such conversion processes; similar studies using optical information have in the past been plagued by uncertainties in the oscillator strength of the Eu^{2+} configurations.[12] These studies suggest that almost all production techniques of divalent-rare-earth ions in CaF_2 do lead to the introduction of oxygen into the lattice, so that these oxygen-stabilized configurations must be generally taken into account. Further studies on these complexes and their relationship to the anomalous isomer shifts reported in Refs. 1–3 are in process.

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Magnetic Field and Stress-Induced Splitting of the Novel Sharp Emission Line Series in Silicon Associated with P, Li, or B: No Bound Multiple-Exciton Complexes

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We report Zeeman spectra and the stress-induced splitting of the three novel sharp emission line series in silicon which are associated with the neutral impurities phosphorus, lithium, or boron. The splitting of all lines belonging to one series is identical except for thermalization effects. This result cannot be explained by the model of bound multiple-exciton complexes but rather suggests the radiative decay of single bound excitons as the origin of the lines. A detailed new model is not evident.

New photoluminescence line series spectra in phosphorus-, lithium-, or boron-doped silicon a few milli-electron-volts below the associated bound-exciton emission were recently attributed to the radiative decay of excitons from multiple-exciton complexes bound to the isolated neutral impurities.[1-4] This model suffered from a number of features which could hardly be understood. Among these are the strong binding of excitons in the complexes (up to ≈15 meV) by far exceeding the ≈5-meV binding of the single exciton to the neutral impurity, and the simplicity of the experimental spectra which is surprising compared to what was expected from such complicated many-particle complexes. New decisive experiments were desirable in order to efficiently support or contradict this model.

In this paper we present Zeeman spectra and stress-split spectra of the line series for phosphorus-, lithium-, or boron-doped silicon which unambiguously contradict the model of bound-multiple-exciton complexes.

In Si:P we have measured the Zeeman spectra of the first four lines of the phosphorus-induced series (Fig. 1). The bound-exciton (BE) line splits into six components, and the whole pattern highly resembles the BE spectrum of Si:As at 91.6 kG which was described by Cherlow, Aggarwal, and Lax[5] in their investigation of electronic g values. In a few details only, e.g., concerning the polarization features, those spectra differ from ours. Therefore, the pattern is sim-

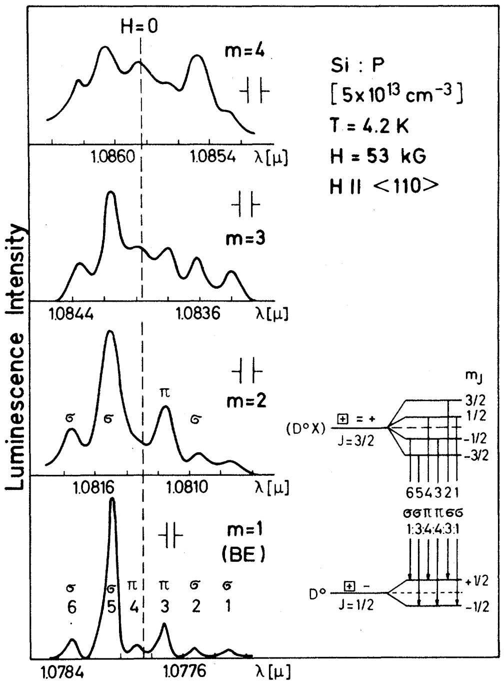

FIG. 1. Zeeman spectra of the $m_1, \ldots, m_4$ no-phonon-(NP)-assisted emission lines of the phosphorus-induced series. The line positions at zero magnetic field (dashed line $H = 0$) are taken as the basis for the comparison. Polarizations refer to transverse observation and could not be measured if not indicated. Relative line intensities as given in the transition level scheme refer to nonthermalized upper states (Ref. 6). All spectra were excited by an unfocused Ar-ion-laser beam ($\lambda = 514$ nm, ≤ 1.5 W).

ilarly well explained by the simple level scheme used by these authors (Fig. 1), which couples "atomic" angular momenta without regard for the special band-structure symmetries.[6] Using the g values of Ref. 5, the magnetic field strength $H = 53$ kG, and an effective temperature $T = 5$ K, we construct a spectrum in good agreement with the observed BE Zeeman pattern.

The Zeeman spectra of the low-energy m_2, m_3, m_4 lines are quite similar to the BE pattern. Each line splits into six components, and corresponding components have comparable spacings to their associated zero-field lines. The polarization features, as far as measurable because of weak intensities, are identical to the BE pattern (Fig. 1). The relative intensities of the components belonging to adjacent m lines resemble each other, but the principal component 5 systematically decreases for the higher indexed lines whereas the high-energy components increase. These properties suggest that the transition level scheme as shown in Fig. 1 applies to *all m* lines. Different thermalization in the fourfold, upper $J = \frac{3}{2}$ states tending to a more uniform occupation of the sublevels for higher line indices m may be responsible for the intensity variation of the m-line components.

Evidently, the model of bound multiple-exciton complexes cannot account for the simplicity and similarity of these spectra. Instead, we conclude that all lines originate from the radiative decay of a single exciton bound to the impurity. In comparison to the BE line, the energy loss of the residual m-line transitions has to be explained. This point is discussed below.

The investigation of the lithium- and boron-induced line series confirms the conclusions drawn above. For lithium which couples only weakly to the NP-line series,[3] we have investigated four lines of the TO spectrum which by phonon participation are considerably broadened. The Zeeman pattern which we observed is identical to the corresponding spectra of P considering that the linewidths are much larger and that therefore weak components are not resolved. As a difference we note that all observed Li lines are unpolarized. For boron the TO-phonon-assisted spectrum predominates the NP-line series by a factor of ≈ 20.[7,8] Nevertheless, we were able to obtain a well-resolved Zeeman pattern of three lines in the NP spectrum (Fig. 2). The BE line splits into four essentially unpolarized components equally spaced from each other. At a temperature of $T = 2$ K component 4 becomes about 1.7 times larger than component 3, whereas component 2 only slightly increases. This temperature dependence indicates that at least two magnetic sublevels are involved in the initial state of the BE transition. On the other hand, from the properties of the bound hole we expect the final state to be a fourfold-split $J = \frac{3}{2}$ level. For excitons bound to neutral acceptors (Cd, Zn) in GaP, Dean *et al.*[9] could interpret the Zeeman spectra with transitions between magnetically split $J = \frac{1}{2}$ and $J = \frac{3}{2}$ states. Although our data suggest a similar transition level scheme, we cannot explain the spectra by such a simple model. This point needs further investigation. Despite the lack of an appropriate level scheme, in connection with our problem only, the striking similarity of the m_2 and m_3 line spectra with the BE spectrum is important. Again, we note identical features, viz. number of components, equal spacings of corresponding components relative to

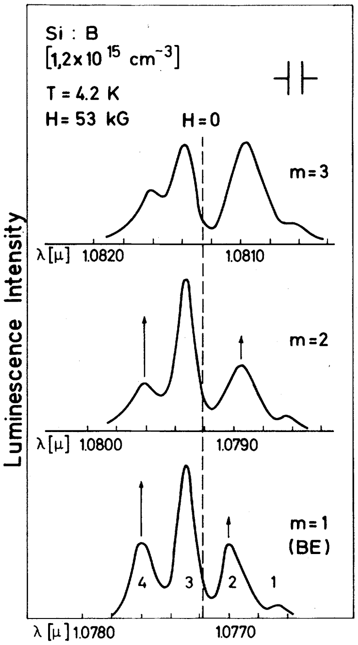

FIG. 2. Zeeman spectra of the m_1, m_2, and m_3 no-phonon-assisted boron lines. m_1 and m_2 line components are not polarized; the polarization of the m_3 line components was not measured. The arrows indicate the increase of components 2 and 4 relative to component 3 at a temperature of 2 K (see text).

the zero-field lines, identical polarization properties, a systematic shift of maximum intensity to the high-energy components, and the same temperature dependence of the spectra.

Finally we have measured identical splittings of the first four lines of the phosphorus-induced series when uniaxial stress is applied. Part of our results are presented in Fig. 3. Details are given in the figure caption. We emphasize that apart from the double splitting the same particular properties are found as were pointed out for the Zeeman measurements. We have further performed experiments with nonuniaxial stress and have observed three components in all lines with identical polarization and temperature-dependent features. These triplets showed again similar spacings and the patterns were uniformly positioned relative to the zero-stress lines.

Although the data presented discard the model of bound multiple excitons in favor of the radiative decay of single-bound excitons, we have not yet succeeded in substituting a complete new model. If we take for granted that isolated impurities are responsible for the lines as was pointed out previously, the different dependence on the excitation level and the distinct decay times of the m-line intensities prove that different initial states are involved.[2-4] Since the BE line originates from the recombination of an exciton bound to the impurity ground state, the other m lines must be associated with excited states of the exciton-impurity complex. Excited states would be consistent with the observed temperature-dependent behavior of the lines which vanish in succession more rapidly for increasing m indices when the temperature is raised.[3,8] Considering the energy loss of these lines relative to the BE transition, we must assume that the impurity is left in an excited state after the excitonic recombination. With regard to such a model we neglect vibrational or rotational levels of the impurities. The former is excluded since the line series are similarly observed for phosphorus, lithium, boron, and arsenic,[10] and their energetic positions and kinetic properties are essentially in-

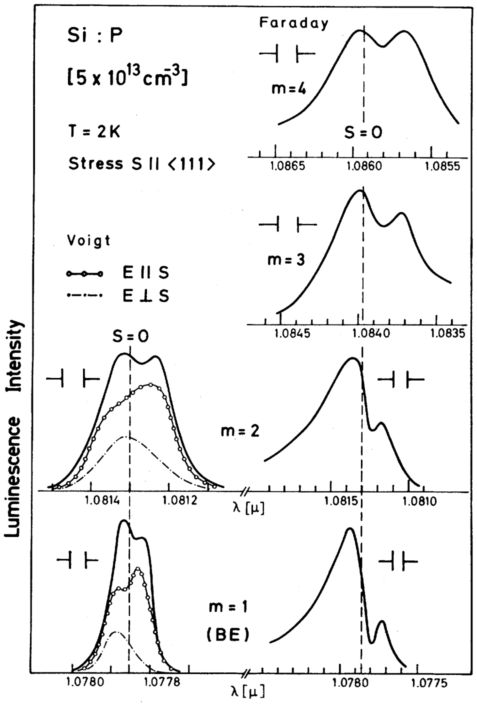

FIG. 3. Uniaxial stress-induced splitting of the $m_1, \ldots, m_4$ no-phonon-assisted phosphorus lines. The spectra are shown relative to their zero-stress positions (dashed line $S=0$). Spectra in Faraday configuration show no linear polarization; they refer to an applied stress of ≈ 59 kp/cm^2 [1 kp (kilopond) = 9.806 65 N]. Spectra in Voigt configuration are polarized as indicated; full lines give the total emission intensities; the polarization of the weak m_3 and m_4 line components could not be measured. The stress is ≈ 10 kp/cm^2. For all lines the high-energy components increase when the temperature is raised to 4.2 K.

sensitive to the mass defect or to the lattice site of the impurity. For phosphorus, e.g., we would under no circumstances expect individual local vibrational modes. The latter is excluded since the 1*s* (A_1) ground states of P or As would not allow the existence of rotational levels by the dynamic Jahn-Teller effect.[11] Finally, we also encounter difficulties with a model of excitons which are bound to electronically excited impurities forming an excited impurity-exciton complex and leave the impurity in the excited state after the radiative transition[12]: From the level scheme applying to the magnetic splitting of the phosphorus lines (Fig. 1), *s* levels of the impurity have to be adopted. Since for donors these expected final excited *s* states are accurately known theoretically[13] and experimentally,[14] we can calculate the energies of the corresponding initial states by simple addition of the related line-emission energies. By this procedure the initial-state energies in the model turn out to be nonmonotonic as a function of the line number, but have a maximum at line indices 3 or 4. This seems very strange to us. Obviously, severe difficulties also arise with an interpretation when interactions of the bound-exciton complex with adjacent impurities or coupling to phonons are taken into account.

To conclude, we have shown that the model of bound multiple-exciton complexes has to be discarded, but that the novel line series originate from the radiative decay of an exciton bound to the impurity. By consideration of the excitation-, doping-, temperature-, and time-dependent behaviors of the line intensities a model of excited exciton-impurity complexes seems to be favored, but the detailed nature of the initial and final states involved is far from being transparent.

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