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Localization of excitons to Cu-related defects in GaAs

B. Monemar and H. P. Gislason
Department of Physics and Measurement Technology, Linköping Institute of Technology, S-581 83 Linköping, Sweden

Z. G. Wang
Institute of Semiconductors, Chinese Academy of Sciences, Peking, China

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The possibility of binding excitons to complex-type defects in GaAs is discussed, with reference to recent photoluminescence data on Cu-related centers. The apparent absence of bound excitons (BE's) associated with certain complex-type acceptors may be explained as a consequence of a local compressional strain field at the defect. Such a field will decrease the binding energy of electron states derived from the Γ_1 conduction-band minimum in GaAs, so that they ultimately become resonant with the band in the limit of a strong field. A similar effect on the BE electron state is expected for neutral-complex defects, particularly if they involve two interstitial species. Only one example of an exciton binding to a neutral complex was found in GaAs so far. It involves a tensional local strain field, in which case the BE electron localization becomes stabilized.

I. INTRODUCTION

Cu introduces several deep acceptor states when incorporated in GaAs;[1–4] additional states of this type are introduced by co-doping with Li.[5] In addition, neutral complexes of isoelectronic character involving Cu are expected to form, as observed in other III-V compounds, such as GaP (Refs. 6–8) and InP (Ref. 9), and recently also in GaAs (Ref. 10). The latter class of defects is detectable in photoluminescence (PL) via bound-exciton (BE) spectra at low temperatures. In GaAs very few examples of Cu-related BE spectra are found, suggesting that there must be fundamental reasons why these excitations cannot be observed. In this paper we discuss the possibilities of binding excitons to deep-level defects in GaAs, with particular emphasis on Cu-related defects, which are, in most cases, of a complex nature. The optical spectra of these Cu-related defects are discussed in considerable detail separately.[4,5,10] Therefore the experimental data here will be shown only for easy reference, when necessary for the discussion. A simple model is provided for the physical explanation of the lack of binding of BE states observed in most of the cases discussed.

II. ACCEPTOR STATES AND THE BINDING OF EXCITONS

Several Cu-related acceptor levels are reported for GaAs, the most prominent being those at 0.15 and 0.45 eV.[3,4] The 0.15-eV acceptor level is usually dominant, and rather convincing arguments have been put forward for its identification as substitutional $\mathrm{Cu_{Ga}}$, distorted in the $\langle 100 \rangle$ direction.[3,4,11] No bound excitons have been found to be associated with this level.[4] The 0.45-eV acceptor level was previously thought to be the second ionization level $(2-/-)$ of the double acceptor $\mathrm{Cu_{Ga}}$ [the 0.15-eV level was then suggested to be the first $(-/0)$].[1] Since these levels were recently reported to occur at different unrelated concentrations,[3] this assignment seems to be in error. The 0.45-eV acceptor level is therefore referred to here as an unidentified Cu-related acceptor complex. Two prominent BE lines are seen in Cu-doped GaAs (Fig. 1): the so-called *C* and *F* lines,[4,12–14] which have recently been shown to be associated with acceptors.[15] The *C*-line acceptor has a $\langle 111 \rangle$-oriented symmetry axis,[12] and has been shown to be unrelated to the 0.15-eV acceptor.[4] The *F* line has a lower symmetry (orthorhombic),[12] and is also associated with a Cu-related acceptor complex (i.e., different from the 0.15-eV one[4]). The association of the *F* line with the 0.45-eV acceptor level cannot be definitely ruled out, but is unlikely from studies of diffusion profiles. The Cu-Li co-doping creates a 0.11-eV acceptor-complex level as evidenced via the 1.41-eV PL band in Fig. 2. No bound-exciton state is found to be associated with this acceptor (Fig. 2). We therefore have the somewhat disturbing experimental situation that of the three Cu-related acceptor levels (0.11, 0.15, and 0.45 eV) discussed here none seems to bind excitons. On the other hand, bound excitons for deep acceptor states do indeed exist in GaAs, as manifested by the observation of the strong *C* and *F* lines[15] (Fig. 1).

The binding of excitons to neutral single acceptors in GaAs should be possible even in the case of low-symmetry defects, since two holes should still (via incomplete screening) be attracted to a hole-attractive (Cu-induced) central-cell potential for such an acceptor.[16] The electron in the BE state should be bound by a secondary Coulomb attraction to the center, once it is charged by the presence of two bound holes.[16] This "pseudodonor" model has been experimentally verified for excitons bound to acceptors (acceptor-bound excitons, or ABE's) in several cases for GaAs.[15,17,18] Therefore, the presence of the *C* and *F* lines is not surprising, if the corresponding center are single acceptors. Rather, the lack of observation of ABE lines associated with other deep acceptors will have to be understood in terms of the electronic properties of hole and electron states bound to complex defects in GaAs.

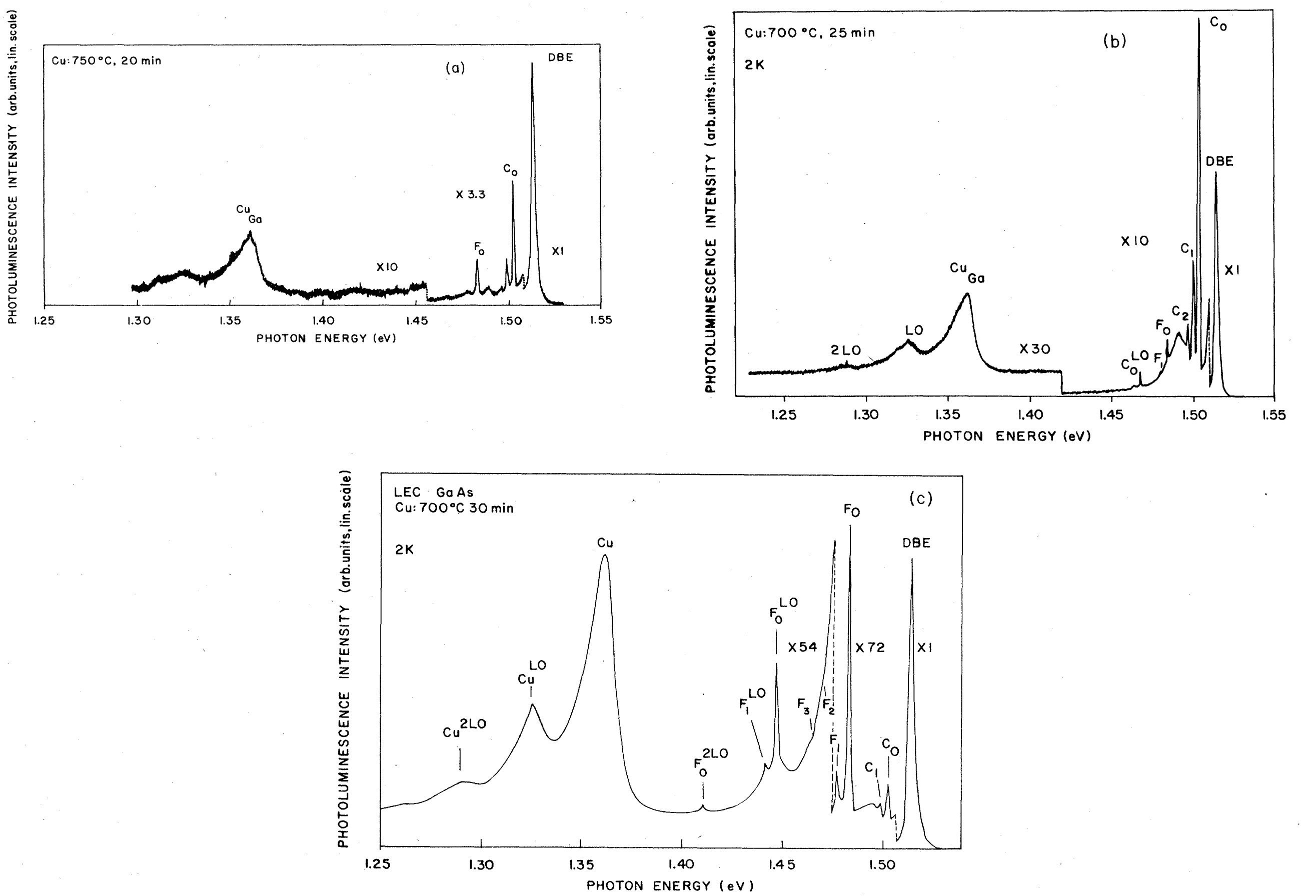

FIG. 1. (a) Near-band-gap photoluminescence (PL) spectrum (2 K) from an originally n-type vapor-phase-epitaxy (VPE) GaAs sample, with an 18-μm-thick epilayer of an original uncompensated doping level $n_{300\,\mathrm{K}} = 3\times10^{14}$ cm^{-3}, which has been Cu-diffused at 750 °C for 20 min in vacuum and rapidly quenched in water. Apart from the slightly broadened DX band at 1.5135 eV due to residual shallow donors, the spectrum is dominated by the rather strong C_0 and F_0 lines and their phonon sidebands. These lines are both connected with Cu-related deep acceptors (Ref. 4). The presence of the $\mathrm{Cu_{Ga}}$ double acceptor is manifested by the broad band at 1.36 eV. No additional Cu-related BE lines apart from the C and F series are observed. (b) Similar near-band-gap PL spectrum (2 K) as for an originally n-type liquid-phase-epitaxy (LPE) GaAs sample, with a 28-μm-thick epilayer of an original uncompensated doping level $n_{300\,\mathrm{K}} = 1\times10^{15}$ cm^{-3}. The sample was Cu-diffused in vacuum at 700 °C for 20 min and rapidly quenched in water. The spectrum is very similar to the one in (a), apart from a higher radiative efficiency in this LPE sample. The C and F BE spectra are seen, together with the $\approx$1.5135-eV broadened donor BE band and the 1.36-eV $\mathrm{Cu_{Ga}}$ band. No additional bound excitons are seen that could be related to the $\mathrm{Cu_{Ga}}$ acceptor. (c) Similar near-band-gap PL spectrum (2 K) as in (a) and (b) for an originally semi-insulating undoped liquid-encapsulated-Czochralski (LEC) bulk GaAs sample, which was diffused with Cu in vacuum at 700 °C for 30 min and rather slowly quenched in air. Similar spectra as in (a) and (b) are produced in this case also, and no bound exciton is observed associated with the $\mathrm{Cu_{Ga}}$ acceptor.

A. Double-acceptor case

We shall first discuss the special case of the $\mathrm{Cu_{Ga}}$ acceptor, which is a *double* acceptor. There is substantial evidence that the 0.15-eV level is associated with the $(-/2-)$ transition of the defect,[4] i.e., the capture of the first hole into the doubly ionized acceptor. Furthermore, there is a static Jahn-Teller (JT) relaxation taking place upon this hole capture, so that the Cu atom is distorted in a [100] direction.[19] This will affect the binding energy of the 0.15-eV acceptor state, being partly derived from this static relaxation.

Binding of a second hole by the singly ionized Cu acceptor is, in general, expected to be possible. However, an electron would not easily bind to the resulting neutral Cu acceptor state, due to the repulsive effect of the strongly hole-attractive core potential of the $\mathrm{Cu_{Ga}}$. In general, binding of excitons to charged acceptors (which is the case discussed here) is not expected to be possible for GaAs, due to the large m_h^*/m_e^* effective-mass ratio,[16] and has not been reported in the literature. With this assignment of the 0.15-eV level to the $\mathrm{Cu_{Ga}}$ $(-/2-)$ transition, the absence of a BE state associated with this state is therefore simply what is expected.

A related problem is whether $\mathrm{Cu_{Ga}}$ actually binds a second hole. Only one level has been observed in our experimental data. The $(0/-)$ level is expected to be much shallower than the 0.15-eV level, of the same order as the shallow acceptor levels in GaAs (25—35 meV).[20] This is inferred from recent studies of other double acceptors in

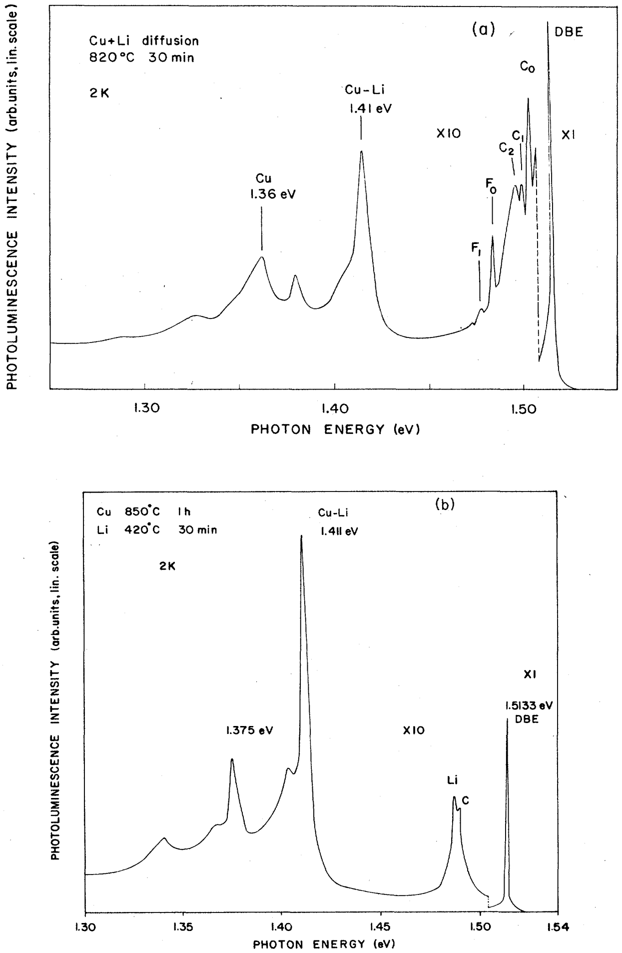

FIG. 2. (a) Photoluminescence spectrum at 2 K from a horizontal-Bridgman (HB) GaAs sample diffused with Cu and Li simultaneously at 820 °C for 30 min in an evacuated ampoule. Note the presence of the $\mathrm{Cu_{Ga}}$ acceptor from the 1.36-eV emission, and likewise, the Cu-Li acceptor from the 1.41-eV emission. BE spectra from the *C* and *F* centers dominate, together with the ≈ 1.5135-eV peak related to shallow donors. (b) PL spectrum from another HB sample doped with Cu at 850 °C for 1 h and subsequently with Li at 420 °C for 30 min. The dominant deep acceptor is the Cu-Li acceptor associated with the 1.41-eV peak. Other shallow acceptors give rise to the peak at ≈ 1.49 eV, but the only BE-related peak is the near-band-gap peak at ≈ 1.5135 eV due to residual donors.

GaAs, such as $\mathrm{Ga_{As}}$ (Ref. 21) and $\mathrm{Li_{Ga}}$ (Ref. 5). We see no such additional acceptor level revealed as, e.g., donor-level—acceptor-level (*D*-*A*) or conduction-band—acceptor-level transitions (free to bound) in the photoluminescence spectra. Therefore the binding of the second hole by the defect is not observed so far, but cannot be excluded for lack of more extensive experimental data from sufficiently pure starting material. A speculative solution to the possibility that the second hole is not bound would be that the neutral Cu^0 state is actually resonant with the valence band. The first charge state Cu^- is stabilized in the gap by a distortion off the tetrahedral lattice site to produce a level in the band gap, as observed in the $(2-/-)$ 0.15-eV level. The piezospectroscopic data on the 1.36-eV emission only indicate a small (JT) relaxation, however.[19]

The case of binding an exciton to a neutral double $\mathrm{Cu_{Ga}}$ acceptor in GaAs is of particular interest, if there is an axial distortion connected with this state as well. In tetrahedral symmetry a neutral double acceptor is expected to be able to accommodate a third bound hole (actually, four holes are possible in a closed-shell multiple-bound-exciton model[16,22]). This has been verified in other materials, such as Ge (Ref. 23) and Si (Ref. 24). Once an axial stress field is applied, this hole shell is split up, so that the lowest-energy branch can hold only two holes, as has been demonstrated by piezospectroscopic data in Ge.[25] A local axial strain field at the defect usually has a large effect on hole states, as observed, e.g., in GaP (Refs. 7 and 8) and ZnTe (Ref. 26). Consequently, the strain field of an axial defect could easily drive the split-off hole state (accommodating two holes) to higher hole energies, i.e., into the valence band. Therefore the fact that no bound exciton has so far been observed connected with a neutral $\mathrm{Cu_{Ga}}$ acceptor in GaAs might not give any indication as to whether the corresponding acceptor level [the $(-/0)$ transition] is in the band gap. In addition, the possibility of a strong Auger effect in the recombination of excitons bound to a neutral double acceptor cannot be ruled out as an alternative explanation of the absence of an ABE associated with neutral $\mathrm{Cu_{Ga}}$ in PL spectra. The Auger effects will be further discussed below.

B. Single-acceptor case

The absence of binding of excitons for neutral *single* acceptors seems to call for a different explanation. We have interpreted the Cu-Li 0.11-eV acceptor level as due to the $(-/0)$ transition of an axial $\mathrm{Cu_{Ga}}$-Li_i single acceptor.[5] The case of the deep 0.45-eV acceptor level mentioned above is more uncertain and will therefore not be discussed in detail here. [We just note that it is most likely to be a $(-/0)$ transition of a single acceptor complex, since the attribution of the 0.15-eV level to the $\mathrm{Cu_{Ga}}$ acceptor exhausts the choice of simple Cu-related double acceptors.] No bound exciton is seen associated with the 0.11-eV Cu-Li acceptor (Fig. 2). We cannot completely rule out that an ABE state could be resonant with other shallow ABE states in this case, but the overall weakness of such shallow ABE spectra in Cu-Li—doped GaAs does not support this point. We shall therefore search for a model to explain the absence of a bound ABE state in this case. We believe that the second hole would be bound comfortably in this case, as normally expected for a BE bound to a neutral acceptor. The binding of the electron to the positive $(\mathrm{Cu\text{-}Li})^+$ state could be more difficult for an axial defect, since the electron binding energy for donor states is so small in GaAs (<6 meV).[27] Furthermore, the electron states are very sensitive to a strain field.

The Cu-Li acceptor complex is expected to cause an overall compressive local strain field as a result of the presence of an interstitial Li in the complex. A field of this sign is expected to raise the shallow bound-electron states in energy, in the same way as the Γ_1 conduction-band minimum behaves with stress in GaAs.[28] Local strain fields around defects in semiconductors are found to be quite large.[29] Furthermore, it has been demonstrated in GaP that such local fields may have substantial effects on the binding energy of both electrons and holes to complex defects.[6–8] We therefore suggest that a compressional local strain field could render a shallow Γ_1 electron state in GaAs resonant with the conduction band, in which case no BE state can be seen in optical spectra for such defects.

All substitutional acceptor dopants from groups II and IV in GaAs are found to produce bound excitons. Even the deep $\mathrm{Sn}_{\mathrm{As}}$ acceptor with a binding energy of 167 meV produces a strong bound-exciton line at 1.507 eV.[17] The case of $\mathrm{Mn}_{\mathrm{Ga}}$ is interesting in connection with this work, however, since it was previously reported not to produce a bound exciton.[30] We have repeated the experiment on Mn doping, and indeed no ABE state related to the Mn was found in our case either; see Fig. 3. The Mn acceptor has a binding energy very close to the Cu-Li acceptor discussed above, just ≈ 3 meV deeper.[5] Mn has been shown to be a single substitutional acceptor on a Ga site,[31] and thus represents an anomalous case. For Mn the influence of the *d*-like character of localized hole states in the band gap may well cause an increase in hole-hole repulsion energy in cases where more than one hole is to be bound to the acceptor (as in an ABE), eventually resulting in an instable ABE state.[32] This problem should not be important for $\mathrm{Cu}_{\mathrm{Ga}}$, where the *d*-like hole states are predicted to be resonant very deep down in the valence band.[33]

At this point it seems appropriate to return to the *C* and *F* complex acceptors in GaAs, where indeed deeply bound ABE's are observed.[15] In addition, the acceptor hole state is in this case found to be a spinlike hole, which is only possible in a rather strong compressive local field of symmetry lower than tetrahedral.[15] The electron of the ABE is found to be loosely bound, like a shallow donor electron, from the observed diamagnetic shift in Zeeman data for both these ABE's.[15] This means that the binding of the electron is delicate, since a rather strong compressive local stress field does not necessarily render the electron unbound. In addition, the local complex-defect potential, in this case thought to be dominated by the *hole-attractive* $\mathrm{Cu}_{\mathrm{Ga}}$ site, may, in these cases, contain substantial *electron-attractive* contributions from a localized donorlike part (such as a rather deep interstitial donor level),[4] which could compensate for the action of the local compressive strain field on the electron state of the ABE, discussed above.

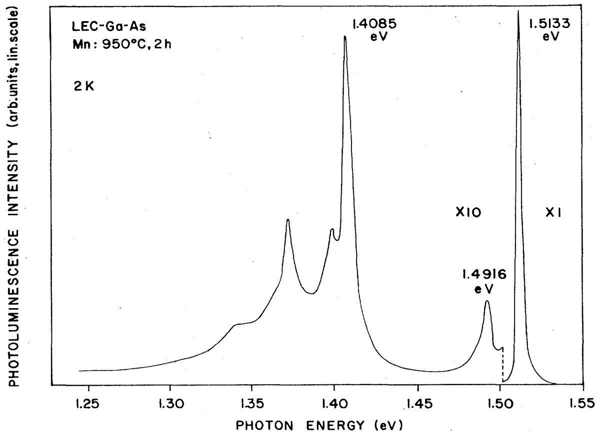

FIG. 3. Near-band-gap photoluminescence spectrum (2 K) from an originally nominally undoped semi-insulating sample of LEC GaAs, Mn-diffused at 950 °C for 2h. The peak at ≈ 1.4085 eV is due to the Mn acceptor, occurring at about 3 meV lower energy than the corresponding Cu-Li peak in Fig. 2. [It has been carefully checked that no Mn contamination was present in the (Cu-Li)-doping experiments (Ref. 5).] No bound-exciton line is seen associated with the Mn acceptor, only the broad line at ≈ 1.5135 eV related to shallow donors.

III. NEUTRAL (ISOELECTRONIC) COMPLEXES

In GaP a large number of neutral-complex defects occur with Cu doping[6–8] and an additional number of such centers are created upon Li co-doping, as manifested from BE spectra taken at low temperature.[8] There is no reason to believe that the defect chemistry upon Cu and Li diffusion is drastically different in GaAs and GaP, and we consequently believe that neutral Cu complexes and Cu-Li complexes are formed also in GaAs upon proper diffusion treatments, as discussed separately.[4,5] No bound-exciton spectra associated with such defects in GaAs are observed, however. This should thus be taken as evidence that BE's are, in general, not bound to such defects in GaAs, while the corresponding neutral-complex defects indeed are expected to exist in the material. In fact, no BE's related to simple substitutional isovalent atoms have been observed in GaAs at normal pressures.

Since these neutral-complex Cu-related defects should have an overall hole-attractive central-cell potential being dominated by $\mathrm{Cu}_{\mathrm{Ga}}$, a hole may be bound to such a defect at low temperature, making it positively charged. An electron might subsequently be bound as a result of the Coulomb attraction from the bound hole. The usual effective-mass-like binding energy of ≤ 6 meV of an electron in GaAs will certainly in such a case be reduced by the electron-repulsive (hole-attractive) $\mathrm{Cu}_{\mathrm{Ga}}$ central cell, but this effect might be insufficient to render the electron state unbound. The above-mentioned local strain field might be much more potent in this respect, however. As explained above, it should take just a moderate local strain field of a compressive sign to raise a shallow bound-electron state a few meV in energy to make it resonant with the Γ_1 conduction band in GaAs.[28] It is believed that most Cu-related (or Cu-Li—related) neutral-complex defects in GaAs actually give rise to a strong compressive strain field, as they are deduced to do in GaP.[6–8] This is easy to imagine for the model case of a $\mathrm{Cu}_{\mathrm{Ga}}$ (double acceptor) and *two* additional interstitial atoms accommodated around the same site. Therefore our conclusion is that such "compressive" neutral complexes exist in GaAs, although they are unable to bind excitons in the cases studied in our work.

In contrast, we believe that there is a case in Cu-diffused Zn-doped GaAs where a neutral complex actually binds an exciton deeply. As described in detail separately, an emission with the lowest electronic line at

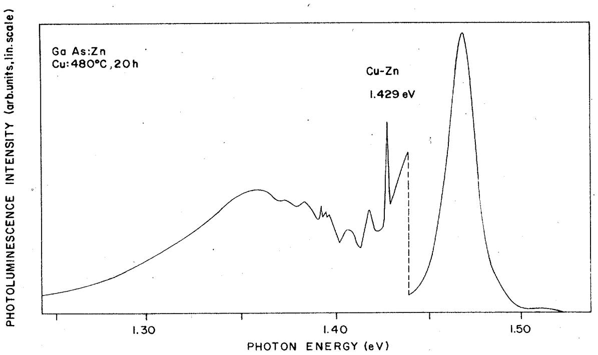

FIG. 4. Near-band-gap photoluminescence spectrum (2 K) from a bulk GaAs sample originally doped with Zn to a concentration $\approx 1.5\times10^{16}$ cm^{-3}, and subsequently Cu-diffused at 480 °C for 20 h. This procedure produces a rather strong line at 1.429 eV, a bound-exciton line related to a neutral defect containing Cu and Zn (Ref. 10). A rich spectrum of phonon replicas accompany this line. The broad band at $\approx$1.47 eV is related to Zn_{Ga} acceptors.

1.429 eV is ascribed to an exciton bound to a neutral complex involving both Cu and Zn (Ref. 10) (Fig. 4). The electronic structure revealed by Zeeman data shows that the defect gives rise to a local strain field of tensional sign.[10] In such a case the binding energy of the electron is actually increased by the strain field. This is consistent with the strongly reduced diamagnetic shift observed in Zeeman data for this BE state,[10] which is only expected when the electron state is strongly localized.

Therefore a consistent picture emerges from these data for the possibility of binding excitons to neutral-complex defects with a hole-attractive central-cell potential in GaAs. If the local strain field at the defect is tensional, BE states are expected to be bound. On the contrary, defects causing a compressive local strain field may, in general, not bind excitons.

IV. ABSENCE OF STRONG AUGER EFFECTS IN BOUND-EXCITON SPECTRA

Auger effects in BE recombination are known to be quite important in indirect-band-gap materials, both for donors and acceptors.[16] The effect is particularly strong for deeply bound excitons where the bound particles are more localized. For direct-band-gap materials the radiative lifetime is usually much shorter ($\approx$1 ns, or even less), meaning that Auger processes could be of less importance. Detailed investigations on the Auger effects in DBE or ABE spectra for GaAs are not known to the authors, but have recently been reported for ZnTe.[34] In the ZnTe case Auger effects were concluded to strongly influence the observed BE lifetime for ABE's with rather high binding energy.[29] Returning to GaAs, our PL spectra show that the rather deep C and F ABE's are indeed quite strong in intensity (Fig. 1), indicating that Auger effects are *not* important for excitons bound to a single-acceptor ABE's in GaAs. This is contrary to the predictions made in recent literature.[16] As discussed above, the possible role of Auger effects for the double acceptor Cu_{Ga} is less obvious. Naturally, no Auger effects can occur with BE recombination for neutral defects, and therefore we would expect to see BE spectra from neutral defects in GaAs in PL spectra once they are able to bind excitons.

V. SUMMARY AND CONCLUSIONS

Bound excitons in GaAs have previously been studied in detail mainly for shallow substitutional donors and acceptors. From a study of Cu-related defects in GaAs, several conclusions can be drawn on the ability of deep-level defects as well as complex-type defects to bind excitons in GaAs. For double acceptors such as Cu_{Ga}, no ABE's are observed in PL spectra. For the case of the 0.15-eV acceptor level, interpreted as being connected with the second ionization level of Cu_{Ga}, the absence of a corresponding BE state is consistent with the general rule that charged acceptors do not bind excitons in GaAs. No evidence of a BE associated with the neutral state of Cu_{Ga} is seen either, which can be explained if this state also experiences a Jahn-Teller distortion such as that experienced by the singly ionized charge state. In any case, the third hole necessary for creation of a bound-exciton state for neutral Cu_{Ga} might easily be split off to be resonant with the valence band by the action of an axial strain field. With other Cu-related acceptors, BE's are observed in two cases, the so-called C and F lines, due to two different Cu-related complex-type acceptors. These are believed to be single acceptors, and the excitons are bound to their neutral charge states. Other Cu-related complex-type acceptors observed in our study probably do not bind excitons, however. This is believed to be caused by the delicate shallow binding of the electron in the BE state. This electron state is believed to be easily driven up in the conduction band by the action of a compressional local strain field at the defect.

For the case of neutral (so-called isoelectronic) defects, it is well known that such single substitutional atoms do not bind excitons in GaAs, i.e., the central-cell potentials for binding the primary particles are not strong enough to localize a state in the band gap. For complex-type defects consisting of more than one impurity atom, we have found one case where a neutral-(Cu-Zn)—related defect binds an exciton deeply (1.429 eV) in GaAs, in a *tensional* local strain field. This sign of the crystal field seems to be required in GaAs, in order to stabilize the electron state in the band gap for neutral complexes with hole-attractive central-cell potentials. In the cases where neutral complex defects are believed to give rise to a *compressive* local strain field, BE states are not seen in our study.

The experimental data for the cases where bound excitons associated with neutral acceptors are actually observed in this work are interesting since a high intensity of ABE lines is consistently seen. This means that Auger effects for ABE's associated with deep acceptors are not very important in GaAs, contrary to previous belief.

ACKNOWLEDGMENTS

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