Excited states of donor bound excitons in GaP

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We have investigated excited states of bound excitons in GaP:S and GaP:Se. The states were observed by using a near-band-gap tunable laser source to create a bound exciton in an excited state and by then monitoring the luminescence due to radiative recombination. The $S_{O'}$ line observed previously is identified to be an excitation of the hole to a higher-lying state about a core consisting of two electrons bound in a D^- configuration. We find two other transitions which are relatively weak in the no-phonon transition while they are comparatively strong in the phonon-assisted transitions. We interpret these lines as being due to excitations of the core electrons in the bound exciton.

I. INTRODUCTION

Luminescence from excitons bound to donors in GaP has been investigated by Dean.[1] This study primarily dealt with the ground state of the bound exciton for S-, Se- and Te-doped GaP. However, Dean also observed in absorption several excited states of the bound exciton. These included two states labeled by Dean as $S_{O'}$ and $S_{O''}$ which has a large oscillator strength compared to several other transitions which were observed and labeled S_A, S_B, S_C, and S_D. The nature of these states as well as the observed S_A, S_B, S_C, and S_D transitions has not been explained.[1]

Recently several authors have attempted to calculate the excited-state spectrum of bound excitons. Chang *et al.*[2] have calculated the energies of excited states of bound excitons in a spherical model as a function of the mass ratio of the electron and hole. Herbert has developed a detailed theory for which he presents results for the observed spectra in GaAs.[3] Ruhle and Klingenstein have applied a nonrigid rotational model to describe states observed in InP and GaAs.[4]

In this paper we report the results of a luminescence excitation spectroscopy which has enabled us to observe more detail in the excited-state structure of the bound exciton for donors in GaP. By monitoring bound-exciton luminescence and luminescence from deeper impurities we are able to distinguish between photon-assisted and no-phonon transitions of the lines previously observed by Dean. By considering the form of the optical transition matrix elements we can identify the nature of most of these transitions. The $S_{O'}$ line is identified to be an excitation of the hole to a higher-lying state about a bound-exciton core consisting of two electrons bound in a D^- configuration. The $S_{O''}$ is observed to be the sum of two lines, one of which is observed to be the TA-phonon-assisted complement of the S_A transition while the other line is also identified as being due to the excitation of the hole about a D^- bound-exciton core. The S_C line is the TA-phonon-assisted replica of a weak no-phonon transition which we observe. The S_D line is apparently the LA-phonon-assisted replica of the S_A transition. The S_A and S_C transitions are apparently associated with states which have an electron core with different symmetry than the ground state. We can show that such states may have weak transitions in the no-phonon region while having comparatively large transitions rates in the phonon-assisted regions. In addition, we also observe a peak which we interpret as being due to the no-phonon creation of free excitons (FE) associated with donor impurities.

This paper contains a description of the experimental procedure in Sec. II. The experimental results are presented in Sec. III. These results are interpreted and discussed in Sec. IV.

II. EXPERIMENT

The method used in this experiment is virtually identical to that used by Cohen and Sturge to study the excited states of excitons bound to nitrogen pairs in GaP.[5] A sample is optically pumped with a tunable source and the luminescence from a particular transition is monitored as the source is tuned. In this experiment the tunable source is a continuous dye laser with constant output power operating with the dye sodium fluorescein pumped with an Ar^+ laser.

The output of the laser was focused on the sample which was placed in a variable temperature Dewar. Luminescence from the sample was collected and passed through a grating monochromator and monitored with a C31034 photomultiplier. This method has several advantages over normal methods of studying the absorption. First, the laser has a very narrow linewidth with high brightness compared to a thermal source filtered with a spectrometer. Second, the luminescence is proportional to the light absorbed and is relatively insensitive to small fluctuations in the laser inten-

sity. This feature makes it possible to observe weak features in the absorption spectrum. Finally, the laser spot can be focused to a very small size and thus samples a very small region within the material. Thus small regions in bulk material which have less internal strain and yield better overall spectra can be studied.

In this study we present results from samples doped with S and Se. The spectrum of the GaP:Se indicated that there was a negligible contribution from sulfur in this sample. Because additional structure was observed in samples contaminated with nitrogen which made the results uninterpretable, only samples with low nitrogen content were used. The doping level was estimated to be approximately 3×10^{17} for the GaP:Se sample and 5×10^{16} for the GaP:S sample.

III. RESULTS

Figures 1 and 2 show the luminescence excitation spectra obtained from GaP doped with S and Se. We monitored the luminescence from two spectral regions. In the top panel of each figure the luminescence, which is monitored as the dye laser is scanned, appears at 1.77 eV. This luminescence occurs in a broad emission band peaked at approximately 7000 Å which was observed in both the case

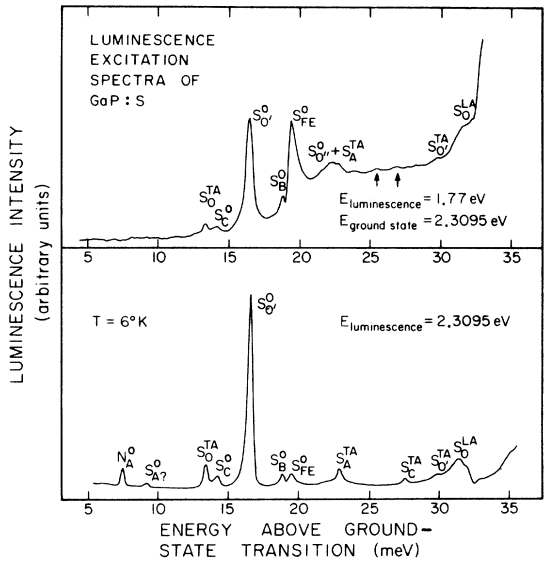

FIG. 1. The luminescence excitation spectra for GaP:S. In the top panel luminescence is monitored at 1.77 eV. In the bottom panel luminescence is monitored at 2.3095 eV, the energy of the ground-state transition. The two small arrows in the top panel point out weak features in the spectra which are discussed in the text. The ground-state transition is not shown in this figure but is approximately four times stronger than the $S^0_{0'}$ transition.

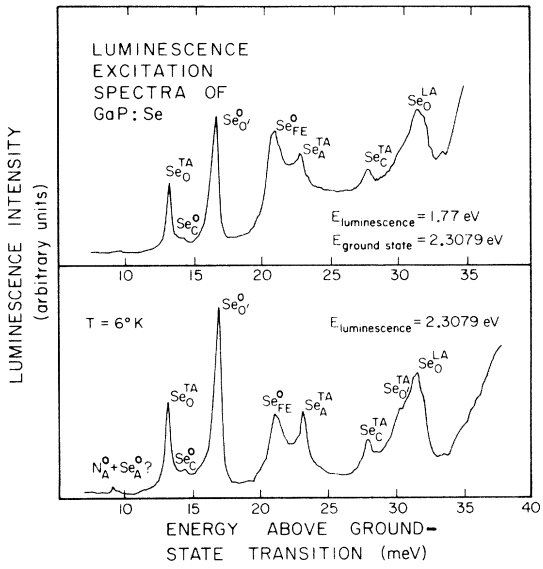

FIG. 2. The luminescence excitation spectra for GaP:Se . In the top panel luminescence is monitored at 1.77 eV. In the bottom panel luminescence is monitored at 2.3079 eV, the energy of the ground-state transition. The ground-state transition for GaP:Se is about 1.6 meV lower in energy than the corresponding transition in GaP:S. The ground-state transition is not shown in this figure but is approximately four times stronger than the $Se^0_{0'}$ transition.

of GaP:S and GaP:Se. Such an emission band is probably associated with recombination from a deep center.[6] Presumably, excitations, which are created by light absorbed in the sample, decay rapidly into states producing the broad band in the spectrum. If such excitation transfer occurs with uniform efficiency for all excitations created in the crystal, then the observed luminescence spectrum accurately portrays the absorption spectrum. If such excitation transfer does not occur with uniform efficiency, then the observed spectrum will be influenced by the details of the transport process of the excitation to the luminescent center. To evaluate the effects of such transport, we measured the optical absorption spectra directly in the GaP:Se sample. Although the quality of the absorption spectra was not as good as that obtained by using the tunable laser, the details of the spectra were basically the same in both cases. In addition the observed spectra are relatively independent of temperature. We thus must assume that the details of the spectra are not influenced by the transport process but rather reflect changes in the optical absorption.

We also measured the luminescence of the ground-state no-phonon bound-exciton transition as a function of pump wavelength from the dye laser. For GaP:S this peak occurs at 2.3095 eV and for GaP:

Se it occurs at 2.3079 eV. In the bottom panels of Figs. 1 and 2 we show these spectra. Such luminescence is also affected by excitation transfer, especially from highly excited states. Excitation into a state which is not bound has a large probability of decaying to a deeper level, whereas a state which is tightly bound has a much lower probability of such a transfer. States which are loosely bound have a higher probability of thermalizing to the free-exciton continuum than deeper lying states. By comparing these spectra with those obtained from the previous method where the luminescence from deeper levels was monitored, valuable information on the nature of the states can be obtained.

Figure 1 shows the spectra for GaP:S. In addition to features previously observed by Dean, these spectra contain additional lines which enable us to interpret much of the structure. We have labeled these lines in accordance with the interpretation which we give later in the paper. In the top panel of Fig. 1 we show a spectra for GaP:S where luminescence at 1.77 eV was monitored. At energies 13.4±0.2 meV and 14.2±0.2 meV above the ground state, we see two weak lines labeled S_O^{TA} and S_C^o. The energy of the first line matches that of the TA-phonon-assisted ground-state transition. In addition, at higher energies we see two strong lines labeled $S_{O'}^o$ and $S_{I\text{-}F}^o$, a weaker line labeled S_B^o and a broad feature labeled $S_{O'}^o + S_A^{TA}$. There is a structure at 31.4 meV above the ground state which is apparently associated with the LA-phonon-assisted bound-exciton transition. The line labeled $S_{O'}^o$ is apparently the same as the S_0, transition observed by Dean.[1]

In the bottom panel of Fig. 1 we show a spectrum for GaP:S, where luminescence at 2.3095 eV was monitored. This spectrum is more detailed than the first and in addition the strong continuum above 2.33 eV is much weaker. We interpret this as being due to the fact that this continuum is primarily due to the no-phonon creation of free excitons scattering off the neutral donor. Such states will have a high probability of decaying to deeper levels and only give rise to weak luminescence at 2.3095 eV. In addition to lines observed in the top panel we see lines at 7.7±0.2 meV, 9.2±0.2 meV, and 27.5 meV above the ground state. The latter two lines are most likely the S_A and S_C lines observed by Dean.[1] The 7.7 meV line is most likely associated with the nitrogen N_A^0 line which occurs at the same energy. This line is weak in the top spectra and is in general stronger in all samples when donor bound-exciton luminescence is monitored than when the deeper luminescence is monitored. This may indicate that nitrogen preferentially transfers to the deeper donor bound-exciton levels. We have observed considerable structure in the exciton spectra at the energy of the N_A line in samples with a higher nitrogen content. A line in the region of the $S_{O''}^o$ line occurs at 22.9 ±0.2 meV above the ground state. This energy is slightly higher than the $S_{O''}^o$ transition observed in the top panel and the line is much sharper. It appears likely that the line observed in the top panel is actually the sum of two lines $S_{O''}^o$ and S_A^{TA} of which the $S_{O''}^o$ is relatively broad and at slightly lower energies than the S_A^{TA} which is the line observed at 22.9 meV. In addition the line labeled $S_{I\text{-}F}^o$ at 19.5 meV is much weaker in there spectra.

This behavior suggests that these peaks are associated with states which decay into free excitons with high probability since they are weak in the lower spectra. The higher-lying transitions which are observed at 22.9 and 27.5 meV and labeled S_A^{TA} and S_C^{TA} are not particularly weak and thus are more likely due to TA-phonon-assisted transitions. The separation in energy between these lines and those at 9.2 and 14.2 meV suggests that the lower-energy lines are possibly the corresponding no-phonon transitions. In addition the S_D line matches the energy of the LA-phonon replica expected for the S_A transition and, as mentioned by Dean, appears to track the phonon-assisted components in intensity.[1] As we will discuss later the interpretation of the higher states as being phonon assisted in strongly supported by similar data observed for GaP:Se. In Fig. 1 we have labeled these transitions in accordance with this interpretation, and we listed the corresponding lines and their positions in Table I.

Figure 2 shows the luminescence excitation spectra for GaP:Se. As in the spectra for GaP:S the top panel shows the spectrum for luminescence monitored at 1.77; the bottom panel shows the spectrum for luminescence monitored at 2.3079 eV, the energy of the ground-state no-phonon bound-exciton transition. Close inspection shows that much of the structure in this spectrum matches that of the GaP:S spectra if the energy scale is shifted by 1.6 meV, the difference in energy between the GaP:S and GaP:Se bound-exciton ground states. We thus note a one-to-one correspondence between the S_A^{TA}, S_C^{TA}, $S_{O'}^o$, and S_C^o transitions and corresponding transitions for GaP:Se. A simple observation confirms that the S_A^{TA} and S_C^{TA} transitions are phonon assisted. In the Se-doped samples the phonon-assisted transitions are much stronger than the no-phonon components. However, the ratios between the S_A^{TA} and S_C^{TA} line strengths and the S_O^{TA} ground-state transition line strength are nearly the same as for the ratios between the corresponding lines observed in GaP:Se even though the ratio between no-phonon and TA

TABLE I. Energies measured for features in the luminescence excitation spectra of GaP:S. The maximum possible error in the measurements is 0.2 meV. The separation between the weak feature labeled S_A^o and S_A^{TA} is slightly larger than the observed TA-phonon energy and therefore the S_A^o transition is labeled with a question mark.

	Energy (eV)	Energy relative to S_O^o (meV)
S_O^o	2.3095	0
S_B^o,	2.3261	16.6
S_O^o"	2.3319	22.4
S_{FE}^o	2.3290	19.5
S_A^o	2.3187	9.2?
S_B^o	2.3283	18.8
S_C^o	2.3237	14.2
S_O^{TA}	2.3229	13.4
S_O^{TA},	2.3394	29.9
S_A^{TA}	2.3324	22.9
S_C^{TA}	2.3370	27.5
S_O^{LA}	2.3409	31.4

components has changed by a factor of 5. This suggests very strongly that these transitions are phonon assisted and explains why such states can appear above the FE threshold in the spectra shown in the lower panels of Figs. 1 and 2.

The peak which is labeled Se_{FE}^o appears in the spectra for GaP:Se at 21.1 meV above the ground state and occurs at the same position in energy as the peak in GaP:S which is labeled S_{FE}^o which occurs 19.5 meV above the ground state. Both of these peaks occur very close to the free-exciton threshold. It is likely that these peaks are associated with the creation of free excitons in the vicinity of the donor impurity. The fact that both of these are weaker than the S_O^o, peak in the spectra shown in the lower panels of Figs. 1 and 2 than in the top panels supports this conclusion. We have listed the lines which we have observed for the GaP:Se and their positions in Table II.

IV. DISCUSSION

Of the available calculations for the excited-state structure of bound excitons, Chang's calculation[2] is the only one carried out for the appropriate mass ratio. Chang's theory suggests that for mass ratios of electron to hole greater than 0.5, the excited-state spectrum closely resembles that of a neutral acceptor. In such a case the excited state can be roughly pictured as a hole excited about the two electrons which are bound in a

TABLE II. Energies measured for features in the luminescence excitation spectra of GaP:Se. The maximum possible error in the measurements is 0.2 meV. The feature Se_A^o is very weak and occurs near the N_A^o nitrogen no-phonon bound-exciton transition. Therefore the assignment of this feature as being due to Se must be considered speculative.

	Energy (eV)	Energy relative to S_O^o (meV)
Se_O^o	2.3079	0
Se_O^o,	2.3248	16.9
Se_{FE}^o	2.3289	21.0
Se_A^o	2.3175	9.6?
Se_C^o	2.3224	14.5
Se_O^{TA}	2.3211	13.2
Se_O^{TA},	2.3380	30.1
Se_A^{TA}	2.3309	23.0
Se_C^{TA}	2.3357	27.8
Se_O^{LA}	2.3393	31.4

D^- core. In GaP we can make an estimate of the appropriate mass ratio to use in the calculation of Chang by considering the ratio of the donor binding energy to the acceptor binding energy for simple impurities. If we take 100 meV as a typical donor binding energy and 50 meV as the simple acceptor binding energy we obtain a mass ratio of 2 which is considerably greater than 0.5. Thus it seems most likely that we can explain the data in a model which has the features of the excited states described by Chang.

Since the band structure is considerably more complicated than the spherical nondegenerate bands used by Chang in his calculation, it is necessary to consider what effect the details of the band structure will have on the excited-state spectrum of the bound exciton. Since the conduction band in GaP has several equivalent minima, it is possible to construct several D^- states of different symmetry. In addition the excitations of the neutral acceptor are not simply described by a hydrogenic spectrum but have a more complicated form. From Chang's calculation we expect that the nature of the excited states in the bound excitation will be similar to the excited states of the neutral acceptor and can therefore be classified according to the scheme developed by Baldereschi and Lipari.[7]

If we consider the form of the optical transition matrix element we see that some of these states are expected to have relatively large oscillator strengths in no-phonon transitions, whereas, others are expected to have relatively weak transitions in the no-phonon region.[8-10] The neutral

donor and the donor bound-exciton wave functions have the form

$$|\psi_0\rangle = \sum_{\vec{k}_e^0 v_0 \sigma_0} a^0_{v\sigma}(\vec{k}_e^0) C^\dagger_{\vec{k}_e^0 v\sigma} |G\rangle \quad (1)$$

and

$$|\psi^j_{BE}\rangle = \sum_{m, v_1\sigma_1, v_2\sigma_2} \sum_{\vec{k}_e^1, \vec{k}_e^2, \vec{k}_h} A^j_{m, v_1\sigma_1 v_2\sigma_2}(\vec{k}_e^1, \vec{k}_e^2, \vec{k}_h) \times C_{\vec{k}_h m} C^\dagger_{\vec{k}_e^1 v_1\sigma_1} C^\dagger_{\vec{k}_e^2 v_2\sigma_2} |G\rangle . \quad (2)$$

$C_{\vec{k}_h m}$ creates a hole with wave vector $\vec{k}_h$ and spin projection m, $C_{\vec{k}_e^1 v_1\sigma_1}$ creates an electron with wave vector $\vec{k}_e^1$ in a valley combination v_1 with spin σ_1, and $|G\rangle$ is the state with all conduction-band states empty and all valence-band states occupied. a^0 is the amplitude function for the ground state of the neutral donor and A^j is the amplitude function for the jth excited state of the donor bound exciton. The transition matrix element becomes

$$\langle\psi_0|P_y|\psi^j_{BE}\rangle = \sum_{m, v_1\sigma_1, v_2\sigma_2} \sum_{\vec{k}_e, \vec{k}_h} \langle\phi_m(\vec{k}_h)|P_y|\phi_{\sigma_2 v_2}(\vec{k}_h)\rangle \times a^{0*}_{v_1\sigma_1}(\vec{k}_e)[A^j_{m, v_1, \sigma_1, v_2, \sigma_2}(\vec{k}_e, \vec{k}_h, \vec{k}_h) - A^j_{m, v_1\sigma_1, v_2\sigma_2}(\vec{k}_h, \vec{k}_e, \vec{k}_h)], \quad (3)$$

where $|\phi_m(k_h)\rangle$ and $|\phi_{\sigma_2 v_2}(k_h)\rangle$ are the periodic parts of, respectively, the pseudo-Bloch function of the hole and the Bloch function of the electron, and P_y is the y component of the momentum operator. We can expand the functions $A^j_{m, v_1\sigma_1, v_2\sigma_2}$ terms of the amplitude functions of the neutral donor $a^p(k)$ (where p labels the state of the neutral donor) and for the single-particle amplitude function the hole $b^r(k)$ (where r labels the basis states).

To illustrate the important features of this equation, we can suppress most of the indices and write

$$\langle\psi_0|P_y|\psi_{BE}\rangle = \sum_{pqr} \left\{ D(p,q,r) \left[\sum_{\vec{k}_e} a^{0*}(\vec{k}_e) a^p(\vec{k}_e) \right] \times \left[\sum_{\vec{k}_h} M(\vec{k}_h) a^q(\vec{k}_h) b^r(\vec{k}_h) \right] \right\}, \quad (4)$$

where $M(\vec{k}_h)$ is the momentum matrix element, and D is the set of expansion coefficients for the bound-exciton wave function. This equation shows that the matrix element is the product of two terms in brackets: the overlap in k space between the initial state of electron in the bound exciton and the electronic state of the neutral donor, and the overlap in k space of the electron and hole amplitude functions weighted by the momentum matrix elements. The first term in the product is zero unless the two single electron states involved are the same. The second term can be approximated by noting that the hole wave functions are relatively localized in k space at $\vec{k}_h = 0$ and that the momentum matrix element is not rapidly varying function of $\vec{k}_h$.[11] Then this term is approximately

$$M(\vec{k}_h = 0) \sum_{\vec{k}_h} a^p(\vec{k}_h) b^r(\vec{k}_h).$$

Hence, the size of this term depends on the k-space overlap of the electron and hole wave functions near the zone center. In addition, if the amplitude functions $a^p(\vec{k}_h)$ and $b^r(\vec{k}_h)$ have opposite parity this sum will be zero.

For GaP, these considerations lead us to expect that only certain bound-exciton states will be involved in the no-phonon absorption process. These states are those with a substantial projection on the single-particle configuration consisting of two electrons in $1s(\Gamma_1)$ states and the hole in an even-parity state. One of the electrons must be in a $1s(\Gamma_1)$ state because the initial state in absorption is the $1s(\Gamma_1)$ ground state of the neutral donor. In addition the $1s(\Gamma_1)$ electron state is the only state which is sufficiently spread out in k space to have a large overlap with the hole. Thus, the second electron must also be in the $1s(\Gamma_1)$ state for the second term in Eq. (4) to be appreciable. The hole must have the same parity as the electron and therefore has even parity. We thus expect transitions to hole levels with p-like rotational symmetry to be very weak.

There are three features in the excited-state spectrum which have fairly large oscillator strengths in the no-phonon region for GaP:S and two such features for GaP:Se. These are the lines $S^o_{O'}$, S^o_{FE}, and $S^o_{O''}$ for GaP:S and the lines $S^o_{O'}$ and Se^o_{FE} for GaP:S. On the basis of the preceding arguments, we would expect the $S^o_{O'}$ and the $Se^o_{O'}$ lines to be due to the creation of a bound exciton with a $D^-(\Gamma_1)$ core and the hole excited in a $2s_{3/2}$ state. The $S_{O''}$ line, as pointed out earlier in this paper, appears to be the sum of two lines which we have relabeled S^{TA}_A and $S^o_{O''}$. This transition is possibly due to the creation of a bound exciton with a hole excited in a $3s_{3/2}$ state.

Cohen and Sturge[5] have observed acceptor-like excited states of excitons bound to nitrogen pairs

in GaP. Since we expect the highly excited hole states of both the N and S bound excitons to have the same relative energy positions, we can compare our results with theirs. Cohen and Sturge observed a $3s$ state which was consistently between 6 and 7 meV above the $2s$ state. Such a separation agrees with our assignment of the S_0^0 and $S_{0''}^0$ transitions.

In Fig. 1 we have marked two features with small arrows in the luminescence excitation spectrum for GaP:S which match the same relative positions as the $4s$ and $5s$ states reported by Cohen and Sturge, and must tentatively be considered as candidates for such transitions for the case of GaP:S.[5] Such states are technically resonant states since they lie above the threshold for free-exciton formation. If we assume that the relative energies of the excited-state spectrum for GaP:S match that of the acceptor-like spectrum observed by Cohen and Sturge, we obtain a hole ionization energy of 30 ± 1 meV. Assuming a free-exciton binding energy of 22 ± 2 meV and a bound-exciton binding energy of 20 ± 1 meV, we obtain a value of approximately 12 meV for the binding energy of an electron in the $D^-(\Gamma_1)$ state.

The bound-exciton excited states associated with the S_A^{TA}, S_C^{TA}, Se_A^{TA}, and Se_C^{TA} transitions are most likely states with an electron core with overall symmetry different from Γ_1. Such states have been previously reported for excitons bound to donors in silicon.[12,13] Preliminary calculations by Chang indicate that D^- states with Γ_3 symmetry and a strong projection on the single-particle product $1s(\Gamma_1)\ 1s(\Gamma_3)$ have the correct energy to be responsible for these transitions.[14] Such states are expected to have weak no-phonon components since the $1s(\Gamma_3)$ state is not sufficiently localized to have significant overlap with the hole in momentum space. If a transition is phonon assisted, such an overlap is no longer necessary and the oscillator strength should be comparable to the phonon-assisted ground-state transition.[8,9] Two such states occur for the $D^-(\Gamma_3)$, one being an electron-spin singlet, the other being a spin triplet. When coupled to a Γ_8 hole these states split in energy into a multitude of possible final states for the bound-exciton transitions. It is also possible that these states are associated with p-like rotational states associated with the hole. However, there is no compelling reason to believe that such rotational states should have large oscillator strength in phonon-assisted transitions.

In conclusion, we have observed the excited-state structure of bound excitons in GaP:S and GaP:Se and are able to distinguish between phonon-assisted and no-phonon transitions of lines which were previously observed by Dean. We can identify the nature of most of these lines. There are basically three different types of features in the spectra. We have observed peaks which appear to be due to the no-phonon creation of bound excitons in the vicinity of the impurity. In addition, we have identified several transitions as being due to the excitation of the hole about a $D^-(\Gamma_1)$ electron core. The third type of transition is to states with an electron core with different symmetry. Such transitions are much stronger when a momentum-conserving phonon is involved.

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