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# MAGNETO-OPTIC EFFECTS IN THE EXCITON ABSORPTION SPECTRA OBSERVED IN MEGAGAUSS FIELDS

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Various new aspects have been observed in the exciton absorption spectra for GaSe, $PbI_2$ and CdS in very high magnetic fields in the megagauss range.

## I. Introduction

The recent development of techniques for generating the megagauss fields greatly extended the available magnetic field range for investigation of magneto-optic effects in the exciton spectrum of semiconductors [1]. In such high magnetic fields, we can readily achieve the condition that the dimensionless parameter $\gamma = \frac{1}{2}\hbar\omega_c$ / Ry* (where $\hbar\omega_c$ is the energy of the cyclotron motion and Ry* is the effective Rydberg energy of excitons) is much larger that unity in many semiconductors. Thus the megagauss fields are useful to study the problem of hydrogen atom in high magnetic fields, in particular, to study the joining relationship between the states of the hydrogenic series at low field limit and the Landau-like states at high field limit [2,3].

Moreover, in such high fields, the diamagnetic shift becomes so large that we can study the diamagnetic shift of the ground state in detail, which provides a great deal of information on the energy band structure, the effect of the anisotropy or the exciton-phonon interaction on exciton spectrum and so on. In this paper, we present various new aspects observed in the spectra for excitons in GaSe, $PbI_2$ and CdS.

## II. Experimental Procedure

Pulsed high magnetic fields up to a few megagauss were produced by the electromagnetic flux compression method. The rise time of the fields was several microseconds. For observing the magneto-optic spectrum of excitons, a streak spectrometer with an image converter camera was used. A xenon flash lamp was used as a light source. The sample, about 2mm in diameter, was set at the center of the field, and a pick-up coil was wound on it to measure the field intensity. The temperature of the samples was lowered by flowing liquid helium in the vicinity of the sample through the sample holder. The lowest available temperature ranged 4-24K. Examples of the streak photographs of the optical spectra are shown in Fig.1 for three materials. The sharp pulses at both sides of the photographs represent event marker signals. The exciton absorption lines are seen as dark lines in the photographs.

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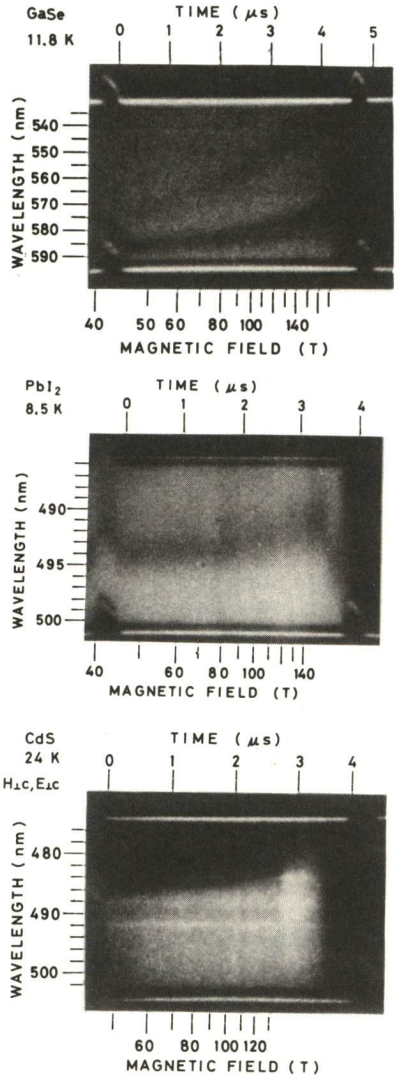


**Fig.1 Streak photographs of the magneto-optical spectra of excitons, Top: GaSe. (H// c), middle: $PbI_2$, bottom: CdS ($H_{\perp}c$, $E_{\perp}c$)**

## III. Results and Discussion

### 1. GaSe

The experiment was performed on thin samples cleaved from a single crystal, and magnetic fields were applied parallel to the c-axis. Figure 2 shows the photon energy of the observed exciton absorption lines as a function of magnetic fields. In addition to the strong N=1 line (the ground state), N=2, N=3 lines were observed just below the lines $L_1$ and $L_2$. A weak line was also observed between the N=1 and 2 lines. The ground state showed a Zeeman splitting. A better resolution was obtained by expanding the wavelength scale using a grating with finer grooves. The result of such a measurement is shown in Fig.3 for the diamagnetic shift of the ground state. The energy splitting $\Delta E$ is almost linear against the field with a slope corresponding to $g \simeq 2.9$, which is in agreement with $g \simeq 2.7 \pm 0.2$ reported by Mooser and Schlüter [4]. The midpoints between the two split lines are almost in good agreement with the theoretical lines for the 1s state by Cabib et al. (CFF) [5] and by Yafet et al. (YKA) [6], as shown in Fig.2.

As for the excited states, however, the observed lines are not those which are the extension of the hydrogenic 2s, 3s ... states lying below the lowest Landau transition $L_0$, but those which are associated with the Landau transitions $L_1$, $L_2$, the lines which tend to $(100^+)$, $(200^+)$ ... in the high field limit. The observation of the states which are associated with the higher Landau states instead of the 2s, 3s ... lines which lie below $L_0$ in the present experiment for $\gamma > 1$ is consistent with the theoretical prediction by Baldereschi and Bassani [7]. However, comparing the present result with the previous data at lower fields by Aoyagi et al. [8], it seems as if the observed lines N=2 and N=3 in the present experiments are extrapolated to the 2s, 3s states, at low field limit. On the other hand, the non-crossing rule predicts that the lines extrapolated from the m=0 hydrogenic states should lie, in the high field limit, below the lowest Landau state [3]. Therefore, the behavior of the N=2, N=3 lines at fields lower than 50T, particularly, their relationship with the 2s and 3s levels is still an open question to be solved with a precise measurement at lower fields around $\gamma=1$.

### 2. $PbI_2$

The measurement was performed on samples 140 nm in thickness prepared by the vacuum deposition on quartz substrates. The observed

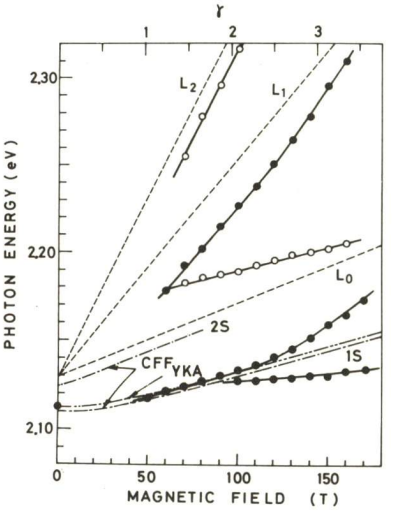


Fig.2 Photon energy of exiton lines as a function of magnetic fields for GaSe: Lines with closed and open points indicate strong and weak absorption lines, respectively. T=11.8K. The broken lines $L_0$, $L_1$, $L_2$ denote the transition energy between the Landau levels.

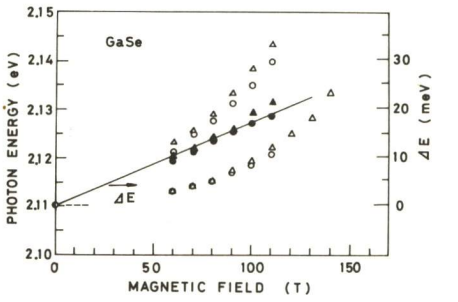


Fig.3 Details of the diamagnetic shift and the Zeeman splitting of the ground state exciton line in GaSe: The circles and the triangle points represent the data on two different samples. The open points (the left ordinate) show the photon energy of the lines and the closed points (the right ordinate) show the amount of the Zeeman splitting ΔE.

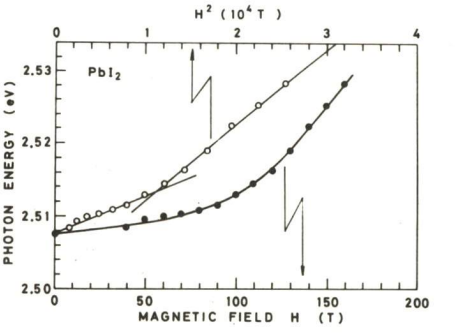


Fig.4 Photon energy of the ground state exciton line against the magnetic field (closed points and the lower scale) and against the square of the magnetic field (open points and the upper scale) for $PbI_2$: T=8.5K.

diamagnetic shift of the ground state is shown in Fig.4 as a function of magnetic field H and the square of the magnetic field $H^2$. For H<100T, the shift is almost proportional to $H^2$. The amount of the shift is 5.4meV at 100T. From this value the reduced mass of the exciton $\mu_\perp$ is obtained as $0.21m_o$, [1]. At fields higher than 100T it was found that the shift increased abruptly. The larger diamagnetic shift than expected from the $H^2$ dependence at higher fields cannot be explained by the theoretical field dependence for the 1s state [5,6]. It has been shown that the exciton-phonon interaction and the anisotropy have a large effect on the diamagnetic shift in $PbI_2$ [9]. However, these effects should quench the diamagnetic shift, and they will not cause the increase of the shift either.

There has long been a controversy on the deviation of the ground state energy of the exciton from a single Wannier series for $PbI_2$. Harbeke and Tosatti explained the ground state anomaly by a repulsive central-cell correction due to the cationic nature of the exciton in $PbI_2$ [10]. The observed sudden increase of the diamagnetic shift above 100T may be somehow related to the central cell correction combined with the effect of the shrinkage of the exciton wave function at high fields. However, it is not clear why the amount of the shift changes abruptly near 100T. More experimental and theoretical work will be required to clarify this puzzling behavior of the ground

state above 100T.

3. CdS

Measurement was performed on flakes of samples which were grown by a vapor growth. At zero magnetic field, the absorption peak for the N=1 state of the A-exciton was observed at 485.4nm (2.5543ev) for $E_{\perp}c$. Figure 5 shows the observed diamagnetic shift of the ground state of the A-exciton. In Fig.5, we plotted also theoretical lines of the diamagnetic shift after Gerlach and Pollmann [11] for CdS taking into account of the anisotropy effect. The broken lines are the calculated diamagnetic shift without the spin Zeeman energy. The ground state of the A-exciton is split into two states $\Gamma_5$ and $\Gamma_6$ for $H_{\perp}c$ due to the exchange and spin Zeeman energy [12]. The $\Gamma_6$ state lies below the broken line by the amount of spin Zeeman energy, and is shown by thin solid lines in Fig.5. If we assign the observed line as the $\Gamma_6$ state, the observed line should be compared with the thin solid lines.

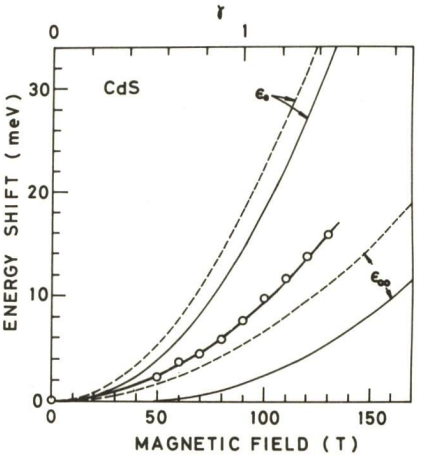


**Fig.5 The shift of the ground state of the A exciton v.s. magnetic field ($H_{\perp}c$, $E_{\perp}c$): The thick line with open circles is the experimental line. The broken lines and the thin lines are the theoretical lines for the 1s state, with the static ($\varepsilon_o$) and the high frequency ($\varepsilon_\infty$) dielectric constans.**

It should be noted that the observed diamagnetic shift is remarkably smaller than the theoretical value calculated with the static dielectric constants $\varepsilon_o$ ($\varepsilon_{o\parallel}$ and $\varepsilon_{o\perp}$). The observed line lies between the two theoretical lines, one calculated with $\varepsilon_o$, and the other with $\varepsilon_\infty$. Considering the facts that the polar electron-phonon interaction is considerably large in CdS and that the exciton binding energy of the ground state (29.8meV) is close to the LO phonon energy (37.8meV), we can expect a considerable amount of quenching of the diamagnetic shift in CdS. The observed quenching of the diamagnetic shift at $\gamma$=0.9 (H=80.8T) is ~37%, which is in agreement with the theoretical value of about 39% calculated by Behnke et al. [13].

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