Absorption due to the creation of bound excitons in phosphorus-doped silicon

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LETTER TO THE EDITOR

Absorption due to the creation of bound excitons in phosphorus-doped silicon

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Abstract. New lines have been detected in the absorption spectrum of P doped Si associated with the creation of bound excitons in an excited state. The positions and strengths of these lines are in agreement with corresponding features recently identified in the luminescence spectrum. These results support the shell model for bound exciton and multiple bound exciton complexes.

The luminescence due to the recombination of excitons bound at neutral donor centres in Si takes the form of a series of sharp no-phonon (NP) lines a few meV below the free exciton (FE) energy gap with momentum conserving phonon replicas occurring at lower energies (Dean *et al* 1967a, Kaminskii *et al* 1971, Sauer 1973, 1974, Kosai and Gershenzon 1974, Sauer and Weber 1976, Thewalt 1977a, b). The strongest of these lines is associated with the recombination of a single bound exciton (BE) whilst the weaker satellite lines have been ascribed to exciton recombination within multiple bound exciton (MBE) complexes. A model capable of accounting for all the experimentally observed phenomena, in particular Zeeman and uniaxial stress splitting patterns, has proved to be elusive (Dean *et al* 1976 and references therein). Recently, however, Kirczenow (1977a, b) has proposed a new model for BE and MBE complexes, incorporating the tetrahedral symmetry of the donor site and the band structure, which appears to account for the Zeeman and uniaxial stress data and the large apparent binding energies of the MBE complexes. In this shell model, BE and MBE complexes are considered to have bound excited states, and the model predicts that the luminescence spectra should contain more lines than had hitherto been observed. Detailed high-resolution measurements by Thewalt (1977a, b) on the BE and MBE luminescence from lightly doped Si : P have revealed the existence of these previously unreported lines. In addition, the relative strengths and the temperature dependence of the BE and MBE lines were shown to be consistent with the shell model.

Figure 1 shows the energy level scheme proposed by Kirczenow (1977a, b) for the neutral donor D^0 and the first bound exciton D^0X. Electrons occupy states with Γ_1, Γ_3 or Γ_5 symmetry and the holes occupy Γ_8 states. A splitting between the Γ_3 and Γ_5 states is shown for D^0 but not for D^0X—this point will be discussed later. Only the valley–orbit split 1s states are shown for D^0 and these are identified by the notation used in earlier studies (Kohn 1957). Higher excited states of D^0 are not shown.

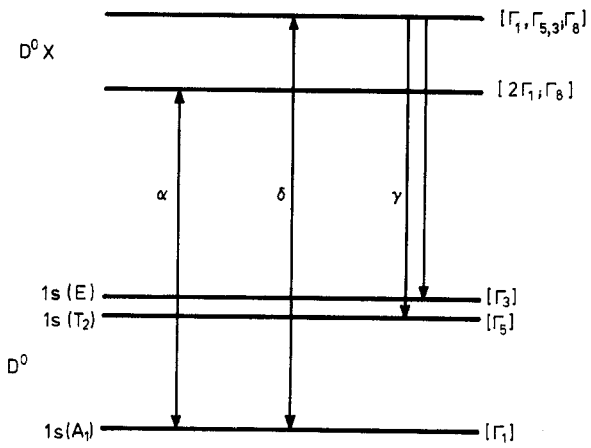

Figure 1. Energy level scheme for the neutral donor and single bound exciton predicted by the shell model (Kirczenow 1977a).

On this model the BE luminescence should consist of the α, δ and γ transitions, as shown in the figure, with the δ and γ lines both showing thermalisation with respect to the α line. The α line is the well known principal BE line. The δ line, however, had not been previously reported but was identified by Thewalt (1977b) in the TA and TO replicas with a peak which occurs on the side of the FE(TA) and FE(TO) bands, respectively. The TO replica was also observed by Vouk (1976) in P, As and Sb doped Si but was identified with FE recombination in the vicinity of a neutral donor (Dean *et al* 1967a, Nishino and Hamakawa 1975). Lines previously interpreted as FE two-electron transitions (Dean *et al* 1967a) were identified by Thewalt with the γ transitions. According to the measurements reported by Thewalt, the infinite temperature intensity ratio of the δ and α lines in the TA replica is $\delta(\text{TA})/\alpha(\text{TA}) = 9{\cdot}2 \pm 0{\cdot}8$. The no-phonon δ(NP) line was not observed by Thewalt.

Since the α and δ transitions both terminate on the ground state of the neutral donor, they should both be present in absorption and, on the basis of Thewalt's luminescence measurements, with the δ line stronger in the phonon replicas than the α line. Both α(NP) and α(TO) have been observed in the absorption spectrum of P doped Si by Dean *et al* (1967b,) but the δ line was not reported. Nishino and Hamakawa (1975) Nishino *et al* (1977) have reported structure in the wavelength derivative absorption spectrum close to the FE(TA) and FE(TO) thresholds, but with insufficient resolution to determine the intensities or exact transition energies. They identified the structure with the creation of FE in the vicinity of donor centres. Clearly, a crucial check on the shell model of BE and MBE complexes is to establish whether the δ transition takes place in absorption and with what strength relative to the α transition.

In this Letter we report absorption measurements on a P doped Si sample (Wacker) with $N_D \sim 8 \times 10^{22}\ \text{m}^{-3}$. Since the absorption is known to be very weak (the total oscillator strength for the α lines is $\sim 7 \times 10^{-6}$—Dean *et al* 1967b) a large optical path length is necessary. We employed a sample 49 mm in length immersed in liquid helium pumped below the λ point. The radiation source was a tungsten iodine lamp and the transmitted radiation was dispersed by a Spex 1701 monochromator, with a 1200 lines/mm grating blazed at 1·2 μm, and detected by a cooled S1 cathode photomultiplier (EMI

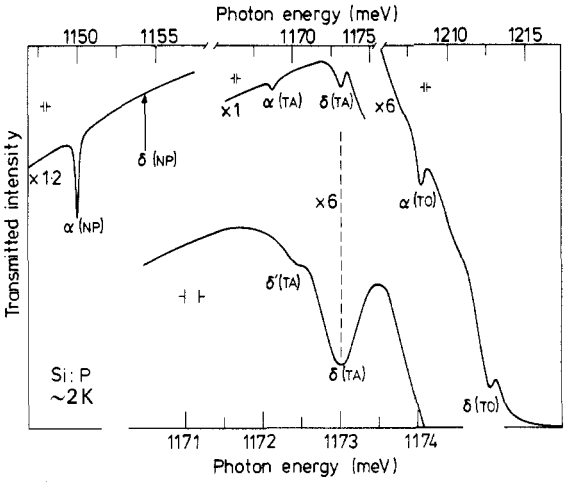

Figure 2. Transmission spectrum of P doped Si ($N_D \sim 8 \times 10^{22}$ m^{-3}) with a path length of 49 mm, uncorrected for system response. The transmitted intensity is measured in arbitrary units which are different for each of the four spectra shown. Approximate sensitivity changes are indicated in the figure.

9684) with magnetic focusing. Figure 2 shows the NP, TA and TO regions of the transmitted spectrum, not corrected for system response. Each region was investigated on an expanded transmission scale and the absorption spectrum was calculated taking full account of multiple internal reflections. The peak energies of the absorption transitions and their integrated areas are listed in table 1.

Table 1. Energies and integrated areas of the BE absorption lines. The area of the δ lines includes that of the δ' components which occur 0·6 ± 0·1 meV lower in energy. The areas are considered to be accurate to ±10%.

	α(NP)	δ(NP)	α(TA)	δ(TA)	α(TO)	δ(TO)
Line energy (meV)	1150·00 ±0·05	1154·35 ±0·10	1168·68 ±0·05	1173·02 +0·05	1208·10 ±0·15	1212·50 ±0·15
Integrated area (10^{-6} cm^{-1} eV)	≲5·9	0·15	0·6	5·0	17	130

The α(NP) and α(TO) lines have similar oscillator strengths to those reported by Dean *et al* (1967b) and in addition the α(TA) line, which is about ten times weaker than α(NP), can be clearly seen. The δ(NP) line cannot be seen in figure 2 but can just be detected with a 10× expansion of the ordinate scale and has an integrated area $\lesssim \frac{1}{40}$ of the α(NP) line. The δ(NP) line is predicted to be very weak (Kirczenow 1977b) and the emission line was not reported by Thewalt (1977b), but it has been detected in recent luminescence measurements by the present authors. The TA and TO replicas of the δ line are clearly seen in the transmission spectrum and their strengths relative to the α lines are δ(TA)/α(TA) = 8·3 ± 1·2 and δ(TO)/α(TO) = 7·6 ± 1·1 which are close to the value of 9·2 ± 0·8 derived by Thewalt (1977b) from the temperature dependence of the TA components in the emission spectrum.

In the luminescence measurements Thewalt (1977a, b) observed a second component in the δ(TO) line. This feature he labelled δ', and it peaks $0{\cdot}6 \pm 0{\cdot}1$ meV lower in energy than the principal component δ. The δ' component has been detected in both the TA and TO replicas in the absorption spectrum and a high-resolution trace is shown for the TA replica in figure 2. It might be considered that the δ–δ' separation represents the splitting of the $[\Gamma_1, \Gamma_3; \Gamma_8]$ and $[\Gamma_1, \Gamma_5; \Gamma_8]$ levels of the BE. However, the splitting of the γ lines in the emission spectrum ($1{\cdot}17 \pm 0{\cdot}1$ meV) is in close agreement with the separation ($1.33 \pm 0{\cdot}1$ meV) of the 1s (E) and 1s (T_2) levels observed in infrared absorption measurements (Aggarwal and Ramdas 1965) which would appear to rule out this possibility.

The very substantial agreement that exists between the absorption results reported here and those previously reported for luminescence provides further strong support for the shell model of BE and MBE complexes.

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