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Zeeman photoluminescence spectroscopy of isoelectronic beryllium pairs in silicon

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ABSTRACT

Isoelectronic beryllium (Be) pair centers in silicon have been studied by photoluminescence spectroscopy under a magnetic field. The photoluminescence of the bound-exciton recombination at this center shows that the number of Zeeman split peaks is the smallest for the magnetic field applied along a ⟨100⟩ direction. This result provides direct evidence that the Be pairs orient themselves in ⟨111⟩ directions. The *g* values of the hole and electron in this bound exciton determined by fitting of the Zeeman diagrams support the shallow acceptor character of this isoelectronic center.

1. Introduction

Readout of individual quantum states is one of challenges towards the realization of quantum information processing that employs nuclear spins in silicon (Si) as qubits. A representative example of such qubits is ^{31}P nuclear spins of phosphorus donors. Optical initialization [1] and readout [2] of an ensemble of ^{31}P nuclear spins have been demonstrated recently utilizing isotopically enriched ^{28}Si crystals [3,4]. However, due to the predominance of a non-radiative Auger process [5], the quantum efficiency of photon emission from the phosphorus bound exciton is very low $\approx 10^{-4}$, i.e., the success probability of only one out of ten thousand trials makes this readout scheme unrealistic for a single ^{31}P qubit. While electrical readout via electrons emitted by the Auger process is shown to be much more useful for the case of phosphorus [2], an optical readout is desired especially when transmission of quantum information from one place to another and/or formation of entanglement between spatially separated qubits is required. Therefore, it is meaningful to turn our attention to isoelectric centers in Si that are expected to have nearly 100% radiative efficiency due to the absence of the Auger process. Such high emission efficiency was shown for the excitons bound to isoelecronic centers in indirect GaP semiconductors [6]. Optical readout of nuclear spins contained in an isoelectric center will enable quantum information processing based on nuclear spins and photons in Si [7].

One of the isoelectronic centers that may allow optical detection of their nuclear-spin states is a beryllium (Be) pair center in Si [8–18]. Beryllium is a monoisotopic element (^{9}Be) with the nuclear spin $I = 3/2$. In order to derive nuclear spin states from the luminescence arising from the beryllium pair, one has to identify the microstructure of the center which is reflected by the electronic structure of the bound exciton. Interestingly, the same group led by Lightowlers reached two different conclusions on the symmetry axis of the Be pairs in Si by photoluminescence (PL) spectroscopy: ⟨100⟩ from Zeeman study of the no-phonon (NP) lines [8,11] and ⟨111⟩ from a local-mode-phonon-assisted line [14]. In addition, this group suggested that the Be pairs appeared to favor ⟨111⟩ in a piezospectroscopic study of near-infrared absorption measurement [13]. However, Ref. [13] did not present the actual data leading to the ⟨111⟩ character. Furthermore, an *ab initio* calculation of the total energy of the Be pairs in Si supported the ⟨111⟩ interpretation [15].

The present work revisits the Zeeman PL spectroscopy of Be pairs to determine unambiguously that the two beryllium atoms configure themselves along ⟨111⟩ directions. Furthermore, we obtain *g*-values that support the shallow acceptor character of this center [13,16].

2. Experiments

Beryllium was introduced by thermal diffusion into Si following the method employed by Crouch et al. [10]. Si substrates were cut

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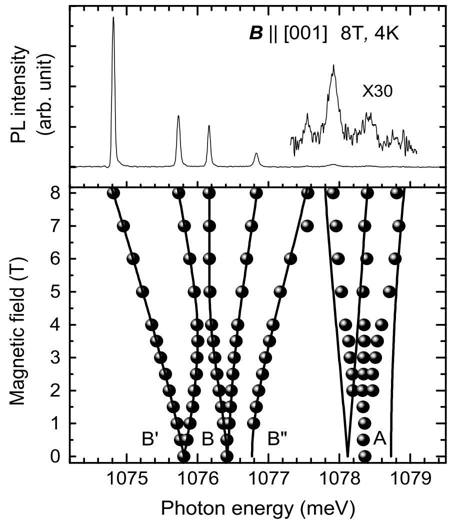

Fig. 1. The Zeeman spectroscopy of a Si:Be bound exciton at $T = 4$ K and $\boldsymbol{B} \parallel [001]$. The upper panel shows the PL spectrum at $B = 8$ T. The lower panel shows the NP peak energies as a function of the magnetic field up to 8 T. Solid lines are fitting curves based on the method described in the text.

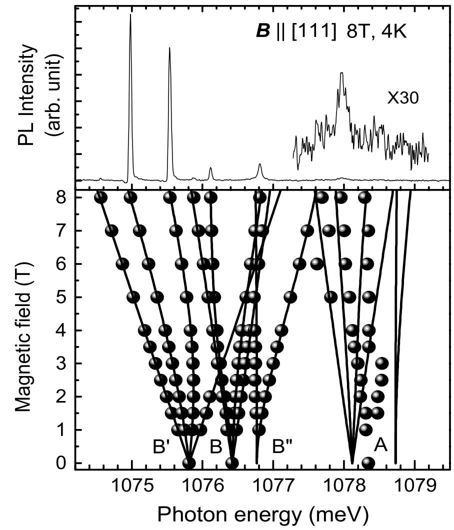

Fig. 2. The Zeeman spectroscopy of a Si:Be bound exciton at $T = 4$ K and $\boldsymbol{B} \parallel [111]$. The upper panel shows the PL spectrum at $B = 8$ T. The lower panel shows the NP peak energies as a function of the magnetic field B up to 8 T. Solid lines are fitting curves based on the method described in the text.

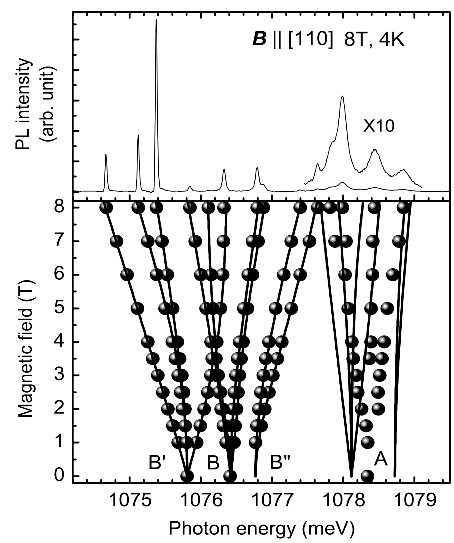

Fig. 3. The Zeeman spectroscopy of a Si:Be bound exciton at $T = 4$ K and $\boldsymbol{B} \parallel [110]$. The upper panel shows the PL spectrum at $B = 8$ T. The lower panel shows the NP peak energies as a function of the magnetic field up to 8 T. Solid lines are fitting curves based on the method described in the text.

from n-type Fz–Si wafers (the resistivity higher than 2 kΩ cm) having three different surfaces: (001), (111), and (110). After deposition of Be films on these Si surfaces, the samples were sealed in Ar filled ampoules and annealed at 1000 °C for 1 h. The samples were cooled down to room temperature by simply taking the ampoule out of the furnace at the end of the annealing. Finally, the sample surfaces were lapped with SiC powders and etched with a mixture of hydrofluoric and nitric acids. The concentration 10^{14} cm^{-3} of the isoelectronic Be pairs in these samples were estimated by near-infrared absorption spectroscopy [9].

For PL measurements, the Be-doped samples were mounted in a strain-free manner and immersed in liquid helium. PL spectra with excitation by a 1047 nm Nd:YLF laser were recorded with a BOMEM DA8 Fourier transform interferometer. A superconducting magnet was used to apply magnetic field parallel both to the optical axis and to the sample surface normal directions [001], [111] and [110].

3. Results and discussion

Figs. 1–3 are the PL spectra of the bound exciton recombination associated with the isoelectronic Be-pair centers at a temperature $T = 4$ K under magnetic field $\boldsymbol{B}$ up to 8 T in [001], [111], and [110] directions, respectively. The upper panels show the PL spectra in the NP region at the magnetic field of 8 T, and the lower panels give the Zeeman diagrams plotting the NP peak energies as a function of B.

The NP spectrum in the absence of the magnetic field is composed of three lines labeled B′ (1075.81 meV), B (1076.41 meV), and A (1078.36 meV) [8,9], as seen at the bottom of the lower panels in Figs. 1–3. By applying the magnetic field, all these lines split, and another series of lines labeled B″ emerges between the lines B and A. The number of line splittings depends on the series of lines (B′, B, B″, or A), and the number of B′, B and B″ lines doubles for the field directions [110] and [111] compared to that for [001]. This doubling is explained by assuming that each Be pair is oriented in one of ⟨111⟩ directions. Fig. 4 shows all possible Be-pair orientations satisfying this assumption. When a magnetic field $\boldsymbol{B}$ is applied, the four orientations are distinguishable and separated into two nonequivalent groups for $\boldsymbol{B} \parallel [111]$ and $\boldsymbol{B} \parallel [110]$, while they remain equivalent for $\boldsymbol{B} \parallel [001]$. This is consistent with our Zeeman diagrams, which indicate that the bound exciton energy levels for $\boldsymbol{B} \parallel [111]$ and $\boldsymbol{B} \parallel [110]$ are twice as many as those for $\boldsymbol{B} \parallel [001]$. If the Be pairs were oriented to the ⟨100⟩ directions, the simplest Zeeman diagram would be obtained for $\boldsymbol{B} \parallel [111]$ instead of $\boldsymbol{B} \parallel [001]$. If the Be pair were oriented in any other directions, no field direction would give a Zeeman diagram as simple as we obtained for $\boldsymbol{B} \parallel [001]$ in Fig. 1. Thus, our PL results unambiguously show that the Be pair center has the ⟨111⟩ axis of symmetry in agreement with Refs. [13–15].

To quantitatively analyze the magnetic field dependence of the PL spectra, we use the Hamiltonian for a bound exciton with a uniaxial stress at the isoelectronic center with an external magnetic field [19–21]. This Hamiltonian is described with the electron and hole angular momenta $\boldsymbol{s}_e$ and $\boldsymbol{j}_h$ ($s_e = 1/2, j_h = 3/2$) and divided into two parts: $H = H_0 + H_{LZ}$. Here, H_0 is the Hamiltonian in the absence of the magnetic field;

$$H_0 = -a\boldsymbol{j}_h \cdot \boldsymbol{s}_e - D\left(j_{hz'}^2 - \frac{j_h(j_h+1)}{3}\right). \quad (1)$$