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Zeeman splitting of double-donor spin-triplet levels in silicon

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ABSTRACT: Observation of the Zeeman effect confirms the identification of spin-triplet terms for double donors in silicon. The theory of Landé g-factors fits the data well when the orbital magnetic moment of the $1s(A_1)1s(T_2)$ configuration is zero as predicted by effective-mass theory. The spin-orbit interaction parameter is determined from the non-linear part of this splitting, and the Zeeman splitting of the singly-ionized double-donor, spin-orbit-split $^2T_2$ line is studied in order to confirm the validity of this method.

1. INTRODUCTION

New infra-red absorption lines, attributed to spin-forbidden transitions to spin-triplet states of the $1s(A_1)1s(T_2)$ configuration of the double donors Se^0 and Te^0 in silicon, have been observed (Bergman *et al.* 1986, 1988) in experiments employing uniaxial stress to tune these lines and thereby enhance their intensities. In previous work (Peale *et al.* 1988) we have observed the Zeeman effect of the corresponding lines in zero stress in fields up to 4 T, and from this Zeeman splitting we have confirmed the identification of these lines with the spin-triplet states. We have shown that these lines result from transitions to components of the $J=1$ spin-orbit level of the $^3T_2$ term and that the Zeeman splitting of this level is described well by effective-mass theory.

The purpose of the present paper is to report the results of new experiments that extend the earlier Zeeman measurements on $Si:Se^0$ and $Si:Te^0$ to fields of 9 T in order to provide a more exacting test of the theoretical model. The lines are not linear in the field but instead show a curvature which we attribute to magnetic coupling between the $J=0,1,2$ spin-orbit levels of the $^3T_2$ term and which we use to obtain the value of the spin-orbit parameter λ that gives the separation of these levels. We have been unable to check the validity of our procedure by direct observation of the transitions to the $J=0$ and $J=2$ levels, since these transitions are strictly forbidden at low fields. We have therefore applied the same theoretical procedure to interpret the Zeeman splitting of the $1s(A_1) \rightarrow 1s(T_2)$ transition of the singly ionized donors S^+ and Se^+ in silicon, the spin-orbit splitting of which is directly observed. The Zeeman splitting of S^+ and Se^+ has not been reported previously and is described in this paper. The strength of the spin-orbit coupling obtained from our results for the $^3T_2$ term of Se^0 and Te^0

is found to be larger than that obtained by Bergman *et al.* (1986) from the interaction of the $^3T_2$ and $^1T_2$ terms.

2. THEORY

The theory of the spin-orbit and Zeeman splitting of the $^3T_2$ term of the $1s(A_1)1s(T_2)$ configuration has been developed (Peale *et al* 1988) using the analogy of a ^3P term of a free atom and the formalism of the theory of the Landé g-factor. Only the results are given here.

Within the $^3T_2$ term, the spin-orbit interaction is described by the parameter λ. As in the Landé interval rule for the relative energies of the spin-orbit levels of a ^3P term there are three levels with J=0, 1 and 2 and energies -2λ, $-\lambda$ and $+\lambda$, respectively. The Zeeman energies are found by solving the secular equation for each value M_J of the component of the total angular momentum J along the field direction. The relative energies of the two states with $M_J = +1$ are given exactly by

$$E_{+1} = \frac{1}{2}(g_S + g_L)\,\mu_B B \pm \left[\lambda^2 + \frac{1}{4}(g_S - g_L)^2\,(\mu_B B)^2\right]^{1/2} , \qquad (1)$$

and those for M_J=-1 by

$$E_{-1} = -\frac{1}{2}(g_S + g_L)\,\mu_B B \pm \left[\lambda^2 + \frac{1}{4}(g_S - g_L)^2\,(\mu_B B)^2\right]^{1/2} . \qquad (2)$$

The lower sign in both Eqs. (1) and (2) corresponds to the states originating in the J =1 level, the only level of $^3T_2$ to which optical excitations from the $^1A_1$ ground state are allowed in low fields. States with M_J=0 are given by the three roots of the equation

$$E^3 + 2\lambda E^2 - [\lambda^2 + (g_S - g_L)^2\,(\mu_B B)^2]\,E - 2\lambda^3 = 0 . \qquad (3)$$

If ξ denotes the one-electron spin-orbit parameter in the $1s(T_2)$ state, we should have

$$\lambda = \xi/2 . \qquad (4)$$

For the singly-ionized double donor, the Zeeman splitting of the $1s(A_1) \rightarrow 1s(T_2)$ transition $[^2A_1 \rightarrow {}^2T_2]$ is found by the same method. The $^2T_2$ term comprises two spin-orbit levels Γ_7 and Γ_8, with J = 1/2 and 3/2, respectively. The Γ_7 line splits into two lines for Faraday geometry in a magnetic field, with transition energies given by

$$E_{\pm 1/2} = -\frac{\lambda}{4} \pm \frac{1}{2}(g_S+g_L)\mu_B B - \frac{3}{4}\lambda\left[1 \pm \frac{4(g_S-g_L)}{9\lambda}\mu_B B + \frac{4(g_S-g_L)^2}{9\lambda^2}\mu_B^2 B^2\right]^{1/2} , \qquad (5)$$

where the notation $E_{\pm 1/2}$ indicates the M_J value of the final state. The Γ_8 line has in general four components in Faraday geometry, given by

$$E_{\pm 1/2} = -\frac{\lambda}{4} \pm \frac{1}{2}(g_S+g_L)\mu_B B + \frac{3}{4}\lambda\left[1 \pm \frac{4(g_S-g_L)}{9\lambda}\mu_B B + \frac{4(g_S-g_L)^2}{9\lambda^2}\mu_B^2 B^2\right]^{1/2} , \qquad (6)$$

$$E_{\pm 3/2} = \lambda/2 \pm g_L \mu_B B . \qquad (7)$$

We note that the $\pm 3/2$ transitions coincide if the orbital g-factor g_L is zero as predicted by

effective-mass theory. In Eqs. (5)-(7) we have $\lambda=\xi$, and we assume that to a sufficient approximation we may take the spin g-factor g_S to be the same in the initial and final states.

3. MEASUREMENTS

All spectra were taken with an IBM Fourier transform infrared interferometer with the sample immersed in pumped liquid helium at a temperature of 1.7K. A wire-grid polarizer was used to polarize the beam when necessary. Magnetic fields were applied by placing the sample in the bore of 9T solenoid. Voigt geometry was achieved by placing a reflection device inside the magnet bore.

The center frequencies vs. magnetic field of the $^3T_2$ (J=1) spin-triplet Zeeman components for Si:Se^0 are plotted in Figure 1a, and the same for Si:Te^0 appears in Figure 1b. The data for the component of the split absorption line with π polarization are denoted by triangles and those for the σ polarized components are plotted as circles. We also plot $E_{\pm1}$ from Eqs. (1) and (2), and the solution E_0 to Eq. (3) corresponding to J=1. For Figure 1a, Eq. (3) was solved numerically for discrete values of the magnetic field B. For Figure 1b, E_0 was assumed to have a series form and the first three terms were found. For all curves in Figs. 1a and 1b, we took g_L = 0 and used the experimentally determined spin-orbit parameter λ extracted from the curvature of the data (described below) and appearing in the Table.

We obtain the spectroscopic splitting factor g_J = (g_S+g_L) / 2 for the J=1 level from the linear part of the Zeeman effect by fitting $(E_{+1} - E_{-1})$

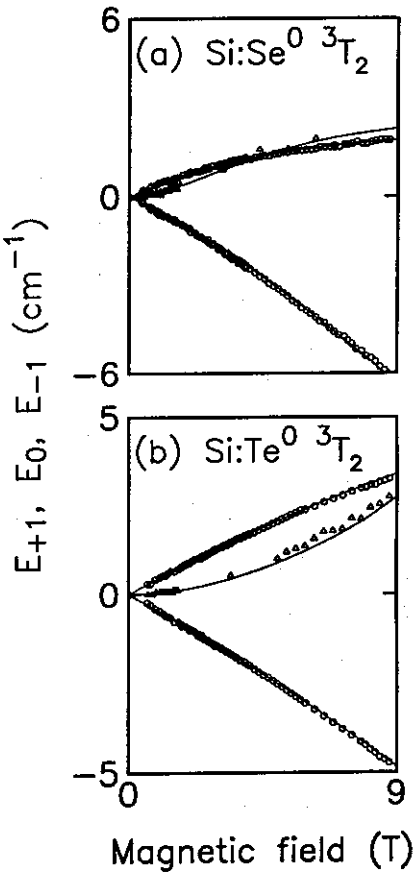

Figure 1. Theory curves (solid lines) and center frequencies of the π (Δ data) and σ (O data) Zeeman components vs. magnetic field for the J=1 spin-orbit component from the $^3T_2$ term of (a) Si:Se^0 and (b) Si:Te^0.

Table	Present work		Bergman *et al* (1986)	
	Se^0	Te^0	Se^0	Te^0
λ(cm^{-1})	2.93 ± 0.02	11.18 ± 0.06		
ξ(cm^{-1})	5.86	22.36	3.2	12.4
g_J (J=1)	0.983 ± 0.002	0.980 ± 0.001		

vs. magnetic field. The slope of the fit equals $2g_J\mu_B$. These data yield g_J-values of 0.983±0.002 (Se^0) and 0.980±0.001 (Te^0).

The spin-orbit interaction parameter is determined from the curvature of the Zeeman data by fitting $(E_{+1} + E_{-1})$ to the sum of Eqs. (1) and (2). We obtain the values for λ of 2.93 ± 0.02 cm^{-1} (Se^0) and 11.18 ± 0.06 cm^{-1} (Te^0). From Eq. (4) and our values for λ we determine values for the parameter ξ, which are also determined by Bergman *et al.* (1986, 1988) from the stress-tuned interaction between $^3T_2$ and $^1T_2$ terms. These values are given in the Table.

The values for ξ that we obtain for Se^0 and Te^0 from the spin-orbit splitting of the $^3T_2$ term are nearly twice as large as those obtained by Bergman *et al.* (1986, 1988) from the avoided crossing of the components of the $^1T_2$ and $^3T_2$ terms under applied stress. The simplest model of the spin-orbit coupling, on the other hand, predicts that these two determinations of ξ should agree. Further evidence that such a difference is real is found for Se^0 and Te^0 from the relative intensities of the transitions to the $^3T_2$ and $^1T_2$ states in zero stress (Peale *et al.* 1988) and, for Te^0, from the size of the stress-induced splitting of the $^3T_2$ term at high stress for uniaxial compression along the [110] crystal axis (Bergman et al. 1988).

In Figure 2a we plot the Zeeman data for the $Si:S^+$ $1s\ ^2A_1 \rightarrow 1s\ ^2T_2$ (Γ_7, Γ_8) line along with Eqs. (5)-(7). Here g_L is assumed to be zero and the value of λ used was determined from the Γ_7, Γ_8 zero-field splitting to be 2.0 cm^{-1}. For g_S we used the value 2.0054 (Ludwig 1965). In Figure 2b we plot the same for $Si:Se^+$ using g_S=2.0057 (Grimmeiss *et al.* 1981) and λ=11.8 cm^{-1}. For $Si:Se^+$, the weak coupling between the Γ_7 and Γ_8 components and knowledge of g_S allow us to extract a value for g_L of 0.008±0.002, which

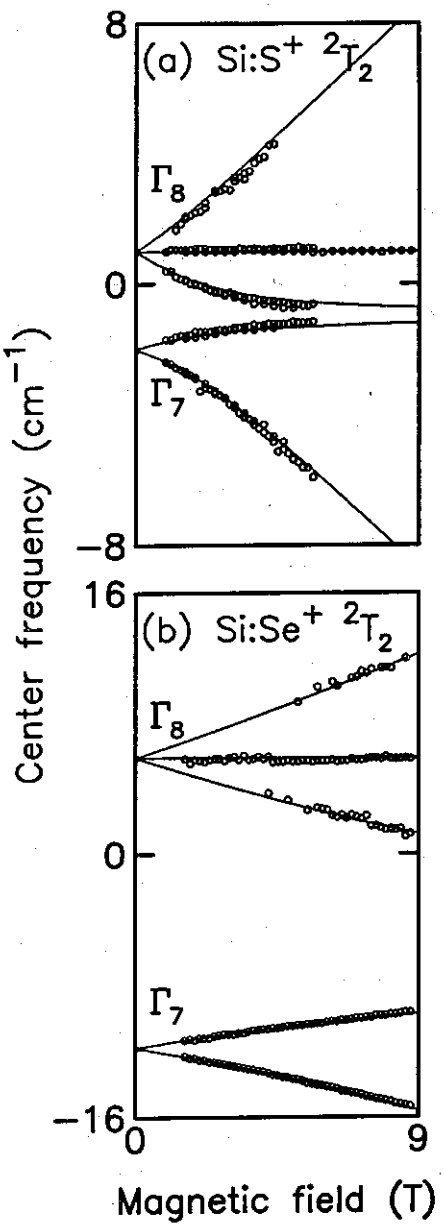

Figure 2. Theory curves (solid lines) and center frequencies (points) of the Zeeman-split $1s(^2A_1) \rightarrow 1s(^2T_2(\Gamma_7))$, and $1s(^2A_1) \rightarrow 1s(^2T_2(\Gamma_8))$ lines for (a) $Si:S^+$ and (b) $Si:Se^+$.

has been used in evaluation Eqs. (5), (6) and (7) for use in Figure 2b. The good agreement of data and theory evident in Figures 2a and 2b leads us to the conclusion that there is no additional source of nonlinearity in these data beyond that described by Eqs. (5) and (6). These data therefore confirm the validity of our method of extracting the spin-orbit interaction parameter λ within the neutral double-donor $^3T_2$ term from the curvature of its Zeeman splitting.

4. DISCUSSION

The new data reported in this paper more than double the range of magnetic field over which the Zeeman splitting of the transition into the $^3T_2$ term of the $1s(A_1)1s(T_2)$ configuration of $Si:Se^0$ and $Si:Te^0$ have been observed, as compared to our previous work (Peale *et al.* 1988). These data continue to support the identification of these lines (Bergman *et al.* 1986) with transitions into the spin-triplet states of the neutral double donor, and they increase the accuracy with which we are able to determine the value of the spin-orbit coupling parameter λ effective within the $^3T_2$ term. Indeed our new value of λ for Se^0 is nearly 6% smaller than that obtained in the earlier work, but it remains still nearly twice as large as that inferred from the strength of the spin-orbit coupling between the $^3T_2$ and $^1T_2$ terms as determined in the stress experiments of Bergman *et al.* (1986, 1988). A similar discrepancy occurs for Te^0 as well. The theory we have presented for the Zeeman splitting of the neutral donors gives an excellent fit with our value for λ to all the observed lines over the full range of field, as seen in Fig. 1. Despite the higher fields, however, we have not been able to observe transitions into any of the states derived from the $J=0$ and $J=2$ spin-orbit levels of $^3T_2$, which should borrow intensity from the $J=1$ states with increasing field.

Our new Zeeman spectra for the singly-ionized donors S^+ and Se^+, in which both the $J=1/2$ and $J=3/2$ levels are observed, also give an excellent fit to the theory, as seen from Fig. 2. The near-vanishing of the orbital g-factor $g_L \approx 0$ in these spectra shows that effective-mass theory remains an excellent approximation for this charge state of the donor as well, despite the increased binding energy of the $1s(T_2)$ state in the presence of the doubly charged core.

We conclude from the success of the theory in fitting the S^+ and Se^+ Zeeman data that our use of this model to obtain the value of λ for the neutral donor should be valid. We conclude that a real discrepancy occurs between the strength of the spin-orbit coupling inferred in this way and that obtained from the stress data of Bergman *et al.* (1986, 1988). This difference may reflect a difference in the $1s(T_2)$ wave function between the $^1T_2$ and $^3T_2$ states, as suggested by King and Van Vleck (1939) in accounting for similar, though smaller, discrepancies in the excitation spectra of atoms such as mercury, cadmium and zinc with the electronic ground-state configuration $(ns)^2$.

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