Terahertz Zeeman spectroscopy of boron in germanium to high magnetic fields

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The fundamental absorption lines of substitutional boron acceptor impurity in germanium, which lie between 1 and 3 THz, now have been measured in magnetic fields to 18 T, greatly extending the reach of both previous experiments (to 7 T) and theory (to 10 T). The Faraday configuration was employed with the magnetic field $\mathbf{B}\|\langle 110\rangle$. Unexpected behavior has been observed relating to the magnetic-field-induced splitting of the ground and first two excited states (all of which are fourfold degenerate): (a) One pair of Zeeman ground states splits at only half the rate predicted with field; this behavior continues to high field. The other pair shows a rapid increase with field. (b) The two pairs of Zeeman states emerging from the first excited state initially separate with field, then, above 10 T, converge, almost meeting by 18 T. (c) One pair of the Zeeman states from the second excited state begins to plateau at high field; the other shows a dramatic decrease above 9 T, extrapolated to become zero at around 24 T. Taken together, these results suggest modification to the existing theory is required, and may have implications for quantum computation involving substitutional impurities.

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I. INTRODUCTION

Since the introduction of the quantum computer based on P donor in Si by Kane and co-workers,[1,2] the magnetic-field dependence of energy states associated with substitutional impurities in semiconductors has been of renewed interest. Cole *et al.* have discussed the use of terahertz radiation to manipulate qubits based on donors in GaAs.[3] Even more recently, Haendel *et al.* have described the anisotropic Zeeman splitting of confined acceptors in Si/Ge.[4] In related work, the hole spin splitting in quantum wires[5] and the Zeeman splitting of hole states in GaAs (Ref. 6) and InP (Ref. 7) have recently been reported. In this paper we investigate to unprecedented high magnetic fields the terahertz absorption spectra of a classic system for which detailed calculations are available, B acceptor in Ge. We conclude current theory is inadequate to explain all observations.

The theory of the energy states of acceptors in semiconductors has developed over many years. Lin-Chung and Wallis first gave the intensities of Zeeman components and estimated the g factors.[8] Bhattacharjee and Rodriguez derived the selection rules from group theory and also calculated g factors.[9] Lipari and Baldereschi[10–12] devised a technique to calculate the unperturbed energy states of group III impurities in semiconductors. Two groups applied this technique to the computation and classification of the Zeeman states. Broeckx[13] treated the cases of the $\langle 100\rangle$ and $\langle 111\rangle$ crystallographic directions and included states in the split-off $p_{1/2}$ valence band but only considered fields to 5 T. In contrast, Schmitt *et al.*[14] included the $\langle 110\rangle$ case and extended the calculations to slightly more than 10 T, but did not include any of the $p_{1/2}$ states. Both these calculations included interactions between a number of Zeeman states of the same symmetry. The results obtained by Schmitt *et al.*[14] for the $\langle 110\rangle$ orientation of the magnetic field are shown in Fig. 1. The notation used to label the irreducible representations of the acceptor states follows Koster *et al.*[15] Figure 1 serves as a basis on which we discuss our experimental results in Sec. III.

The experiments have marched to higher magnetic fields over time. Optical measurements have proceeded from 1.6 T,[16] to 2 T,[17,18] 2.5 T,[19] 5.6 T,[20] and 7 T.[21–26] Piezo-Zeeman spectroscopy to 7 T has been reported.[27] Optical studies have now been made of the five acceptors B, Al, Ga, In, and Tl to 7 T for magnetic field $\mathbf{B}\|\langle 100\rangle$ and $\langle 111\rangle$. The splitting of the ground states is small but measurably different for all five acceptors; see Ref. 25 for a full comparison.

In comparing the latest experimental and theoretical data, reasonable agreement is found to 7 T. The predicted behavior of the final Zeeman states of the $1\Gamma_8^-$ state is not in as close agreement as that predicted for the $2\Gamma_8^-$ state. The behavior of the Zeeman states of the ground state, $1\Gamma_8^+$, is poorly described. For $\mathbf{B}\|\langle 100\rangle$, the theory[14] predicts the reverse order from that observed of the splitting of the $1\Gamma_7^+$ and $1\Gamma_8^+$ Zeeman states.[21–25] This is also the case for the $1\Gamma_{3.2}^+$ and $1\Gamma_{4.2}^+$ states with $\mathbf{B}\|\langle 110\rangle$.[26]

The present data extend the experiments by a factor of more than 2.5× in magnetic field, and the theory by a factor of almost 2×, but do we expect any new physics? To answer this question we introduce a dimensionless unit for the magnetic field, the ratio of the magnetic energy $\mu_B B$ (μ_B is the Bohr magneton) to the Rydberg energy Ry. The ratio is 1 at the magnetic field

$$B_0 = \frac{\mathrm{Ry}}{\mu_B} = 235{,}052\ \mathrm{T}. \tag{1}$$

(Equivalently, the magnetic length $\sqrt{\hbar/eB}$ is equal to the Bohr radius, a_0.) In the effective mass approximation we scale using the first Luttinger parameter γ_1 and the dielectric constant ϵ to obtain an effective Rydberg $\mathrm{Ry}^* = \mathrm{Ry}/\gamma_1\epsilon^2$ and effective Bohr magneton $\mu_B^* = \gamma_1\mu_B$. Hence

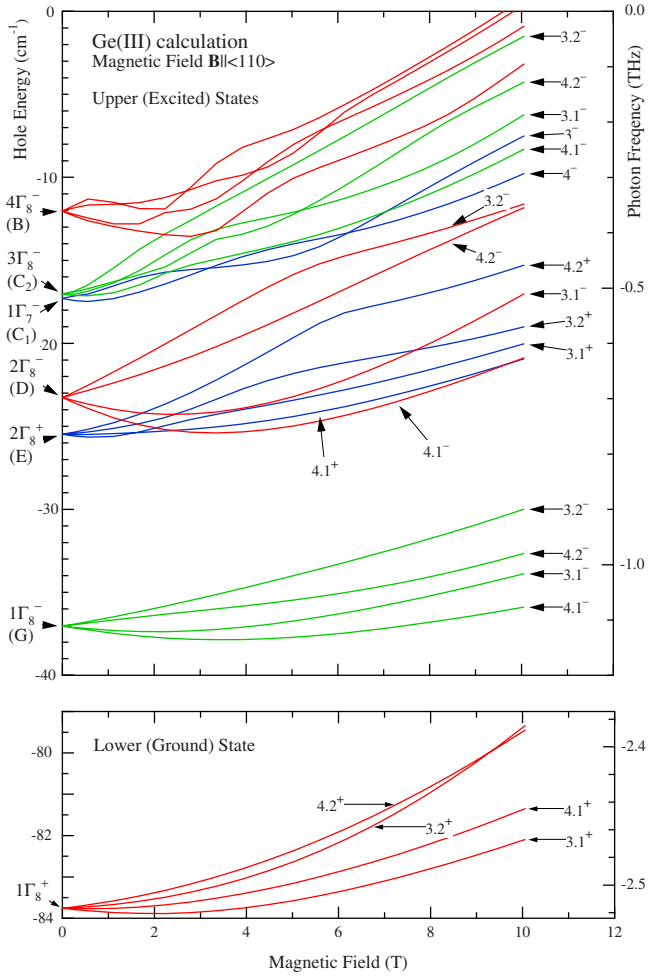

FIG. 1. (Color online) Theoretical Zeeman states (Ref. 14) for a group III acceptor in Ge for magnetic field $\mathbf{B}\|\langle 110\rangle$. The unperturbed states, in order of increasing hole energy, are labeled $1\Gamma_8^+$ (the ground state) then $1\Gamma_8^-$, $2\Gamma_8^+$, $2\Gamma_8^-$,… (the excited states). Transitions from the ground to the excited states are labeled in order G, E, D,…. The ground state and the lowest three excited states are all fourfold degenerate.

$$B_0^* = \frac{1}{\gamma_1^2 \epsilon^2} \frac{\mathrm{Ry}}{\mu_B}. \quad (2)$$

Using the materials parameters for Ge,[14] we find $B_0^*=6$ T. Fields below B_0^* may be termed "low" and fields above B_0^* "high." The highest fields used in optical experiments to date, 7 T, only just break into the "high-field" regime. The data reported here are the only measurements deep into the high-field regime for any group III acceptor in Ge.

II. EXPERIMENTAL PROCEDURE

Two samples were cut from a Ge crystal of B concentration $\sim 2\times 10^{14}$ cm^{-3}. After wedging (to remove interference fringes) and polishing they were treated ultrasonically to remove internal stress produced by the surface damage.[28] The samples were held at $\sim$5 K by He gas from a reservoir or an exchange gas. A 7 T split-coil Oxford Instruments superconducting magnet interfaced to a modified Polytec Fourier transform infrared (FTIR) spectrometer with unapodized resolution of 0.037 cm^{-1} and an 18 T superconducting solenoid coupled with a Bruker FTIR spectrometer with unapodized resolution of 0.18 cm^{-1} were used. The magnetic field **B** was parallel to $\langle 110\rangle$. The Faraday configuration was used and so $\mathbf{E}\perp\mathbf{B}$, where **E** is the electric vector of the radiation.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Unperturbed spectrum

A low-temperature, high-resolution unperturbed spectrum was acquired with a Bomem FTIR spectrometer at a resolution of 0.015 cm^{-1} with the sample immersed in liquid He pumped down to $\sim$1.6 K. This is given in Fig. 2.

As well as B lines, lines appear due to the acceptors Al and Ga,[29,30] and due to the donor P. These do not affect the measurement, as the Al and Ga are of much smaller concen-

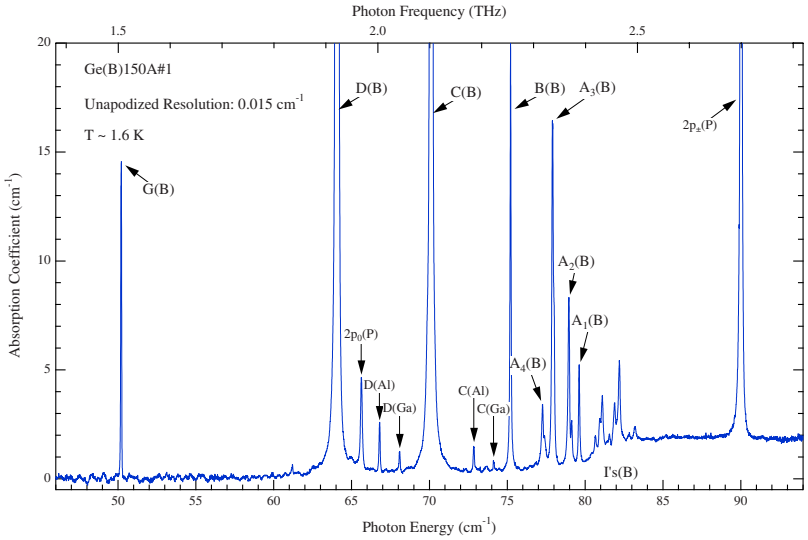

FIG. 2. (Color online) Spectrum of Ge(B). The sample also contains the unintentional dopants Al, Ga, and P, which contribute additional absorption lines.

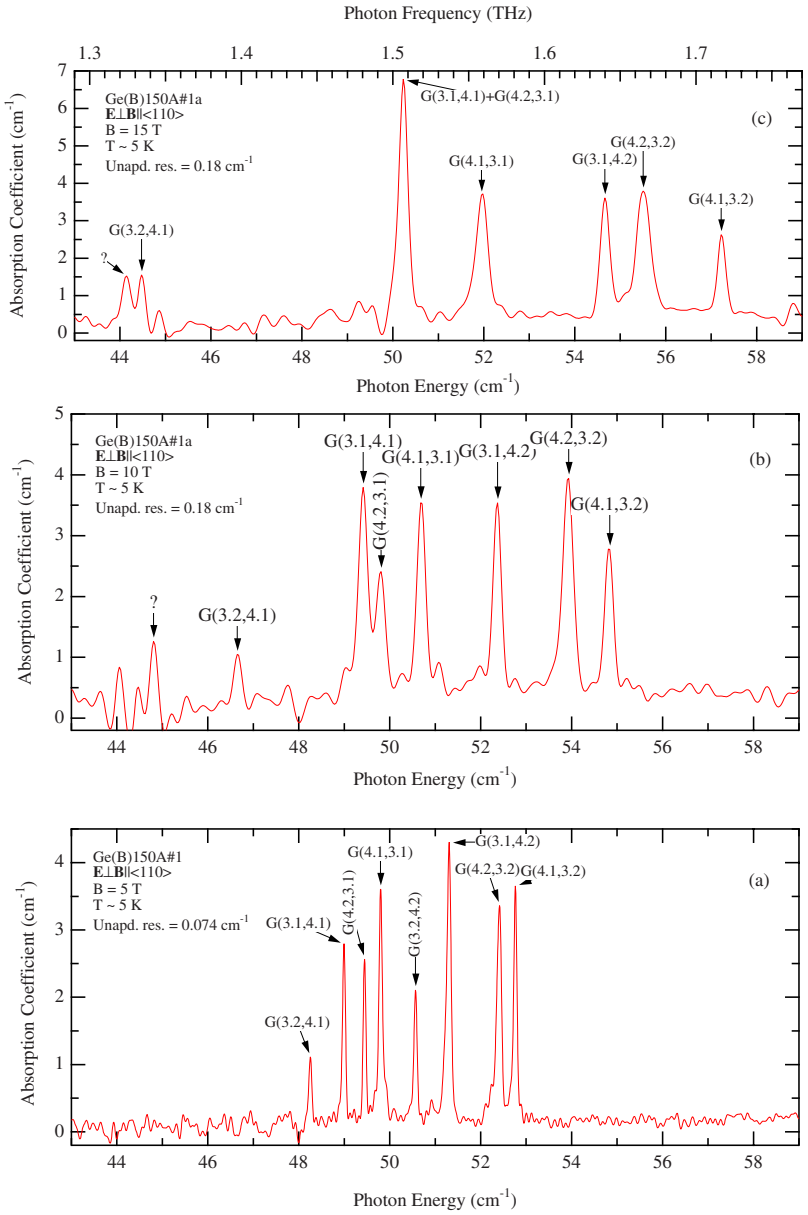

FIG. 3. (Color online) Zeeman spectra of Ge(B) for the transition $1\Gamma_8^+ \rightarrow 1\Gamma_8^-$ (G line) with $\mathbf{B}\|\langle 110\rangle$ for 5, 10, and 15 T.

tration than the B and the P lines are well separated from the B ones.

B. Zeeman spectra

1. The G line and the $1\Gamma_8^-$ and $1\Gamma_8^+$ states

The Zeeman spectra for $\mathbf{B}\|\langle 110\rangle$ of the G line at fields of 5, 10, and 15 T are shown in Figs. 3(a)–3(c), respectively. The labeling of the Zeeman components indicates the initial and final states of the transitions involved. The electric dipole selection rules for the transitions $1\Gamma_i^+ \rightarrow n\Gamma_f^-$, i.e., (i,f), where $i,f=3,4$ are $i \neq f$. Eight components are allowed, all of which are observed. The spectra obtained are shown in Fig. 4. The dependence of the energies of G components on field strength is shown in Fig. 5. The full curves in this figure are polynomial fits to the data. A comparison of the behavior of the G components in Fig. 5 with the data obtained for Al in Ge (Ref. 26) with $\mathbf{B}\|\langle 110\rangle$ and $\mathbf{E}\perp\mathbf{B}$ in the range 0–6 T unambiguously identifies the origin of the transitions.

A comparison between the theoretical and experimental results for the final state of the G line, $1\Gamma_8^-$, can be obtained from the splittings of the $1\Gamma_{3,1}^-(+3/2)$ and $1\Gamma_{3,2}^-(-1/2)$ states and $1\Gamma_{4,1}^-(+1/2)$ and $1\Gamma_{4,2}^-(-3/2)$ states; this is shown in Fig. 6. Since the differences are used, the comparison is independent of the behavior of the ground state, $1\Gamma_8^+$. Figure 6 includes the calculated results obtained for the two splittings displayed. For the $4.2^-/4.1^-$ splitting the agreement is very good for fields up to ~7 T, while beyond that the trend of the calculated results follows the experimental behavior. For the $3.2^-/3.1^-$ splitting, the calculated result has the same qualitative behavior as the experimental result but at all fields is smaller. Spectacularly, beyond ~7 T, the splittings

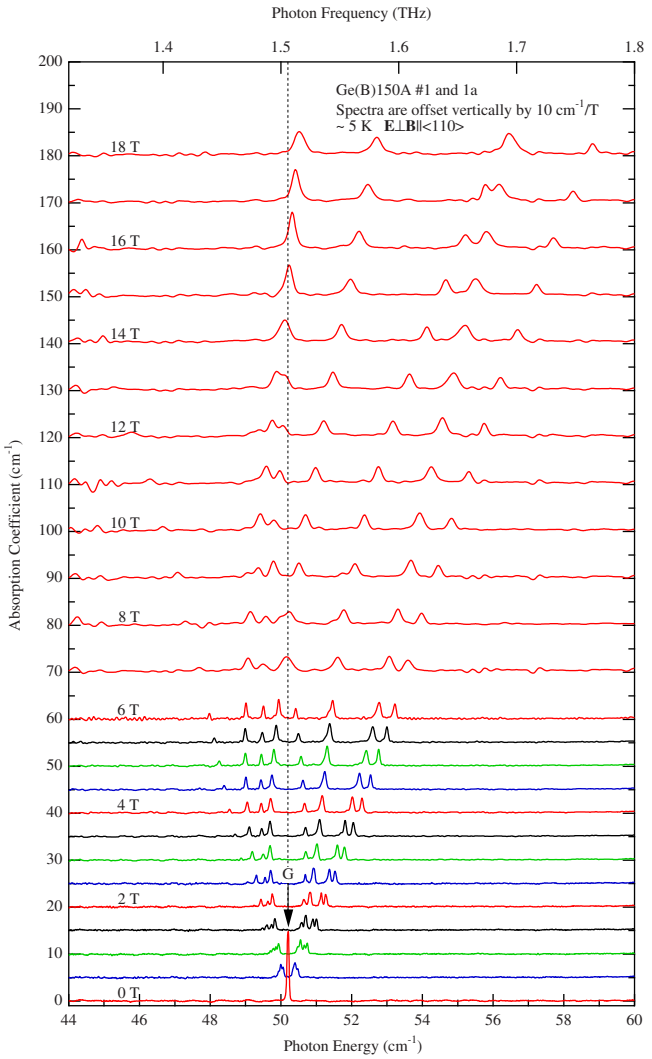

FIG. 4. (Color online) Zeeman spectra of Ge(B) for the transition $1\Gamma_8^+ \rightarrow 1\Gamma_8^-$ (G line) with $\mathbf{B}\|\langle 110\rangle$ from 0 to 18 T.

move toward each other, and by the time the highest field is reached are almost equal.

Figure 7 gives the corresponding splittings of the ground state, $1\Gamma_8^+$; the Zeeman states are $1\Gamma_{3.1}^+(+1/2)$, $1\Gamma_{3.2}^+(-3/2)$, $1\Gamma_{4.1}^+(+3/2)$, and $1\Gamma_{4.2}^+(-1/2)$. Also shown are the calculated splittings.[14] The theoretical splittings are in qualitative but not in good quantitative agreement with the experimental results. Most markedly, the observed $4.2^+/4.1^+$ splitting is much less than the theoretical prediction.

2. *The D line and the* $2\Gamma_8^-$ *and* $1\Gamma_8^+$ *states*

The Zeeman spectra at 5, 10, and 15 T of the D and C components are shown in Fig. 8. The labeling of the D components identifies the initial and final states of the eight allowed transitions. The labels on the C components are according to increasing energy with C_1 being of lowest energy. The set C_1–C_6 is shown clearly in Fig. 8(a). In view of the complexity of the final states of the C line,[31] the states involved in these transitions have not yet been identified.

The dependence of the energies of the D and some of the C components on field is given in Fig. 9(a) for the range

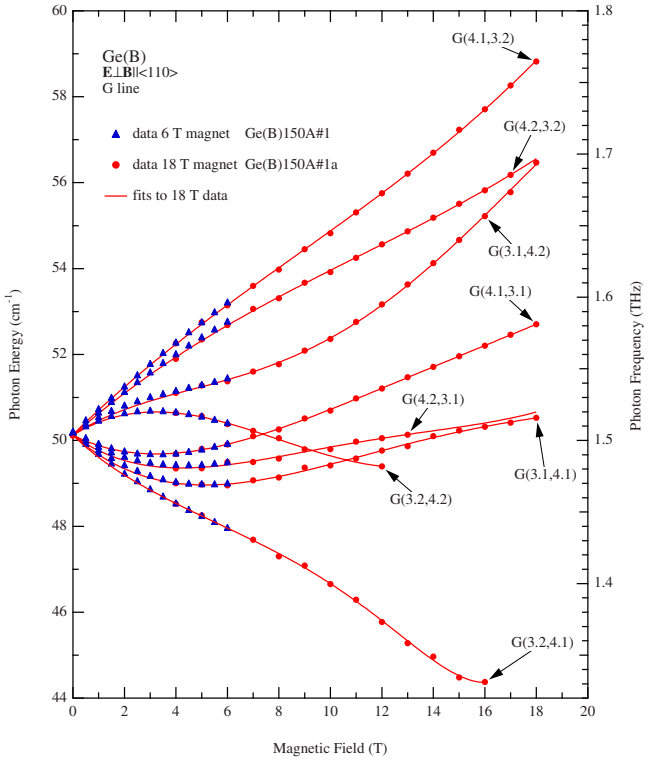

FIG. 5. (Color online) Fan chart for the transition $1\Gamma_8^+ \rightarrow 1\Gamma_8^-$ (G line) of Ge(B) with $\mathbf{B}\|\langle 110\rangle$.

0–6 T. The full lines are polynomial fits to the data points. A similar graph to 18 T is shown in Fig. 9(b). Beyond ~7 T, the D and C components become difficult to separate. Identification of the D components labeled D(4.1,3.2) and D(4.2,3.2) has been accomplished by comparing their spacing with the ground-state splitting of the 4.1^+ and 4.2^+ states as obtained from the data for the G line and lines are drawn between the data points. The other six D components have polynomial fits to the data points.

The splittings of the $2\Gamma_8^-$ and $1\Gamma_8^+$ states as obtained from the D components are given in Fig. 10. The polynomial pass-

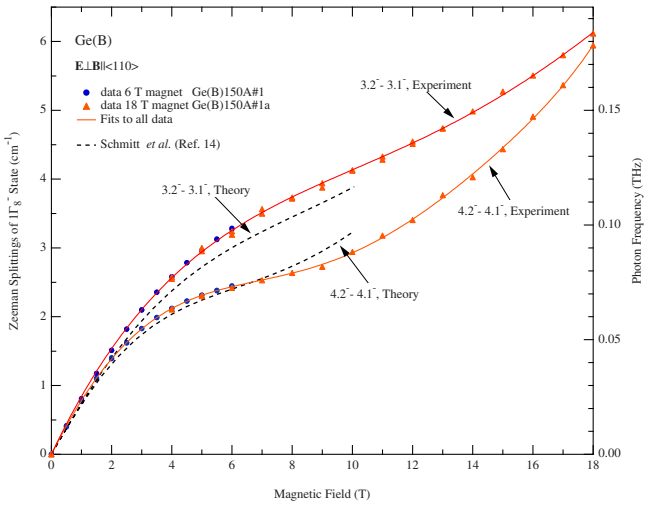

FIG. 6. (Color online) Excited-state splittings for the transition $1\Gamma_8^+ \rightarrow 1\Gamma_8^-$ (G line) of Ge(B) with $\mathbf{B}\|\langle 110\rangle$.

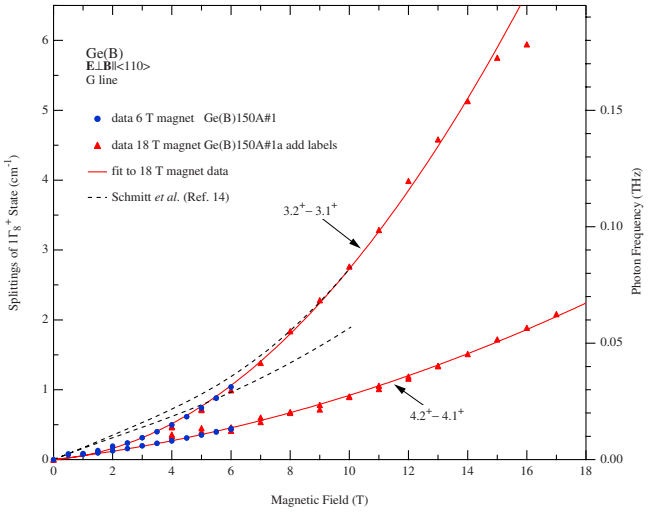

FIG. 7. (Color online) Ground-state splittings for the transition $1\Gamma_8^+ \rightarrow 1\Gamma_8^-$ (G line) of Ge(B) with $\mathbf{B}\|\langle 110\rangle$.

ing through the $3.2^+/3.1^+$ splitting of the Zeeman ground states is a fit to all the D data. The polynomial passing through the $4.2^+/4.1^+$ splitting of the Zeeman ground states is the fit obtained from the G data. The open circles are obtained from the differences in energy of the components D(4.1,3.2) and D(4.2,3.2), which are the most uncertain features in Fig. 9(b). A fit (not shown in Fig. 10) to this splitting using the D data is essentially identical to that obtained from the G data as given in Fig. 10. The fit obtained for the other ground-state splitting from the G data (see Fig. 7) is essentially identical to that obtained from the D data as shown in Fig. 10. No fits have been made to data for the excited-state splittings. Figure 10 also includes the splittings predicted by the calculations.[14]

The splittings of the two pairs of excited states are markedly different from the splittings of the two pairs of the ground states, and from the theory. The $4.2^-/4.1^-$ splitting plateaus at high field and approaches the $3.2^+/3.1^+$ ground-state splitting at the highest field. The $3.2^-/3.1^-$ splitting decreases rapidly beyond 9 T and approaches the $4.2^+/4.1^+$

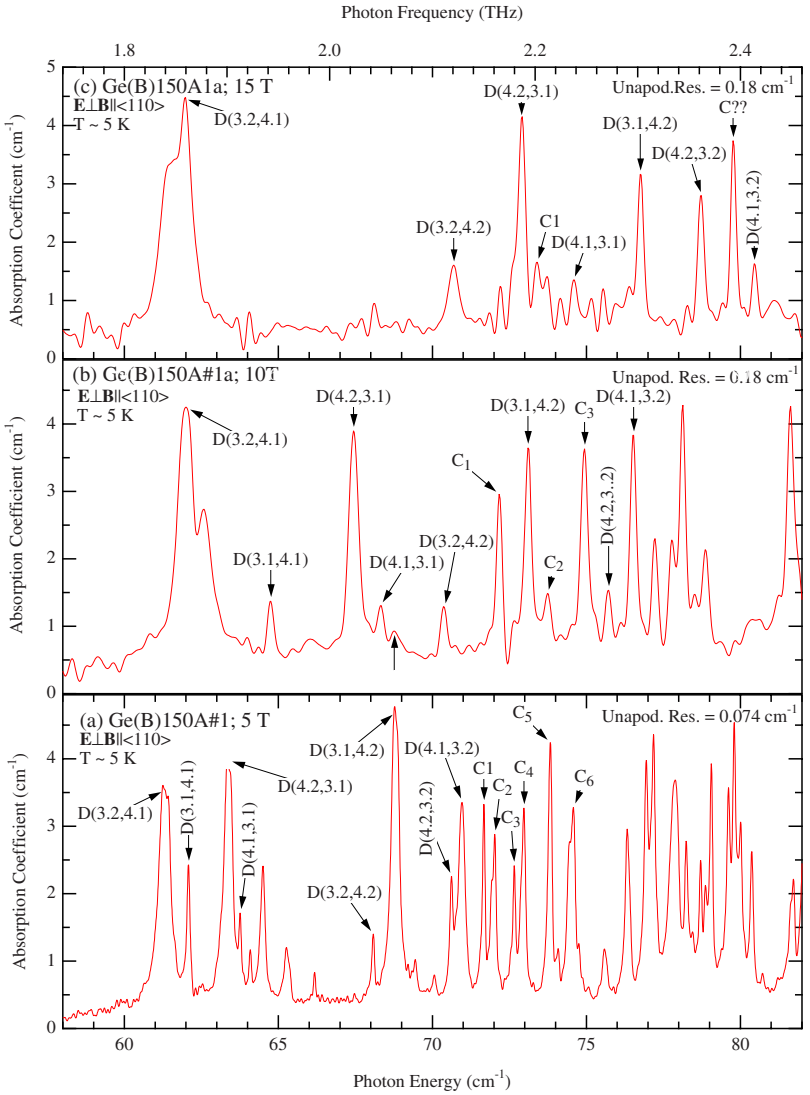

FIG. 8. (Color online) Zeeman spectra of Ge(B) for the transition $1\Gamma_8^+ \rightarrow 2\Gamma_8^-$ (D line) with $\mathbf{B}\|\langle 110\rangle$ for 5, 10, and 15 T. Some C line transitions are also shown.

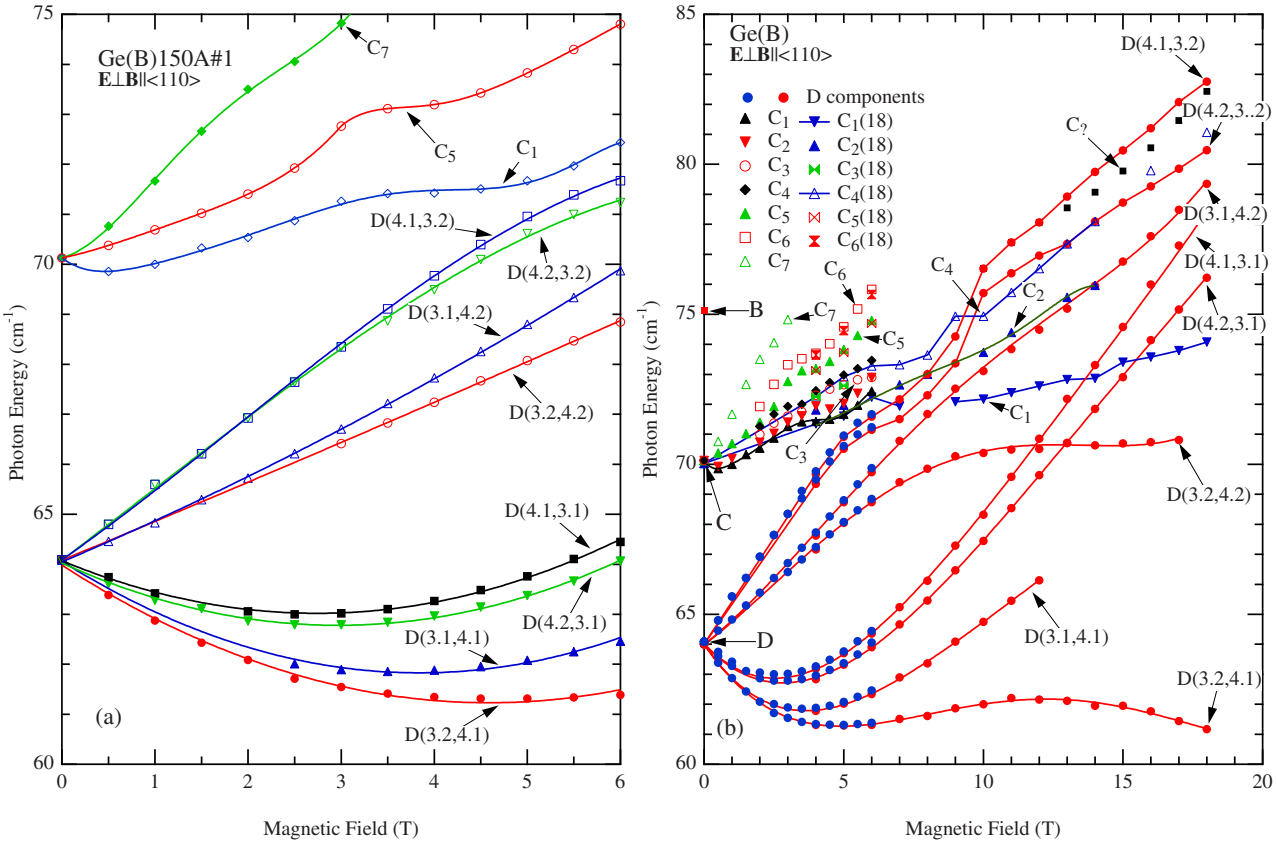

FIG. 9. (Color online) Fan chart for the transition $1\Gamma_8^+ \rightarrow 2\Gamma_8^-$ (D line) of Ge(B) with $\mathbf{B}\|\langle 110\rangle$. Some data are also given for the C line.

ground-state splitting at the highest field. It is extrapolated to become zero at 24 T.

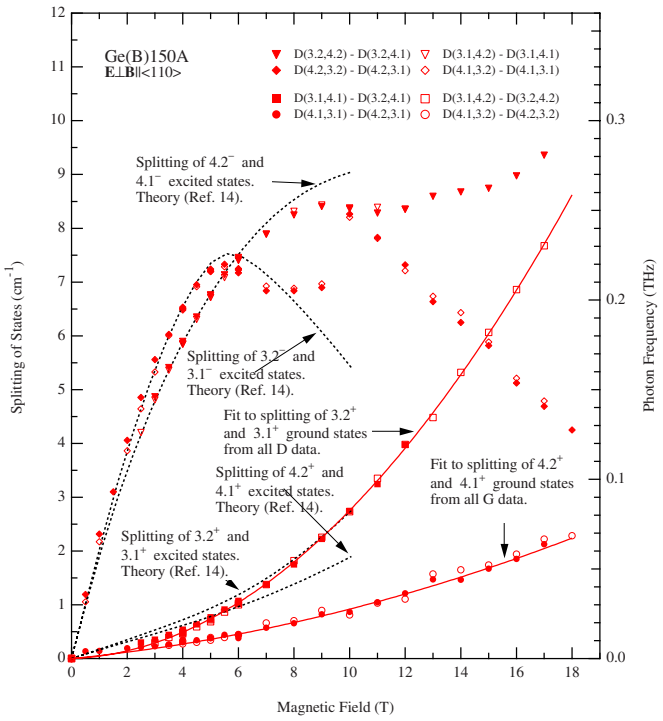

FIG. 10. (Color online) Ground- and excited-state splittings for the transition $1\Gamma_8^+ \rightarrow 2\Gamma_8^-$ (D line) of Ge(B) with $\mathbf{B}\|\langle 110\rangle$.

IV. CONCLUSIONS

The absorption spectrum of the Lyman series of B acceptor in Ge has been measured in the region 1–3 THz. Fields as high as 18 T have been used, approximately three times the crossover value to a high field when regarded in terms of the effective mass theory based on atomic hydrogen. High-precision measurements have allowed splittings of states of <0.01 THz to be resolved. Of the levels studied, the best agreement between the magnitudes of the Zeeman splittings as obtained experimentally and theoretically is for $1\Gamma_8^-$, the excited state of the D line, but this only to about 6 T. The poorest comparison is the splitting of the $1\Gamma_{4.1}^+$ and $1\Gamma_{4.2}^+$ Zeeman ground states. There is a very large splitting of the $1\Gamma_{3.1}^+$ and $1\Gamma_{3.2}^+$ states at high fields. These results are expected to spur further theoretical investigation of the general problem of an acceptor impurity in a semiconductor at a high magnetic field and to be of value in devising schemes for quantum computation.

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