{(2\pi\hbar)^3}[v_x^{(2)}(\mathbf{p}+\hbar\mathbf{k})\tau_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-v_x^{(1)}(\mathbf{p})\tau_{\mathbf{p}}^{(1)}]\times G(\mathbf{p}+\hbar\mathbf{k},\mathbf{p})\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{k}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ . \tag{17}$$

To obtain the y component one should replace $v_x^{(1,2)}$ by $v_y^{(1,2)}$.

This is the general formula that we shall use to investigate the case $l \ll \delta$. The derivation of this formula, however, warrants a comment. In the lowest approximation we assume the nonequilibrium part of the electron distribution function to be small: $|\Delta f^{(1,2)}| \ll 1$. Equation (16) formally violates this condition because it is singular. One can think, however, of a light beam having a finite frequency interval. This would permit us to satisfy the inequality $|\Delta f^{(1,2)}| \ll 1$ and, at the same time, would affect very little Eq. (17) where the singular function is integrated. (We may mention that such an approach would not in general be permissible were we interested in the nonlinear approximation in the light intensity. For nonlinear phenomena the real spectral width of the beam is usually of importance.)

As k is small compared to the typical values of p, we shall expand Eq. (17) in powers of $\mathbf{k}$. The term of the zeroth order vanishes as $v_x^{(1,2)}$ are odd functions of $\mathbf{p}$ while all the rest functions in the integrand are even. Our immediate purpose is to derive an equation for the dc current by calculation of the term proportional to k_x.

V. PROPERTIES OF TRANSITION PROBABILITIES

We are interested in the $\mathbf{p}$ and $\mathbf{k}$ dependence of the matrix element between the Bloch amplitudes $\mathbf{EP}_{21}(\mathbf{p}',\mathbf{p})$, where $\mathbf{p}'=\mathbf{p}+\hbar\mathbf{k}$; we assume that vector $\mathbf{k}$ is oriented along the x direction. The equation for a Bloch amplitude is (we neglect here the spin-orbit interaction for the conduction electrons)

$$(1/2m_0)[(-i\hbar\nabla+\mathbf{p})^2+V(\mathbf{r})]u_{\mathbf{p}}(\mathbf{r})=\varepsilon_{\mathbf{p}}u_{\mathbf{p}}(\mathbf{r}) \tag{18}$$

where we have omitted the superscript indicating the band's number. Here $V(\mathbf{r})=V(\mathbf{r}+\mathbf{a})$ is the periodic self-consistent potential for a conduction electron while $\mathbf{a}$ is a lattice vector, i.e., an arbitrary sum of the primitive translations. In what follows we shall consider crystals with a center of symmetry where $V(\mathbf{r})=V(-\mathbf{r})$.

Let us present relations between the Bloch amplitudes that are due to existence of a center of symmetry.[19] Making use of Eq. (18) one can see that $u_{-\mathbf{p}}(-\mathbf{r})$ satisfies the same equation as $u_{\mathbf{p}}(\mathbf{r})$. For the quasimomentum in an arbitrary position there is, in general, no degeneracy, which means that these two functions should be linearly dependent: $u_{-\mathbf{p}}(-\mathbf{r})=C_{\mathbf{p}}u_{\mathbf{p}}(\mathbf{r})$ where C_p can in general be $\mathbf{p}$ dependent. As the Bloch amplitudes are normalized we have $|C_{\mathbf{p}}|^2=1$.

The transition probability is proportional to the matrix element squared $\mathcal{G}=|\mathbf{EP}_{21}(\mathbf{p}',\mathbf{p})|^2$. Let us investigate properties of this function under variation of the signs of its arguments. Writing down the explicit equation for $\mathcal{G}(-\mathbf{p}',-\mathbf{p})$, replacing the integration variables according to $\mathbf{r}\to-\mathbf{r}$, and taking into account condition $|C_{\mathbf{p}}|^2=1$ we have $\mathcal{G}(-\mathbf{p}',-\mathbf{p})=\mathcal{G}(\mathbf{p}',\mathbf{p})$. Introducing $\mathcal{G}(\mathbf{p};\mathbf{k})=\mathcal{G}(\mathbf{p},\mathbf{p}')$ as a function of variables $\mathbf{p}$ and $\mathbf{k}=(\mathbf{p}'-\mathbf{p})/\hbar$ we have $\mathcal{G}(-\mathbf{p};-\mathbf{k})=\mathcal{G}(\mathbf{p};\mathbf{k})$.

Expanding this function in powers of k_x we get

$$\mathcal{G}(\mathbf{p};\mathbf{k})=\mathcal{G}(\mathbf{p};0)+k_x\partial\mathcal{G}/\partial k_x \tag{19}$$

where the derivative is taken at $k_x=0$. Here the first term is even under the change of the sign of $\mathbf{p}$ whereas the second one is odd. In the following sections we shall extensively use these properties.

VI. ELECTRON CURRENT DENSITY

The holes in the lower band 1 and the electrons in the upper band 2 should contribute to the total current j_x in a similar way. This can be made more transparent by the change of the integration variable in Eq. (17) $\mathbf{p}\to\mathbf{p}-\hbar\mathbf{k}/2$. As k_x is small, one can expand the equation obtained in this way by retaining the first term in k_x. One obtains the current density as a sum of terms that originate in the k_x dependence of the velocities, energies (in the argument of the δ function), probabilities, and times of relaxation. A straightforward calculation gives for the current density (cf. with Ref. 7)

$$j_x=j_x^{(1)}+j_x^{(2)}+j_x^{(3)}+j_x^{(4)}\ , \tag{20}$$

$$j_x^{(1)}=ek_x\int\frac{d^3p}{(2\pi\hbar)^3}\left[\frac{\tau_{\mathbf{p}}^{(2)}}{m_{xx}^{(2)}}+\frac{\tau_{\mathbf{p}}^{(1)}}{m_{xx}^{(1)}}\right]G(\mathbf{p},0)\times\delta(\varepsilon_{\mathbf{p}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ , \tag{21}$$

$$j_x^{(2)}=-ek_x\frac{\partial}{\partial\omega}\int\frac{d^3p}{(2\pi\hbar)^3}[v_x^{(2)}(\mathbf{p})\tau_{\mathbf{p}}^{(2)}-v_x^{(1)}(\mathbf{p})\tau_{\mathbf{p}}^{(1)}]\times G(\mathbf{p},0)[v_x^{(1)}(\mathbf{p})+v_x^{(2)}(\mathbf{p})]\times\delta(\varepsilon_{\mathbf{p}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ , \tag{22}$$

$$j_x^{(3)}=ek_x\int\frac{d^3p}{(2\pi\hbar)^3}[v_x^{(2)}(\mathbf{p})\tau_{\mathbf{p}}^{(2)}-v_x^{(1)}(\mathbf{p})\tau_{\mathbf{p}}^{(1)}]\times\frac{\partial G}{\partial k_x}\delta(\varepsilon_{\mathbf{p}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)}) \tag{23}$$

where the derivative is calculated at $k_x=0$:

$$j_x^{(4)}=ek_x\int\frac{d^3p}{(2\pi\hbar)^3}\left[v_x^{(2)}(\mathbf{p})\frac{\partial\tau_{\mathbf{p}}^{(2)}}{\partial p_x}+v_x^{(1)}(\mathbf{p})\frac{\partial\tau_{\mathbf{p}}^{(1)}}{\partial p_x}\right]\times G(\mathbf{p};0)\delta(\varepsilon_{\mathbf{p}}^{(2)}-\hbar\omega-\varepsilon_{\mathbf{p}}^{(1)})\ . \tag{24}$$

Here we have introduced a notation

$$\frac{1}{m_{xx}^{(1,2)}}=\frac{\partial^2\varepsilon_{\mathbf{p}}^{(1,2)}}{\partial p_x^2}$$

and have made use of the relations $\tau_{-\mathbf{p}}^{(1,2)}=\tau_{\mathbf{p}}^{(1,2)}$ and

$$\frac{\partial G(-\mathbf{p},\mathbf{k})}{\partial k_x}=-\frac{\partial G(\mathbf{p},\mathbf{k})}{\partial k_x}$$

where again the derivative is calculated at $k_x=0$.

In the same way one can calculate the contributions to

j_y, the current along the surface and perpendicular to the plane of light incidence.

VII. EXAMPLE OF ISOTROPIC ELECTRON SPECTRUM

To get a better insight into the physical situation let us consider a simple example. It will enable us to make order-of-magnitude estimates of the phenomenological coefficients, such as ξ and η introduced in Eq. (1). Such estimates need not be particularly sensitive to the actual symmetry of the electron dispersion laws and the transition probabilities. Therefore to investigate a case of a particular symmetry it is sufficient to use these estimates and to generalize the phenomenological relations Eqs. (1), (5), and (6).

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

$$\varepsilon^{(1)}(p)=p^2/2m^{(1)}\ ,\quad \varepsilon^{(2)}(p)=\varepsilon_g+p^2/2m^{(2)}\ . \tag{25}$$

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

$$\mathbf{P}_{21}(\mathbf{p}',\mathbf{p})=\alpha(\mathbf{p}+\mathbf{p}') \tag{26}$$

where α is a dimensionless constant. In reality the angular dependence of 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 whereas to investigate the symmetry of the expressions describing the surface current we can use a generalization of Eqs. (1) or (5). What is of real importance are the numerical values of coefficients α. For instance, in the approximation of almost-free electrons one should consider them as small, the smallness being proportional to the ratio of the pseudopotential constant to some characteristic energy of the order of the Fermi energy. As for the actual dependence of the transition probabilities on the absolute value of the quasimomentum, it is very important for order-of-magnitude estimates for semiconductors where the values of the involved quasimomentum are usually small. There one should discriminate between the cases where (depending on the symmetries of the bands involved) the expansion of $\mathbf{P}_{21}$ begins with the zeroth and the first powers of $\mathbf{p}$. In metals where all the relevant values of p are of the order of the Fermi quasimomentum p_F, the actual form of the functional dependence is of not so great importance for the estimates (cf. with Refs. 18 and 16).

To further simplify our task we shall assume that the $\mathbf{p}$ dependence of $\tau_{\mathbf{p}}^{(1)}$ and $\tau_{\mathbf{p}}^{(2)}$ is of no importance so we shall consider these two relaxation times as constants. We are giving here the result of a straightforward calculation:

$$\mathbf{j}=\mathbf{j}^{(1)}+\mathbf{j}^{(2)}\ . \tag{27}$$

Here only the two first terms of Eq. (20) are present; $\mathbf{j}^{(3)}=0$ as after the change of the variables of integration we get the function $\mathbf{P}_{21}$ given by Eq. (26) independent of $\mathbf{k}$,

$$\mathbf{j}^{(1)}=\frac{e^3\alpha^2 m p_{\omega'}^3}{3\pi\hbar^3 m_0^2\omega^2}\left[\frac{\tau^{(2)}}{m^{(2)}}+\frac{\tau^{(1)}}{m^{(1)}}\right]|\mathbf{E}|^2\mathbf{k}\ , \tag{28}$$

$$j_i^{(2)}=A_{il}k_l\ , \tag{29}$$

$$A_{xx}=-\frac{e^3\alpha^2 m^2 p_{\omega'}^3}{3\pi\hbar^3 m_0^2 m^{(+)}\omega^2}\left[\frac{\tau^{(2)}}{m^{(2)}}-\frac{\tau^{(1)}}{m^{(1)}}\right]\times(3|E_x|^2+|E_y|^2+|E_z|^2)\ . \tag{30}$$

Here we have introduced notations

$$\frac{1}{m}=\frac{1}{m^{(2)}}-\frac{1}{m^{(1)}}\ ,\quad \frac{1}{m^{(+)}}=\frac{1}{m^{(2)}}+\frac{1}{m^{(1)}}\ ,$$

and $\hbar\omega'=\hbar\omega-\varepsilon_g, p_{\omega'}=\sqrt{2m\hbar\omega'}$. Now,

$$A_{xy}=A_{yx}=-\frac{2e^3\alpha^2 m^2 p_{\omega'}^3}{3\pi\hbar^3 m_0^2 m^{(+)}\omega^2}\times\left[\frac{\tau^{(2)}}{m^{(2)}}-\frac{\tau^{(1)}}{m^{(1)}}\right]\mathrm{Re}(E_x^*E_y)\ . \tag{31}$$

Comparing these results with the phenomenological equation (1) we get

$$\xi=\frac{2e^3\alpha^2 m p_{\omega'}^3}{3\pi\hbar^3 m_0^2\omega^2}\frac{\tau^{(2)}-\tau^{(1)}}{m^{(2)}-m^{(1)}}\ , \tag{32}$$

$$\eta=-\frac{2e^3\alpha^2 m^2 p_{\omega'}^3}{3\pi\hbar^3 m^{(+)} m_0^2\omega^2}\left[\frac{\tau^{(2)}}{m^{(2)}}-\frac{\tau^{(1)}}{m^{(1)}}\right]\ . \tag{33}$$

Here we assume an effective mass to be positive if the curvature of the band is positive, or, in other words, $\partial^2\varepsilon/\partial p_x^2>0$.

VIII. PHOTOGALVANIC ELECTRON CURRENT

To make a general analysis and order-of-magnitude estimates we are going to calculate this contribution for the simplest possible situation. We shall again assume the isotopic quadratic electron spectrum given by Eq. (25). As for the transition amplitude, we can now neglect a small item $\hbar k$ in comparison to p and assume

$$\mathbf{P}_{21}=2\alpha\mathbf{p}\ . \tag{34}$$

This contribution cannot be properly treated without taking into account the variation of the electron distribution function with the distance from the surface z. Here we need an assumption in regard to the electrons' reflection from the metal surface. Let us assume that the reflection is elastic and completely diffuse, so that

$$\Delta f(0)=0\quad \text{for } v_z>0\ . \tag{35}$$

As is well known, in general the condition for diffuse reflection should have for $v_z>0$ a more complicated form, $\Delta f(0)=\mathcal{C}$, where constant $\mathcal{C}$ depends on the electron energy. $\mathcal{C}$ should be chosen to satisfy the condition that the normal component of the electron current should vanish at the surface. However, one can easily check that to calculate the components of surface current g_x and g_y with the accuracy accepted throughout the paper it is sufficient to consider the boundary condition in the simplified form (35).

We shall analyze the Boltzmann equation (15) for the electrons; the superscript (2) at the distribution function and other quantities will be omitted. We have

$$v_x \frac{\partial \Delta f}{\partial z} + \frac{\Delta f}{\tau} = D(z) \tag{36}$$

where, within the accepted approximation, $D(z) = G(z)\delta(\varepsilon_{\mathrm{p}}^{(2)} - \hbar\omega - \varepsilon_{\mathrm{p}}^{(1)})$. Its solutions are

$$\Delta f(z) = \frac{1}{v_z}\exp\left[-\frac{z}{v_z\tau}\right]\int_0^z dz'\exp\left[\frac{z'}{v_z\tau}\right]D(z')\ ,\quad v_z > 0$$

$$\Delta f(z) = -\frac{1}{v_z}\exp\left[-\frac{z}{v_z\tau}\right]\int_z^\infty dz'\exp\left[\frac{z'}{v_z\tau}\right]D(z')\ ,\quad v_z < 0\ . \tag{37}$$

Assuming an exponential variation of D with the distance $D(z) = D_0 e^{-\kappa z}$ we get

$$\Delta f(z) = \frac{1}{v_z}\frac{e^{-\kappa z} - e^{-\beta z}}{\beta - \kappa}D_0\ ,\quad v_z > 0 \tag{38}$$

$$\Delta f(z) = -\frac{1}{v_z}\frac{e^{-\kappa z}}{\beta + \kappa}D_0\ ,\quad v_z < 0 \tag{39}$$

where we have introduced the notation $\beta = 1/|v_z|\tau$. We shall use here the same equation for G as in Sec. VII setting $\mathbf{k} = 0$. Assuming that the wave is polarized in the xz plane we can write

$$G_0 = \frac{2\pi}{\hbar}\alpha^2\left[\frac{e}{m_0\omega}\right]^2[|E_{x0}|^2 p_x^2 + |E_{z0}|^2 p_z^2 + 2\,\mathrm{Re}(E_{x0}E_{z0}^*)p_x p_z]\ . \tag{40}$$

Here E_{i0} stands for the amplitude of a field component at $z = +0$, i.e., inside the metal just near the surface. It is the last term in the square brackets that is responsible for the effect we consider in this section, so we shall retain only this term in the equation for G_0. The electron current density can be calculated by integration over d^3p of the electron distribution function, Eq. (39), times the electron velocity v_x:

$$j_x = \frac{e\alpha^2 l m p_{\omega'}^3}{\pi\hbar^4}\left[\frac{e}{m_0\omega}\right]^2 \times \int_0^1 d\zeta\,\zeta(1-\zeta^2)\left[\left[\frac{1}{1-\kappa l\zeta} - \frac{1}{1+\kappa l\zeta}\right]e^{-\kappa z} - \frac{e^{-z/l\zeta}}{1-\kappa l\zeta}\right]\mathrm{Re}(E_{x0}E_{z0}^*)\ . \tag{41}$$

Here $l = p\tau^{(2)}/m^{(2)}$ is the electron mean free path.

Integrating this equation over z we get for the density of the surface current carried by the electrons

$$g_x = \frac{e\alpha^2 l m p_{\omega'}^3}{4\pi\hbar^4}\left[\frac{e}{m_0\omega}\right]^2\mathrm{Re}(E_{x0}E_{z0}^*)\mathcal{Z}(\kappa l)\ , \tag{42}$$

where

$$\mathcal{Z} = 4\kappa l\int_0^1 \frac{d\zeta\zeta^2(1-\zeta^2)}{1+\kappa l\zeta}\ . \tag{43}$$

For $\kappa l \ll 1$, $\mathcal{Z} = 8\kappa l/15$ while for $\kappa l \gg 1$, $\mathcal{Z} = 1$.

The physics of the result is rather transparent. If the electron mean free path l is much smaller than the electromagnetic field penetration depth $\delta = 1/\kappa$, the electron gas is sensitive to the diffuse reflection from the surface only within a thin layer of the width l near the surface. Elsewhere there is a cancellation of the contributions of the electrons with $v_z > 0$ and $v_z < 0$. Thus the result is proportional to the small parameter κl.

In the opposite case all the electrons with $v_z > 0$ excited by the light carry the current, in contrast to the electrons with $v_z < 0$, which are strongly scattered at the surface. As a result, there is no balance between the two electron groups and one gets 1 instead of the factor proportional to κl.

In a more general case where the reflection of the electrons is partly specular, so that $\mathcal{P}$ is the part of specularly reflected electrons, the net photogalvanic current would be proportional to $1-\mathcal{P}$ (see Ref. 15), in strong contrast to the current due to the quasimomentum transfer which is much less sensitive to the character of the reflection (see Sec. IX).

Let us now give the result for the case where contributions of two bands are of importance, the conditions $l \gg \delta$ being valid for both bands,

$$g_x = \frac{\alpha^2 e m p_{\omega'}^4}{4\pi\kappa\hbar^4}\left[\frac{e}{m_0\omega}\right]^2\left[\frac{\tau^{(2)}}{m^{(2)}} - \frac{\tau^{(1)}}{m^{(1)}}\right]\mathrm{Re}(E_{x0}E_{z0}^*)\ . \tag{44}$$

This means that we have

$$\lambda = \frac{\alpha^2 e p_{\omega'}^4}{4\pi\kappa\hbar^4}\left[\frac{e}{m_0\omega}\right]^2\left[\frac{\tau^{(2)}}{m^{(2)}} - \frac{\tau^{(1)}}{m^{(1)}}\right] \tag{45}$$

for a parameter λ as defined by Eq. (6). It is useful to give for the analysis of the photogalvanic contribution the product $E_{x0}E_{z0}^*$ at a surface of an optically isotropic metal as a solution of a standard boundary problem

$$E_{x0}E_{z0}^* = -\frac{32\pi\sqrt{\epsilon - \sin^2\theta}\cos^2\theta\sin\theta}{|\epsilon\cos\theta + \sqrt{\epsilon - \sin^2\theta}|^2}Q^{(e)}\ . \tag{46}$$

Here θ is the angle of light incidence, ϵ is the (complex) dielectric susceptibility of the metal, and $\mathbf{Q}^{(e)}$ is the Poynting vector of the wave *falling from the vacuum* on the metal surface.

It is worthwhile to note that one could have entirely different phase relations between x and z components of the field for a circular[20] (generally, elliptical) polarization of the wave. It means, in particular, that by variation of the ratio of the polarization ellipse axes and of their orientation one can vary the photogalvanic contribution to the current. In such a case one can also expect a nonvanishing value of the y component of the surface current g_y. It is determined by the same equation as g_x with the replacement of $\mathrm{Re}(E_{x0}E_{z0}^*)$ by $\mathrm{Re}(E_{y0}E_{z0}^*)$.

IX. SURFACE CURRENT DUE TO QUASIMOMENTUM TRANSFER: LARGE ELECTRON MEAN FREE PATHS

We start with a general equation for the electron current density that after the appropriate coordinate transformation in the integrand takes the form

$$j_x = 2e\int \frac{d^3p}{(2\pi\hbar)^3}[v_x^{(2)}(\mathbf{p})\Delta f_{\mathbf{p}}^{(2)} - v_x^{(1)}(\mathbf{p})\Delta f_{\mathbf{p}}^{(1)}] \ . \quad (47)$$

In principle, we are interested in the case of any relation between the penetration depth δ and the electron mean free paths. Again we shall calculate the contribution of electrons in band 2; the contribution of band 1 we shall write by analogy.

To calculate the surface current density we actually need not know the distribution function but rather its integral over z,

$$F_{\mathbf{p}}(z) = \int_0^\infty \Delta f_{\mathbf{p}}(z)dz \ . \quad (48)$$

Here, to calculate the surface current with regard to the boundary conditions (35) we shall use the following equations for the integrals of the type (48). By the same method as in Sec. VIII we get

$$F_{\mathbf{p}} = \frac{\tau}{\kappa}D_0 \ , \quad v_z > 0 \ , \quad F_{\mathbf{p}} = \frac{\tau}{\kappa[1+\kappa\tau|v_z(\mathbf{p})|]}D_0 \ , \quad v_z < 0 \quad (49)$$

where

$$D(\mathbf{p},\mathbf{p}-\hbar\mathbf{k}) = D_0 e^{-\kappa z} = G(\mathbf{p},\mathbf{p}-\hbar\mathbf{k})\delta(\varepsilon_{\mathbf{p}}^{(2)} - \hbar\omega - \varepsilon_{\mathbf{p}-\hbar k}^{(1)}) \ . \quad (50)$$

Again we assume that the relaxation times are independent of $\mathbf{p}$. We will solve the Boltzmann equation for $\mathcal{P}=0$. We shall consider here a limiting case of a comparatively large electron mean free path $l \gg \delta$ and shall retain the lowest approximation in δ/l. In this approximation we should consider as nonvanishing only the solution of Eq. (49) for $v_z > 0$. It gives

$$g_x(z) = \frac{2e\tau}{(2\pi\hbar)^3\kappa}\int d^2p \int_0^\infty dp_z v_x D_0 \ . \quad (51)$$

Here we assume that the metal surface coincides with its plane of symmetry, so that $v_z > 0$ corresponds to $p_z > 0$. Further on in this section, for simplicity, we shall again use Eq. (26) for $\mathbf{P}_{21}$ and assume, for the same reason, that the times of relaxation are independent of $\mathbf{p}$.

Calculating the surface current we shall expand it in powers of k_x retaining the terms of the zeroth and the first order. The zeroth-order term is the usual photogalvanic current given by Eq. (44) while for the first-order term we get

$$g_x = -\frac{ek_x\alpha^2 m p_{\omega'}^3}{3\pi\kappa\hbar^3}\left[\frac{e}{m_0\omega}\right]^2 \times \left[\frac{m}{m^{(+)}}\left[\frac{\tau^{(2)}}{m^{(2)}} - \frac{\tau^{(1)}}{m^{(1)}}\right]|E_{x0}|^2 - \frac{\tau^{(2)}-\tau^{(1)}}{m^{(2)}-m^{(1)}}|\mathbf{E}_0|^2\right] \ . \quad (52)$$

The physics reflected by this equation can be visualized as follows. All the electrons excited by the light can be subdivided into two groups, i.e., those moving towards the surface and those moving from the surface. Because of the condition $l \gg \delta$ the electrons of the first group experience a strong scattering from the surface. As the scattering is assumed to be completely diffuse, these electrons give a negligible contribution to the current. To the contrary, the contribution of the electrons of the second group is conditioned by the scattering in the bulk of the metal, i.e., by the same mechanism as for $l \ll \delta$. Therefore the general expression for the integral over z of their distribution function remains the same, however, with an extra factor $\frac{1}{2}$.

X. TWO CONTRIBUTIONS TO THE SURFACE CURRENT AND THEIR COMPARISON

For the electric field polarized in the xz plane one may give the following order-of-magnitude estimate of the two contributions. Let us denote by $g^{(q)}$ the surface current proportional to wave vector k and by $g^{(p)}$ the photogalvanic current. Then we have

$$|g^{(p)}/g^{(q)}| \approx \gamma(1-\mathcal{P})\frac{p_{\omega'}}{\hbar k}\frac{|\mathrm{Re}(E_{z0}^* E_{x0})|}{|E_{x0}|^2} \ . \quad (53)$$

The ratio of the two current densities contains a numerical factor γ, a factor $(1-\mathcal{P})$ depending on the reflection of the electrons from the metal surface, a ratio of two characteristic wave vectors $p_{\omega'}/\hbar k$, and a field factor that is determined by the ratio of certain amplitudes of the electric fields within the metal, with regard to the phase relations between the components.

A theory based on oversimplified Eq. (26) cannot give the exact value of the factor γ for it is rather sensitive to the actual form of the dispersion laws and, especially, transition probabilities. It would be most interesting to investigate in the future the relative role of both contributions for more realistic models. (For a particular case of semiconductors where light induces intraband transitions a comparison of the two contributions to the surface current has been done in Ref. 13.)

Now, the factor $1-\mathcal{P}$ usually can be replaced by 1 unless the surface current is determined mainly by the skipping trajectories for which in the case we consider here we see no special reasons. The ratio $p_{\omega'}/\hbar k$ is of the order of 10^3 for the visible light.

As for the field factor, it should be discussed more carefully. It is determined by dielectric susceptibility of the metal which, for a visible light, is usually determined by free electrons. We have

$$\epsilon=\epsilon_0-\omega_p^2/\omega^2 \tag{54}$$

where ϵ_0 is a contribution due to atomic core polarization (usually comparatively small within the frequency range we are interested in) whereas the plasma frequency ω_p is given by $\omega_p^2=4\pi ne^2/m$ where n is the free-electron concentration. For n of the order of 10^{23} cm^{-3} and m of the order of free-electron mass $\omega_p^2\approx 3\times 10^{32}$ s^{-2}, which means that for a visible light the absolute value of the second term in Eq. (54) is at least larger than 10.

Now, the ratio

$$\frac{|\mathrm{Re}(E_{x0}E_{z0}^*)|}{|E_{x0}|^2}=\frac{\sin\theta|\mathrm{Re}\sqrt{\epsilon-\sin^2\theta}|}{|\epsilon-\sin^2\theta|} \tag{55}$$

is small for two reasons. First, $|\epsilon|$ is large. Second, for a real negative dielectric susceptibility [Eq. (54)] we have $\mathrm{Re}(E_{x0}E_{z0}^*)=0$. Therefore to obtain a finite result one should take into account an imaginary part (usually small) that is due to the light absorption, i.e., proportional to the transition probabilities. The best way to make estimates of these is to use the experimental data (for instance, on light absorption). All this means that it is difficult to state in advance which contribution would prevail under given circumstances and their relative values should be investigated carefully in every particular case.

XI. CONCLUSION

One can see on the basis of the calculation presented that there are two contributions to the surface current. One of them is initiated by the quasimomentum transfer. Another one (the photogalvanic current) is due to the asymmetry in the electron distribution brought about by a surface scattering of the conduction electrons.

Both contributions exist for interband as well as for intraband electron transitions induced by light. We are treating here the case of relatively high energies of light quanta. Therefore we consider interband transitions.

As we have seen in Sec. X, depending on the particular experimental situation, the photogalvanic contribution to the surface current can be larger than the part determined by the quasimomentum transfer, or of the same order, or can even vanish. Indeed, this contribution is sensitive to the way the electrons are reflected at the metal surface; it vanishes for the specular reflection. It is more likely, however, to expect diffuse reflection, except for the electrons whose velocities are at very small angles to the surface. The polarization of the light may be of major importance for this part of the current. For instance, there would be no contribution for a monocrystal provided the plane of light incidence coincides with a plane of high symmetry of the crystal and the electric field vector of light is perpendicular to the plane.

If the light is polarized in the xz plane, the theory predicts an effect. The effect is proportional to $\mathrm{Re}(E_{x0}E_{z0}^*)$, i.e., it should be sensitive (as has already been mentioned) to the phase relations between the components of the electric field. According to Eq. (46) the product is proportional to $\sqrt{\epsilon-\sin^2\theta}$. If the frequency of light ω is well below the plasma edge (which is likely to be the case for the experiments in Ref. 1) ϵ is in the first approximation real and negative, and probably much larger than unity by its absolute value. In the next approximation there is an imaginary part which determines the light absorption. It may be considered as small if on the experiment the absorbed intensity is small in comparison to the reflected one.

An experiment of the polarization dependence of the photomagnetism has been done by the authors of Ref. 21. A distinct polarization dependence of the effect has been observed. However, these results should be considered as preliminary, as they were obtained on polycrystalline samples. The experiments on monocrystals are now in progress.

Investigation of the photomagnetism 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 high 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 in the future may provide much useful information.

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 light absorption.

Third, this could provide a way to study various aspects of the 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.

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