Direct evidence for the acceptorlike character of the Cu-related C and F bound-exciton centers in GaAs

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Zeeman measurements have been performed on Cu-doped GaAs in order to reveal the nature of the centers binding the Cu-related excitons C_0 at 1.5030 eV and F_0 at 1.4839 eV. Thermalization between the magnetic subcomponents in Zeeman transmission spectra shows that a splitting occurs in the ground state of the bound-exciton system in each case. In photoluminescence Zeeman spectra no thermalization is observed. From the diamagnetic shift observed for both centers it is concluded that each excitonic complex contains a weakly bound electron, localized by the Coulomb field of more tightly bound holes. Consequently, we definitely identify the C and F centers as neutral acceptor centers, which has hitherto been arbitrarily, albeit correctly assumed. The magnetic measurements lead to effective g values for the C_0 and F_0 bound excitons, $g_{\text{eff}}=2.44$ and $g_{\text{eff}}=2.30$, respectively. In both cases the splitting is almost isotropic. A model is proposed for the electronic configurations of these excitons in which the isotopic g value is explained by a quenching of the orbital angular momentum of the bound hole states through the action of a strong compressive strain field locally at the defect site. In this model the bound exciton consists of two spinlike holes with their spins coupled off, and an electron. The bound-exciton transition occurs between the $S=\frac{1}{2}$ state of the exciton to a spin-only $j_z=\pm\frac{1}{2}$ hole state at the neutral acceptor. This state has a hole g value close to the electron value $g=2$ for both acceptor centers. In addition we report a new unrelated line in the transmission spectra close to the C_0 line. We assign this line to an unidentified neutral isoelectronic donor complex from the thermalization behavior and the diamagnetic shift measured for this center.

I. INTRODUCTION

Cu is known to give rise to several defect centers in GaAs, the interrelation of which has been the cause of a long-standing confusion in the literature.[1] The most persistent of these defect centers are the 0.156-eV and the 0.45-eV acceptors, which have been studied using junction techniques,[2] Hall measurements,[3] and in the case of the former, infrared-absorption measurements.[4] These acceptors are generally accepted to be related to Cu, although their microscopic identities have been subject to various hypotheses.[1,5] The 0.156-eV acceptor gives rise to a photoluminescence (PL) band peaking at 1.36 eV upon recombination of an electron from the conduction-band or shallow donor states. Through piezospectroscopic measurements of this PL band the 0.156-eV acceptor level has been assigned to a $\langle 100\rangle$-oriented Jahn-Teller-distorted Cu_{Ga}.[6] On the other hand, the identity of the 0.45-eV acceptor center is more uncertain.

In this paper we focus on the two bound-exciton (BE) lines C_0 and F_0 at 1.5030 and 1.4839 eV, respectively.[7] (Other values have been reported, differing less than 0.1 meV from ours, which were checked with calibration lines.) In the literature the centers binding these excitons have often arbitrarily been assumed to be the two Cu-related acceptors discussed above. However, it has recently been shown that there exists no correlation between the occurrence of the C_0 line and the 1.36-eV PL band in the photoluminescence spectra of Cu-doped GaAs.[1,8] Furthermore, the symmetry found for the C_0 line (trigonal $\langle 111\rangle$-oriented[7]) does not agree with the recently reported symmetry of the 1.36-eV center.[6] Similarly, it is by no means certain or even likely that the 0.45-eV acceptor center is responsible for the F_0 bound exciton. In fact, it has never been proved that the centers binding these excitons are just acceptors.

In this paper we report new experimental results from magneto-optical spectroscopy on the C_0 and F_0 lines. Our Zeeman data for magnetic fields up to 10 T definitely prove that both BE systems involve acceptor associates. Further, we discuss the electronic structure of the BE states for these acceptors. In particular, the occurrence of a spinlike hole in the ground state of both BE systems is explained in a simple perturbation scheme. It is proposed that a local strain field is the primary perturbation, dominating over the spin-orbit splitting at the neutral Cu-acceptor associates.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

The GaAs samples used in this part of a more extended investigation of Cu-doped GaAs (Ref. 1) were made from liquid-encapsulated Czochralski (LEC)-type bulk material. The starting material was semi-insulating GaAs, nominally undoped with electron concentration $n_{300}=1.8\times10^{12}$ cm^{-3} and mobility $\mu_{300}=4200$ $\text{cm}^2/\text{V s}^1$. To create

strong Cu-related BE lines the Cu diffusion procedure was as follows: A thin film of Cu was evaporated on the polished and etched sample surface, typically 600 Å, and the samples were immediately sealed in evacuated quartz tubes. The C_0 and F_0 lines are strongest after a Cu diffusion at about 700 °C for typically 30 min. A crucial condition for a strong C_0 line is a fast quenching to room temperature either in water or liquid nitrogen, while the F_0 line tends to become stronger if the quenching rate is slower. A necessary condition for the appearance of the F_0 line is the absence of Ga-GaAs melt in the ampoule during the Cu diffusion. The intensity of the C_0 line is also affected by the presence of melt, although less. The samples used in this work show simultaneously strong C_0 and F_0 lines. This is accomplished through an optimization of the above conditions, that is, Cu diffusion at about 700 °C for 30 min in vacuum followed by a rapid quenching to room temperature. The most favorable doping conditions for the different Cu centers in GaAs are discussed in more detail in a separate publication.[1]

The magneto-optical measurements were performed at the Max Planck Hochfeld Magnetlabor, Grenoble, employing a 10-T superconducting magnet in the Voigt configuration. Measurements were usually made at 2 K, but also in the temperature range up to 10 K in order to study the thermalization behavior between the magnetic subcomponents. The signal was dispersed and recorded through a 1.5-m Jobin-Yvon monochromator with high resolution. The photoluminescence was excited with the 5145-Å Ar^+ laser line, and for zero-field PL measurements the signal was recorded through a 0.75-m Jarrell-Ash double-grating monochromator.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 illustrates the PL spectrum of a sample showing strong C and F lines, that is, the electronic lines C_0 and F_0 with phonon replicas. A phonon replica C_1 represents a low-energy phonon mode of 3.6 meV coupling to the C_0 line, with second and third replicas clearly resolved in the spectrum, C_2 and C_3, respectively. The LO replicas C_0^{LO} and C_1^{LO} are also observed in the spectrum. In the F spectrum a low-energy mode of 6.2 meV, F_1, and the LO replica, F_0^{LO}, are present.

In order to tell whether an exciton is bound to an isoelectronic center or a neutral donor or acceptor associate, the thermalization behavior in both the excited state of the system (the center with the bound exciton) and the ground state of the system (the center without the bound exciton) has to be investigated. In addition, for the latter type of BE system it must be possible to distinguish between a loosely bound and a tightly bound particle to decide whether a center is an acceptor or a donor.

The C center binding the C_0 exciton has previously been shown to possess a trigonal ⟨111⟩-oriented symmetry axis, while the F center binding the F_0 exciton is orthorhombic with one axis oriented in the ⟨100⟩ direction.[7] A magnetic field in the ⟨100⟩ direction should give the simplest splitting pattern for the trigonal C center since all four possible lattice orientations of the defect are equivalent in that case. For the F center this direction of the magnetic field is parallel with one of the defect orientations, e.g., [100], and perpendicular with two, [010] and [001].

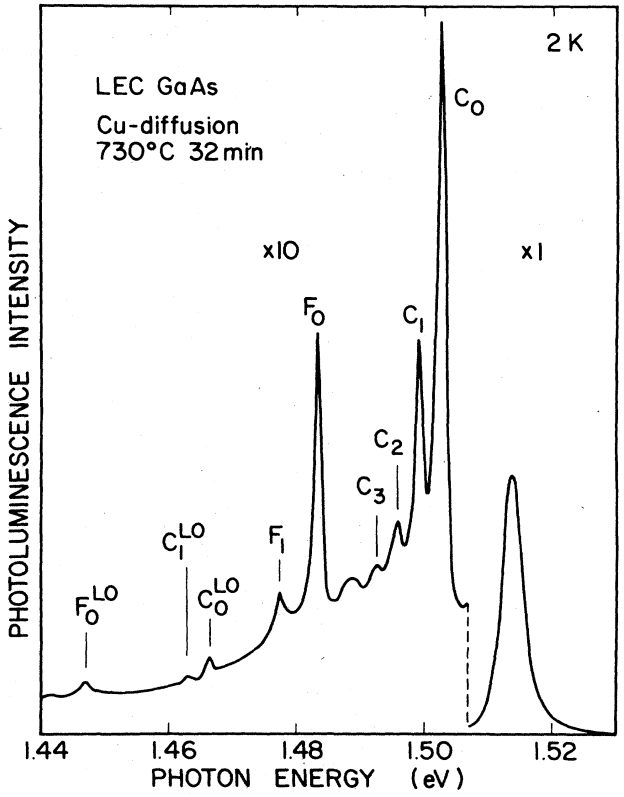

FIG. 1. Typical photoluminescence spectrum of a semi-insulating GaAs sample of Czochralski type, Cu-diffused at 730 °C for 32 min in vacuum. Both the C_0 and F_0 BE lines are strong in this spectrum. Three local-mode replicas of the C_0 line are resolved, labeled C_1, C_2, and C_3, as well as the LO replicas C_0^{LO} and C_1^{LO}. In the F spectrum a local-mode replica F_1 is observed together with the LO replica of F_0.

The transmission spectrum of the same crystal as in Fig. 1 is shown in Fig. 2. The zero-field spectrum is shown as well as the Zeeman spectrum for magnetic field $\mathbf{H}\|\langle 100\rangle$ and $\mathbf{k}\perp\mathbf{H}$ for fields up to 10 T. In the zero-field transmission spectrum the C_0 line and the F_0 line are clearly resolved at the same energy positions as in the PL spectrum of Fig. 1. In addition, a new line of unknown identity appears in the spectrum at 1.5022 eV, or about 1 meV below the C_0 line position. This line does not appear in the PL spectrum of Fig. 1, nor has it been observed in PL spectra of higher resolution. Consequently, it cannot be an electronic line belonging to the C spectrum, since it is not observed in emission despite its energy being lower than that of the C_0 line. It is not a phonon replica either because it appears in the Stokes wing on the low-energy side of the C_0 line in absorption. Further, as discussed below, its thermalization behavior rules out any connections with the C_0 line.

As is obvious from Fig. 2 and, in greater detail, Fig. 4 below, the splitting and thermalization of this new line in magnetic field differ from those of the F_0 line, which makes it unlikely that it is a high-energy component of

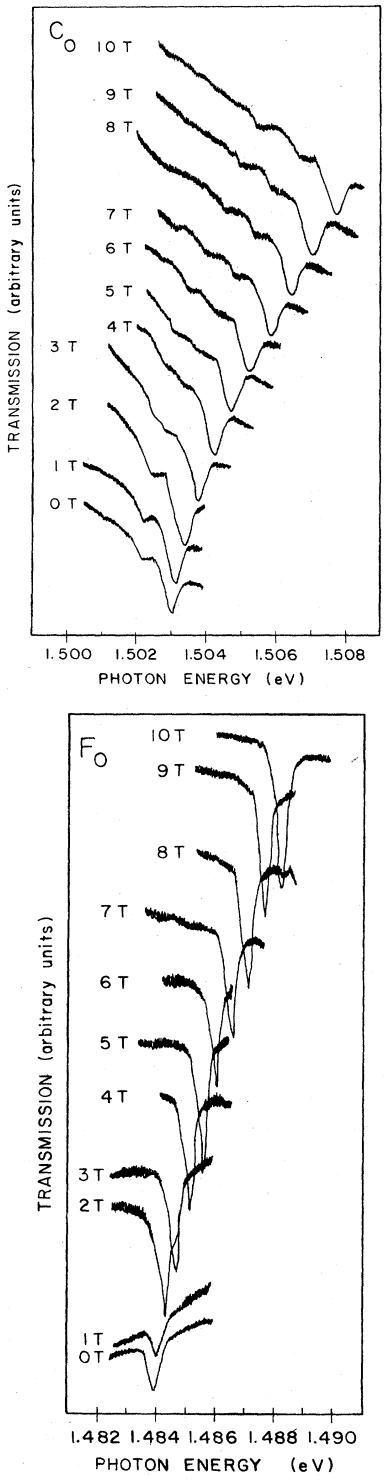

FIG. 2. Transmission spectra of the sample in Fig. 1 in magnetic fields from 0 to 10 T, with $\mathbf{H}||\langle 100\rangle$ and $\mathbf{k}\perp\mathbf{H}$. Three different lines are observed at zero field, the Cu-related BE lines C_0 at 1.5030 eV and F_0 at 1.4839 eV, together with a new unidentified line at 1.5022 eV. Strong thermalization is observed for the Cu-related lines, where only one magnetic subcomponent is observed at all fields (the one at highest energy). The 1.5022-eV line shows negligible thermalization.

the F spectrum. The 1.5022-eV line is not detectable in the absorption spectrum reported by Gross *et al.*[7]

At first sight perhaps one does not realize the strong thermalization between the magnetic subcomponents of both the C_0 and F_0 lines in Fig. 2. Only a single highest-energy component is observable in each case, whereas two components of the 1.5022-eV line can be traced up to 10 T. The thermalization behavior becomes obvious upon comparison with the PL spectrum measured at 10 T for the same magnetic field configuration as presented in Fig. 3. Here two strong components of approximately equal strength are present. The energy position of the high-energy component in PL coincides with the transmission line at 10 T for both centers.

An immediate consequence of this thermalization behavior is a firm exclusion of a hypothetical isoelectronic center binding either of the C_0 or F_0 excitons. Since the ground state of such a system contains no electronic particles, the magnetic splitting must occur in the excited state and thermalization will therefore be observable only in luminescence. The assignment of both centers to acceptors but not donors is not possible without taking into account the diamagnetic shift of the Zeeman components, however. This shift is obvious already in Fig. 2, and will be discussed in connection with Fig. 4 below.

The arbitrary assignment of the C and F lines to the 0.156-eV and the 0.45-eV acceptor centers[9] in previous work possibly made comments on the negligible thermalization in the Zeeman split PL spectrum unnecessary. The published spectra of Willmann *et al.*[9] are, however, not decisive as to whether the centers are, in fact, acceptors or isoelectronic associates. They observe four lines at the relatively small magnetic field of 5.7 T for $\mathbf{H}||\langle 100\rangle$ in the case of the C center and $\mathbf{H}||\langle 110\rangle$ for the F center. In our measurements we only resolve two rather broad components. This is due to the lower resolution of our experiment, which was designed for transmission measurements but not for PL measurements. Consequently, bulk material was used in this work instead of epitaxial wafers, which give smaller PL linewidths.

The two broad components of the C_0 line in PL at 10 T

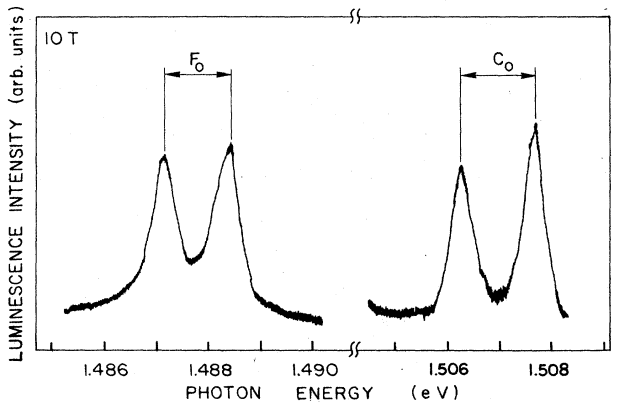

FIG. 3. Zeeman splitting of the C_0 and the F_0 lines at 10 T in photoluminescence ($\mathbf{H}||\langle 100\rangle$, $\mathbf{k}\perp\mathbf{H}$). Obviously no thermalization between the two resolved magnetic subcomponents is observed in contrast to the transmission spectra. This splitting, consequently, occurs in the ground state of the BE system.

for $\mathbf{H}||\langle 100\rangle$ in Fig. 3 have half-widths of 0.48 meV and consist of two unresolved subcomponents each. These are separated by approximately 0.2 meV at 10 T, the inner components being weaker according to Willmann *et al.*[9] The separation between the two outer Zeeman components of the C_0 line in Fig. 3 is 1.4 meV. This can be interpreted as the splitting of the hole states of the neutral acceptor in a magnetic field of 10 T at an angle of 54.7° to the defect axis. The smaller unresolved splitting within each broad component in Fig. 3 would then be the corresponding splitting of the electron, as will be postulated below in view of the collected evidence.

This interpretation of the PL data is by no means the only possible one, since a similar pattern of Zeeman components is expected for an exciton bound to an isoelectronic center in case both the electrons and holes are spin-$\frac{1}{2}$ particles. The former has a negative g value, as is usual for shallow donors in GaAs, and the latter a g value close to the spin value $g=2$. In the Paschen-Back limit, where the Zeeman splitting exceeds any electron-hole exchange splitting, the transition probabilities are governed by the selection rule $\Delta m_s=0$, and a four-line pattern with the outer pair of subcomponents being the strongest is expected. Such a splitting has been reported for an isoelectronic Cu center in InP (where the g values of both particles are positive, however).[10] In the case of an isoelectronic center, thermalization would be expected in PL measurements given that the sample heating from the laser excitation is negligible and that the spin relaxation time is shorter than the recombination rate. This possibility has to be disregarded for the C and the F centra in view of the transmission measurements reported here, however.

The Zeeman pattern for the F_0 line is similar to that of the C_0 line. Here the direction of the magnetic field is not equivalent for all defect orientations and different from that in Ref. 9. This is unimportant, however, because of the isotropic behavior of the splitting observed. We return to the question of g values below.

In Fig. 4 the Zeeman splitting of the three BE lines in the transmission spectrum is plotted for magnetic fields from 0 to 10 T, $\mathbf{H}||\langle 100\rangle$ and $\mathbf{k}\perp\mathbf{H}$, as in Fig. 1. The experimental points at 10 T from the PL measurements are included in the figure. A striking feature of the magnetic field splitting pattern is the large diamagnetic shift observed for all three lines, about 4 meV at 10 T. This is close to the shift of electrons bound to effective-mass-like donors in GaAs.[11] In Fig. 4 the diamagnetic shift for such electrons in GaAs as obtained from a calculation of the diamagnetic shift of the $1s$ hydrogenic ground state of shallow donors[12] has been included. For scaling this calculation to fit shallow donors in GaAs, the effective Rydberg $R^*=m^*e^4/2\epsilon^2\hbar^2=5.52$ meV, with the conduction-band effective mass $m^*=0.067m_0$, was used as scaling parameters.

The shift of the center of gravity of the Zeeman patterns for the C_0 and the F_0 lines (measured from the PL data at 10 T) agree well with the calculated diamagnetic shift of a shallow donor electron.

For the F_0 BE the diamagnetic shift is about 0.16 meV larger than for the effective-mass donor electron as calculated above. This corresponds to a slightly smaller effective mass, $m^*=0.064m_0$, if m^* is taken as an adjustable parameter. For the C_0 BE the observed shift is 0.25 meV larger than the donor-electron value, corresponding to $m^*=0.063m_0$. This close agreement strongly suggests that a pseudodonor model is valid for both centers. In this model an electron is loosely bound in the Coulomb field of more tightly bound holes at the center, which, consequently, is an acceptor in view of the thermalization behavior. Hence, the combined information from the thermalization and the diamagnetic shift allows us to reach the conclusion that both the C and the F centers

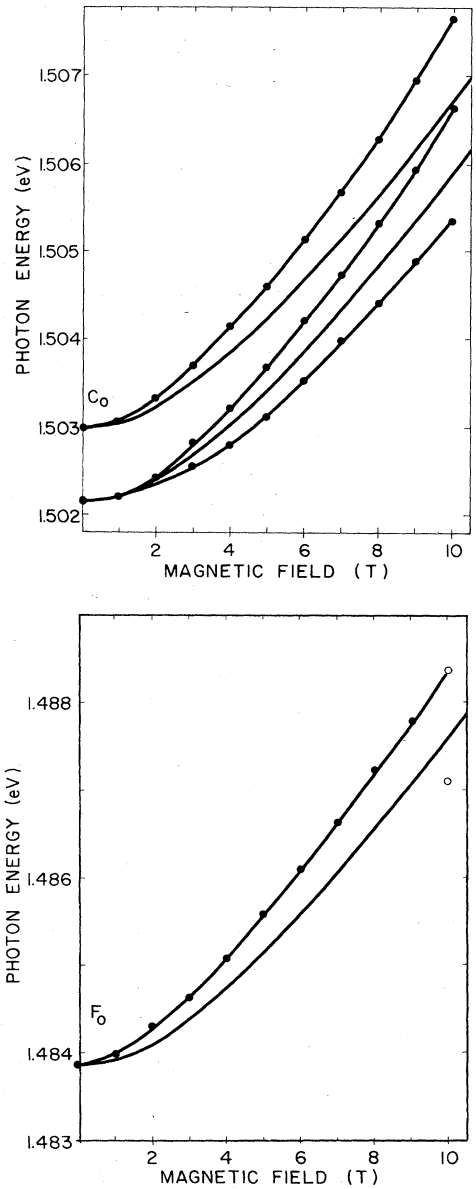

FIG. 4. Zeeman splitting of the transmission spectrum of Fig. 2 for magnetic fields from 0 to 10 T, with $\mathbf{H}||\langle 100\rangle$ and $\mathbf{k}\perp\mathbf{H}$. Solid circles are experimental points from the transmission measurements, whereas the open circles inserted for 10 T are the corresponding experimental points from PL measurements. The solid lines are the diamagnetic shift for an effective-mass donor electron calculated from Ref. 12.

are, in fact, acceptors. Both the Zeeman splitting in luminescence and the diamagnetic shift could, however, be in agreement with isoelectronic donors, that is, isoelectronic centers with hole-attractive central cells. The transmission measurements thus provide the necessary piece of evidence in form of the thermalization behavior (which in itself is insufficient, since it is consistent with a model of excitons bound to either donors or acceptors).

The diamagnetic shift of the C_0 and the F_0 lines has been reported previously[13] from Zeeman PL measurements giving a similar value to the one we find. In that work it was, however, presupposed that both excitons were bound to acceptors. The interpretation was also different, since the diamagnetic shift was ascribed to a free exciton which has been pointed out to be incorrect if the exciton-localization energy exceeds the free-exciton binding energy.[11]

As far as the 1.5022-eV line is concerned, no thermalization is observed between its two magnetic subcomponents in the transmission measurements. No corresponding BE line was resolved in PL measurements in our samples. In the absence of thermalization in the ground state of the BE system, it is then natural to assume that the center binding this exciton is of isoelectronic nature. In luminescence from an excited state of a BE system, spin-lattice relaxation has to be faster than the recombination rate for thermalization to occur. Thermalization in the ground state of an acceptor-related BE system should always be observable, on the other hand, since the ground state has a practically infinite lifetime.

In view of the diamagnetic shift of the Zeeman components of the 1.5022-eV line it can be concluded that a pseudodonor model is also applicable in this case. The agreement with the expected value for an effective-mass donor is even closer in this case than in the previous ones, a perfect fit to the center of gravity corresponds to $m^*=0.065m_0$. The center consequently is an isoelectronic donor, consisting of a hole-attractive central cell binding the hole in a short-range potential and the electron by the Coulomb potential of the hole. The effective g value for this exciton is $g=2.2$.

In Fig. 5 a schematic level diagram for the case of excitons bound to neutral acceptors is shown. The small g value for weakly bound electrons in GaAs, $g_e=-0.46$, causes only a small splitting, about 0.27 meV at 10 T or below our resolution limit. Thus the major part of the splitting is caused by the hole bound at the neutral acceptor. This is illustrated in Fig. 5. The outer components are shown stronger in the diagram, in agreement with the spin electron rule $\Delta m_s=0$, which is valid in the electron picture (the hole corresponds to a missing electron with $m_e=-m_h$). The level diagram is drawn in this picture.

The Zeeman splitting of the C_0 and the F_0 lines has been reported to be isotropic in PL data with a small anisotropy present in the case of the C_0 line.[9] Since both centers have noncubic symmetry,[7] it is natural to assume that the angular orbital momentum of the hole states is quenched as a result of a compressive low-symmetry strain field.[10,14] This would explain the absence of anisotropy in the Zeeman splitting. For the C center the angular dependence of the hole states as a function of the angle

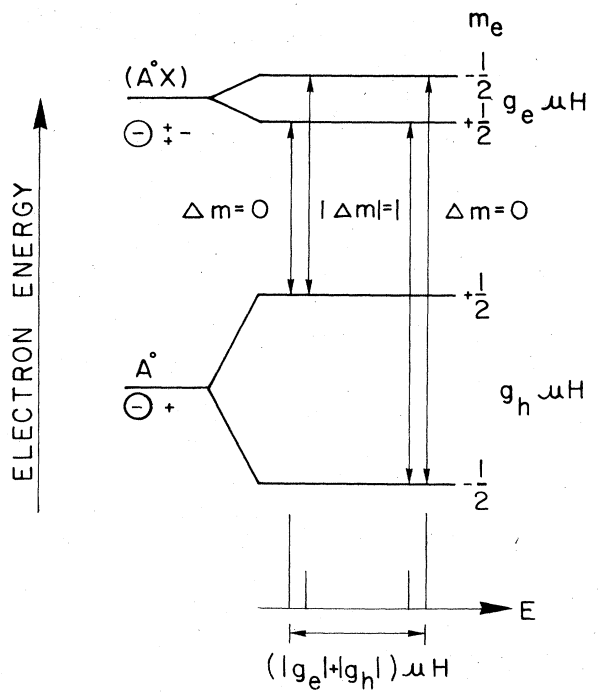

FIG. 5. Schematic level diagram illustrating the Zeeman splitting of the C_0 and the F_0 BE lines. The small (unresolved) splitting in our data is ascribed to the electrons with g value $g_e=-0.46$, while the holes have g value close to $g=2$. The figure illustrates the spin selection rule $\Delta m_s=0$ in the electron picture ($m_e=-m_h$).

θ between the direction of the magnetic field and the trigonal axis can be written $\Delta E=g(\theta)\mu H$, where the g value for the holes, $g(\theta)$, involves the angular dependence. A similar expression is valid for the orthorhombic symmetry of the F center. In the case of a compressive trigonal field, the Kramers doublet $|\frac{3}{2},\pm\frac{1}{2}\rangle$ in $|J,M_J\rangle$ notation would be the ground state of the neutral acceptor (see below) with the g value $g(\theta)=(g_{\parallel}^2\cos^2\theta+g_{\perp}^2\sin^2\theta)^{1/2}$. For a tensional axial crystal field the ground state of the holes would be $|\frac{3}{2},\pm\frac{3}{2}\rangle$ and the angular dependence of the g value is then simply $g(\theta)=g_h\cos\theta$.[15]

From the absence of anisotropy in the hole g value in the Zeeman data, it is clear that neither of these two cases applies to the two acceptors of this study. Instead, the simplest expression for isotropic splitting is valid both for electrons and holes: $E=g_{\text{eff}}\mu H$, where g_{eff} is the effective g value. The total splitting is therefore, as is obvious from Fig. 5, $E=(|g_e|+|g_h|)\mu H$, which gives for the C center $g_h=2.44-0.46=1.98$ and for the F center $g_h=2.30-0.46=1.84$. Both g_h values are relatively close to the spin value $g=2$, which agrees with the hypothesis of a quenched angular orbital momentum of the holes bound to the defects. A necessary condition for this to occur is the combination of a strong noncubic strain field of a compressive sign and a localized hole wave function, overlapping the strain field created locally at the defect. This is reasonable for the case of Cu-related defects, which are expected to have strongly hole-attractive central cells. Similar effects have been observed for com-

plex Cu-related defects in GaP (Refs. 16 and 17) and in InP (Ref. 10). Another condition for a complete quenching of the hole orbital angular momentum is that the spin-orbit interaction of the hole states derived from the valence band can be regarded as a small perturbation of the dominating local strain field. In GaAs the spin-orbit splitting is 340 meV, or 4 times larger than that of GaP. This requires a strong local strain field and highly localized hole wave functions. In ZnTe, however, a similar phenomenon has been observed for Cu-related centers, despite the much larger spin-orbit splitting 0.9 eV.[18] Also, InP has a much larger spin-orbit splitting, or close to 1 eV.[19]

For the case of a compressive sign of the local strain field, but of a smaller magnitude, so that the spin-orbit interaction would be the primary perturbation, the hole states of the neutral acceptor are properly described in terms of the projection M_J of the total angular momentum J along the defect axis. Then the $P_{3/2}$ hole state of cubic symmetry splits into the Kramers' doublets $|\frac{3}{2},\pm\frac{3}{2}\rangle$ and $|\frac{3}{2},\pm\frac{1}{2}\rangle$ in $|J,M_J\rangle$ notation. The sign of the strain field raises the energy of the former doublet and lowers the energy of the latter one, $|\frac{3}{2},\pm\frac{1}{2}\rangle$.[20] Willmann *et al.*[9] ascribe the $|\frac{3}{2},\pm\frac{1}{2}\rangle$ state to the bound hole state of the neutral acceptor for both the C and F centers, while the $|\frac{3}{2},\pm\frac{3}{2}\rangle$ state is suggested to merge into the valence band by the action of the strain field. This model does not explain the isotropic behavior of the Zeeman pattern, however, but would instead give the anisotropic g value given above. In view of the spinlike hole states expressed by the isotropic g value for both centers, it must instead be concluded that the strain field is the primary perturbation, with the smaller spin-orbit interaction being superimposed, as illustrated in Fig. 6 for both the C and the F centers. In the level diagram the BE energy is drawn so that transitions to acceptor states of higher binding energy have higher transition energy and, consequently, the BE energy increases vertically downwards as the hole energy. This simplified diagram (which is valid only for the $J=0$ hole state of the BE, where the hole spins are coupled off) accounts for the increase of the magnetic subcomponents of higher transition energy upon thermalization to hole states of lowest hole energy in transmission measurements at low temperatures. In this picture we assume that the compressive strain field increases the hole energy of the $p_\pm$ states, which are proposed to merge into the valence band. The ground state

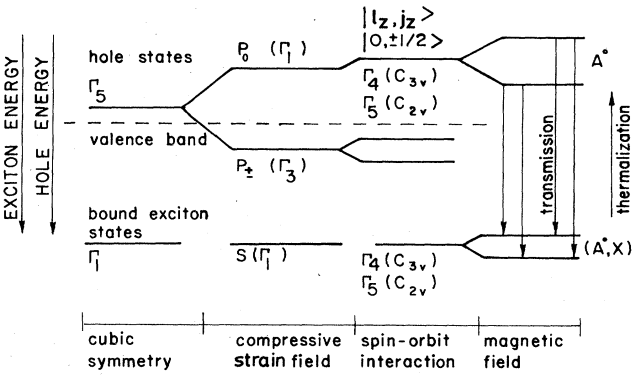

FIG. 6. Schematic level diagram for the C_0 and the F_0 centers as explained in the text. The primary perturbation is ascribed to the crystal field with the spin-orbit interaction of the valence band being superimposed. The BE states are drawn towards higher hole energy, which illustrates the thermalization in the ground state of the BE system favoring higher-energy components at low temperature.

of the neutral acceptor is derived from the p_0 state, which is lowered in energy by the strain field. Upon spin-orbit coupling these states are denoted $|0,\pm\frac{1}{2}\rangle$ in the $|l_z,j_z\rangle$ notation,[14] where l_z and j_z are projections along the defect axis z. The exciton is formed by the coupling of two such spinlike holes, which pair off their spins, in addition to an electron, resulting in a $S=\frac{1}{2}$ BE state. The electronic transitions occur between this BE state and the spinlike $|0,\pm\frac{1}{2}\rangle$ hole states of the neutral acceptor.

This picture supports the model for the C and the F centers put forward in a separate publication, suggesting the involvement of Cu interstitials in both. Such defect associates can naturally be assumed to create compressive strain locally at the defect sites through the presence of the intersitial species.[1]

To summarize, we have shown that both the C_0 and F_0 excitons in Cu-doped GaAs are bound to complex acceptors, creating strong compressive local strain field of noncubic symmetry. We have also shown that previous assignments of these spectral features as excitons bound to acceptors were only coincidentally true, since the formerly available data were not conclusive on this point. The erroneous assignment of these bound excitons to well-known Cu acceptors was possibly misleading in this respect as well.

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