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VALIDITY OF THE CONSERVATION RULE OF NODAL SURFACE NUMBERS IN THE WAVE FUNCTION OF HYDROGENIC EXCITON IN MAGNETIC FIELD

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Validity of the Conservation Rule of Nodal Surface Numbers in the Wave Function of Hydrogenic Exciton in Magnetic Field

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Magnetoabsorption experiments on the A exciton in InSe have been performed in search of a unified correspondence relation between low and high magnetic field levels of a hydrogenlike state. Results show that the nodal-surface conservation rule of Shinada *et al.* is a more appropriate representation of this relationship than the so-called noncrossing rule.

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It has been recognized as an everlasting problem of both theoretical and experimental studies to solve for the energy eigenvalues of Wannier exciton in the intermediate magnetic field range, $\gamma \equiv \hbar\omega_c/2\mathcal{R}^* \cong 1$, where $\hbar\omega_c$ is the cyclotron energy of the exciton and $\mathcal{R}^*$ the effective Rydberg constant. For tracing the transition in characters of hydrogenic exciton levels with respect to the field, Kleiner[1] has proposed a topological concept that for any strength of the field the number of nodal surfaces of the wave function for a given level has to be kept constant. Elliott and Loudon[2] were the first to refer to this rule in their theoretical analysis of absorption spectrum in a high magnetic field of $\gamma > 2$. Subsequently, Shinada, Akimoto, Hasegawa, and Tanaka (SAHT)[3] have modified Kleiner's concept in such a way that the excitonic states with a common magnetic quantum number m and a parity would interact with each other in the intermediate field range if their total (polar and radial) nodal-surface numbers are equal, and derived a modification of Kleiner's correspondence relations between low- and high-field levels. Boyle and Howard (BH),[4] on the other hand, have proposed that the correspondence should be established only on the

basis of the noncrossing rule for such levels, thus proposing a new correspondence scheme.

There have been a number of works, especially of Wagner and McCombe,[5] on the magnetospectroscopy of shallow donors in various semiconductors in which they claim quantitative consistency with BH's correspondence relations. It should be pointed out, however, that BH's relations are identical with SAHT's for $1s$, $2p_{0,\pm1}$, $3p_{0,\pm1}$, $4p_{0,\pm1}$, etc. Thus no optical transition involving any of these levels can discriminate SAHT's relations from BH's. The magnetospectroscopy of a donor is inherently not an appropriate probe in search of the level correspondence between low- and high-field levels.

An essential spectroscopic difference between SAHT's relations and BH's is found in the behavior of nd_0 levels. According to SAHT, a level which originates from a d_0 should cross over the $N=0$ Landau edge to track with the $N=1$ edge in a high-field limit: The $3d_0$ level, for example, forms a hybridization with the $2s$ level so that the upper branch connects to the (100^+) state, where the notation of the high-field state implies $(Nm\nu^\pm)$ of SAHT. According to BH's relations, on the other hand, all the $m \leq 0$ bound states should lie below the $N=0$ Landau edge at any field strength; the (100^+) state develops out of continuum states of the exciton as the field increases. It is clear that a magnetospectroscopy of an exciton is much more appropriate than that of an impurity for tracing such levels experimentally. It is for this reason that we report on the magnetoabsorption of the A exciton in InSe in fields up to 39.5 T. The results show that the most reasonable correspondence is given by SAHT.

One cannot be too careful on the choice of the substance with its electronic band structure as simple as possible. According to Lipari and Altarelli,[6] the diamagnetic shift of the ground state in the group IV, III-V, and II-VI compounds cannot be described in terms of a simple hydrogenic exciton model, but exhibits a complicated behavior due to the cubic anisotropy, the k-linear term, and the intervalence band couplings. Tl-halide compounds have been examined by Kobayashi[7] up to the field of 15 T ($\gamma \cong 0.25$). Only the $1s$ and $2s$ lines in TlCl show the field dependence satisfying the noncrossing rule. But there are several levels which obviously cross over the Landau edges (see Fig. 5 of Ref. 7). The prescence of a strong exciton-phonon coupling as well as a comparatively large exciton reduced mass makes further detailed investigation difficult. A typical layer semiconductor, GaSe, has been a material of great interest for both theoretical[3] and experimental[8,9] analyses. Our recent magneto-optical absorption experiments[8] of the B exciton in GaSe up to a field of 38 T ($\gamma \cong 0.7$) clearly shows that the line originating from the $2s$ level crosses over the $N=0$ Landau edge at $\gamma = 0.65$. Furthermore, Miura *et al.*[9] have measured the absorption spectra in the extremely high-field range between 50 and 170 T. In fields above 60 T, they have found an unidentified line below the $N=1$ Landau edge which seems to extrapolate to our $2s$-like line toward lower field. Under these circumstances, the experiment on the details of level connections around $\gamma = 1$ is highly desirable as has been pointed out by Miura *et al.*

To the best of our knowledge, the only case which barely satisfies all of the physical and technical requirements as mentioned above is the A exciton in $3R$-InSe. The second valence-band edge, A, is well decoupled from the B and C band edges since the crystalline field anisotropy is as large as -1.33 eV in the quasicubic model and the spin-orbit coupling constant of the Se atom is 0.47 eV, both of which are much larger than $\mathcal{R}^* = 15$ meV.[10] Although the reduced-mass parameter is somewhat anisotropic, the anisotropy parameter for the exciton, $\epsilon_\perp \mu_\perp / \epsilon_\parallel \mu_\parallel$, is very close to unity.[11] The above value of $\mathcal{R}^*$ together with $\mu_\perp \cong 0.12 m_0$ (Ref. 11) gives $\gamma = 1$ for a field of ~30 T which may be reached by our pulsed method without considerable difficulty.

A single crystal of $3R$-InSe has been grown by the Bridgman method. The sample is cleaved to a thickness of 1–2 μm. Measurements have been performed by the same method as described in Ref. 8.

Two typical sets of transmission spectra measured at 4.2 K in the Faraday configuration with magnetic field parallel to the c axis are shown in Figs. 1(a) and 1(b). The $1s$ line which lies at 2.5475 ± 0.0005 eV at zero field exhibits a Zeeman splitting as recognized by the difference between right-circular polarization and left-circular polarization curves. The center of the splitting shows a diamagnetic shift. The second weak absorption, though unobservable in the zero field, appears already in the field of 7.3 T at 2.564 ± 0.001 eV as shown in Fig. 1(b), and becomes stronger toward higher field. The third absorption at higher photon energy becomes observable at the field of ~12 T near 2.59 eV. A significant Zeeman splitting is also observable for the sec-

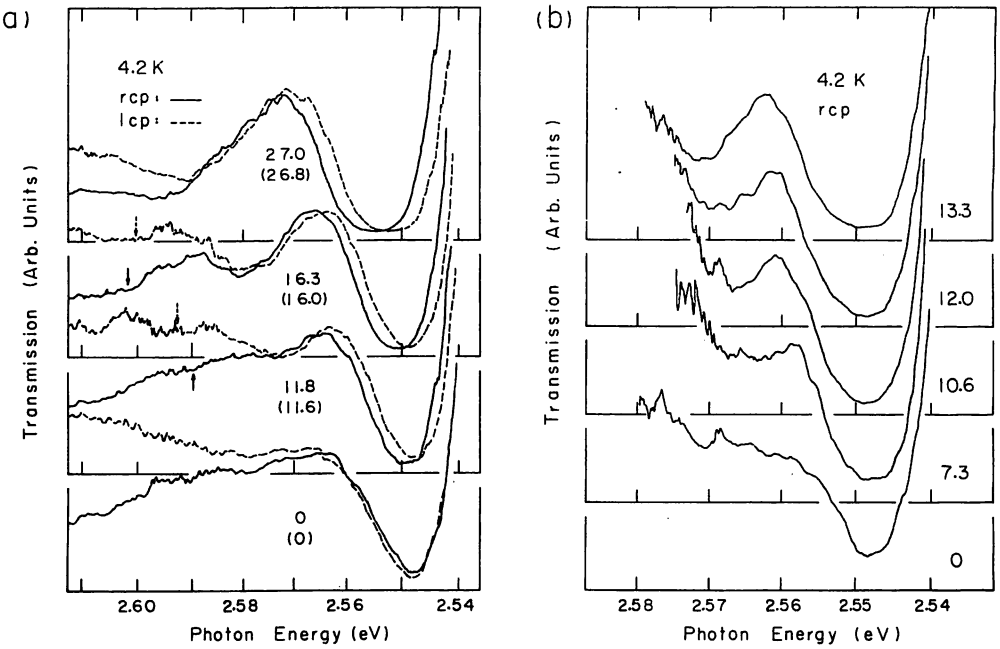

FIG. 1. Typical transmission spectra of InSe at 4.2 K in the Faraday geometry with external magnetic field parallel to the c axis of the hexagonal unit cell. The third absorption bands quoted in the text are shown by arrows. The magnetic field in teslas for right-circular polarization (rcp: solid line) is given by the numbers, and that for left-circular polarization (lcp: dashed line) by those in parentheses. The sample thickness is (a) (No. 3) 1.25 μm and (b) (No. 4) 1.70 μm. The ordinate is the ratio of the transmitted to the incident light intensity in arbitrary units.

ond line. The photon energies for these absorption peaks are plotted as a function of magnetic field in Fig. 2.

The ground state of the A exciton consists of parallel and antiparallel configurations of electron and hole spins. In the Faraday geometry, only the exciton with antiparallel spins may be optically created.[12] Their energy eigenvalues, $E_{1,2}$, in a magnetic field, H, parallel to the c axis are expressed[13] as follows:

$$E_{1,2}=E_0+D+\Delta_{ex}+(\tfrac{1}{2})\Delta_{LT} \pm(\tfrac{1}{2})[\Delta_{LT}^2+(g_A-g_e)^2\mu_B^2H^2]^{1/2}, \quad (1)$$

where E_0 is the ground-state energy at zero field, D the diamagnetic energy, Δ_{ex} the exchange energy, Δ_{LT} the longitudinal to transverse splitting of the exciton, g_A the effective g factor of the A valence band in the direction of the c axis, g_e that of the conduction band, and μ_B the Bohr magneton. The Lyddane-Sachs-Teller relation applied to the A exciton of InSe gives $\Delta_{LT}=0.3$ meV. It follows from Eq. (1) that the Zeeman splitting of 3.8 ± 0.4 meV at 30 T deduced from Fig. 2 for the $1s$ line gives $g_A-g_e=2.2\pm0.2$.

In order to estimate the value of D, we make a fit of the theoretical results of Cabib, Fabri, and Fiorio (CFF)[14] to our experimental points with $g_A-g_e=2.2$ obtained above. The best fit is obtained with $\mu_\perp=0.102m_0$ as shown in Fig. 2 by dash-dotted lines. Then the deduced value of $\mu_\perp$ allows us to scale the magnetic field in terms of γ, and thereby to compare the observed energies of the second and third absorption lines with theoretical values. The magneto-optical dispersion curves derived by Tanaka and Shinada (TS)[15] and by CFF are shown by solid and dash-dotted lines, respectively, in Fig. 2.

We notice that our second and third absorption lines are in excellent agreement with the (100^+) and (200^+) curves, respectively, in magnetic fields above 12 T. According to TS, the adiabatic potential employed in their calculation gives inaccurate solutions in the low-field range. They argue that the (100^+) line should extrapolate to the $2s$ level at zero field and (001^+) to $3d_0$, if the potential is adequately modified and no interaction between these levels takes place. Because of the presence, however, of the interaction as proposed by SAHT and supported by the exact calculation of CFF, these two levels admix together and repel each other. Consequently, the first excited state, (001^+), which corresponds to the lower hybrid branch would change its character from $2s$-like to $3d_0$-like as the field

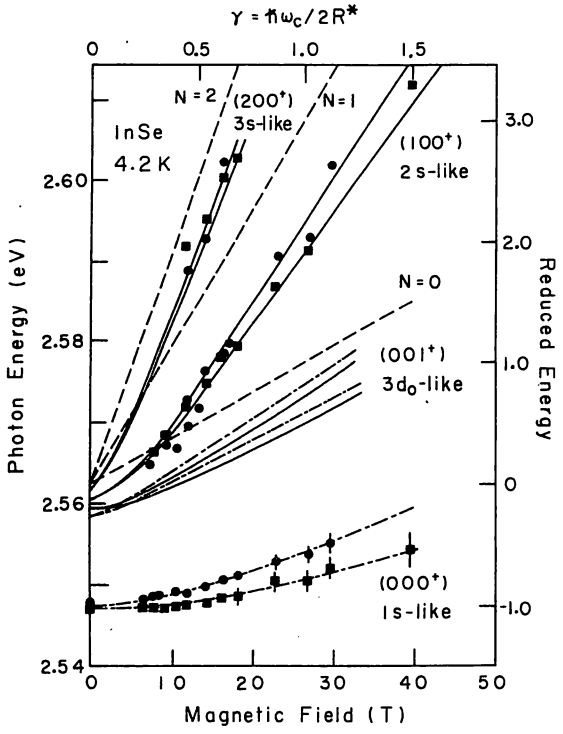

FIG. 2. Magnetic field dependences of energies of absorption peaks measured with rcp (circles) and lcp (squares) light. Theoretical curves of the lowest three Landau edges and excitonic levels calculated by Tanaka and Shinada and those by Cabib *et al.* are shown by dashed, solid, and dash-dotted lines, respectively. The reduced energy in the ordinate is scaled in units of $\hbar\omega_c^*$ with its origin at the direct energy gap in the zero field.

increases from $\gamma = 0$ through $\gamma \cong 0.3$. Hence its oscillator strength remains as weak as 11% to 18% of the $1s$-like (000^+) line at any field strength,[14] and thereby its observation is extremely difficult. On the other hand, the upper branch, (100^+), has the $3d_0$ character at zero field, but would be transformed to the $2s$-like as a result of the interaction, so that its oscillator strength increases almost linearly with magnetic field to exceed that of the (001^+) line above $\gamma \cong 0.5$.[15] Such a field dependence as expected for the $2s$-like state is in good agreement with our experimental result on the second absorption band. Furthermore experimental points along the (100^+) line do not exhibit any evidence of repulsion by the $N = 0$ Landau edge, but rather intersect it near the field of 10 T, thus disproving BH's correspondence relation. In the light of this result, the unidentified line observed by Miura *et al.*[9] below the $N = 1$ Landau edge in GaSe may be attributed to the (100^+) level which connects to the $2s$-like line observed by the present authors.[8] If one applies the same argument to the third absorption band, it is assigned to the $3s$-like (200^+) state after experiencing the interaction with $4d_0$ and $5g_0$ levels.[3]

From the above results, one can conclude that the level connection of a hydrogenic exciton upon the Rydberg-to-Landau transition satisfies the nodal-surface-number conservation rule, although the detail of interactions between excitonic levels is not clear yet. Besides the works of TS and CFF, numerical calculations of energy levels have been reported by a number of groups.[4,16-18] Apart from Ref. 16, their results show obvious violations of the noncrossing rule [see the (210) level in Ref. 4, the $3d_0$ level in Ref. 17, and the (210) and ($1\bar{1}0$) levels in Ref. 18], but no contradiction with SAHT's rule. Furthermore SAHT's rule can explain all of the experimental results cited[4,5,7,8,9,18] in this paper.

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