EXPERIMENTAL DETERMINATION OF THE POLARON MASS IN CUPROUS OXIDE

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The effective mass of the current carriers in cuprous oxide was determined at helium temperatures by the method of diamagnetic resonance. The value obtained is close to the polaron mass calculated for cuprous oxide using Pekar's theory.

1. STATEMENT OF THE PROBLEM

A number of investigations on the theory of polarons have appeared, but so far as we are aware there has been to date no direct experimental confirmation of the existence of mobile polarons. The experimental observation of a carrier mass equal in order of magnitude to the calculated polaron mass would provide such confirmation. We set ourselves the problem of measuring the polaron mass in Cu_2O, using the method of diamagnetic (cyclotron) resonance.

It should be noted that, because the infra-red oscillation frequencies of ions in a crystalline lattice are large in comparison with resonant frequencies in the centimeter band, the polarization of the crystal is able to follow the high frequency field of the absorbed radiation, and, consequently, diamagnetic resonance will reveal the mass of the polaron, if it exists, and not the effective mass of an electron moving relative to the polarization well.

For the polaron mass Pekar[1] has given a formula (10.35) which, using the relationship (10.37) in the same work, can be written in the form

$$M = 9.08\cdot 10^3 (m^*/m)^3 c_0^4/\omega_1^2, \tag{1}$$

where m^* is the carrier effective mass, m is the electronic mass; ω_1 is the limiting frequency of the polarization oscillations of the ions;

$$c_0 = 1/n^2 - 1/\varepsilon_0, \tag{2}$$

n is the refractive index, and ϵ_0 is the static dielectric constant. Equation (1) was obtained for a crystal of the rock-salt type, but for other crystals it can only differ by a numerical factor of order unity.

If, following Pekar (reference 1, pp. 210 and 214), we take for cuprous oxide

$\varepsilon_0 = 9$, $n^2 = 4$, $c_0 = 0.139$, $m^*/m = 1.81$, $\omega_1 = 4.73\cdot 10^{13}$,

then formula (1) gives

$$M = 9\cdot 10^{-27}, \quad M/m = 9.8.$$

These values should, however, be considered in the light of more recent investigations. The ratio $m^*/m = 1.81$ used by Pekar was obtained indirectly and is unreliable. Gross and Pastrnyak,[2] from measurements of the diamagnetic Landau levels, adopt for cuprous oxide $m^*/m = 0.80$; they obtain approximately the same value from the exciton spectrum.[3] In reference 2 are also given more accurate values of ϵ_0 and n^2, viz: $\epsilon_0 = 7.5$ $n = 1.8$, which lead to the value $c_0 = 0.176$. Using these values of m^*/m and c_0, we obtain $M/m = 2.20$.

However, as Haken[4] has remarked, because of the rapid motion of the electron, the polarization of the neighboring electronic orbits does not take place completely. Consequently, instead of the dielectric constant n^2, some effective smaller value, κ, should be taken, because the frequency of oscillation of the electron in the polaron well is much larger than optical frequencies. This viewpoint is confirmed by the calculations of Muto and Okuno[5] who, for the x-ray exciton in KCl, obtained better agreement with experiment by taking $\kappa = 1.5$ instead of $n^2 = 2.12$.

The idea of retarded electronic polarization was used by Toyozawa[6] for a calculation on the electron polaron.

Thus, the creation of the polaron well results not only from ionic polarization, but in part also from the electronic polarization; this leads to an increase in the parameter c_0, to which the polaron mass is very sensitive. It is sufficient, for example, to take $n^2 = 1.6$ and $c_0 = 0.258$ to obtain $M/m = 7.5$.

It should also be remarked that according to Feynman[7] formula (1) for the polaron mass is inaccurate and its error is difficult to estimate. So, even if we take the correct value of c_0, the calculated ratio M/m is correct only with an accuracy of tens of percent.

2. EXPERIMENTS

The study of the absorption of high-frequency energy was carried out in a microwave-spectrometer of the superheterodyne type similar to that described by Manenko and Prokhorov[8] with an intermediate frequency of 60 Mcs. The wavelength of the radiation used was $\lambda = 3.27$ cm. The range of magnetic fields in which the resonance absorption of the polaron is expected was determined from the resonance condition

$$M = eH / \omega c, \tag{3}$$

in which, having substituted the theoretical estimates of the polaron mass, values of from 7×10^3 to 3.3×10^4 oe were obtained.

The resonator consisted of a section of rectangular waveguide with a diaphragm of diameter 6.3 mm placed in a liquid-helium Dewar.

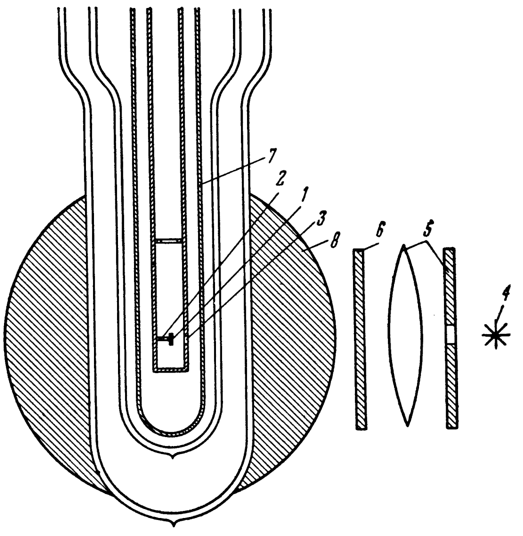

FIG. 1. Experimental apparatus.

The Cu_2O specimen (1 in Fig. 1) in the form of a plate $3 \times 3 \times 1$ mm was affixed to a polystyrene support, 2, situated at an antinode of the electric field of the high frequency wave. The specimen was illuminated through the aperture, 3, of 2 mm diameter in the wide wall of the resonator by a 340-watt PZh lamp (4) through a lens and diaphragm system, 5, and an SZS-14 filter, (6). To avoid contact with the liquid helium, the resonator and the waveguide carrying the energy were placed in a thin-walled glass tube, 7. The electromagnet, 8, with pole diameter 150 mm, produced fields up to 26,000 oe. Modulation of the magnetic field was produced in a number of experiments by additional coils at a frequency of 50 cps. The magnetic field was measured by the ballistic method.

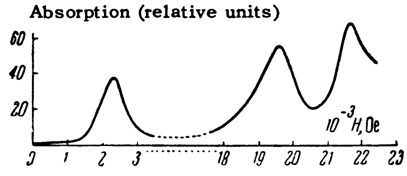

FIG. 2

Polycrystalline specimens of Cu_2O were used; they were optically transparent and aged for a while. A number of curves were obtained for the absorption of electromagnetic energy as a function of the external magnetic field. One of the curves is given in Fig. 2; the others had similar forms. Three absorption maxima were observed at magnetic field values of H = 2350, 19,600 and 21,600 oe, which, according to formula (3), correspond to ratios of carrier masses to the electronic free mass of 0.7, 6, and 6.6.

It should be mentioned that the effect of illumination on the crystal was already very large at H = 0. No maxima were observed on the curve in the absence of illumination at T = 4.2°K. Control experiments also showed that illumination without the filter did not essentially change the results, and the support without the specimen did not give absorption maxima.

3. DISCUSSION OF RESULTS

As was shown by Dresselhaus, Kip, and Kittel,[9] a clearly-displayed maximum of the diamagnetic absorption can only be observed under the conditions

$$\omega\tau > 1, \tag{4}$$

where ω is the resonance frequency and τ is the free path time of the carriers.

The photomagnetic effect in Cu_2O has been studied in this laboratory at helium temperatures,[10] and it was shown that the effect has a maximum in fields of the order of 6000 oe. This maximum occurs near $\omega\tau = 1$. Consequently, for fields greater than 6000 oe, condition (4) is satisfied. Thus, the two maxima obtained in large magnetic fields are, in fact, due to diamagnetic resonance. The corresponding values of the ratio M/m = 6 and 6.6 lie within the limits obtained by theoretical estimates for polarons. From what has been described, it follows that our experiments confirm the existence of polarons in cuprous oxide crystals.

The maximum at small magnetic fields H = 2350 oe gives a carrier mass close to the effective mass of band electrons or holes obtained by Gross and Pastrnyak.[2,3]

The simultaneous observation of the effective masses of the band electrons and the polarons indicates the existence of some equilibrium between the number of electrons in the band and number of polarons when the crystal is illuminated. It should be remembered that light excites electrons into the conduction band and polarons are formed subsequently. The existence of two polaron masses can, apparently, be explained by the presence of n- and p-type polarons. According to Gross and others,[11] the effective masses of electrons and holes in Cu_2O are approximately the same. But it should be noted that even a small difference in effective masses for the electrons and holes leads to a marked difference in the mass of the polarons, since the polaron masses are proportional to the cubes of the effective masses m^*.[1] This explains the presence of one maximum for electrons and holes and two maxima for polarons. Apparently the maximum of the photomagnetic effect[10] is determined by the polarons, since their concentration is greater than the concentration of electrons in the band; this follows from the larger height of the absorption maxima caused by polarons. So, in fact, $\omega\tau = 1$ at $H = 6000$ oe for polarons, and for electrons in the band this probably occurs at much smaller fields, because the maximum of diamagnetic absorption at $H = 2350$ oe is quite clearly displayed.

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