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IV-4. The Exciton and Magneto-Oscillatory Absorption of Cu_2O

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The effects of strong magnetic fields on the yellow exciton series of Cu_2O were studied by using pulsed fields up to 200 kG. With the increase of the field, each line moves towards high energies quadratically. High members cross the series limit, and above the limit the shift becomes almost linear to the fields. The effective reduced masses of an electron and a hole were estimated from a coefficient of the diamagnetic shift and separations between neighboring lines in the linear region to be 0.32 m and 0.29 m, respectively.

§ 1. Introduction

The effect of the coulomb force on the Landau level is an interesting problem from the experimental as well as from the theoretical point of view. The purpose of the present investigation is to study this effect through the behavior of exciton levels consisting of a hydrogen-like series under strong magnetic fields.

With application of the magnetic field to the Wannier type exciton, the line would shift towards high energy side because of the diamagnetic effect, and if the field is strong enough, high members of the exciton lines would cross the series limit into the continuum region and tend to interband magneto-oscillatory absorption lines.

As a typical Wannier type exciton, we chose the yellow series of cuprous oxide, which has been widely studied. According to the band scheme proposed by Elliot,[1] the yellow series corresponds to a forbidden transition between Γ_7^+ and Γ_6^+. Gross[2] studied the Zeeman effect up to 30 kG, and he also observed a few tens of magneto-oscillatory absorption lines.[3]

In this paper the results obtained by using pulsed magnetic fields up to 146 kG (200 kG in the case of 77°K) will be reported.

§ 2. Experimentals

The absorption spectra were obtained photographically. A spectrograph used is of medium size, equipped with four prisms, F:5.5, and has dispersion of 28 A/mm at 5,700 A. Because of the low intensity of a light source, a rather wide slit width such as 0.02 or 0.05 mm was used. The magnetic field is produced in a coil immersed in liquid nitrogen by discharging a condenser bank of 4,800 μF, and it varies sinusoidally with time with a half period of 1.5 msec. The maximum field attained is 200 kG for 77°K and 146 kG for 4.2°K. A light source used for absorption spectra is a Xenon flash lamp with a duration time of about 30 μsec, which is triggered at the peak of the magnetic field. The direction of the light beam was set parallel to the axis of the coil and an unpolarized light is used, so that the absorption spectra correspond to σ-components of the Zeeman pattern or the arrangement is the so called Faraday configuration.

Specimens are of polycrystalline ones of a few μ in thickness,* and they are immersed in the coolant.

§ 3. Results and Discussions

The spectrograms were taken at 77°K and 4.2°K. Main features were the same for both temperatures except the positions and widths of

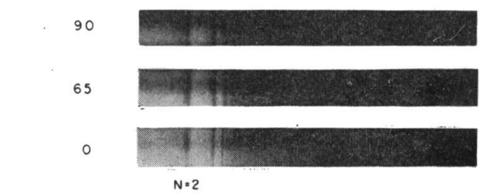

Fig. 1. Absorption spectra of Cu_2O at 4.2°K.

* Specimens were kindly furnished by Prof. Hayashi.

absorption lines, so that in the following the results at 4.2°K will be mainly described.

An example of the absorption spectra under various magnetic fields is shown in Fig. 1. Without the field, four absorption lines are observed with the slit width of 0.02 mm, which correspond to $n=2$, 3, 4 and 5 of the hydrogen series. With

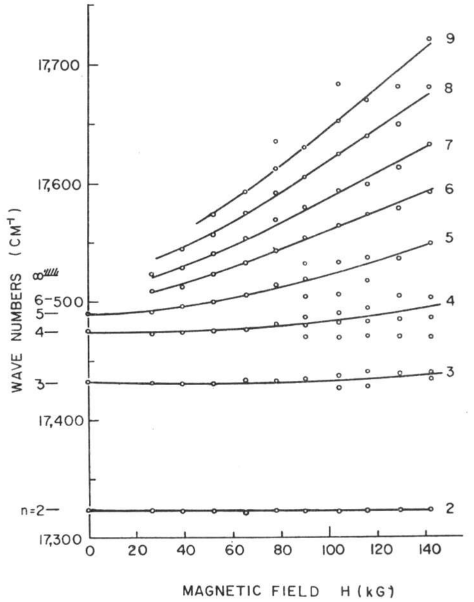

Fig. 2. The positions of the absorption lines of Cu_2O at 4.2°K vs. magnetic fields H.

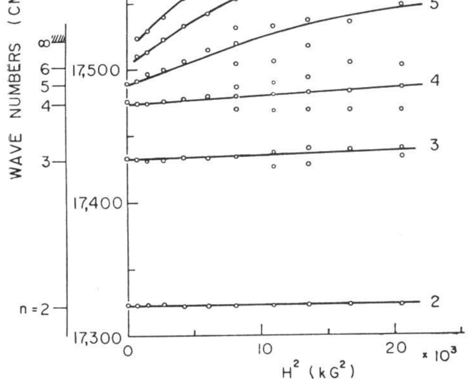

Fig. 3. The positions of the absorption lines of Cu_2O at 4.2°K vs. the square of magnetic fields H^2.

application of magnetic fields, number of lines become visible near and at the region beyond the series limit. These lines are numbered successively as $n=6$, 7, 8 and so on in the order of increasing energy. As the magnetic field is increased, all lines move towards higher energy side.

In Fig. 2, the positions of the absorption lines are plotted as a function of the magnetic field H. The lines with large quantum numbers show larger amount of shift than the lines with small quantum numbers, and at high magnetic fields the shift is nearly proportional to the magnetic fields for lines with large n.

In Fig. 3, the positions are plotted *versus* the square of the magnetic fields $(H)^2$. For lines with small n or at low fields the shift is proportional to H^2, and it is approximately expressed as

$$\Delta\nu=4.2\times10^{-12}n^{3.8}H^2\ \mathrm{cm}^{-1}.$$

The exponent of n is not really a costant, but it further decreases for large n. This might be an indication of deformation of exciton orbits caused by the magnetic fields.

Both quadratic and linear dependence of the shift on the magnetic field are clearly observed for the lines with $n=5$, 6 and 7. The transition from the quadratic to the linear relation appears to start at a magnetic field where each line crosses the series limit.

With the increase of the magnetic field, all lines, especially members above $n=5$, become broad. Broadening is probably due to unresolved structures. With our experimental conditions, the line $n=3$ is observed to split into a doublet, and the line $n=4$ into a triplet. Within our accuracy, the amount of the splitting is linear to the magnetic field, and at 100 kG, it is 1.04 cm^{-1} for $n=3$, 1.10 cm^{-1} for a low energy component and 0.88 cm^{-1} for a high energy component of the line $n=4$.

The intensities of the Zeeman components are not equal, that is, the high energy component is stronger than the lower energy one. The intensity ratios are approximately 1:2 and 2:5:7 for the doublet and triplet at 100 kG. Though the ratios change with the magnetic field, the high energy component is stronger throughout our experimental range, so that if the structure is not resolved the line shape would be asymmetric and the peak would always be located in the high energy side of each line in the case of $n=3$ and

4. The line $n=5$, whose structure can not be resolved, has asymmetric shape, which changes remarkably with the magnetic field. This line shades off towards longer wavelength below 62 kG, towards shorter wavelength between 91 and 104 kG, and it becomes almost symmetric above 117 kG. The line $n=6$ appears to be symmetric above 60 kG. The lines $n=3$ and 4 does not cross the series limit below the maximum value of the magnetic field in the present experiment, while the lines $n=5$ and 6 cross the limit at around 100 and 60 kG, respectively. The other higher members in the continuum region have symmetric shapes and appear to consist of two components of nearly the same intensity.

The experimental results described above can be summarized as follows. The behavior of each exciton line under magnetic fields is divided into two regions.

In region I, the shift of the line is quadratic with respect to the magnetic fields, and the structure changes rather remarkably with the field.

In region II, the shift is nearly proportional to the field, and the structure change is rather simple, *i.e.* the line width or the amount of splitting increases with the magnetic field, the shape or the intensity ratio remaining constant.

The transition from region I to II occurs at a magnetic field, where each line crosses the series limit or exceeds the limit by a few tens in wave number.

In region I, the actual shifts determined in the present experiment were compared with the corresponding values extrapolated quadratically from the data of Gross[2] below 30 kG. They agreed fairly well with each other at 100 kG for $n=4$ and 5, and at 90 kG for $n=6$.

In region I, the exciton nature is still predominant and the effects of the magnetic field, *i.e.* the Zeeman (Paschen Back) effect and the diamagnetic effect can be taken into account as perturbations. While in region II, the motions of an electron and a hole are mainly governed by the magnetic field, so that the absorption line in this region corresponds to the inter-band magneto-oscillatory absorption.

The effective reduced mass of an electron and a hole μ can be estimated by two ways.

One is to determine from the coefficient of the diamagnetic shift by fitting the values for small n to the formula

$$\Delta\nu=(\hbar^3\varepsilon^2/16\pi e^2\mu^3c^3)n^4H^2\ \mathrm{cm}^{-1},$$

where ε is the dielectric constant.

Another is to calculate from the separation between successive absorption lines in region II, which is assumed to be given by

$$\Delta\nu=eH/(2\pi\mu c^2)\ \mathrm{cm}^{-1}.$$

They are found to be $0.32\,m$ and $0.29\,m$, respectively, which are to be compared with $0.25\,m$ determined from the Rydberg constant of the exciton series.[3]

References

1) R. J. Elliot: Phys. Rev. **124** (1961) 340.
2) E. F. Gross and B. P. Zakhartchenia: J. Phys. Radium **1** (1957) 68; Soviet Physics—Doklady **1** (1957) 678.
3) E. F. Gross: J. Phys. Chem. Solids **8** (1959) 172.

DISCUSSION

Grosmann, M. H.: Which is the precision of your measurements of wave-length? Is it possible to deduce effective masses of electron and hole from your measurements?

Kuwabara, G.: The accuracy of wave-length is one or two cm^{-1} for sharp lines. It is possible to obtain effective masses of an electron and a hole separately from the low field as well as from the high field region, if we can assign the observed components.

Mooser, E.: What is your explanation for the small dependence on the magnetic field of the lowest exciton state?

Kuwabara, G.: In our case, $(1/2)\,\hbar\omega_c/R_y$ is 0.05 for 200 kG, so that the effect of the magnetic field for the 2nd lowest state is quite small.