Observation of photoinduced bulk current in metals

R. Laiho
Wihuri Physical Laboratory, University of Turku, 20500 Turku, Finland
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Observation of photoinduced bulk current is reported in Cu and Zn illuminated with visible light at normal incidence. The current is detected by measuring the magnetic moment created in a superconducting loop connecting the surface exposed to the light and the rear surface of the specimen. A correlation of the photoinduced signal and the absorption coefficient of the metal is observed. In Cu the current is as large as 23 μA over a distance of 1 mm when the optical power density is 1 W/cm^2 at λ=514.5 nm and the temperature is 4.2 K.

In a recent paper[1] an observation of photoinduced surface current in copper and aluminium specimens was reported. In that experiment the light falled obliquely on the surface of a cylindrical sample so that a circular current could exist. The effect has been attributed to transfer some of the quasimomentum of light to conduction electrons when the light is reflected from a metal surface.[1,2] This creates a surface current which, for an appropriate geometry brings about a magnetic moment. There is another contribution to this current due to anisotropy of the electron transition probabilities induced by light, in combination with diffuse reflection of the electrons at the surface.[2]

In Ref. 3 was predicted an appearance of bulk current in a metal exposed to the light propagating in the direction perpendicular to the surface. It was argued[3] that even a larger effect can be achieved for normal incidence of light than for illumination falling obliquely on the metal surface. In the present paper we report the observation of the photoinduced bulk current in a metal for near normal incidence of light.

The simplest experimental arrangement to observe the predicted effect[3] is shown in Fig. 1. The L-shaped specimen prepared from a 99.999% pure Cu single crystal is fixed to an end of an optical fiber which has the core diameter of 1 mm. Then they are assembled aside a rf superconducting quantum interference device (SQUID) detector located at the lower end of a stainless steel coaxial cable. The surrounding of the SQUID is carefully shielded against stray electromagnetic and magnetic fields and is immersed in a conventional helium transport dewar.

Details of the specimen are shown in Fig. 1. For detection of the photoinduced bulk current the front surface (excluding the area to be illuminated) and the rear surface of the sample are connected with a 0.2 mm thick superconducting Pb stripe. The length of the Cu layer through which the current should pass is 1 mm. The area of the loop formed in this way is $\simeq$0.4 cm^2. The sample is placed immediately on the signal coil of the SQUID (eight turns of 0.05 mm thick NbTi wire, the diameter of the coil is 8 mm). In addition to the L-shaped specimen a U-shaped sample of polycrystalline oxygen-free copper ($\rho<1\times10^{-10}$ Ω cm at 4 K) containing a similar Pb short circuit (see Fig. 2) and another U-shaped sample of 99.999% pure polycrystalline zinc were constructed. The U-shaped specimens were used to investigate the symmetry of the currents flowing in the Pb loop when they were illuminated through one of the two optical fibers fixed on their top (see Fig. 2).

As light sources were used an Ar, Kr, and Ar-pumped dye laser, covering the range of 1.8–2.6 eV. To avoid overheating of the sample by light it was in a direct contact with liquid helium and the light was chopped at frequency of 14 Hz. For observation of any slow drift of the signal detected by the SQUID the light was periodically interrupted for a time of 30 s.

At the output of the optical fiber the light is spread into a cone with opening $\simeq$35°, resulting in a reduction of the Poynting vector in the direction perpendicular to the surface.

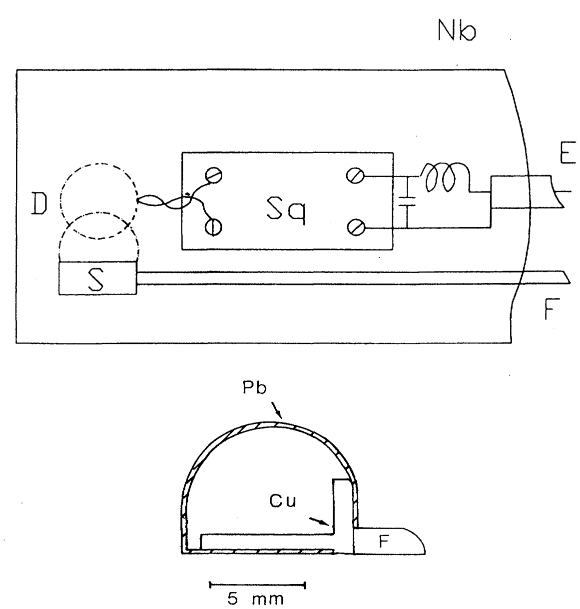

FIG. 1. Construction of the probe used for detection of the photoinduced bulk current. In the upper part of the figure Sq denotes a rf-SQUID, S is the sample, D the signal detection coil, F an optical fiber, E the input cable of the SQUID and Nb a niobium shield. In the lower part are shown details of the L-shaped Cu sample and the lead stripe (Pb) connecting its front face and the region situated 1 mm from the illuminated area.

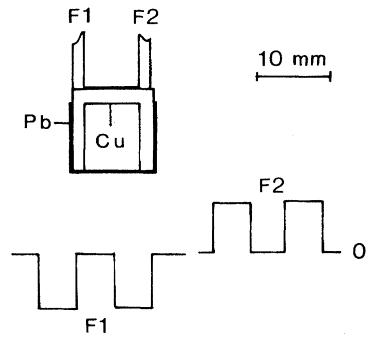

FIG. 2. U-shaped Cu specimen on top of which two identical optical fibers, F1 and F2, are glued. The thick lines show the superconducting coatings (Pb) forming the low resistance current loop together with the 1 mm thick unplated Cu sections immediately below the ends of F1 and F2. In the lower part of the figure are shown the SQUID output signals when the specimen is illuminated either through F1 or through F2.

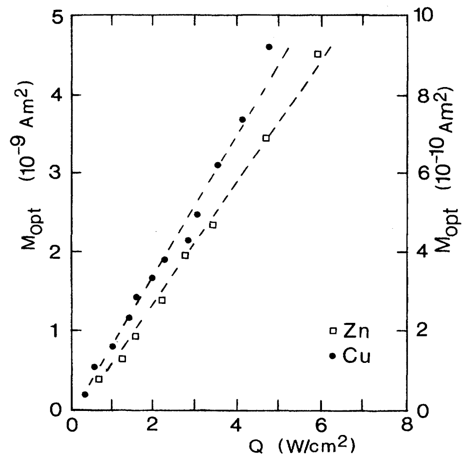

FIG. 3. Dependence of the magnetic moment M_{opt} on the optical power density Q at $\lambda=514.5$ nm applied on the Zn (right-hand scale) and the Cu (left-hand scale) specimen at 4.2 K.

Consequently part of the energy flux is lost as photoinduced surface currents spreading symmetrically around the illuminated spot. This part is proportional to an integral over a function $\sin\theta \cdot I(\theta)$ where $I(\theta)$ represents for the distribution of the light intensity on the surface of the specimen. We assume that $I(\theta)$ has a Gaussian shape around the center of the cross section of the light cone. Finally the loss due to the surface currents is estimated to be about 20%.

When using the optical fiber the specimen can be immersed into liquid helium and illuminated without disturbing effect of boiling coolant. Another advantage is that several optical contacts can be made simultaneously on its surface. In the lower part of Fig. 2 are shown the magnetic moments observed by the SQUID when illumination is switched from one fiber to another. The accompanying reversal of the sign of the signal suggests that after passing the 1 mm thick bridge of copper the current induced by light in these two cases flows in the opposite directions through the detection loop, i.e., we are dealing with bulk currents. Possible surface currents entering from the illuminated spot to opposite directions in the detection loop do not have any net influence to its magnetic moment because they suffer the same scattering loss within the normal regions of the loop.

In Ref. 3 is derived a theoretical expression for the photoinduced bulk current, J, in a metal under normal incidence of illumination,

$$J = e\eta \frac{rQ_z}{\hbar\omega} \frac{l}{L} S, \tag{1}$$

where $\eta=0.5$ is a numerical factor, $rQ_z/\hbar\omega$ is the absorbed photon flux, l/L is the ratio of the electron mean free path to the sample length, and S is the illuminated area. According to Eq. (1) J is directly proportional to (i) the applied power density Q_z, (ii) the absorbed photon flux, and (iii) the reciprocal of the sample length.

As shown in Fig. 3 the magnetic moment observed from illuminated Cu and Zn samples increases linearly with increasing pumping power. As shown for photoinduced surface current [1] heating of the specimen by light decreases the value of M_{opt} by losses due to enhanced electron-phonon interaction. This would bring about nonlinear behavior of $M_{opt}(Q)$. To avoid excessive optical heating the measurements were made in the range of $Q<6$ W/cm^2. Corresponding to the optical power density $Q=1$ W/cm^2 applied over an area of 0.7 mm^2 on the Cu surface we observe the moment of the closed loop formed by the L-shaped Cu part and the superconducting short circuit to be $M_{opt}=0.9\times10^{-8}$Am^2. Then the current flowing through the 1 mm thick Cu bridge is $\sim$23 μA at the photon energy 2.4 eV.

At the values of $Q_z=1$ W/cm^2, $1/L=0.1$, $\omega=3\times10^{15}$ s^{-1},

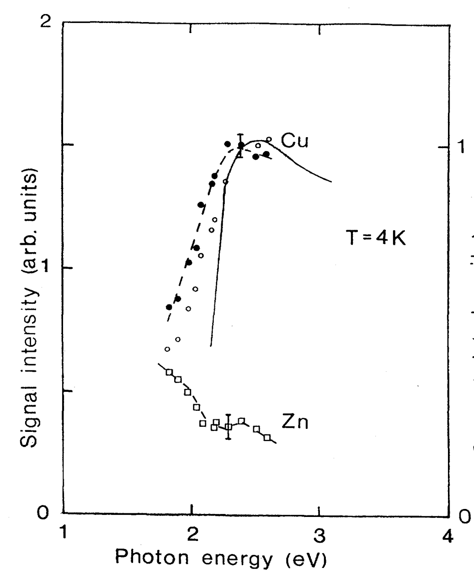

FIG. 4. Dependence of the M_{opt} signal for Cu and Zn (left-hand scale). The dashed lines are to guide the eye and the solid line shows the absorption edge of Cu at visible wavelengths 6. The open circles represent the values of the current J calculated for the Cu specimen from Eq. (1). The data of J are normalized to unity at the point with the error bar (2.41 eV). All measurements are made at the power density of 3 W/cm^2 from the output of the optical fiber.

$r=0.1$, and $S=0.1$ cm the prediction from Eq. (1) is $J\simeq0.3$ mA.[3] Assuming that these parameters apply for our experimental conditions except that according to reflectivity data[4] of Cu we have $r\simeq0.4$ and the cross section of the fiber is $S=0.007$ cm^2 the predicted result is 92 μA. This is about 4 times the value of the observed current, 23 μA. It is likely that this discrepancy can be attributed both to the uncertainty of the electron mean free path[5] in the specimen and to the fact that the geometry considered in the present paper is somewhat more complicated than the comparatively simple situation discussed in Ref. 3 (where the current was assumed to depend on a single spatial coordinate).

In Fig. 4 are shown the dependences of the magnetic moments (signal intensity) induced in the Cu and Zn samples on the energy of the photons used for illumination. The current induced in Cu in the blue-green region of the spectrum (~2.3–2.6 eV) is about five times larger than the current created in the Zn sample. In the red region the two curves approach each other. On the other hand they resemble the absorption edge of Cu due to interband transitions[6] or to the increase of the optical absorption in Zn at red wavelengths.[7] This agrees with the general statement made earlier that the current J should be directly proportional to the absorbed photon flux. However, as may be observed from Fig. 4 our data of J for Cu, normalized against the change of the laser frequency ω, differ to some extent from the experimentally determined absorption edge of Cu.[6] The more slow decrease of the current with decrease of the photon energy as compared with the absorption data obtained from light reflection measurements[6] gives evidence that the electronic processes involved in the two types of experiments are not exactly the same. The main difference is that J is proportional to the electron mean free path. The latter may be very sensitive to the excess electron energy above the Fermi level which is determined by the wavelength of the light. Indeed, the bigger is the energy, the more channels for the electron scattering are opened. At the same time, the absorption coefficient of light is much less sensitive to the electron scattering.

One of the predictions of Eq. (1) is that J is proportional to the inverse of the length of the specimen, $1/L$. This was tested by increasing gradually the length of the current path inside Cu before it could reach the end of the Pb stripe below the illuminated spot on the sample surface. In this way the value of L was varied from 1 to 8 mm. As shown in Fig. 5 the observed signal obeys well the L^{-1} dependence when the distance from the top of the specimen is clearly larger than the thickness of its upper leg (~1 mm). The breakdown of this behavior at small values of L is also in agreement with our considerations. It can be explained by the influence of the geometrical structure of the specimen on the current flowing through the superconducting loop.

In a metal with a temperature gradient thermoelectric current may be generated between cold and hot regions. The effect can be attributed to difference between the electron energy distributions at the hot and cold ends of the specimen.[8] To investigate a possible contribution of the thermoelectric effect to our results a specimen containing two junctions of Cu and Pb was prepared. The junctions were situated on opposite sides of a square-frame construction 8 mm apart from each other. One of the connections between the two metals was covered with a 0.25 mm thick plate of resistive Si fixed on it by a thin layer of glue. When this plate was illuminated through the optical fiber we could detect a signal which was about 5% from that obtained when the specimen was partially covered by Pb and one corner consisting of Cu was exposed to light as in the U-shaped samples shown in Fig. 2. Another difference was that the sign of the signal observed in the thermoelectric configuration was opposite to that observed for the normal specimen. This difference may be caused by the ambiguity of the sign of the thermoelectric power in Cu. Depending on the strain in the specimen this sign may be either positive or negative.[5] These results suggest that the contribution of the thermoelectric effect to our data is not significant. It should also be noted that the concept of thermopower may not be well defined for photoexcited electrons having the energy distribution other than the near equilibrium electrons.

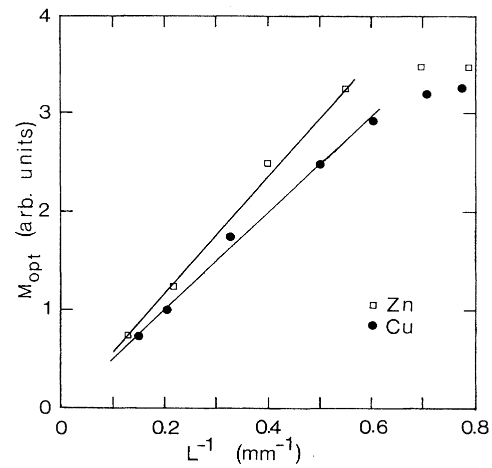

FIG. 5. Dependence of the magnetic moment of the detection loop on the inverse length of the current path in Zn and Cu.

To summarize, we have observed the photoinduced bulk current in Cu and Zn specimens illuminated with visible light at the temperature of 4.2 K. The strength of the current and its dependence on the experimental conditions agree reasonably with the theoretical predictions given in Ref. 3. An interesting result is that the observed signal correlates with the absorption spectra determined mainly by interband electronic transitions in the both investigated metals. However, utilizing the steep dependence of the absorption edge of Cu on the photon energy in the vicinity of 2 eV it is shown that the photoinduced current diminishes more slowly with decreasing the photon energy than the tail of the absorption band. This behavior is tentatively attributed to modification of the electron scattering processes when they are heated sufficiently above the Fermi level. On the other hand, careful measurements of the photoinduced bulk current in metals may provide a method to investigate these electrons.[9]

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