Cyclotron resonance and strong phonon coupling in n-type ZnS at high magnetic fields up to 220 T

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(Received 31 May 1994)

We have observed cyclotron resonance in the infrared wavelength range between 10.6 and 16.9 μm with pulsed high magnetic fields up to 220 T. The band-edge mass of n-type ZnS is $m^*=0.20\pm0.01m_0$ and the Fröhlich electron-phonon coupling constant α is 0.63 from a comparison with the experimental results of cyclotron resonance and a theoretical calculation based on the Wigner-Brillouin second-order perturbation theory. We have also observed phonon-assisted cyclotron resonance at the low-field side of the cyclotron resonance.

I. INTRODUCTION

Recently, ZnS has attracted much attention for its potential applicability to optoelectronic devices at short wavelengths. It is also an interesting material from a basic physics point of view because of the large electron–LO-phonon interaction. The Fröhlich electron-phonon interaction coupling constant

$$\alpha=\frac{e^2}{2\hbar\omega_{\rm LO}}\left[\frac{2m^*\omega_{\rm LO}}{\hbar}\right]^{1/2}\left[\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right] \quad (1)$$

is the largest among II-VI compounds. In polar materials with large α, significant polaron effects are observed. In a previous paper, we have studied a very large resonant polaron effect in n-type ZnSe, which has $\alpha=0.42$.[1] In ZnS, much larger polaron effects are expected to occur due to a larger value of α.

However, only very few reports on the effective mass of n-type ZnS have so far been made.[2-4] No studies on cyclotron resonance have ever been tried. This is because its low mobility and large effective mass have made it difficult to perform cyclotron resonance experiments. Recent advance of high-field magnet technology has enabled us to study cyclotron resonance in ultrahigh magnetic fields above 100 T (MG fields).[5] Far-infrared transmission experiments over a wide range of energy have now become possible.[1] In the present paper, we present an observation of cyclotron resonance (CR) in n-type ZnS above the LO-phonon energy ($\hbar\omega_{\rm LO}=43.4$ meV). The CR data were compared with a theoretical calculation based on the Wigner-Brillouin second-order perturbation theory and the electron band-edge mass is found from this comparison.

II. EXPERIMENTAL PROCEDURE

In the present work, infrared transmission experiments were performed in n-type ZnS by using CO_2 and H_2O lasers under pulsed high magnetic fields up to 220 T. Infrared laser lines at 10.6 μm (117 meV), 10.8 μm (115 meV), and 16.9 μm (73.4 meV) were used as a radiation source. The transmission signal was detected by using the extrinsic photoconductivity of a Cu-doped Ge detector cooled to 4.2 K for a H_2O laser, and a HgCdTe photovoltaic detector cooled to 77 K for a CO_2 laser. These detectors have sufficiently fast response time for the measurements in very short pulsed magnetic fields. Pulsed high magnetic fields up to 220 T were generated by the single turn coil technique with 6-mm bore coils.[5] A fast pulsed current is discharged from a 100-kJ capacitor bank into a single turn coil. The pulse duration was about 7 μs. The absolute field strength can be determined to an accuracy of better than 3% by measuring the induced voltage in a calibrated pickup coil wound around the sample. n-type ZnS was grown by iodine transport method with a normal parallel to $\langle 110\rangle$. Its resistivity is 3 Ω cm. n-type ZnS has two crystal structures, wurtzite and zinc blende. The crystal structure of our sample is zinc blende. More details about the single turn coil technique and the data-acquisition system are described in Ref. 5.

III. RESULTS AND DISCUSSION

Figure 1 shows a wavelength dependence of the transmission spectra in n-type ZnS. The resonance peak is observed twice on rising and falling slopes of the magnetic field. It should be noted that the good coincidence of the two slopes ensured a sufficiently fast response of the detector system and the accuracy of the measurement. A very broad cyclotron resonance was observed even at room temperature, owing to very high magnetic fields which easily satisfy the condition for observing cyclotron resonance, i.e., $\omega_c\tau\gg 1$. The mobility of this sample is 200 cm^2/V s at room temperature, which is estimated from cyclotron resonance linewidth. We also found another resonance at the low-field side of the cyclotron resonance.

Figure 2 shows a temperature dependence of the cyclotron resonance spectra at a wavelength of 10.6 μm. The cyclotron resonance absorption is observed around 190 T and its absorption intensity is less than 20%. This indicates that the number of free electrons contributing to cyclotron resonance is not so large even at room temperature because of its large donor ionization energy (≥ 50 meV). The absorption intensity of cyclotron resonance drastically decreases as the temperature is lowered.

Below 250 K, there is no response in the transmission spectra any more. Besides the main peak at 190 T, we can see a smaller subsidiary peak (peak X) at a lower field of 130 T. The lower-field resonance X is not the impurity cyclotron resonance ($1s$-$2p^+$ donor transition), because the intensity becomes weaker as the temperature is reduced, contrary to the case of the impurity resonance. The resonance field was determined by decomposing two peaks using Lorentzian curves, as shown in Fig. 3. Although the resonance peaks are broad, we found that this procedure provides an experimental error less than 0.1% in the determination of the two resonance peak positions.

In Fig. 4, we plotted the peak positions of the cyclotron resonance and the lower-field resonance X. A horizontal dashed line is the LO-phonon energy ($\hbar\omega_{\rm LO}=43.4$ meV). A solid curve is a calculated result of the Wigner-Brillouin second-order perturbation theory.[6] We found the calculation result is in very good agreement with the experimental data, when we employed the band-edge mass of $m^*=0.20m_0$. Fröhlich electron-phonon interaction coupling constant α is obtained as 0.63 using formula (1) where ϵ_∞ and ϵ_0 are 5.0 and 8.3, respectively.[7]

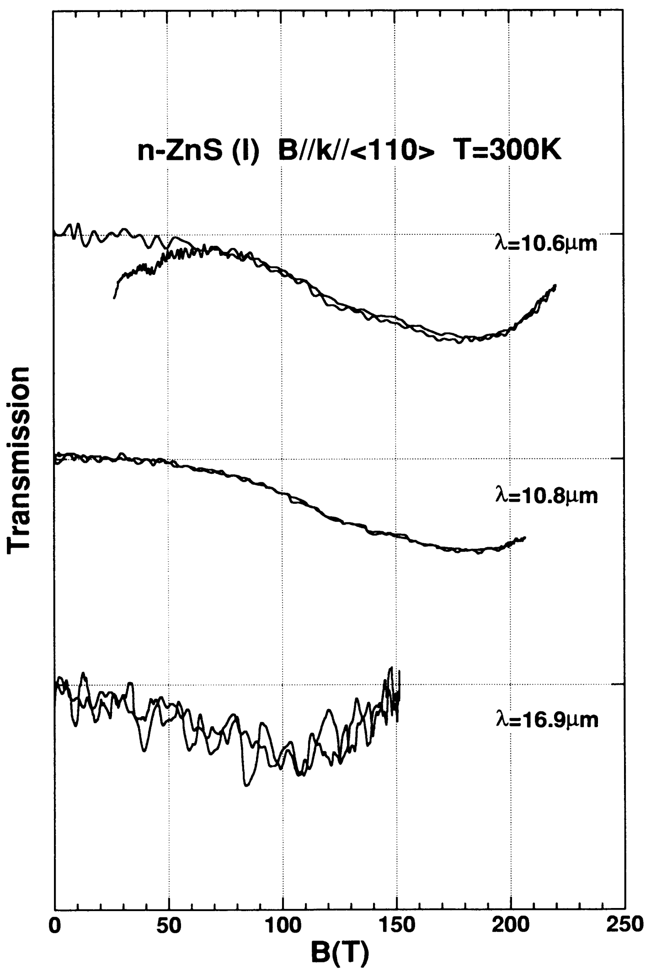

FIG. 1. Wavelength dependence of the magnetotransmission spectra in n-type ZnS. The cyclotron resonance (CR) and the phonon-assisted cyclotron resonance are observed.

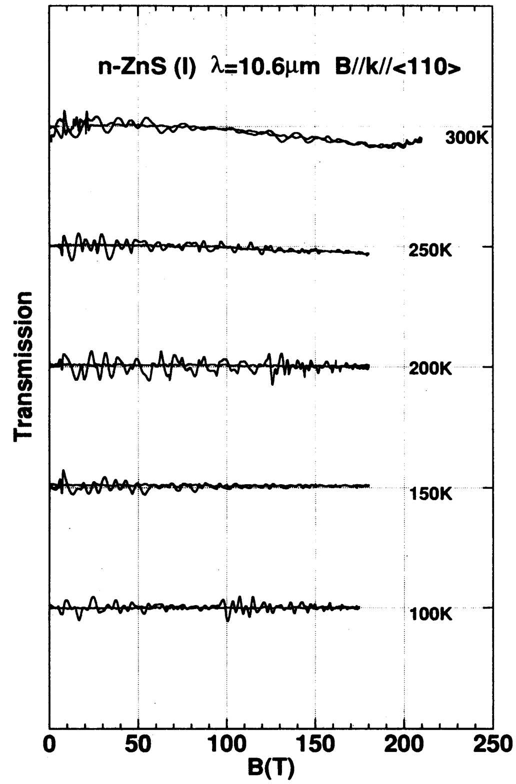

FIG. 2. Temperature dependence of the magnetotrnasmission spectra in n-type ZnS at 10.6 μm.

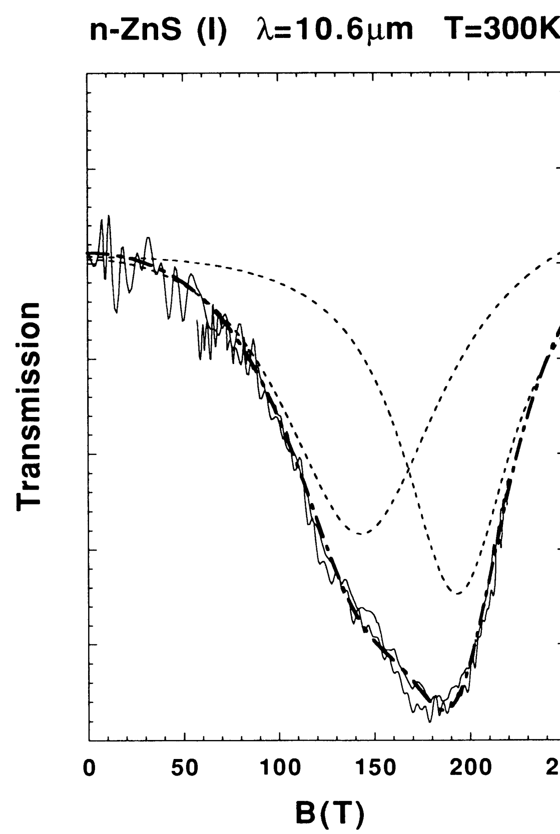

FIG. 3. The magnetotransmission spectra at a wavelength of 10.6 μm decomposed into two peaks by using Lorentzian curves.

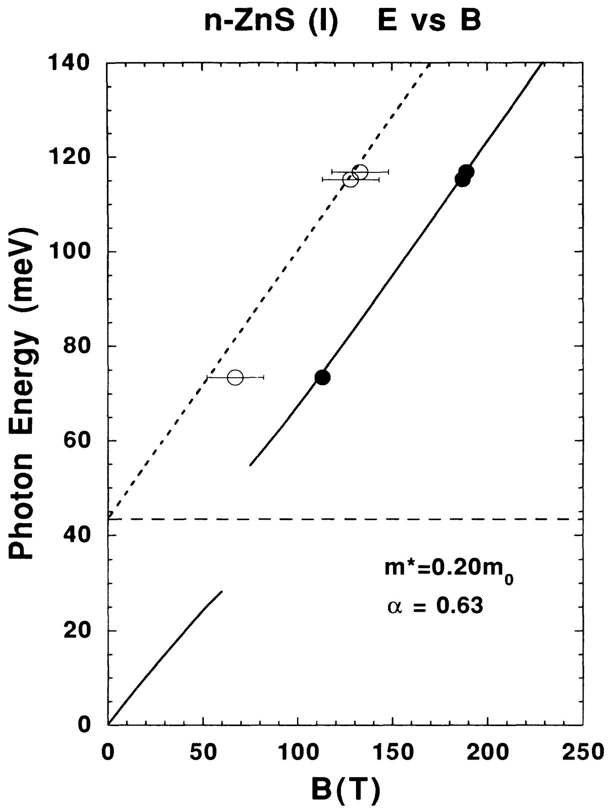

FIG. 4. Transition photon energies of the resonance peaks against magnetic field as compared with the Wigner-Brillouin second-order perturbation theory. The solid circles denote the CR data and the open circles denote the lower-field resonance. The LO-phonon energy is represented by the horizontal dashed line at 43.4 meV. The solid line is the theoretical line calculated by the Wigner-Brillouin second-order perturbation theory with $\alpha=0.63$. The broken line is a guided line representing $E=\hbar\omega_c+\hbar\omega_{LO}$.

The apparent cyclotron effective mass at various wavelengths is summarized in Table I, together with the data which have already been reported. The top of the data is a result of a magneto-optical experiment for wurtzite crystals by Miklosz and Wheeler.[2] Other results are calculated values by Cardona[3] and Lawaetz.[4] Cardona calculated the band parameter by using a three-level $\mathbf{k}\cdot\mathbf{p}$ perturbation theory for wurtzite crystals. The method of Lawaetz's calculation is also similar to that of Cardona except employing a five band model for zinc-blende crystals. A calculated result of Lawaetz is in good agreement with an experimental result of Miklosz and Wheeler. The effective mass of the present work is, however, significantly lighter than these values. Especially, the effective mass at 16.9 μm is much lighter owing to the resonant polaron effect. This is because the present experiments are performed in a higher-energy region than LO-phonon energy. If a cyclotron resonance experiment is performed at the low-energy region ($\hbar\omega \ll \hbar\omega_c$), the polaron mass ($m^*_{pol}$) should be obtained. It is deduced as $0.22m_0$ using the formula

$$m^*_{pol}=m^*(1+\alpha/6) . \quad (2)$$

TABLE I. The effective mass of the conduction band in n-ZnS (I).

λ (μm)	$\hbar\omega_0$	T (K)	m^*/m_0	Reference
			0.28	Miklosz and Wheeler[a]
			0.39	Cardona[b]
			0.28	Lawaetz[c]
10.6	117	10–300	0.188±0.005	
10.8	115	300	0.188±0.005	Present work
16.9	73.4	300	0.178±0.002	(Observed mass)

[a]Reference 2.
[b]Reference 3.
[c]Reference 4.

This value is very close to the result of Lawaetz and Miklosz.

The broken line is a line representing $E=n\hbar\omega_c+\hbar\omega_{LO}$ for $n=1$. This line is in good agreement with the positions of the peak X at lower fields. Thus the peak X can be assigned as the phonon-assisted cyclotron resonance, where a simultaneous emission of a LO phonon is involved in cyclotron resonance. Such phonon-assisted cyclotron resonances have been reported for n-type InSb.[8,9] Bass and Levinson predicted a phonon-assisted cyclotron resonance by using a perturbation theory.[10] Peeters and Devreese calculated the magneto-optical absorption of a polaron by using an anisotropic Feynman polaron model and also predicted that a series of phonon-assisted lines should be observed in strongly polar materials.[11] The Fröhlich electron-phonon coupling constant α of n-type ZnS is very large so that it is very reasonable to observe a phonon-assisted cyclotron resonance. The resonance absorption peaks are very broad because of the low mobility, so that other harmonic phonon-assisted cyclotron resonances ($n \geq 2$) are hidden by the cyclotron resonance peak.

IV. CONCLUSION

High-field magnetotransmission experiments up to 220 T allowed the first observation of cyclotron resonance and phonon-assisted cyclotron resonance in n-type ZnS. A significant resonant polaron effect was observed near the LO-phonon energy. The Wigner-Brillouin second-order perturbation theory for cyclotron resonance explained the experimental results well. The band-edge mass was determined as $m^*=0.20\pm0.01m_0$ from this comparison. The polaron mass to be observed in the low-field range, $m^*_{pol}=0.22m_0$, is estimated from this value and it is very close to previously reported data. We have also observed phonon-assisted cyclotron resonance at the low-field side of the cyclotron resonance, because of large Fröhlich electron-phonon interaction coupling constant α.

ACKNOWLEDGMENTS

The authors are obliged to Y. Yamada and T. Taguchi for providing the ZnS sample.

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