Photomagnetism of metals. First observation of dependence on polarization of light

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We report first observation of the polarization dependence of the d.c. photocurrent induced by illumination in Cu. The dependence of d.c. photocurrent on the direction of the plane of light polarization is measured. In agreement with the theoretical considerations, the current parallel to the plane of light incidence is a symmetric function of the angle between this plane and the plane of light polarization. The angular-dependent part of the current perpendicular to the plane of light incidence is an antisymmetric function of the angle.

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1. Introduction

The purpose of the present paper is to investigate the dependence of the photoinduced magnetic flux in normal metals on the polarization of light. For the first time the photoinduced magnetic flux in a metal has been observed by Lashkul and the present authors in [1]. Further experimental investigations of the effect have been made in [2]. The foundations of the theory of the surface photocurrent in metals are developed in [1,3]. It is shown that a circular photocurrent excited in a sample of special geometry builds up a magnetic flux. The effect appears to be rather big and can be easily measured by a SQUID magnetometer [1]. Like the second harmonic generation, a well-known phenomenon investigated in detail in a great number of papers, this is a nonlinear effect proportional to the electromagnetic amplitude squared, i.e. to the intensity of light Q. Its observation and particularly investigation of its dependence on the polarization of light can give much valuable information about the properties of metal including unique data concerning the behavior and properties of electrons excited by light well above the Fermi surface.

2. Physical considerations

There are two contributions to the d.c. surface current responsible for the photomegnetism. They may be named the photogalvanic current and the quasimomentum transfer current (see [3]). Both contributions exist for any electron spectrum and interaction of conduction electrons with light. In our theoretical analysis we will consider the simplest form of the spectrum and interaction and give only brief comments concerning the general case. The first contribution is due to the anisotropy of electron transition probability in regard to the direction of light polarization, in combination with the diffuse reflection of electrons at the metal's surface. The second one is due to the fact that the light reflected from a metal surface transfers to the conduction electrons not only some of its energy but also some of its quasimomentum and thus creates a d.c. photocurrent.

As we have mentioned, the photogalvanic contribution is due to a diffuse scattering of the electrons from the surface (cf. [4,5] where such effects have been considered for semiconductors). In general, the transition probability is an anisotropic function of the electron quasimomentum. The anisotropy exists even in the case of isotropic electron spectrum due to the directional asymmetry brought about by the electric a.c. field vector, $\mathbf{E}$. Let us assume that $\mathbf{E}$ lies in the plane xz where z is perpendicular and x is parallel to the metal's surface (Fig. 1). In the simplest (isotropic) case, the transition probability can have an item proportional to $(\mathbf{Ep})^2$, $\mathbf{p}$ being the electron quasimomentum. As a result of the invariance of the transition probability to the change of the

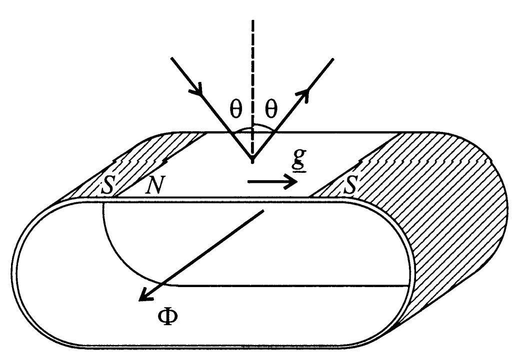

Figure 1. Arrangement for detection of a current $\mathbf{g}$ excited by light. The light falls at an angle θ to the perpendicular of a normal metal surface N and is partly refelected. The current is short-circuited with a superconductor S, so that the current encircles the orifice. The magnetic flux Φ created by the current within the loop is detected with a SQUID magnetometer.

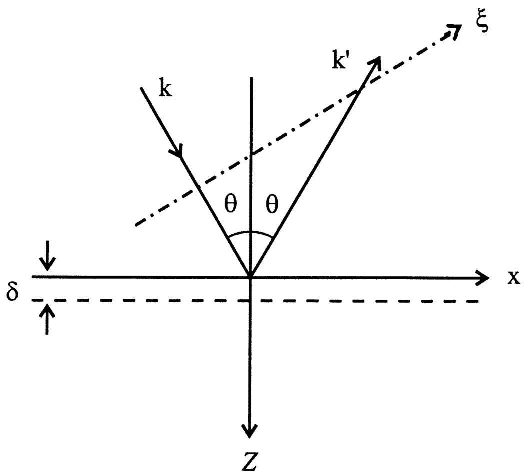

Figure 2. The plane of light incidence and relfection. The metal's surface coincides with the x-axis; the z-axis is perpendicular to the surface. $\mathbf{k}$ is the wave vector of the ingoing light while $\mathbf{k}'$ is the wave vector of the refelected light, ξ-axis is perpendicular to the direction of ingoing light propagation. y and η-axes coincide and are perpendicular to the plane of the figure. δ is the depth of light penetration.

sign of quasimomentum, the number of electrons generated by light and moving, say, along the positive direction of x axis and, at the same time, towards the surface would be equal to the number of electrons moving along $-x$ and away from the surface. The electrons of the first group would be scattered from the surface and if the scattering is 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 [5]. It should not exist for polarization of a. c. electric field, $\mathbf{E}$, in y direction, i. e. perpendicular to the plane of light propagation. The last statement is valid for an isotropic case, which can be seen directly from the form of angular dependence of the surface current (see Section III). It should also be true if, for instance, the plane of light propagation coincides with a plane of symmetry of the crystal or the direction of surface current coincides with an axis of symmetry of the crystal (provided it has a center of symmetry as is usually the case with most metals).

To understand the second contribution consider a plane surface of uniformly illuminated metal. The light falls at angle θ to the perpendicular (Fig. 2). It penetrates into a thin layer at the metal surface. Due to interaction with the conduction electrons within this layer, a part of the light energy is absorbed by the electrons (usually in the course of their interband transitions as we assume in the present paper) so that one can write for the Poynting vectors $\mathbf{Q}$ and $\mathbf{Q}'$ of the wave falling from vacuum onto the metal surface and the wave reflected from the surface

$$Q'_z = -(1-r)Q_z.$$

Here $r > 0$,

$$\mathbf{Q} = (c/4\pi)\left[\overline{\mathbf{E}, \mathbf{H}}\right],$$

where the bar means a time average. Along with energy, light carries a momentum. The average flux of the x-component of momentum of light through the metal surface can excite a d. c. surface electron current.

This reasoning should be modified in one point. As the electrons move in a spatially periodic field, their momentum is not conserved. It means that one should discuss the phenomenon in terms of quasimomentum (rather than momentum) both for electrons and for light propagating in optically homogeneous media and in vaccum (where the quasimomentum equals the ordinary momentum). This is why we call this contribution the quasimomentum transfer (QT) contribution.

The physical picture can be described as follows, There is a perpetual influx of the quasimomentum into the system of conduction electrons. As in vacuum the quasimomentum turns into the ordinary momentum, the quasimomentum flux for this case is just the momentum flux. Its average zx-component is given by $(Q/c)\sin\theta\cos\theta$. If the illuminated part of the sample is bigger than the electrons' mean free path, the balance is established due to the scattering of electrons (for example, by the impurities). Therefore the effect is proportional to the electron mean free time [1,3]. (It should be very interesting to investigate this effect in metallic nanostructures where there is no scattering of electrons and the balance is established due to the fact that the path of an electron within the illuminated part of the nanostructure is finite — cf [6].)

The photoinduced d. c. current depends on the polarization of light. For an appropriate polarization there is a component of the current not only in the x- but also in the y-direction. One can visualize one of the sources of the polarization dependence as follows. As the effect exists already for an isotropic case, let us discuss it for this simplest situaiton. As is already indicated, the interband transition probability depends on the direction of electron quasimomentum. It has a maximum if the quasimomentum is oriented along the oscillating (a. c.) field and a minimum if it is perpendicular to the field. It means that the transition probability has the term $\mathrm{Re}(E_x^* E_y p_x p_y)$ [3]. Assume that the electric field lies in the xy-plane. Let it be, because of the quasimomentum transfer from the reflected electromagnetic wave to the electrons a surplus of the electrons with $p_x > 0$. Then, because of the aforementioned term, there should be also a surplus of the electrons with $p_y > 0$ which would create a net d. c. current in the y-direction. These considerations show, in particular, that g_y should vanish both for the cases where the a. c. electric field is polarized either in the plane xz (where z is perpendicular to the metal's surface) or along y-axis. The microscopic calculation (see Section IV) shows that g_y is indeed proportional to

$\sin 2\phi$ whereas g_x is proportional to $\cos 2\phi$. Here ϕ is the angle between the electric field vector and the line of intersection of two planes, i. e. the plane $\xi\eta$ perpendicular to the wave vector of the falling light and the plane of light incidence. This line we will choose as the ξ-axis.

3. Photogalvanic contribution

We start with the phenomenological equation (see [3]) describing one of the two contributions to the surface current density **g**

$$\mathbf{g} = \frac{\lambda}{2}\Big\{\big[\mathbf{E}_0 - \mathbf{n}(\mathbf{n}\cdot\mathbf{E}_0)\big]\big(\mathbf{n}\cdot\mathbf{E}_0^*\big) + \text{c.c.}\Big\}, \tag{1}$$

where $\mathbf{E}_0$ is the electric field amplitude, $\mathbf{n}$ is the normal to the metal surface while λ is a factor depending on the electron spectrum (see below) and the frequency of light.

Let the reference frame x, y, z be directed so that the z-axis is parallel to the inner normal to the metal surface while the x-axis is parallel to the plane of light propagation. We will also need another reference frame ξ, η, ζ which will be chosen in such a way that the ζ-axis will be parallel to the direction of light incidence while the η-axis will coincide with y-axis. One can write for the light polarized in the plane making angle ϕ with the ξ-axis

$$E_{0\xi} = \mathcal{E}_0 \cos\phi,$$
$$E_{0\eta} = \mathcal{E}_0 \sin\phi, \tag{2}$$

where $\mathcal{E}_0$ is the electric field amplitude in the light falling on the metal surface.

Now

$$E_{0x} = \mathcal{E}_0 \cos\theta\cos\phi,$$
$$E_{0y} = \mathcal{E}_0 \sin\phi,$$
$$E_{0z}^{(\text{out})} = \mathcal{E}_0 \sin\theta\cos\phi, \tag{3}$$

where θ is the angle between the wave vector of light and the z-axis. Here $E_z^{(\text{out})}$ is the z-component of the electric field just outside the metal surface. The z-component of the electric field just inside the metal near the surface is given by

$$E_{0z} = (1/\varepsilon)\mathcal{E}_0 \sin\theta\cos\phi, \tag{4}$$

where ε is the (complex) dielectric susceptibility of the metal.

Finally, one gets

$$g_x = \frac{\lambda}{2}|\mathcal{E}_0|^2\,\mathrm{Re}(\varepsilon^{-1})\sin 2\theta(1 + \cos 2\phi) \tag{5}$$

and

$$g_y = \frac{\lambda}{2}|\mathcal{E}_0|^2\,\mathrm{Re}(\varepsilon^{-1})\sin\theta\sin 2\phi. \tag{6}$$

We will give order-of-magnitude estimate of λ. For the electron spectrum in the lower band 1 and in the upper band 2 we assume

$$\varepsilon^{(1)}(p) = p^2/2m^{(1)}; \qquad \varepsilon^{(2)}(p) = \varepsilon_g + p^2/2m^{(2)} \tag{7}$$

and we use the notation

$$\frac{1}{m} = \frac{1}{m^{(2)}} - \frac{1}{m^{(1)}};$$

and

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

In the present paper we assume for simplicity that the electrons of the upper band 2 give the principal contribution to the current as $m^{(2)} \ll m^{(1)}$, so that with the accepted accuracy $m_2 \approx m$. According to [3] we have

$$\lambda \approx \frac{e\alpha^2\tau^{(2)}p_{\omega'}^4\delta}{4\pi m\hbar^4}\left(\frac{e}{m_0\omega}\right)^2. \tag{8}$$

Here m_0 is the free electron mass, ω is the frequency of light, δ is the penetration depth of light into the metal, we assume that it is much smaller than the electron mean free path $\tau^{(2)}p_{\omega'}/m$, α is a constant (it is a measure of interaction of electrons with light). By order-of-magnitude we have

$$\alpha \approx P_{21}(\mathbf{p}', \mathbf{p})/(p + p'),$$

where P_{21} is the matrix element of interband itransition induced by light, p and p' are the initial and final values of the electron quasimomentum, respectively.

The principal result of this consideration can be formulated as follows. The x-component of the surface current is proportional to $\cos 2\phi$ while the y-component is proportional to $\sin 2\phi$.

4. Quasimomentum transfeer current

In the present section we are interested in the QT surface current **g** where for $l \gg \delta$ [3]

$$g_\mu = g_\mu^{(1)} + g_\mu^{(2)}. \tag{9}$$

Here [7]

$$g_\mu^{(1)} = \frac{4\pi\zeta_1\omega}{c^2}\,Q_\mu, \tag{10}$$

$$g_\mu^{(2)} = \frac{i\zeta_2}{2}\left(E_\nu\frac{\partial E_\mu^*}{\partial x_\nu} - E_\nu^*\frac{\partial E_\mu}{\partial x_\nu}\right), \tag{11}$$

where

$$\mathbf{Q} = \frac{c}{4\pi}\,\mathrm{Re}\big[\overline{\mathbf{E}, \mathbf{H}^*}\big] \tag{12}$$

is the time-averaged Pointing vector inside the metal. A summation over the repeated indices is implied in (11), μ, ν run through values x, y; E_μ are the components of the electric field at the surface.

For the simple model of electron spectrum adopted in [3] we have

$$\mathbf{g}^{(1)} = \frac{e^3\alpha^2p_{\omega'}^3\tau^{(2)}\delta}{3\pi\hbar^3m_0^2\omega^2}\,|\mathbf{E}|^2\mathbf{k}, \tag{13}$$

and

$$g_i^{(2)} = \sum_l A_{il}k_l, \tag{14}$$

$$A_{xx} = -\frac{e^3\alpha^2 p_{\omega'}^3 \tau^{(2)}\delta}{3\pi\hbar^3 m_0^2\omega^2}\left(3|E_x|^2 + |E_y|^2 + |E_z|^2\right), \tag{15}$$

$$A_{xy} = A_{yx} = -\frac{2e^3\alpha^2 p_{\omega'}^3 \tau^{(2)}\delta}{3\pi\hbar^3 m_0^2\omega^2}\,\mathrm{Re}(E_x^* E_y). \tag{16}$$

More detailed equations valid for a two-band model one can find in [3].

One can easily analyze the polarization dependence of the QT current using (3), (4), (14), (15) and (16). One can also take into consideration that as usually in metals

$$|\varepsilon| \gg 1, \tag{17}$$

one can neglect the contribution due to the terms proportional to E_{0z} as compared to other contributions. One gets almost the same ϕ-dependence of the surface currents as in the previous section, i. e.

$$g_x \propto (\mathrm{const} + \cos 2\phi),$$

$$g_y \propto \sin 2\phi, \tag{18}$$

where the constant depends on the spectrum of electrons and on their interaction with the light. Of course, the two contributions essentially differ by their θ-dependence. However, most important feature is vanishing of g_y for the light polarized in the plane xz resulting from the symmetry of the metal's surface.

One can add the following consideration. In general η_1 and η_2 are tensors. Their symmetry for monocrystals depends on the symmetry of the crystal surface under consideration. In particular, η_2 is a tensor of the 4th rank. If its symmetry is low enough, one can have a non-vanishing g_y-component together with g_x-component for the light polarized in the plane xz.

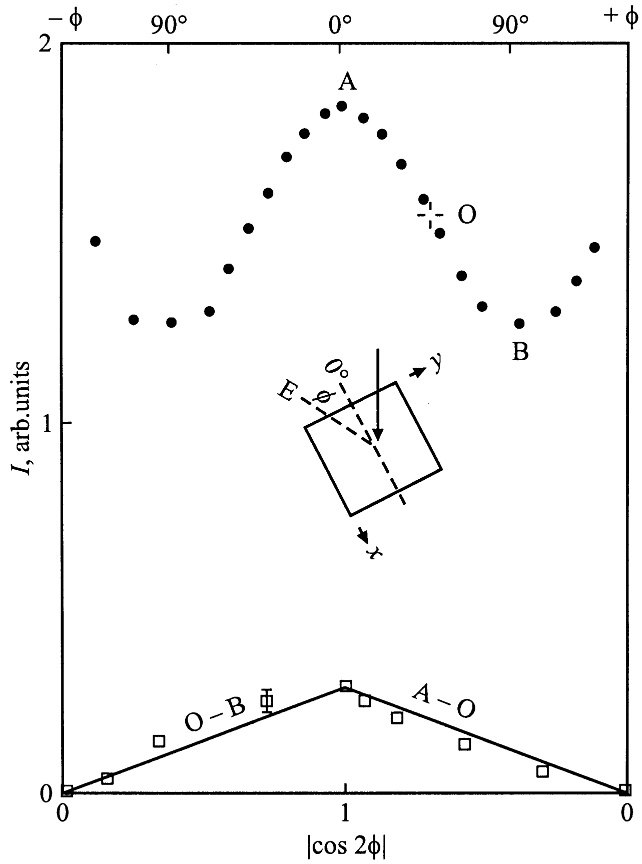

Figure 3. Polarization dependence of the x-component of the surface current g_x. In the upper part of the figure is shown the variation of the total current ($I \propto$ SQUID output signal) when the angle ϕ between the direction of the ξ-axis and the electric vector of the incoming light **E** is changed (dots). In the lower part are shown the amplitudes of the ϕ-dependent parts of I, O–A and O–B, plotted as a function on $\cos 2\phi$. The solid lines are for guiding the eye.

5. Experimental

A square-shaped sample of the size $5 \times 5 \times 1\,\mathrm{mm}^3$ was cut from a 99.999% pure polycrystalline Cu boule. One of the broad faces of the sample plate was polished to optical grade. The average flux of quasimomentum of light through the metal surface can excite a d. c. surface electron current. When short-circuited within a loop (Fig. 1) the current creates a magnetic flux. For observation of this flux two opposite ends of the Cu plate were connected with a 0.2 mm thick Pb band of semicircular shape to form what we call the sample coil. At the temperature of measurements. $\approx 4.2\,\mathrm{K}$, the Pb band is in the superconducting state. This assembly was enclosed in a chamber containing elements for control of the sample temperature and provided with a window for the laser light. The window of the chamber was brought to a close contact with the bottom window of a quartz tube (ID = 10 mm, length = 80 cm) fixed from the upper end to the top flange of a He bath cryostate. Outside the sample chamber, was fixed a pair of signal coils (8 turns of $5\,\mu$m thick NbTi wire, the diameter of the coil was 10 mm), forming part of the flux transformer of a quantum interference device (rf SQUID). The planes of the sample coil and of the signal coils were parallel. The system allowed us to investigate the photoinduced current on the Cu plate by measuring the related magnetic flux within the sample coil. For adjusting the direction of the light polarization plane on the Cu plate, a precisely oriented polarization rotator was fixed to the top flange of the cryostate.

The whole upper surface of the Cu plate was illuminated with linearly polarized light from an Ar ion laser ($\lambda = 514.5\,\mathrm{nm}$). The light falls on the Cu plate at the angle of 30° to the perpendicular of its surface. To minimize heating of the sample by light, the laser beam was chopped at the frequency of 30 Hz and the applied power density was limited to $\approx 100\,\mathrm{mW/cm^2}$. Before the measurement it was confirmed that within this power range the SQUID output was directly proportional to the intensity of the laser beam. Chopping of the light also allowed recording of the SQUID output signal by using the lock-in technique.

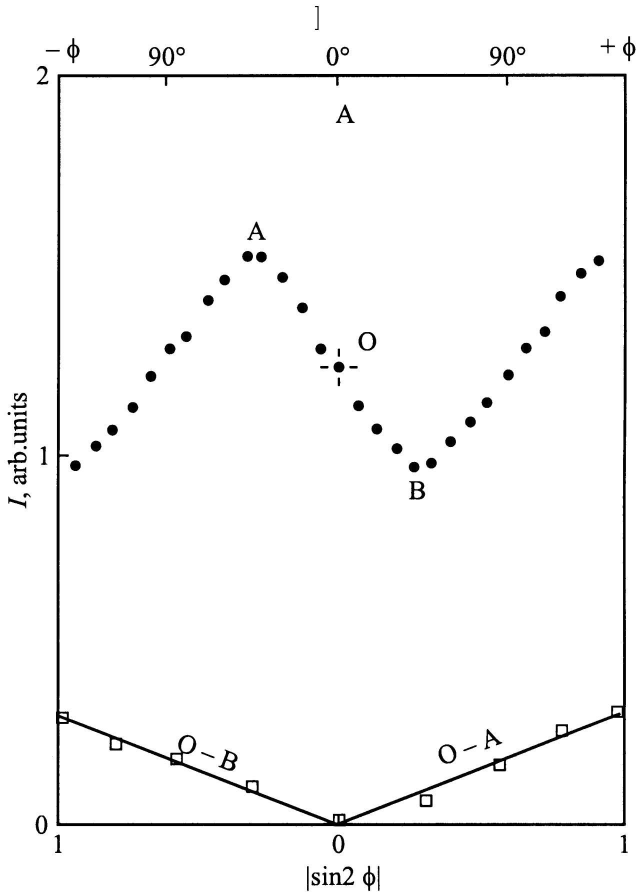

Figure 4. Polarization dependence of the x-component of the surface current g_y. In the upper part of the figure is shown the variation of the ϕ-dependent portion (dots) of the current I. In the lower part this portion is plotted as a function of $\sin 2\phi$ for the sections O–A and O–B. The solid lines are for guiding the eye.

In Fig. 3 are shown the data of the x-component of the surface current I obtained from the output signal of the SQUID electronics as the function of the light polarization direction. Here ϕ is the angle between the electric field **E** and the ξ-axis. As shown by the dots, in the upper part of the figure the polarization-dependent surface current oscillates between the values defined by the points A and B when ϕ is changed from 0° to 90°. The midpoint O between A and B, shown by the cross, is observed for $\phi = 45°$. In the lower part of the figure are depicted the sections O–A and O–B, showing within the accuracy of the measurement a linear dependence of the x-component of the surface current on the value of $|\cos 2\phi|$. The oscillating part of I between points A and B, that is $I_A - I_B$, amounts to 45% of the I_B. We will call I_B the background current as it shows no dependence on the light polarization. Both the angular variation of I and the existence of the polarization-independent component are in agreement with the prediction $g_x \propto (\mathrm{const} + \cos 2\phi)$ in (18).

In Fig. 4 is depicted the polarization dependence for g_y observed under the same temperature (measured with a thermometer fixed inside a copper pillar holding the specimen) and the same laser light power density as for g_x. Also for g_y a strong modulation of I is observed, but now the points A and B related to the extremal values of the current are shifted to ±45°. The data between the sections O–A and O–B can be well fitted with a linear relationship with $\sin 2\phi$. This agrees with the prediction for g_y in (18) excluding the fact that no polarization-independent current should exist in this case. The value of $I_A - I_B$ in Fig. 4 corresponds to 60% of I_B.

At least a part of the background current can be attributed to changes in the polarization state of the light induced by the strain in the windows of the sample chamber and on the bottom of the quartz tube after cooling them from the room temperature to 4.2 K. Under uneven distribution of the strain this effect may lead to locally varying ellipticity of the light on the surface of the sample and to a nonvanishing background current I_B in the case of g_y (see below).

6. Discussion

We have observed the polarization dependence of the surface photocurrent in the polycrystalline samples of copper. This should be considered as the first effort to observe the effect which proved to be successful. We believe that this is related to the high symmetry of copper due to which the faces [100] and [111] may form an essential part of the surface.

The x-component of the surface current has a part proportional to $\cos 2\phi$ superimposed on a ϕ-independent background, which is in accordance with the theory (18). The plot of the y-component has a part proportional to $\sin 2\phi$ which is also superimposed on a ϕ-independent background. To explain this behavior we can offer the following considerations.

As an example let us consider the photogalvanic contribution (Section III). According to (5) and (6) g_x and g_y are proportional to $\mathrm{Re}(E_{0x}E^*_{0z})$ and $\mathrm{Re}(E_{0y}E^*_{0z})$, respectively. This means, in particular, that a lot depends on the phase relations between E_{0x} and E_{0y} on the one hand and E_{0x} and E_{0z} of the other hand. As we have seen, for a linearly polarized light there should be no contribution to g_y for both $\phi = 0$ and $\pi/2$. However, as indicated in [3], for a circular (generally, elliptical) polarization of the light there can be such contribution. The observed background for g_y, as well as a part of the background for g_x may be due to ellipticity of the light polarization; this possibility is indicated in Section V.

Another contribution to the background may be associated with the following phenomenon. Under illumination, the electrons make transitions from the lower band 1 to the upper band 2 within a thin layer of the width δ near the surface. Under a stationary illumination, the influx of the electrons into band 2 should be counterbalanced by their transitions back to band 1. Estimating the rate of transitions, one should take into consideration that in our experiment the energy of the light quanta $\hbar\omega = 2.41$ eV is so large that the electrons in band 2 are well above the Fermi level.

The rate of the $2 \to 1$ transitions may be comparatively low as they mostly take place via phonon emission. The electron energy loss after such a transition cannot exceed the maximal phonon energy $\hbar\omega_D$. (The electron-electron collisions can also play a role in this process.) As a result, the electrons, while still in the band 2, can diffuse (together with the holes of band 1 — to maintain neutrality) out of the layer of the width δ into the bulk of the copper sample. Because of nonhomogeneity of the sample their spatial distribution should also be nonhomogeneous. This means effectively thata temperature distribution over the sample can bring about a thermoelectric current which is short-circuited by the superconductor. This can also result in the background that is superimposed on the $\sin 2\phi$-dependence of g_y.

7. Conclusion

Actually λ, the coefficient in (1), is a tensor of the 2nd rank. Our considerations are valid for the simplest case where the tensor is equivalent to a scalar. This is true, for instance, for the surfaces [100] or [111] of a cubic crystal. One can use the phenomenological considerations to analyze more complicated geometries, i.e. crystalline surfaces of lower symmetry.

The QT current has in general a more complicated structure as ζ_2 is, as we have already indicated, a tensor of the 4th rank. This means that a careful investigation of the polarization dependence of the photocurrent may permit to separate the photogalvanic and QT contributions.

Extremely interesting phenomena, as we have already seen, can be predicted and observed for a circularly polarized light. These we hope to discuss in detail in a separate paper.

In general, the observed effect is a powerful way to study the interaction of the electrons with the light in metals. It can also provide a way to study various aspects of interaction of conduction electrons with the surface of a metal.

In summary, we have measured the polarization dependence of the surface photocurrent in polycrystalline high purity samples of copper. In accordance with the results of the theory, the current parallel to the plane of light incidence is a symmetric function of the angle between this plane and the plane of light polarization. The ϕ-dependent part of the current perpendicular to the plane of light incidence (apart from the ϕ-independent background which most probably originates in a secondary effect) is an antisymmetric function of the angle.

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[7] There are three terms in Eq. (5) of Ref. [3] for the surface QT current. Actually the two last terms can be merged into one and therefore there are only two terms in Eq. (9).