High-field exciton spectrum of gallium selenide

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The excitation spectrum of a free exciton in GaSe has been measured in magnetic fields up to nearly 200 T. The measured spectrum compares favorably with a theoretical calculation done within the effective-mass approximation, consistent with a recent interpretation that the mass anisotropy is small, $\mu_{\parallel}/\mu_{\perp} \doteq 1$.

INTRODUCTION

The energy spectrum of excitons in GaSe has been of considerable interest both theoretically and experimentally. In early experiments Halpern[1] and Aoyagi *et al.*[2] measured the absorption spectra in magnetic fields up to about 18 T. Theoretically the exciton spectrum has been investigated by calculating an approximate solution within the effective-mass approximation. Since the effective mass tensor was thought to be very anisotropic, these experiments were initially interpreted using essentially two-dimensional models.[2,3] To fit the experiments, a mass anisotropy of $\mu_{\parallel}/\mu_{\perp} \doteq 5$ was used. The inability of these two-dimensional models to explain the observed spectrum was pointed out by Brebner *et al.*[4] More recent measurements and calculations have shown GaSe to be much more isotropic than was originally thought.

Several authors[5] have performed calculations for the energy spectrum in the absence of an external field using variational techniques. These calculations show a strong dependence of the spectrum on the anisotropy of the mass and dielectric tensors. Fitting the experimental data of Brebner and Mooser,[6] Baldereschi and Diaz[7] found a smaller anisotropy was needed.

More recent measurements of the exciton spectra for both zero field and small magnetic fields were made by Mooser and Schlüter.[8] For small magnetic fields, perturbation theory is valid. By calculating the small-field behavior of the ground-state energy (diamagnetic shift) in the Voigt and Faraday geometries and fitting to the low-field experiments, a value of the mass anisotropy of $\mu_{\parallel}/\mu_{\perp} = 1 \pm 0.2$ was found. This value also agreed well with their pseudopotential calculations of the band structure.

In the present paper using magnetic fields perpendicular to the planar structure (parallel to the c axis), the absorption spectrum of excitons in GaSe has been measured for fields up to nearly 200 T. At the higher fields the energies become largely insensitive to the value of the anisotropy, allowing a more or less independent determination of the exciton mass perpendicular to the c axis. Within the effective-mass approximation using very general trial wave functions a theoretical fit is obtained for the experimental data. The values found in the fit are in good agreement with the mass anisotropy found by Mooser and Schlüter.[8]

EXPERIMENTAL PROCEDURES

Figure 1 is a sketch of the experimental arrangement used. Shown at left is an argon light source. When the detonator is fired the shocked gas produces an intense, reasonably white light source. This light, after transmission through the GaSe sample, is reflected through a porthole mirror into a rotating-mirror spectrographic camera where it is time and wavelength resolved and recorded on film. Spectrograms from two such experiments or shots are shown on Fig. 2.

Magnetic fields are determined as functions of time from small probes placed in the field coils. Relative times are determined on the spectrograms from "film writing speeds" calculated from the measured mirror rotation rates. Absolute film times are usually established by obtaining the times of first light appearance on the films from photomultiplier tube signals that also arise from the light sources. Wavelength calibrations are obtained from electrically actuated bridgewires fired late in the shot time sequences, whose known spectra are then displayed on the records. Some wavelengths are noted on Fig. 2 alongside the spectra. The 632.8-nm He-Ne laser line is sometimes superimposed upon the films as in Fig. 2(a).

The magnetic field coils, 76 mm long and 17 mm in diameter, were large enough to house the double-walled cryostats required to cool to about 6.5 K the 6–7-mm diameter samples used in the experiments. The fields were produced by two kinds of explosive flux compression systems: a simple strip or bellows generator that produced peak fields of about 100 T and a two-stage device

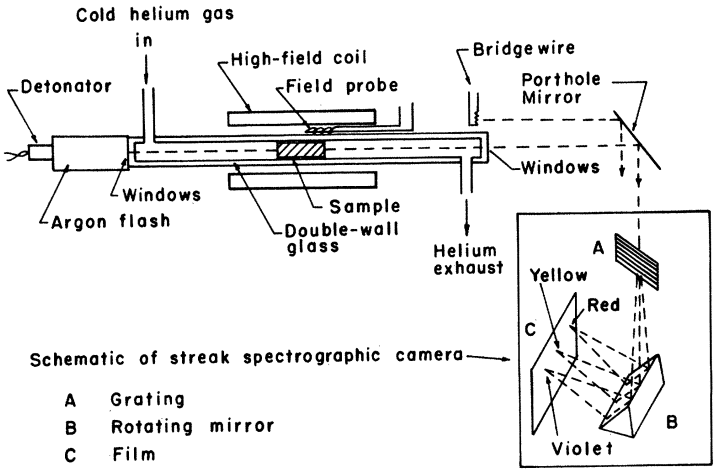

FIG. 1. Schematic drawing of high-field optical experiment.

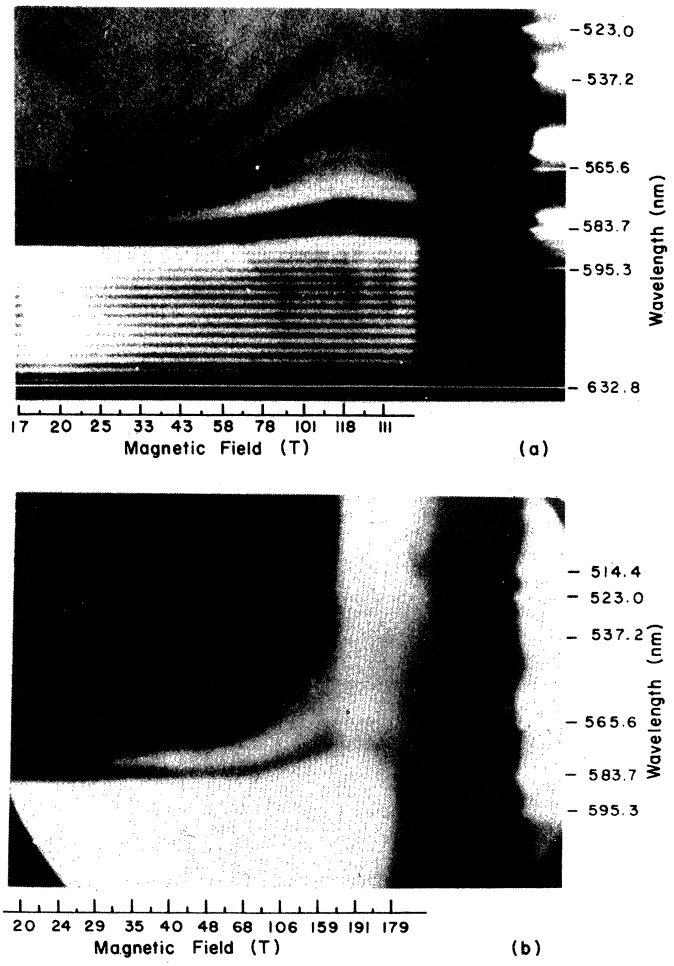

FIG. 2. Spectrograms of GaSe exciton absorption spectra in varying magnetic fields. (a) The upper spectrum was obtained from a thin crystal and also shows interference fringes; (b) the lower spectrum was obtained from a thick crystal and required use of an image intensifier to get sufficient light intensity.

that produced peak fields around 200 T. References 9–12 contain discussions of these and many other kinds of flux compression systems. Additional details of the complete experimental arrangement are given elsewhere.[13,14]

Crystals of GaSe were prepared by a chemical vapor transport method suggested by Mooser. In our work equivalent amounts of elemental Ga and Se (total 12 g), rather than reacted GaSe, were introduced into a quartz ampoule 30 cm long by 4 cm diameter. A sample of three mg/cm^3 of iodine was then added after which the ampoule was evacuated and sealed. Clusters of the crystal were formed by vapor transport, $T_2 \rightarrow T_1$, during a period of 30 h in which $T_2 \doteq 1155$ K and $T_1 \doteq 1105$ K. Suitable crystals were hand selected for the experiments, sandwiched between thin quartz disks, and mounted in the cryostats with c axes parallel to the field and light.

Our current energy-level diagram for GaSe is shown in Fig. 3 and differs mainly from an earlier proposed diagram in the location of the $N=0$ level.[15] Data obtained earlier by Halpern[1] and by Aoyagi *et al.*[2] are contained in the lower left rectangle. The $N=1,2$ levels are shown dashed at higher fields where level splitting was observed, although the amount of splitting was difficult to measure precisely. On some records it appeared that the $N=0$ level showed similar splitting although this is less certain. The weak level $N=0'$ may be the same level noted by Halpern, whose low-field value is about 17 230 wave numbers. As with Halpern, we do not treat this level as part of the exciton spectrum.

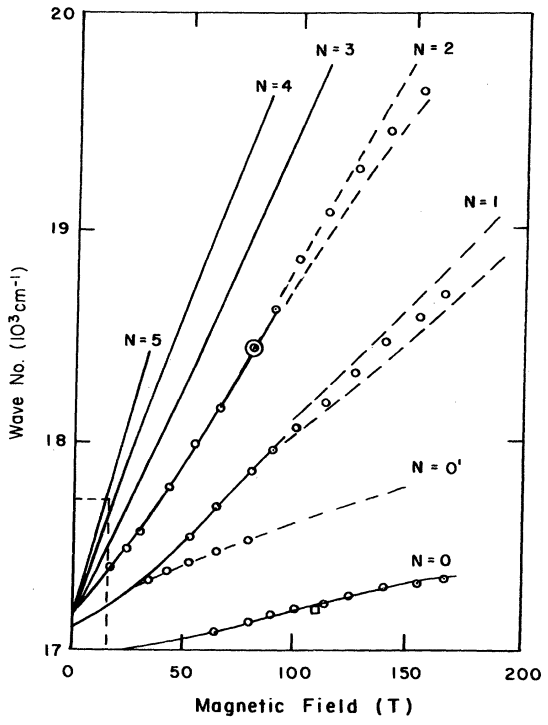

FIG. 3. Exciton spectrum (experimental) of GaSe at 6.5 K. Splitting is observed at higher fields for the $N=1$, 2 levels, as indicated by dashed lines. Experimental points from recent experiments are shown circled.

The earlier energy-level diagram was based upon experiments with crystals 10–20 microns thick. While consistent energy values were obtained for the higher N levels, it was difficult to locate the $N=0$ level because of overlapping crystal interference fringes, as seen in Fig. 2(a). Two recent shots were fired that lessened this problem. In the first, a crystal of only 1 micron thickness was used and the fringes were so far apart that no overlapping with this level occurred. The spectrogram constrast was poor and only the point at peak field, shown as a square, is plotted on Fig. 3. The circled data points were obtained from the second experiment, whose spectrogram is shown on Fig. 2(b). Here, the crystal was quite thick and the interference fringes were so close together as to appear continuous. However, light absorption was large and it was necessary to use an image intensifier to get useful records. The active film area covered by the intensifier was small. Since only a portion of the wavelength time region normally covered could be recorded, it was inconvenient to include the position of first light appearance on the film. Absolute film time was therefore established indirectly by making the magnetic field agree with the established value at one wavelength on the $N=2$ level. This point is shown doubly circled on Fig. 3. All other points were then assigned uniquely from the magnetic field-time record obtained from the probe and the film writing speed. The excellent fit with the rest of the $N=2$ level and the $N=1$ level shows that this procedure was satisfactory. The most important change from the earlier diagram brought about by these data was a general lowering of the $N=0$ level. It was previously placed some 40 wave numbers higher at 100 T, and almost 90 wave numbers at 160 T.

THEORY

This calculation will be presented within the effective-mass approximation. Since the basic Hamiltonian has been derived in some detail by several authors, these details will not be presented here.[5,7] We begin with the Schrödinger equation for the envelope function $F(r)$ for the exciton in the center-of-mass coordinate system. In this approximation, motion of the center-of-mass

frame has been ignored. The energy spectrum of the exciton is then found by solving for the eigenvalues of

$$H_{\text{c.m.}}F(r)=EF(r)\,. \tag{1}$$

In the following calculation all units of length and energy are measured with respect to an effective Bohr radius, a_0, and an effective Rydberg, R:

$$a_0=\hbar^2(\epsilon_\perp\epsilon_\parallel)^{1/2}/\mu_\perp e^2\ , \tag{2}$$

$$R=\hbar^2/2\mu_\perp a_0^2\,. \tag{3}$$

Here $\epsilon_\parallel$ and $\epsilon_\perp$ are the components of the dielectric-constant tensor parallel and perpendicular to the c axis. The components of the effective reduced-mass tensor $\mu_\parallel$ and $\mu_\perp$ are related in the normal way to the electron and hole masses, i.e., $1/\mu_\perp=1/m_\perp^e+1/m_\perp^h$. In a Cartesian coordinate system with the z axis parallel to the crystal c axis the Hamiltonian has the following simple form in the absence of an external field[7]:

$$H_0=p^2-2(x^2+y^2+\beta z^2)^{-1/2}\,. \tag{4}$$

Here β is a measure of the anisotropy of the crystal:

$$\beta=\epsilon_\perp\mu_\perp/\epsilon_\parallel\mu_\parallel\,. \tag{5}$$

In the presence of an external magnetic field an additional term is added to the Hamiltonian.[16] In the case of experimental interest here the field also points along the c axis of the crystal (Faraday geometry). Within a cylindrical gauge where we can write the vector potential $\underline{A}=(\underline{B}\times\underline{r})/2$, we then obtain

$$H_{\text{mag}}=\zeta\gamma L_z+\gamma^2(x^2+y^2)/4+g_z S_z\gamma/2\,. \tag{6}$$

Here L_z is the z component of the angular momentum operator while γ, a dimensionless measure of the strength of the magnetic field, μ_B^*, an effective Bohr magneton, and ζ are defined as follows:

$$\gamma=\mu_B^*B/R\ , \tag{7}$$

$$\mu_B^*=e\hbar/2\mu_\perp c\ , \tag{8}$$

$$\zeta=(m_\perp^h-m_\perp^e)/(m_\perp^h+m_\perp^e)\,. \tag{9}$$

The last term in Eq. (6) is present due to interaction of the electron and hole spins with the magnetic field. The spin component S_z has the value of zero for one of the triplet states and the singlet state, and ±1 for the other two triplet states.

Since the eigenvalues of the total Hamiltonian, $H=H_0+H_{\text{mag}}$, cannot be found in closed form, even in the absence of an external magnetic field, a variational approach is most often adopted. In this paper the adiabatic method as proposed by Elliott and Loudon[17] will be used to describe the high-field excitation spectrum. Since the calculation presented here differs somewhat from that of Elliott and Loudon, some of the details will be presented for clarity.

In the absence of a Coulomb interaction the excitation spectrum (Landau levels) can be obtained in closed form,

$$E_{Nm}(k_z)=\gamma(2N+1+|m|+\zeta m)+k_z^2\,, \tag{10}$$

where the eigenfunctions are[18]

$$\psi_{Nm}(k_z)=e^{ik_z z}X_{Nm}\ , \tag{11}$$

$$X_{Nm}=\frac{1}{(2\pi P_{Nm})^{1/2}}e^{im\phi}\rho^{|m|}e^{-(1/4)\gamma\rho^2}L_N^{|m|}(1/2\gamma\rho^2)\,. \tag{12}$$

Here $L_N^{|m|}(x)$ is the generalized Laguerre polynomial,

$$\rho=(x^2+y^2)^{1/2}\,, \tag{13}$$

$$\phi=\tan^{-1}(y/x)\,, \tag{14}$$

$$P_{Nm}=\frac{\pi(N+|m|)!}{N!\,(\gamma/2)^{|m|+1}}\,. \tag{15}$$

The principle Landau quantum number is represented by N while m is the angular momentum component along the z (magnetic field) axis. Each value of N and m results in a continuum (because of the continuous nature of k_z) extending above the minimum energy

$$\gamma(2N+1+|m|+\zeta m)\,. \tag{16}$$

The basis of the adiabatic approximation is that the presence of a Coulomb attraction will have very little effect on the orbital motion perpendicular to the magnetic field in the limit of high fields (the magnetic energy contributions to the Hamiltonian are much greater than the Coulomb term). The Coulomb attraction, however, will have a significant effect on the motion parallel to the magnetic field. Elliott and Loudon[17] approximate the wave function by a product form.

$$\psi_{Nm}(r)=G(z)X_{Nm}(x,y)\,, \tag{17}$$

where $X_{Nm}(x,y)$ defined in Eq. (12) is the free Landau solution. Movement parallel to the field is then described by motion in an effective adiabatic potential defined by

$$V_{Nm}(z)=\frac{-2(N!)}{(N+|m|)!}\int_0^\infty\frac{d\gamma\,\gamma^{|m|}e^{-\gamma}[L_N^{|m|}(\gamma)]^2}{(2\gamma/\gamma+\beta z^2)^{1/2}}\,, \tag{18}$$

where $G(z)$ then satisfies

$$\left(-\frac{d^2}{dz^2}+V_{Nm}(z)\right)G(z)=\epsilon' G(z)\,. \tag{19}$$

The energy of the state is then

$$E_{Nm}=\epsilon'+\gamma(2N+1+|m|+\zeta m). \tag{20}$$

Qualitatively the excitation spectrum will consist of a range of positive values of ϵ' representing a continuum extending above the minimum energy, Eq. (16), as in the free case. Due to the presence of the Coulomb interaction a discrete set of solutions with the negative ϵ' are pulled below the continuum edge.

Rather than approximate the form of the adiabatic potential, Eq. (18), calculation of the energy eigenvalues will follow the method Altarelli and Lipari[18] used for the isotropic case in which the wave function is expanded in an equally tempered Gaussian basis set,

$$G(z)=\sum_i A_i e^{-\alpha_i z^2}, \tag{21}$$

where

$$[\alpha_i \,|\, i=1,N(\alpha)].$$

Substitution of this expansion into the differential Eq. (19) produces a matrix equation easily solved by a digital computer. The α's are chosen to give accurate values for the first few energy states.

The adiabatic approximation is only good in the high-field limit. Lee, Larson, and Lax[19] in a variational calculation of the first few excited states of the isotropic hydrogen atom showed that this approximation is a reasonable one for the excited states when $\gamma>1.0$. For the ground state, however, the adiabatic approximation requires much higher fields to have sufficient accuracy. For this reason a more general expansion for the exciton wave function will be used for the ground state:

$$X_{gs}(r)=\sum_{i,j} A_{ij} e^{-\alpha_i \rho^2-\beta_j z^2}, \tag{22}$$

$$[\alpha_i \,|\, i=1, N(\alpha)],$$

$$[\beta_j \,|\, j=1, N(\beta)].$$

The basis set is taken equally tempered and the α's and β's chosen to give the lowest energy.

ACCURACY-CONVERGENCE

The convergence of the adiabatic approximation using the basis set in Eq. (21) was discussed for the isotropic case by Altarelli and Lipari.[18] For example, if we calculate the energy of the lowest $N=0$, $m=1$ state at a magnetic field of, say, $\gamma=20.0$ using 15 Gaussians in the wave-function expansion, Eq. (21) $[N(\alpha)=15]$, the calculated energy is accurate to within 0.01%. Fewer than 15 terms give far more than sufficient accuracy for the levels considered in this paper. How accurate the adiabatic approximation itself is at these fields is somewhat hard to estimate with confidence. However, since the fields considered here are of an order of 20 greater than in similar experiments used to determine the mass parameters to date, one expects to determine these parameters with a much higher reliability. For example, the binding energy ϵ' for $N=1$ at $\gamma=4.0$ is only 1.65 rydbergs or 14% of the Landau-level energy.

The ground-state-energy calculation, which uses the expansion of Eq. (22), will converge to the exact eigenvalues of the Hamiltonian at any magnetic field if a large enough basis set is used. In practice this means including about seven Gaussians in each direction to achieve an accuracy of better than 0.1%, or $N(\alpha)\simeq N(\beta)\simeq 7$. This accuracy is confirmed by comparison with several earlier calculations. In the absence of an external field, Faulkner and others[5] have used a variational approach to calculate the ground-state energy as a function of the anisotropy parameter β. The values obtained using the present expansion agree with these calculations to well within the quoted error and are in fact somewhat better since the energies are always slightly more bound (a variational calculation produces an upper bound to the exact eigenvalue). Energy eigenvalues for the isotropic hydrogen atom have been calculated by Lee, Larsen, and Lax[19] for fields up to $\gamma=1.0$. These values agree well with the energies using the present wave function with our eigenvalues being slightly more bound. The calculated eigenvalues go smoothly to the adiabatic values as the magnetic field is increased.

OPTICAL ABSORPTION

Selection rules for the absorption of light in a direct-gap semiconductor by the production of excitons is well known.[20] Since the transition probability is proportional to $[\psi(0)]^2$, only states with $m=0$ are allowed. Thus (neglecting the spin interaction) one expects a series of levels each corresponding to a different value of N with energy

$$\epsilon=\epsilon'+(2N+1)\gamma. \tag{23}$$

In this experiment only the lowest levels in each series are seen. Since the light used to probe the sample is unpolarized, one expects to find a splitting of the levels due to the g factor of the electron and hole. As noted earlier, such splitting has been observed at higher magnetic fields and is indicated on the experimental energy-level diagram of Fig. 3 by dashed lines. However, the degree of splitting could only be obtained qualitatively. Consequently, in our theoretical model

described below, we use the value $g_e = 2.7$ from Mooser and Schlüter[8] to calculate the level splitting.

At the high magnetic fields used in this experiment the binding energy ϵ' is a relatively small fraction of the total energy, at least for the excited states. Since ϵ' tends to be weakly dependent on the anisotropy parameter β, the mass $\mu_\perp$ can be determined independently of β. We find $\mu_\perp = 0.13 \pm 0.01$. This determination is also independent of the choice of the dielectric tensor used to describe GaSe, i.e., the values of $\epsilon_\perp$ and $\epsilon_\parallel$.

We have determined the value for the mass anisotropy from the accurately known ground-state energy measured in the absence of an externally applied magnetic field,[8] $E_g = 19.8 \pm 0.1$ meV. This value, however, is sensitive to the choice of dielectric function. With the measured values of Leung *et al.*,[21] $\epsilon_\perp = 10.2$, $\epsilon_\parallel = 7.6$, we obtain $R = 22.5$ meV and $E_g \simeq 0.88$ Ry. This implies a value of $\beta \simeq 1.45$, or using the anisotropy of the dielectric tensor, $\mu_\parallel/\mu_\perp = 0.9$. We have used these parameters in our model to calculate the energy-level diagram of Fig. 4. For comparison the "free-particle" Landau levels (where the Coulomb interaction potential and spin interactions are neglected) with energy $\gamma(2N+1)$—Eq. (16) with $m = 0$—are plotted along with the full model calculations. The recent experimental points shown circled on Fig. 3 are also plotted on Fig. 4 for comparison.

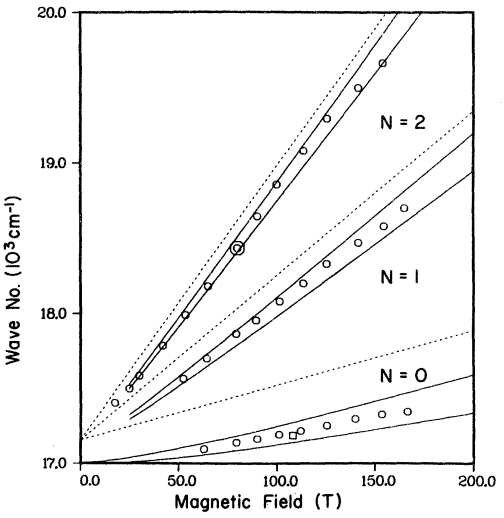

FIG. 4. Exciton spectrum of GaSe. The solid lines are the calculated values from the theoretical model. The dashed lines represent the corresponding Landau levels (see text). The circled experimental points of Fig. 3 are reproduced here for comparison.

Our value of the mass anisotropy compares well with the value of $\mu_\parallel/\mu_\perp = 1 \pm 0.2$ found by Mooser and Schlüter.[8] Their value was found by considering the low-field behavior of the ground-state energy (diamagnetic shift) in both the Voigt and Faraday geometries. This finding, however, is in contradiction with the fits of Fritsche and Heidt[16] who use $\mu_\perp = 0.14$ and $\mu_\parallel/\mu_\perp = 5.0$. This discrepancy may stem from the breakdown of the adiabatic approximation at lower fields. The highest fields considered by Fritsche and Heidt are of the order of 9 T, which is an order of twenty below the fields measured here. Using the fit obtained here, 9 T corresponds to $\gamma \simeq 0.2$. If we choose to use a different set of values for the dielectric constant, say the more recent values of Le Toullec *et al.*,[22] $\epsilon_\perp = 10.6$, $\epsilon_\parallel = 6.18$, we still obtain a value of $\mu_\perp \simeq 0.13$. However, since the rydberg is considerably larger, we find $\beta \simeq 2.1$ and $\mu_\parallel/\mu_\perp = 0.8$.

Other aspects of the experimental data perhaps deserve some qualitative comment: (a) The state labeled $N = 0'$ in Fig. 3 is barely resolvable from photographs such as in Fig. 2; however, it is reproducible and seems to smoothly evolve to the level at 17 230 wave numbers observed by Halpern. The energy for this state is weakly dependent on the magnetic field. This weak dependence may indicate the presence of an additional series of exciton transitions caused by stacking faults of the hexagonal layers and shifted from the one considered here by some 200 cm^{-1}. (b) The $N = 0, 1, 2$ levels all seem to show a consistent departure from linearity for magnetic fields ≥ 100 T, which could be indicative of field dependencies of the electron and hole masses. However, part of these departures could be due to the difficulty of correlating energy level and magnetic field values accurately in this region where, in the experiments, magnetic fields increase rapidly as a function of time.

CONCLUSION

The energies of the ground state and first few excited states of a free exciton in GaSe have been measured as a function of magnetic field up to nearly 200 T. The experimental results agree well with values calculated from the effective-mass approximation. The values obtained for the reduced effective mass and anisotropy are in good agreement with earlier values found by Mooser and Schlüter[8] who used data obtained only at small applied external fields.

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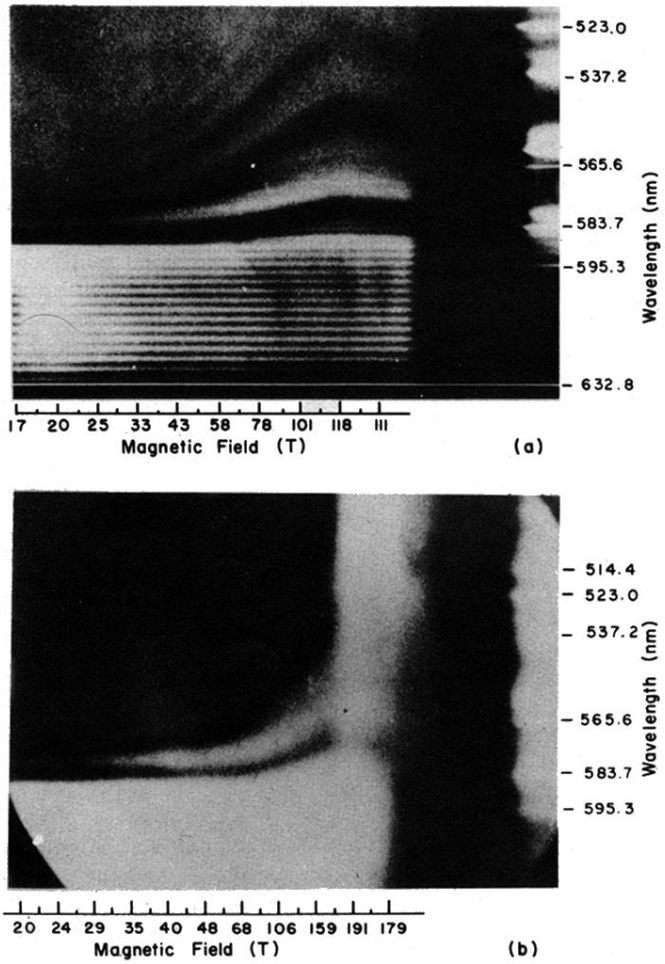

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