"Properties of Crystalline Silicon", EMIS Datareviews Series No. 20
(INSPEC, IEE, London, UK, 1999) p. 669-675

# 11.5 Double donors and acceptors in c-Si

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February 1998

## A INTRODUCTION

Double donors and acceptors usually create an attractive central-cell potential binding electrons or holes in deep bandgap ground state levels. In spite of this situation, the excited states are found to be well described by the effective-mass theory [1,2], both for the neutral and the singly ionized centres. Accurate values for the ground state ionisation energies are derived by adding to a $1s \rightarrow np_\pm$ transition energy, as observed in the experiment, the ionisation energy of the $np_\pm$ state as provided by the effective-mass theory. For n, the highest level with data available is taken; n = 2, 3 or 4.

## B CHALCOGENS

Characteristic examples of double donors are the chalcogen atoms S, Se, Te, from column VI of the periodic table, occupying substitutional sites in a silicon crystal. For homo- and hetero-nuclear pairs and triplets of the chalcogens also double donor action was established [3–5]. Data for the transition energies as directly observed in high-resolution Fourier-transform optical absorption are given in TABLE 1. In the table single sulphur is included to represent the chalcogens; a full review of chalcogen double donors is given in [3].

TABLE 1 Energies of transitions from the 1s ground state to odd-parity excited states and ionization energies ($1s \rightarrow CB$) for the double donor centres S, TDD2 and Mg. States are labelled with effective-mass indices. Energies are given in meV.

| Impurity | $S^0$ | $S^+$ | $TDD2^0$ | $TDD2^+$ | $Mg^0$ | $Mg^+$ |
|---|---|---|---|---|---|---|
| Ref | [5] | [4] | [80] | [81] | [26,30] | [27,30,31] |
| Transition | | | | | | |
| $1s \rightarrow 2p_0$ | 306.84 | 567.6 | 54.8 | 99.97 | 95.83 | 208.63 |
| $2p_\pm$ | 311.93 | 587.5 | 60.5 | 122.96 | 101.15 | 230.24 |
| | | 587.8 | | 123.76 | | 230.47 |
| $3p_0$ | 312.87 | | 61.2 | | 101.98 | 233.90 |
| $4p_0$ | 315.01 | | | | 104.19 | 243.04 |
| $3p_\pm$ | 315.20 | | 63.7 | 137.03 | 104.41 | 243.97 |
| | | | | 137.28 | | 244.06 |
| $4p_\pm$ | 316.13 | | 64.7 | 141.09 | 105.34 | 247.92 |
| $5p_0$ | | | | | 105.33 | 247.92 |
| $4f_\pm$ | 316.41 | | | 142.21 | 105.63 | 249.09 |
| $5f_\pm$ | 317.04 | | | | 105.86 | |
| $5p_\pm$ | 316.86 | | 65.5 | 143.92 | 106.06 | 250.85 |
| $6p_\pm$ | 317.24 | | | 145.51 | 106.45 | 252.29 |
| | | | | | | |
| CB | 318.32 | 613.2 | 66.8 | 149.78 | 107.53 | 256.65 |

## C THERMAL DOUBLE DONORS

Upon heat treatment in the temperature range 450 to 550°C the oxygen-related thermal double donors (TDDns) are formed in Czochralski silicon [6]. This series of double donor centres consists of up to, at present, 16 observed species with very similar, yet distinct, spectroscopic character. For these TDDns data are included in TABLE 1 for the species TDD2 only as a typical example; the whole range of TDDns, n = 1 to 16, is given in Datareview 11.4 of this volume.

## D COLUMN IIa AND IIb IMPURITIES

Impurities with an outer $ns^2$ electron configuration on substitutional silicon sites, lacking two electrons to form four covalent bonds, behave as double acceptors. In contrast, when incorporated in the silicon crystal on an interstitial site the two outer electrons are easily ionized and the impurity acts as a double donor. For the alkali earth elements (Be, Mg, Ca, Sr and Ba) the electronic properties have been best investigated for beryllium and magnesium. Among the IIb elements (Zn, Cd, Hg) most is known about zinc. Data for the IIa and IIb impurities are summarized below.

### D1 Beryllium

Incorporated on the substitutional site, beryllium is a double acceptor. A spectrum labelled Be-I is assigned to this single substitutional impurity in its neutral state [7–10]. The transition energies from ground state to several excited states, with effective-mass labels for the two-hole system, observed in optical absorption are given in TABLE 3. The ground state binding energy is determined as 192.08 meV [9] or 191.9 meV [10]. Data on the second ionization level at $E_v$ + 0.43 eV, i.e. for $Be^-/Be^{2-}$, are more tentative [11–13]. Corresponding data for Be levels based on DLTS are given as $E_v$ + 190 meV and $E_v$ + 440 meV [14]; based on surface capacitance as $E_v$ + 0.42 eV [15]. Another frequently reported spectrum, labelled Be-II, and also revealing a double acceptor centre, is located at $E_v$ + 145.8 meV [10,16]. It is related to a Be-complex [7], presumably a pair of Be atoms on nearest-neighbour substitutional sites with the trigonal symmetry [8,10,16]. By photoluminescence, a triplet of emissions from an exciton bound to an axial isoelectronic centre is observed around 1077 meV. The isoelectronic centre is identified as the double donor-double acceptor pair formed by a substitutional $Be^{2-}$ next to an interstitial $Be^{2+}$ in a <111> geometry [17–20] and accounting for about 90% of the beryllium present [21]. Alternatively, a <100> interstitial configuration has been proposed [22]. Several other Be-related acceptor centres have been reported [8,9,23–25].

### D2 Magnesium

The magnesium impurity in silicon occupies interstitial sites of tetrahedral symmetry and has an electronic 1s(A1) ground state. It behaves as a double donor with ionization energies of 107.53 or 107.51 meV for $Mg^0$ and 256.65 or 256.68 meV for $Mg^+$ according to [26,27] or [28]. Optical absorption spectra, based on high-resolution Fourier-transform spectroscopy, have shown a set of excitation levels forming a solid-state analogue of helium [26–31]. Results for both $Mg^0$ and $Mg^+$ are shown in TABLE 1. For the higher-lying states, such as $6p_\pm$, the detection is more sensitive by photothermal excitation [31]. Data in TABLE 2 illustrate the validity of the effective-mass description for the excited states. For both the neutral and the positively charged centres the spacings of levels follow the effective-mass predictions closely. The four-fold multiplication for $Mg^+$ compared to $Mg^0$ reflects the effective core charge Z = 2 of the former state. Small splittings are observed, by about 0.1 meV, for the transitions to the $2p_\pm$ and $3p_\pm$ states. Careful analysis has shown this to be due to valley-orbit together with central-cell interactions [28]. Taking account of this splitting the above-reported ionization energies were calculated based on the 1s to $4p_\pm$ transition. The $T_d$ symmetry, ground state assignment and extended wavefunction character were confirmed by magnetic resonance [32].

Besides optical spectroscopy, the double donor activity and ground state ionization levels of magnesium were also observed by Hall effect [33,34] with $E_{0/+} = E_c - 115$ meV and $E_{+/2+} = E_c - 227$ meV [33] and deep-level transient spectroscopy (DLTS) [35,36] with $E_{0/+} = E_c - 107$ meV and $E_{+/2+} = E_c - 310$ meV [36]. In general, these techniques reveal a multitude of magnesium-related levels and do not demonstrate full agreement, most probably related to a tendency of magnesium to form complexes in a sample dependent manner. This also applies to the introduction of magnesium by neutron transmutation doping, exploiting the reaction Si(n,α)Mg [37–41]. Specific searches for magnesium-related acceptor activity [42,43] have revealed a level at $E_v + 0.34$ eV, which is considered a good candidate for identification with magnesium on the substitutional lattice site [42].

## D3 Calcium

Data on the calcium impurity in silicon are scarce. It has been reported to be an interstitial double donor with levels at $E_c - 0.22$ eV and (probably) at $E_c - 0.45$ eV [13]. In more recent DLTS (deep-level transient spectroscopy) and optical DLTS experiments only one level at $E_c - 550$ meV related to the $Ca^{0/+}$ ionization was detected [36].

## D4 Strontium

For the strontium impurity in silicon two donor levels are reported at energy positions $E_c - 0.28$ eV and $E_v + 0.5$ eV, respectively [11,13].

## D5 Barium

Double donor activity of barium in silicon was observed by the surface capacitance technique with levels at $E_c - 0.32$ eV and $E_v + 0.50$ eV [13,15]. With more conventional C–V measurements the donor levels were found at $E_c - 0.28$ eV and $E_v + 0.45$ eV [11,13].

## D6 Zinc

Already in the first year of its observation zinc in silicon was firmly identified as a substitutional double acceptor impurity [44]. In many following experiments by the Hall effect [45], photoconductivity [46,47], photothermal ionization [48,49], transient capacitance techniques [50–57] and infrared absorption [58,59] the conclusion was essentially confirmed. A summary of results, presented in [57], gives the values $E_v + (0.31 \pm 0.01)$ eV for the ionization level $Zn^{0/-}$ and $E_v + (0.62 \pm 0.04)$ eV for the second ionization $Zn^{-/2-}$. In view of the propensity of zinc to form complexes with other impurities, as, for example, following from electron-paramagnetic-resonance (EPR) studies [60,61], the results show a remarkable consistency. The most accurate values for the ionization of the neutral zinc were obtained from high-resolution infrared absorption studies and are given as $E_{0/-} = E_v + 319.53$ meV [58] or $E_v + (319.1 \pm 0.3)$ meV [59]. By this infrared spectroscopy also several transitions to bound excited states in a helium-like pattern were observed. These confirmed the validity of the effective-mass theory [2] in understanding the electronic structure of the centre. Results for these transitions are given in TABLE 3.

At slightly elevated temperatures, in the 6 to 30 K range, so-called hot lines appear in the spectra, resulting from levels which become thermally occupied at these higher temperatures. Analysis reveals a four- or five-fold splitting in the ground state with additional levels at 2 to 4 meV higher in energy. The effect has been interpreted as a manifestation of the dynamical Jahn–Teller effect [62]. The ground state structure was also investigated by phonon spectroscopy [63] and by electric-dipole spin resonance [64,65], however without a definite identification of the single zinc impurity as the object of study.

Theoretical calculations of the total energy of zinc in silicon have indicated the cubic substitutional site as the more stable one compared to the interstitial positions with an enthalpy difference of 1.79 eV [66]. Normal ordering of the charge states with Fermi level was found. More speculatively therefore, a level at $E_c$ – 0.47 eV has been put forward for zinc in an interstitial donor position [53,54]. Metastability of zinc between stable lattice positions and negative-U ordering of electronic levels has also been considered [67,68]. The diffusion of zinc in silicon by the dissociative or kick-out mechanisms has been quantitatively described with the implication of zinc in the transient state of a doubly ionized donor on an interstitial position [66,69,70].

TABLE 2 Spacings of donor excited states as obtained by experiments for $Mg^0$ and $Mg^+$ and as provided by the effective-mass theory (EMT); energies in meV.

| Impurity | $Mg^0$ | $Mg^+$ | $Mg^+/Mg^0$ | EMT |
|---|---|---|---|---|
| Ref | [26] | [27] | | [1] |
| States | | | | |
| $2p_0$–$2p_\pm$ | 5.32 | 21.73 | 4.08 | 5.11 |
| $2p_0$–$3p_0$ | 6.15 | 25.27 | 4.11 | 6.03 |
| $2p_0$–$4p_0$ | 8.36 | 34.41 | 4.12 | 8.18 |
| $2p_0$–$3p_\pm$ | 8.58 | 35.39 | 4.12 | 8.39 |
| $2p_0$–$4p_\pm$ | 9.51 | 39.29 | 4.13 | 9.32 |
| $2p_0$–$5p_\pm$ | 10.23 | 42.22 | 4.13 | 10.07 |
| $2p_0$–$6p_\pm$ | 10.62 | 43.66 | 4.11 | 10.47 |

TABLE 3 Energies, in meV, for the spectra of beryllium and zinc in silicon. Observed transitions $E_n$, ground state ionisation energies $E_i$, binding energies in excited level $E_b$.

| Impurity | $Be^0$ | | $Zn^0$ | | EMT |
|---|---|---|---|---|---|
| Energy | $E_n$ | $E_b$ | $E_n$ | $E_b$ | $E_b$ |
| Ref | [9] | | [59] | | [2] |
| Transition | | | | | |
| $1S_{3/2} \rightarrow 2P_{3/2}$ | 176.78 | 15.30 | 303.90 | 15.2 | 15.5 |
| $2P_{5/2}$ | 180.80 | 11.28 | 307.79 | 11.3 | 11.4 |
| $3P_{3/2}$ | 184.73 | 7.35 | 311.69 | 7.4 | 7.3 |
| $2P_{5/2}$ | 185.98 | 6.1 | 313.02 | 6.1 | 6.1 |
| | | | | | |
| $E_i$ | 192.08 | | 319.1 | | |

## D7 Cadmium

The substitutional single cadmium impurity in silicon introduces two acceptor levels which, by Hall effect and photoconductivity, were determined as $E_v$ + 0.55 eV for $Cd^{0/-}$ and $E_c$ – 0.45 eV for $Cd^{-/2-}$ [71]. Different values obtained by Hall effect [72] and surface capacitance [15] were also reported. DLTS applied to radioactive impurities placed the levels at $E_v$ + (485 ±27) meV and $E_c$ –(450 ±20) meV [73,74]. Recently, the electron paramagnetic resonance signal of $Cd^-$ was observed and found to be consistent with tetrahedral symmetry of the centre and having an angular dependence typical for spin $J = 3/2$ for the singly ionized acceptor in its ground state [75]. In photoluminescence, three zero-phonon lines related to centres of low symmetry containing one Cd atom were observed, but were present only in Czochralski silicon. The apparent tendency of cadmium to form CdO complexes shows a close parallel to the behaviour of zinc [76].

## D8 Mercury

On the basis of early photoconductivity experiments, mercury is reported to introduce two acceptor levels at $E_c - 0.31$ eV and $E_c - 0.36$ eV, respectively, and two donor levels at $E_v + 0.25$ eV and $E_v + 0.33$ eV, respectively, in silicon [77]. In later work by the surface capacitance technique only levels at $E_c - 0.39$ eV and $E_v + 0.35$ eV were confirmed [15]. Photoluminescence related to mercury was observed, but the identity of the isoelectronic centres was not well established [78,79].

## E CONCLUSION

A variety of double donor and acceptor species are formed in crystalline silicon from incorporation of chalcogen atoms (S, Se, Te), oxygen (thermal double donors) and column IIa and IIb elements (Be, Mg, Ca, Sr, Ba, Zn, Cd and Hg). For the latter category most data exist for Be, Mg and Zn. Optical absorption, deep level transient spectroscopy, Hall effect, surface capacitance, photothermal ionisation, transient and electron paramagnetic resonance techniques have been applied in these studies. Tabulated data for all the species identified are presented.

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