Photomagnetism of Metals

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(Received 21 January 1992)

A photoinduced magnetic moment has been observed in Cu and Al samples exposed to unpolarized visible light at low temperatures. It is shown that the light reflected from a metal surface transfers some of its quasimomentum to conduction electrons. This mechanism creates surface currents which, for an appropriate geometry, bring about the photomagnetic effect.

PACS numbers: 78.20.Ls, 73.20.−r, 75.20.En

Utilizing the high sensitivity of superconducting quantum interference devices (SQUIDs) it is possible to investigate magnetic phenomena induced by light in matter, such as semiconductors [1–4] or high-T_c superconductors [5]. However, observation of photomagnetism in normal metals is a challenging problem because within linear electrodynamics it is not obvious that an interaction between light and free electrons necessary for promotion of a detectable magnetic moment should exist.

In this Letter we report the observation of a magnetic moment in Cu and Al samples exposed to *unpolarized* light. We also furnish a theory of this phenomenon, based on the idea of transfer of the quasimomentum of light to conduction electrons near the metal surface.

The magnetometer used in our measurements is described elsewhere [5]. The first sample was a cylinder (inner diameter $\phi=6$ mm, length $l=20$ mm, and wall thickness 0.5 mm) made out of 99.999% pure Cu. It contained an inclined mirror to reflect the light from the direction of the axis to the inner wall of the cylinder [see Fig. 1(a)]. The angle of incidence θ between the light propagation direction and the normal of the surface of the target varies within the limits of $0<\theta<\pi/2$. Opening to the other end of the cylinder there was an oppositely inclined mirror, so that by turning the target upside down it was possible to change the direction of incidence to $-\pi/2<\theta<0$. The second type of samples were made out of 99.999% pure Cu or technical grade Al. They had two mirrors at the same vertical level, inclined oppositely so that they both give $\theta>0$. Half of the target ($\phi=6$ mm, $l=20$ mm, and wall thickness 0.5 mm) is shown in Fig. 1(b). The third type of target, made out of technical copper, had a half-closed glass prism mirror inside the cylinder [Fig. 1(c)] to eliminate all metallic surfaces, except that of the specimen, from the sample space.

On the outer wall of the fused silica tube containing the sample holder was wound the signal pickup coil ($\phi=12$ mm). The coil had eight turns of 0.05-mm-thick NbTi wire and its ends were connected to the input of an rf SQUID. As shown in Fig. 1(c) the axis of the coil coincided with the axis of the sample cylinder. The external magnetic field was shielded from the sample space by using combined superconducting and Mumetal shields. The residual field was less than 5×10^{-2} Oe.

The samples were illuminated through a bundle of 1-mm-thick optical fibers fixed above the mirrors or the prism inside the sample tube. The light source was an Ar-ion laser working at $\lambda=476.5$ or 514.5 nm. Before experiments the intensity was measured after each element in the system and the power density was determined

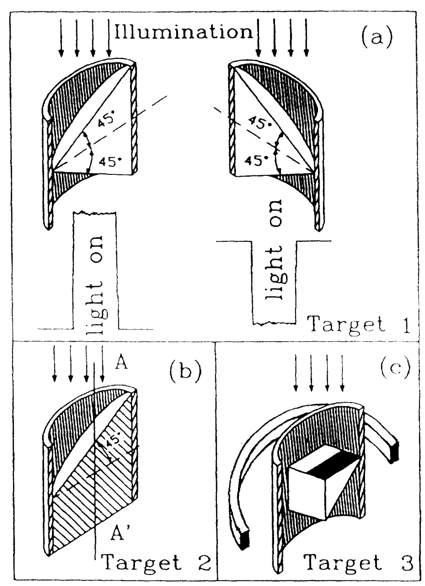

FIG. 1. Samples used to observe the photoinduced magnetic moment. (a) In target 1 light is reflected by one of the two mirrors inclined oppositely by 45° to the axis of the cylinder. The signals from the SQUID are shown for two opposite positions of the cylinders. (b) The target with two simultaneously illuminated mirrors can be imagined by turning the part shown around the AA' axis by 180° and combining it with the original one. (c) In target 3 a half-closed glass prism is used to guide the light. Also shown is the signal detection coil.

by observing the size of the illuminated area on a test cylinder opened from one side. To reduce the heating of samples by light they were anchored firmly to a copper holder and surrounded with He exchange gas at normal pressure. The temperature was measured with a Ge thermometer fixed to the specimen holder. Under illumination the measured temperature rise was less than 10 mK for 1 mW output power from the fibers. If we use the experimental data on the thermal conductivity for copper of various purity then we can give the upper limit of the temperature rise at the surface of the samples as 100 mK.

Magnetic signals from an illuminated metal surface were first observed with the target shown in Fig. 1(a). It is found that when the target is rotated upside down, not only the propagation direction of light in the plane of the detection coil but also the sign of the effect changes. The difference between the sizes of the signals observed in these two symmetries is due to inaccuracies in preparation and assembly of the mirrors. The signal could be nearly doubled when using two mirrors on the opposite sides of the axis of the cylinder [target 2, Fig. 1(b)]. When the cylinder was removed no output from the SQUID could be recorded.

Changing of the metal from Cu ($\rho_{4.2} \leq 1.3\times10^{-10}$ Ωcm [6]) to Al (6063 T5 alloy, $\rho_{4.2} \simeq 2.3\times10^{-7}$ Ωcm) reduced the signal by a factor of $\sim$500 at 4.2 K. This scales roughly with the values of $\sigma \sim \rho^{-1}$ [see Eq. (6)]. However, a straightforward comparison using the bulk resistivities can give only tentative estimates of the signal strength for the following reasons. First, for the case where the electron mean free path considerably exceeds the penetration depth δ, the equation for the current may be affected by the spatial inhomogeneity of the perturbation produced by the light. Second, the contamination of the surfaces, defects, and stresses also influence the electron scattering rate [7]. Using the prism inside the sample tube or changing the light wavelength did not bring new features to the data.

The dependence of the photoinduced magnetic flux on the pumping intensity shows a linear behavior up to the maximum power density, $\simeq$0.7 mW/mm^2, *at the surface of the sample* (see Fig. 2). Below 8 K the photoinduced magnetic moment, as shown in Fig. 3, obeys the temperature dependence of $\sigma(\tau)$ where τ is determined by the electron-phonon and the impurity scattering times, $\tau_{ph} \sim T^{-3}$ and τ_0, as $\tau^{-1} = \tau_{ph}^{-1} + \tau_0^{-1}$. For fitting the data we used the values of $\tau_{ph} = 2.7\times10^{-9}$ s at 4.65 K and $\tau_0 = 3\times10^{-10}$ s [8]. The $\tau_{ph} \sim T^{-3}$ relationship is regularly observed for Cu at low temperatures [8,9], deviating from the T^{-5} law predicted theoretically for the low-angle electron-phonon scattering process [10]. Above 35 K our data can be fitted approximately by using $\sigma(T)$ obtained from the Bloch-Grüneisen law for ρ [11]. Between 8 and 35 K there is a crossover region, starting near the upper limit, 10 K, for the validity of the Debye-Sommerfeld heat capacity $C_V = \gamma T + a T^3$ in Cu [12].

The physics of the effect described above can be visualized as follows. When light falls on the sample part of it is absorbed by electrons and part is reflected at an intensity defined by the coefficient $1-r$. Along with energy some quasimomentum [13] is transferred to conduction electrons [14], exciting a surface current flowing within a layer δ, comparable to the penetration depth of light. Let θ be the angle between the Poynting vector **Q** of the incoming wave and the normal to the surface. For the Poynting vector of the reflected wave, **Q**′, we have $Q'_z = -(1-r)Q_z$. Then the absorbed energy flux is $rQ \times \cos\theta$. Let us assume that the light propagates in the x-z plane. Therefore we are interested in the x component of the current and the flux of the x component of the quasimomentum.

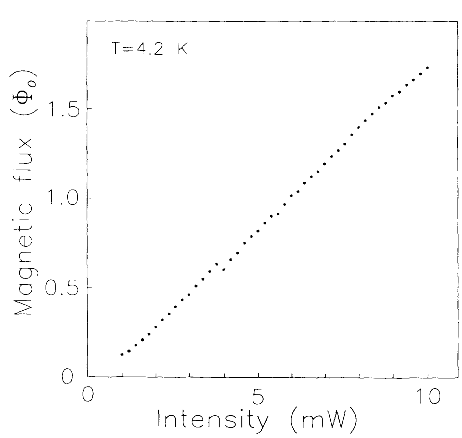

FIG. 2. Dependence of the magnetic flux on the power of the light falling on the inner surface of a Cu sample (target 2), in units of $\Phi_0 = 2.07\times10^{-15}$ Wb. The illuminated area is $S_{eff} \simeq 15$ mm^2 and $\theta_{av} \simeq 40°$.

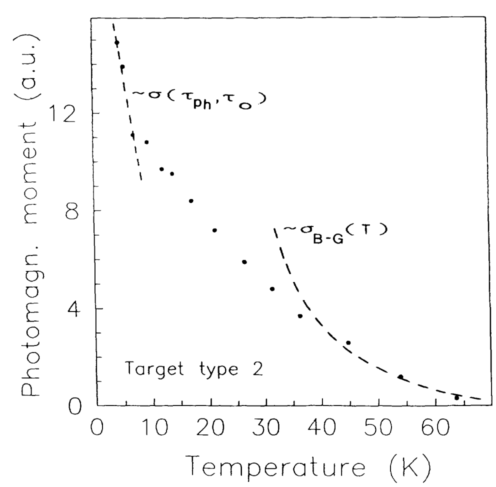

FIG. 3. Temperature dependence of the observed photomagnetic moment (dots). The dashed line below 10 K is a fit by Eq. (6) with $\sigma(\tau)$ calculated by using the values of τ_{ph} and τ_0 from Ref. [8], and that above 35 K is a fit with $\sigma(T)$ determined from the Bloch-Grüneisen approximation for $\rho(T)$.

We consider a macroscopically homogeneous system along the x direction. Because the quasimomentum of light is small in the atomic scale and the umklapp processes are of no significance, the x component of the quasimomentum is conserved in the interaction between light and the conduction electrons. In vacuum, where the quasimomentum and the ordinary momentum are equivalent [13,14], the quasimomentum flux coincides with the corresponding component of the Maxwell stress tensor. Therefore the time-averaged flux of the x component of the quasimomentum through a unit area of the metal surface is $r(Q/c)\cos\theta\sin\theta$.

Light is absorbed within the surface layer defined by δ. Within the same layer the quasimomentum is transferred to the conduction electrons. Denoting the quasimomentum density of the electrons by $\mathbf{P}$ one can write for the rate of variation of P_x

$$\frac{\partial P_x}{\partial t}\delta = r\frac{Q}{c}\cos\theta\sin\theta - \frac{P_x}{\tau}\delta\,. \quad (1)$$

Under stationary conditions $\partial P_x/\partial t = 0$ one gets the estimate $P_x = r(\tau/\delta)(Q/c)\cos\theta\sin\theta$, where τ is the relaxation time of the electron quasimomentum.

Now, one can write the current density $\mathbf{j} = ne\mathbf{v} = e\mathbf{P}/m$, where $\mathbf{v}$ is the electron drift velocity, i.e., an average velocity of the electron system, and m is an average value of the electron effective mass. Then the surface current density can be expressed as

$$g_x = j_x\delta = r(e\tau Q/mc)\cos\theta\sin\theta\,. \quad (2)$$

This equation can be compared with the balance equation for the electron quasimomentum density under the influence of a dc electric field $\mathbf{F}$,

$$\partial\mathbf{P}/\partial t = en\mathbf{F} - \mathbf{P}/\tau_1 = 0\,, \quad (3)$$

where n is the concentration of the conduction electrons. From such a balance we get Ohm's law: $\mathbf{j} = \sigma\mathbf{F}$, where $\sigma = e^2 n\tau_1/m$ (see also footnote [15]).

Equation (2) provides an estimate for the magnetic field variation across the metal surface, H, as

$$H = (2\pi e\tau/mc^2)rQ\sin2\theta\,. \quad (4)$$

If the surface current is excited on the surface of a metallic cylinder one can give the following estimate for the corresponding magnetic flux:

$$\Phi \simeq HS \simeq (2\pi e\tau/mc^2)rQS\langle\sin2\theta\rangle\,, \quad (5)$$

where S is the area of the orifice and the angular brackets denote the angular average. Assuming that r is of the order of 1 (for Cu, $r \simeq 0.5$) or that a significant part of the light is absorbed in reflections of the original beam and the diffusively scattered light returned back to the sample by the mirrors, we get

$$\Phi \simeq (2\pi e\tau/mc^2)\langle\sin2\theta\rangle QS\,. \quad (6)$$

Within the power levels used in our experiments Eq. (6) is in agreement with two important observations: (i) Φ depends linearly on Q (Fig. 2) and (ii) $\Phi \propto \tau(T)$ (Fig. 3). For the case $\tau \simeq \tau_1$ one can insert $\tau \simeq \sigma m/ne^2$. Assuming that $\sigma = 7.7\times10^9$ $\Omega^{-1}\text{cm}^{-1}$, $n = 8.5\times10^{22}$ cm^{-3}, $Q = 0.7$ mW/mm^2, and $S_{\text{eff}} = 15$ mm^2 we have $\Phi \simeq 0.6\Phi_0\langle\sin2\theta\rangle$. In spite of the reasonable agreement with the experimental data in Fig. 3 the estimated value of Φ warrants several comments.

First, in a real situation the inner part of the cylinder is not homogeneously illuminated. Then it is likely that part of the current is not encircling the orifice of the cylinder and therefore either not contributing to the magnetic flux or making a small contribution of the opposite sign. As, however, about a half of the cylinder is illuminated in the plane of the detection coil, an order-of-magnitude estimate should remain valid.

Second, it is assumed in Eq. (6) that τ_1 and τ are of the same order and determined by the same mechanism, for instance by impurity scattering. It is known from magnetoacoustic measurements in Cu [8] that at 4.2 K both the electron-phonon and the impurity scattering contribute to the electronic conductivity. Also the perturbations induced by light and a dc electric field are not similar in their angular and energy dependence. In addition, τ_1 and τ can differ because phonons emitted in the relaxation of the optically excited electrons may provide an extra mode of scattering.

Third, the electrons can transfer part of their quasimomentum to phonons [16] or phonons can be directly excited by light. As a result drag of electrons by the phonons may take place creating an additional current with entirely different relaxational characteristics.

Finally, emission of photoelectrons may occur under illumination. The photoelectrons near the metal surface can also take part in the absorption of light. Then the light could excite photocurrent along the metal surface and just above it, transferring to the photoelectrons the true momentum rather than the quasimomentum. Under such circumstances τ would have no relation to any relaxational characteristics of the conduction electrons inside the metal.

The authors wish to express their sincere gratitude to Armin Thellung for a number of valuable suggestions and critical remarks. One of the authors (V.L.G.) wishes to acknowledge the Forschungszentrum Jülich for the hospitality extended to him while the work of this paper was done. He is also grateful to the Alexander von Humboldt Foundation and to Turku University Foundation for financial support.

[1] D. D. Awschalom, J.-M. Halbout, S. von Molnar, T. Siegrist, and F. Holtzberg, Phys. Rev. Lett. **55**, 1128 (1985).

[2] H. Krenn, W. Zawadski, and G. Bauer, Phys. Rev. Lett. **55**, 1510 (1985).

[3] R. Laiho, E. Lähderanta, E. Supponen, and L. Vlasenko,

Phys. Rev. B **41**, 7674 (1990).
[4] M. Kondo and Y. Nishida, J. Phys. Soc. Jpn. **60**, 2415 (1991).
[5] R. Laiho, E. Lähderanta, L. Saisa, Gy. Kovács, G. Zsolt, I. Kirschner, and I. Halász, Phys. Rev. B **42**, 347 (1990).
[6] Announced by the supplier, Outokumpu Ltd., Finland.
[7] I. Nakamichi and T. Kino, J. Phys. F **18**, 2421 (1988).
[8] B. B. Johnson and J. A. Rayne, J. Low Temp. Phys. **40**, 617 (1980).
[9] R. E. Doezema and J. F. Koch, Phys. Rev. B **6**, 2071 (1972); V. F. Gantmakher and V. A. Gasparov, Zh. Eksp. Teor. Fiz. **64**, 1712 (1973) [Sov. Phys. JETP **37**, 864 (1973)]; R. Stubi, P-A. Probst, R. Huguenin, and V. A. Gasparov, J. Phys. F **18**, 2429 (1988).
[10] A. A. Abrikosov, *Fundamentals of the Theory of Metals* (North-Holland, Amsterdam, 1988).
[11] H. M. Rosenberg, *Low Temperature Solid State Physics* (Oxford Univ. Press, Oxford, 1963).
[12] *Handbook of Chemistry and Physics,* edited by R. S. Weast (Chemical Rubber Company, Boca Raton, 1971), D-139, 51st ed.
[13] R. Peierls, in *Highlights of Condensed Matter Theory,* LXXXIX Corso (Societa Italiana di Fisica, Bologna, Italy, 1985).
[14] V. L. Gurevich and A. Thellung (to be published).
[15] Note that j_z could not be calculated by this method because near a metal surface P_z is not the density of a conserved quantity. Neither could one apply to the calculation the *classical* method proposed by H. E. M. Barlow [Proc. IRE **46**, 1411 (1958)] to calculate the bulk dc electric field produced by an electromagnetic wave traveling within a semiconductor.
[16] V. L. Gurevich and A. Thellung, Phys. Rev. B **42**, 7345 (1990).