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Energy states of Be in GaAs

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Far-infrared absorption due to Lyman transitions of the Be acceptor in GaAs has been measured in bulk molecular-beam-epitaxy-grown samples at various temperatures and under different illumination conditions. Sharp lines of energy (at 1.9 K) of 134.42 ± 0.06 , 166.76 ± 0.05 , and 182.30 ± 0.05 cm^{-1} are observed, being the *G*, *D*, and *C* lines, respectively. Fiorentini's recent calculation [Phys. Rev. B **51**, 10 161 (1995)] of the acceptor energy spectrum in GaAs is in good agreement with these results. At 1.9 K the *G*, *D*, and *C* linewidths are 1.06 ± 0.14 , 1.16 ± 0.04 , and 1.86 ± 0.05 cm^{-1} , respectively, and the integrated intensities are in the ratio $11.5 \pm 5.6:100:57.6 \pm 9.8$. External illumination during cooling or measurement had no effect on the observed spectra.

I. INTRODUCTION

The energies of shallow substitutional acceptor impurities in GaAs have recently been calculated by Fiorentini.¹ The calculation is within the effective-mass theory (EMT) and is based on the method, first proposed by Baldereschi and Lipari, of separating the acceptor Hamiltonian into a spherical² and a cubic term.³ The Baldereschi-Lipari method has since been refined and extended in a number of theoretical investigations.⁴⁻¹⁰ Fiorentini improves on earlier theory for GaAs (Ref. 11) by including in the calculation the cubic term, split-off band coupling, and dispersive impurity screening.

Practical difficulties have prevented the experimental spectra for acceptors in GaAs from being of as high a quality as might be desired for accurate comparison with theory. Early work¹² identified the difficulty in obtaining good absorption spectra, and resorted to the technique of photoconductivity to yield transition energies. While photoconductivity measurements are often more sensitive than absorption measurements, the photothermal ionization process involved requires a relatively high temperature (10–20 K) resulting in rather broad lines and is a complex mechanism to interpret, making it more difficult to obtain, for example, oscillator strengths. Raman spectroscopy has also been used to investigate acceptor spectra in GaAs.^{13,14} This technique is complementary to absorption spectroscopy in that it is sensitive to transitions from the ground state to even-parity excited states. Such excited states have a nonzero amplitude at the impurity site and so require an additional central-cell correction in the calculation. In the present study, far-infrared absorption spectroscopy is used. Absorption spectroscopy yields linewidths and intensities, in addition to energies, and, in detecting transitions to odd-parity final states, provides a direct test of the theory, without the additional complication of chemical shifts of the even-parity final states.

Previous far-infrared absorption measurements of acceptors in GaAs have concentrated on the chief contaminants in

liquid-encapsulated Czochralski- (LEC-) grown samples, C and Zn. Both these impurities are present in the spectra previously reported;^{13,15} the spectral lines from the two impurities overlap, confusing the analysis. In the present study, the Be acceptor alone is examined, permitting a more direct analysis of the spectral features and the determination of linewidths and line strengths. Only one previous set of absorption data for bulk GaAs:Be is known.¹⁶⁻¹⁹ The Be lines shown in the earlier spectrum are much broader and less intense than those observed in the present work.

II. EXPERIMENTAL DETAILS

The samples were grown by molecular-beam epitaxy (MBE) on 450- μm -thick (100) GaAs substrates with Be as the dopant. Details of the thickness of the doped layer and the Be concentration are given in Table I for each sample. The samples were wedged at $\approx 2^\circ$ to suppress optical interference between the front and back faces.

Measurements were made using a Fourier transform spectrometer equipped with a global light source and a He-cooled (4.2 K) Si bolometer detector. From source to detector the beam traversed two polypropylene windows (800 and 50 μm thick) and two Mylar windows (each 23 μm thick). A black polyethylene filter or metal shutter could be inserted into the beam before the sample; additional illumination of the sample could be provided through a side port.

The sample was cooled by exchange gas drawn from a He bath. Temperatures below 4.2 K were achieved by pumping

TABLE I. Be-doped GaAs samples used in this study.

Label	Layer thickness (μm)	Be concentration (atoms cm^{-3})	Be areal density (atoms cm^{-2})
NU650	1.5	3.6×10^{18}	5.4×10^{14}
NU651	4	1.5×10^{15}	6.0×10^{11}
NU652	3	2.3×10^{16}	6.9×10^{12}

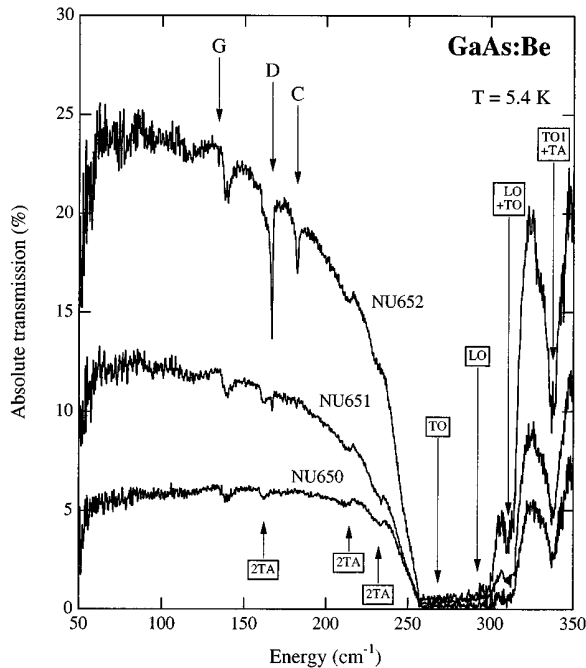


FIG. 1. Transmission of three bulk MBE-grown samples of GaAs:Be at $T = 5.4$ K. See Table I for sample details. The bound-hole to bound-hole transitions are labeled G , D , and C . Phonon absorption bands are identified by boxed labels.

on superfluid He; those above 4.2 K by employing a heater on the sample mount.

The calibration of the spectrometer was confirmed by comparing the measured energies of twelve water vapor lines in the range 120–240 cm^{-1} with reference values.²⁰ In no case did the measured value differ from the reference value by more than 0.049 cm^{-1} .

III. RESULTS AND DISCUSSION

Shown in Fig. 1 are infrared absorption spectra in the region 50–350 cm^{-1} for the three samples investigated. The most prominent feature of the top spectrum (sample NU652) is the sharp line at ≈ 167 cm^{-1} , the D line. The C line (≈ 182 cm^{-1}) and the G line (≈ 134 cm^{-1}) are also evident in this spectrum. The notation used here was first employed for acceptors in Ge (Ref. 21) and is common in the literature for acceptors in GaAs. The transitions are from the ground state, $1s_{3/2}(\Gamma_8^+)$ to the first three excited odd-parity states, $2p_{3/2}(\Gamma_8^-)$, $2p_{5/2}(\Gamma_8^-)$, and $2p_{5/2}(\Gamma_7^-)$, in the commonly used notation.¹² There is no sharp feature at 220 cm^{-1} , which has previously been reported as the location of the A line,^{16–19} nor any sharp transitions of higher energy. The spectrum for sample NU651 (middle spectrum) exhibits a weak D line; neither the C line nor the G line is observed. Sample NU650 (bottom spectrum) shows no electronic transitions.

There are several broad (≈ 5 cm^{-1}) features common to all three spectra in Fig. 1. These are increases in absorption at approximately 140, 162, 214, and 232 cm^{-1} . Given the proximity of the first three of these broad absorption bands to the G and D lines and the previously reported A line, it

might be thought that they are associated with these transitions of the Be impurity. Examination of the pure GaAs spectrum shows that three of these features originate in the GaAs lattice and are not related to the Be impurity. Koteles and Datars²² observe singularities in the absorption spectrum of GaAs at 161/165, 214/216, and 230/235 cm^{-1} , which they assign to two-phonon absorption; Wan and Bray¹⁴ utilize the two-phonon peaks at 162 and 227 cm^{-1} , which they label as 2TA. In the reststrahl region lie the TO (269 cm^{-1}) and LO (292 cm^{-1}) phonons.²³ Lattice absorption at 306 and 333 cm^{-1} is attributed by Cochran *et al.*²⁴ to the two-phonon combinations LO+TA and TO1+TA, respectively. The temperature dependence of all these features (see below) further confirms their origin in lattice, not electronic, excitations. These phonon assignments are indicated in Fig. 1. None of these authors notes a phonon mode near 140 cm^{-1} , where a prominent feature appears in the present spectra. This feature corresponds to a strong dip in the spectrometer instrument function, arising from absorption in the Mylar windows²⁵ and appears not to have been completely ratioed out in calculating the transmission.

The dielectric constant of GaAs in the limit of long wavelength and low temperature is $\epsilon_0 = 12.40$.²⁶ It follows that the reflection coefficient at normal incidence is $R = 0.3110$ and that for a thin transparent sample a maximum transmission of 53% would be realized. A transmission of this magnitude is observed, for example, in Fig. 1 of Ref. 15. In the present Fig. 1, at 120 cm^{-1} , samples NU652, NU651, and NU650 have absolute transmissions of 23%, 12%, and 6%, respectively. We are confident of these values for absolute transmission, even though they stand in the approximate ratios $\frac{1}{2}:\frac{1}{4}:\frac{1}{8}$ to that expected from pure GaAs. In particular, the transmission of sample NU652 has been confirmed on numerous separate experimental runs and mounts. Part of the increased absorption relative to pure GaAs may be due to the substrates on which the layers are grown, which are known to be absorbing in this region. The precise origin of the different transmission observed in the three samples is unknown.

The observed strength of the impurity absorption depends on the Be concentration of the sample (see Fig. 1 and Table I). Sample NU652 has a Be areal density $11.5\times$ that of sample NU651 and consequently exhibits deeper absorption lines. Metallic behavior is expected²⁷ if the Be concentration exceeds $(0.2/a_H)^3 = 1 \times 10^{18}$ cm^{-3} , where a_H is the Bohr radius of Be in GaAs. This is the case for sample NU650 and accounts for the absence of impurity absorption lines in its spectrum.

Only one previous set of absorption data for Be in GaAs is known.^{16–19} The full width at half maximum in transmission of those lines was ≈ 6 cm^{-1} . The present lines are much sharper, of width ≈ 1 cm^{-1} . The present lines are also sharper than those reported previously¹⁵ for C and Zn, which are ≈ 3 cm^{-1} wide.

It might be noted that the spectra of Fig. 1 show no evidence of transitions other than those related to the Be impurity. In particular, the excitation spectra of C and Zn occur in this energy range, and there is no evidence of C or Zn contamination in the present samples.

Samples NU651 and NU650 will not be considered further, and future spectra will be presented over the region in

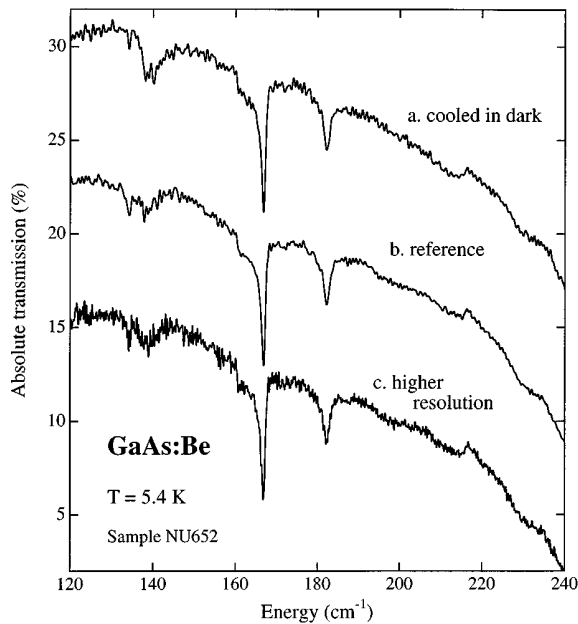


FIG. 2. Transmission of GaAs:Be under different experimental conditions. (a) Sample cooled in the dark, then measured with a black polyethelene filter in the beam. This spectrum is offset in the ordinate by adding 7.5% absolute transmission. (b) Reference spectrum under normal illumination and at resolution of 0.72 cm^{-1} . (c) Spectrum at higher resolution (0.36 cm^{-1}). This spectrum is offset in the ordinate by subtracting 7.5% absolute transmission.

which the Be acceptor transitions of interest occur, $120\text{--}240 \text{ cm}^{-1}$.

Additional illumination was found to have little effect on the spectrum of Be in GaAs. This is in accord with the observation of Reeder, Mercy, and McCombe¹⁷ that, while donor spectra in MBE-grown GaAs exhibit many photoinduced changes, no persistent photoeffects are evident in acceptor spectra. In contrast, in LEC-grown GaAs the significant role of sub-band-gap illumination in revealing the acceptor spectrum has been delineated by Wan and Bray^{14,28} and exploited in Raman and absorption studies.^{13,15,29} The use of band-gap radiation to enhance acceptor spectra in Ge and Si by decompensating acceptor sites and so increasing the concentration of acceptors available for optical transitions, as well as reducing local electric fields and so increasing the lifetimes of acceptor levels, is well known.^{30,31} These effects were examined in the present material.

Figure 2(a) gives the spectrum of the sample cooled in the dark, then exposed to radiation from the globar source only though a black polyethelene filter, which cuts off radiation of energy greater than $\approx 2400 \text{ cm}^{-1}$. Figure 2(b) gives the spectrum of the sample cooled in the light, that is, with the unfiltered radiation from the broadband globar source falling on it during cooling and measurement. It is clear that the spectra are very similar. As the observation of, for example, C and Zn impurities in LEC-grown GaAs depends critically on whether the sample is cooled in the light or in the dark, it is clear that the mechanism of optical quenching in that material, involving metastable *EL2* defect sites, plays no role in the MBE material being studied here. Spectra were also col-

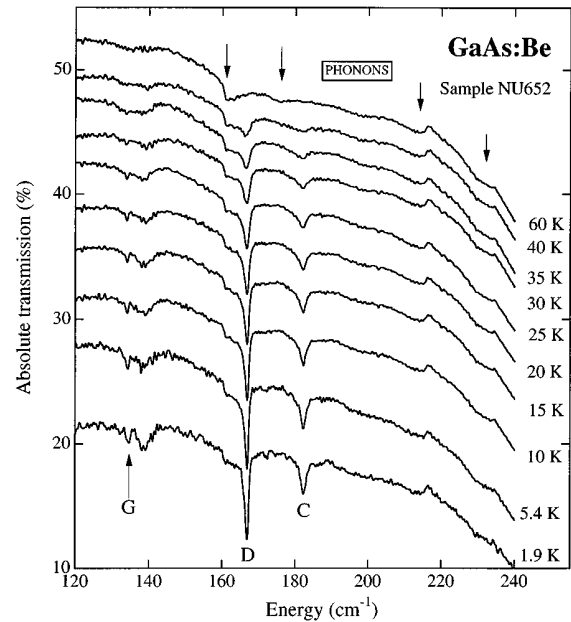


FIG. 3. Temperature dependence of transmission of GaAs:Be. The ordinate applies to the lowest ($T = 1.9 \text{ K}$) spectrum. Spectra at successively higher temperatures have been offset by adding 5%, 10%, 14%, 17%, 20%, 23%, 25%, 27%, and 29% absolute transmission, respectively.

lected after cooling in the dark and then subjecting the sample to midgap radiation (from a 50-W halogen lamp through a $1.18\text{-}\mu\text{m}$ cut-on filter) with no observable effect, further confirming this conclusion. Illuminating the sample with band-gap radiation (again from a halogen lamp) was found to have little effect other than changing the overall absorption (especially in the range $320\text{--}400 \text{ cm}^{-1}$), an effect previously attributed to Drude-like free-carrier absorption.¹³ Thermal cycling — heating an illuminated sample to 128 K, then re-cooling in the dark — was also found to have little effect on the spectrum.

Figure 2(c) gives a spectrum taken at 0.36-cm^{-1} resolution, rather than at the 0.72-cm^{-1} resolution used for the other spectra. (Here resolution is taken to mean the reciprocal of twice the mirror travel of the spectrometer.) It may be observed, most directly in the similar depth of the minima in the transmission of the *D* line in all the spectra in Fig. 2, that the spectrometer resolution used is not limiting the linewidths; that is, the linewidths observed are intrinsic to the sample under the conditions of the experiment. The widths of the lines, and the temperature dependence of the linewidths, are discussed further below.

The spectra of Be in GaAs at the temperatures 1.9, 5.4, 10, 15, 20, 25, 30, 35, 40, and 60 K are shown in Fig. 3. At the highest temperature, broad features are observed due to lattice absorption at 162 , 176 , 214 , and 232 cm^{-1} (see Ref. 22). As noted above, the broad feature at 140 cm^{-1} appears to arise from the Mylar windows, which have a strong temperature-dependent absorption here. The features identified as the *G*, *D*, and *C* lines sharpen and grow in intensity as the temperature decreases, providing further confirmation of their electronic origin.

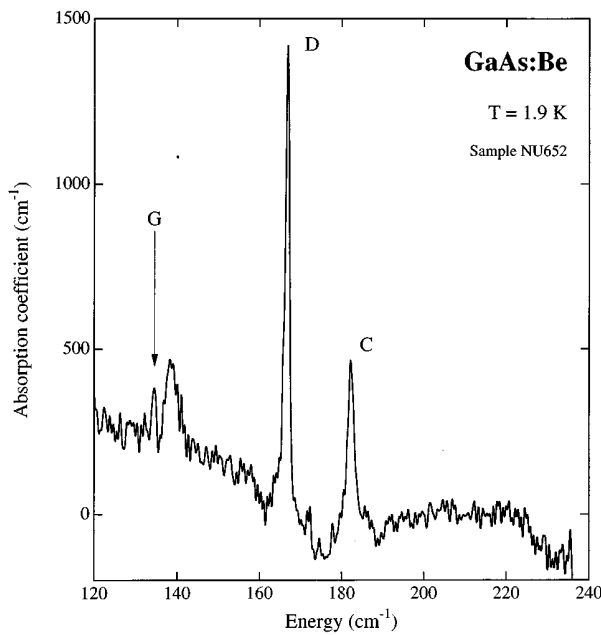


FIG. 4. Absorption coefficient of GaAs:Be at $T = 1.9$ K.

In calculating the absorption coefficient for GaAs:Be, it is assumed that the refractive index, $n=3.521$, is uniform throughout the sample; in particular, it is assumed that there is no refractive index change at the interface between the GaAs:Be layer and the GaAs substrate. The lattice absorption coefficient α_l was calculated from the spectrum taken at $T = 60$ K, in which the acceptor lines are not evident, with the average substrate thickness, $t = 367.5 \mu\text{m}$, employed in the calculation. As an intermediate step, the same calculation was then applied to the low-temperature spectra, to give the pseudoabsorption coefficient α' . Subtracting the first coefficient from the second and multiplying by the ratio of substrate to layer thicknesses then gives the absorption coefficient of the Be impurity: $\alpha = (\alpha' - \alpha_l)t/s$, where $s = 3 \mu\text{m}$ is the Be-doped layer thickness. This method will be inaccurate to the extent that the lattice absorption varies with temperature, but it is expected, and the spectra of Fig. 3 confirm, that this variation is not large in relation to the absorption of the impurity lines. An example of the application of this analysis is shown in Fig. 4 for the spectrum taken at the lowest temperature employed, 1.9 K. The absorption coefficients shown in Fig. 4 are large but not unreasonably so in view of the high concentration of Be. The maximum in absorption coefficient for the strongest line divided by the impurity concentration yields $7.5 \times 10^{-14} \text{ cm}^2$. By way of comparison, for a variety of impurities in Si and Ge (Ref. 32) this ratio lies in the range $(1-8) \times 10^{-14} \text{ cm}^2$.

The G , D , and C lines of the absorption spectra at 1.9, 5.1, 5.4, 10, 15, 20, 25, 30, 35, and 40 K were fitted with Lorentzian curves. This allows precise evaluation of (a) the line positions (b) the linewidths, defined here as the full widths of the lines at the half maximum of the absorption coefficients and (c) the line intensities, that is, the areas under the absorption curves.

The energy of each line was found to decrease slightly with increasing temperature. Data for the D line are given in

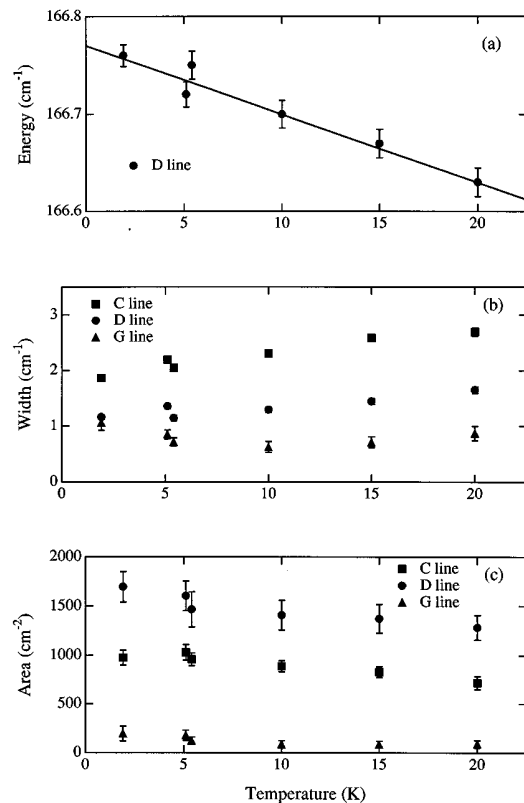


FIG. 5. (a) Temperature dependence of the energy of the D line of GaAs:Be. The error bars indicate the error arising in the curve fitting. The spectrometer calibration is in error by no more than 0.049 cm^{-1} . The solid line indicates the least-squares fit to the data. Its slope is $-0.00704 \pm 0.00075 \text{ cm}^{-1}/\text{K}$. (b) Temperature dependence of the linewidths of the C (squares), D (circles), and G (triangles) lines of GaAs:Be. The error bars indicate the error arising in the curve fitting. (c) Temperature dependence of the integrated areas of the C (squares), D (circles), and G (triangles) lines of GaAs:Be. The error bars indicate the error arising in the curve fitting.

Fig. 5(a). The energy of this transition is seen to decrease by 0.13 cm^{-1} over the temperature range 1.9–20 K. The C and G lines showed similar behavior. The change observed is accounted for within the framework of the EMT, where the energy of each transition varies with dielectric constant as $1/\epsilon^2$. If the dielectric constant in turn varies with temperature as $\epsilon = \epsilon_0(1 + \beta T)$ then, for low temperatures, the transition energy will vary linearly with temperature. A linear fit to the data of Fig. 5(a) yields $\beta = (2.1 \pm 0.2) \times 10^{-5} \text{ K}^{-1}$, which may be compared with the high-temperature (100–300 K) value of $1.2 \times 10^{-4} \text{ K}^{-1}$.²⁶

The energies of the G , D , and C lines measured at 1.9 K are given in Table II. The error in line position resulting from the curve fitting procedure is typically 0.01 cm^{-1} for lines D and C and 0.03 cm^{-1} for line G . These errors, while rather small, are not unreasonable for the fitting of isolated peaks. These fitting errors are smaller than the largest error in the spectrometer calibration of 0.049 cm^{-1} discussed above. The error given when the energies of the various transitions are quoted is obtained by combining the fitting error

TABLE II. Experimental and theoretical energies of Be in GaAs.

Transition label	Transition energy			
	Experiment			Theory Fiorentini ^c (meV)
	Reeder <i>et al.</i> ^a (cm ⁻¹)	This work ^b (cm ⁻¹)	(meV)	
<i>G</i>	135	134.42	16.666	16.05
<i>D</i>	167	166.76	20.676	20.80
<i>C</i>	184	182.30	22.602	22.44

^aReferences 16–19.

^bError estimated to be ± 0.05 cm⁻¹ or ± 0.006 meV for lines *C* and *D* and ± 0.06 cm⁻¹ or ± 0.007 meV for line *G*.

^cReference 1.

and the calibration error and amounts to 0.05 cm⁻¹ for the *D* and *C* lines and 0.06 cm⁻¹ for the *G* line. Also given in Table II are the experimental energies determined earlier by Reeder *et al.*^{16–19} and the theoretical energies calculated recently by Fiorentini.¹ To obtain the transition energies from the theoretical energies, the energies of the excited states given by Fiorentini were subtracted from 28.6 meV, the value this author quotes for the ground state binding energy of Be in GaAs. It is seen in Table II that the recent theory is in good agreement with the present experimental results.

The widths of the *G*, *D*, and *C* lines over the temperature range 0–20 K are shown in Fig. 5(b). The errors arising from the curve fitting procedure are small compared with the resolution of the spectrometer, and it has been demonstrated earlier [see Fig. 2] that the linewidths are not instrument limited. The increase in width for each line is approximately linear in temperature over this temperature range with the width increasing as the final state becomes shallower.

The areas under the *G*, *D*, and *C* line absorption coeffi-

cient curves at $T = 1.9$ K are 196 ± 76 , 1697 ± 156 , and 978 ± 77 cm⁻², respectively. While the areas decrease with temperature [see Fig. 5(c)] as a result of thermal ionization of the acceptor sites, the ratio of line strengths, being $11.5 \pm 5.6:100:57.6 \pm 9.8$ at 1.9 K, remains fairly constant. For many acceptors in a variety of semiconductor hosts the *D* and *C* lines are the most prominent, as observed here. Theoretical estimates have been given for the line intensities of generic acceptors in Si and Ge,⁸ B and Al in Ge and Ga and Al in Ge,³³ and B, Al, Ga, and In in Si,³⁴ but no calculation of oscillator strengths for acceptors in GaAs is known.

Spectra have been taken in the Faraday configuration in magnetic fields of up to 6 T with the electric field vector of the radiation polarized either perpendicular or parallel to the applied magnetic field. The spectral lines split into many low-intensity components that are difficult to resolve. The overall behavior appears to be similar to that previously observed for the *C* and Zn acceptors in GaAs.¹⁵

IV. CONCLUSION

The observation of sharp lines in the far-infrared absorption spectrum of the Be acceptor in bulk MBE-grown GaAs has permitted an accurate determination of the energies of the *G*, *D*, and *C* lines, which is compared with a recent calculation of the energies of acceptors in GaAs. Good agreement is found. The absence of interfering lines from other impurities and subtraction of the lattice absorption have allowed the widths and the strengths of these lines to be determined. A detailed theoretical account of these latter two quantities is looked forward to with interest.

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