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Tin-Vacancy Quantum Emitters in Diamond

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Abstract

Tin-vacancy (SnV) color centers were created in diamond via ion implantation and subsequent high-temperature annealing up to 2100 °C at 7.7 GPa. The first-principles calculation suggested that a large atom of tin can be incorporated into a diamond lattice with a split-vacancy configuration, in which a tin atom sits on an interstitial site with two neighboring vacancies. The SnV center showed a sharp zero phonon line at 619 nm at room temperature. This line split into four peaks at cryogenic temperatures, with a larger ground state splitting (~850 GHz) than that of color centers based on other IV group elements, i.e., silicon-vacancy (SiV) and germanium-vacancy (GeV) centers. The excited state lifetime was estimated, via Hanbury Brown-Twiss interferometry measurements on single SnV quantum emitters, to be ~5 ns. The order of the experimentally obtained optical transition energies, compared with those of SiV and GeV centers, was in good agreement with the theoretical calculations.

The use of point defect-related color centers in solid-state materials is a promising approach for quantum information processing [1,2]. Nitrogen-vacancy (NV) centers in diamond have been studied most intensively from the viewpoint of both fundamental and applied sciences [3,4]. However, the zero-phonon line (ZPL) of an NV center constitutes a fraction of only 4 % in its total fluorescence due to its large phonon sideband (PSB). Additionally, the NV center is susceptible to external noise, leading to instability of the optical transition energy. To overcome these drawbacks, color centers based on IV group elements, i.e., silicon-vacancy (SiV) [5-7] and germanium-vacancy (GeV) [8-11] centers, have attracted attention because of their large ZPLs, structural symmetries that are resistant to external noise, and the availability of quantum emission from single centers. Recently, spin control and spin coherence time have been investigated for these two color centers [12-18], revealing that their spin coherence times are limited to submicroseconds even at cryogenic temperatures of 2-5 K, being much shorter than that of the NV center [19,20]. This limitation originates from phonon-mediated transitions between the lower and upper branches in the ground state [13-16]. Further cooling down to the subKelvin regime or strain engineering has been considered as a potential solution [15,16]. Very recently, both solutions have been attempted for SiV [21-23]. Particularly, in Ref. [23] long spin coherence time in the order of milliseconds was achieved at subKelvin temperatures in a dilution refrigerator. Another possible way to achieve a long spin coherence time even at a high temperature is the creation of a novel color center possessing a larger energy split in the ground state. For this purpose, in this study, we investigated the utilization of a IV group atom of tin (Sn, Fig. 1a). We fabricated tin-vacancy (SnV) centers in diamond in both ensemble and single states via a combination of ion implantation and subsequent high-temperature annealing up to 2100 °C under a high pressure of 7.7 GPa. Low-temperature optical measurements revealed that the ground state splitting of the SnV center was much larger than that of the SiV and GeV centers. The atomic structure and optical transition energy were calculated via first-principles calculations, with comparison to SiV and GeV centers.

Figure 1b shows a room temperature photoluminescence (PL) spectrum from ensemble SnV centers, annealed at 2100 °C at 7.7 GPa after Sn ion implantation, with a dose of $2 \times 10^{13} \text{ cm}^{-2}$, into diamond (see Supplemental Materials [24] for experimental details). The SnV centers exhibit a sharp and strong ZPL at 619 nm, with a full width at half maximum (FWHM) of 6.2 nm and with the accompanying PSB. We investigated the effect of the post-implantation annealing temperature on the activation of the SnV centers. Figure 1c shows the temperature dependence of the ZPL linewidth in the SnV spectrum. We performed the annealing under vacuum or high-pressure conditions for 30 min. The increase in the annealing temperature effectively reduced the FWHM irrespective of the annealing environments. Significantly, the high pressure of 7.7 GPa enabled us to reach a temperature of 2100 °C while avoiding graphitization of the diamond [29,30]. Consequently, the

linewidth was reduced from ~ 12 nm at 800 °C to ~ 6 nm at 2100 °C by suppressing inhomogeneous broadening due to surrounding defects and strain in the atomic structure induced during ion implantation. Furthermore, such a high-temperature treatment is important for suppressing the formation of undesirable fluorescent structures. Although two prominent peaks with unknown origins were observed at 595 and 646 nm at temperatures of 1500 °C and lower, they were annealed out by the treatment at 2100 °C (Supplemental Material [24]). The large Sn atom is thought to be less movable in the diamond lattice and to produce a number of vacancies during ion implantation. Thus, the high-temperature treatment is required to selectively activate the high-quality SnV centers.

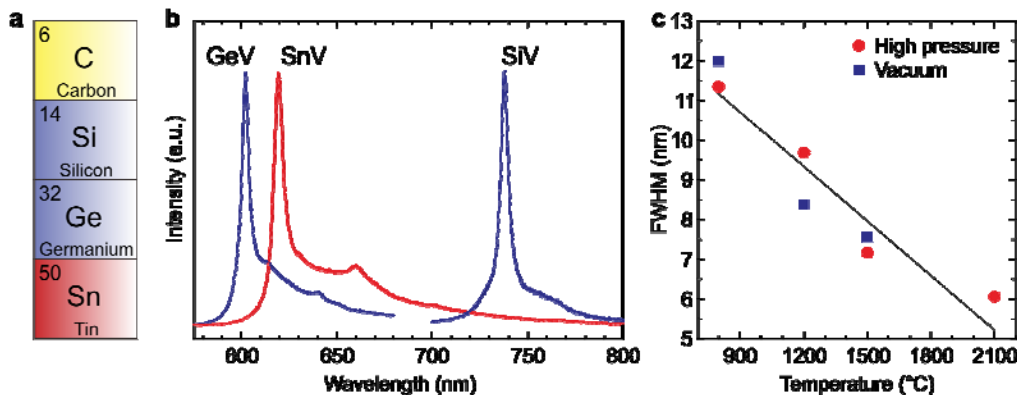


Figure 1. Activation and optical properties of ensemble SnV centers. (a) The IV group periodic table. (b) Room temperature PL spectrum from ensemble of SnV centers, annealed at 2100 °C after ion implantation with a dose of 2×10^{13} cm $^{-2}$ at an acceleration energy of 130 keV. PL spectra from ensemble of SiV and GeV centers are also shown. (c) Dependence of linewidth of ZPL from SnV on the post-implantation annealing temperature under a high vacuum of $\sim 10^{-5}$ Pa or high pressure of 7.7 GPa.

Low-temperature PL measurements were performed to reveal the fine structure of the SnV center (Fig. 2a). Only one peak was observed at room temperature; it split into two peaks with an FWHM of approximately 0.2 nm (155 GHz) at 4 K. This linewidth is broader than the lifetime-limited linewidth of 32 MHz. This was caused by the remaining implantation damages created via the high-dose process, not being fully recovered even via the high-temperature treatment under high pressure. The linewidth is expected to be further reduced by a longer annealing time and/or higher-temperature treatment. Additionally, in further studies, photoluminescence excitation spectroscopy on a low-dose sample will be useful to reveal the natural linewidth.

To investigate the fine structure in detail, the temperature dependence of the PL spectrum was recorded (Fig. 2b). When increasing the temperature, another two peaks (denoted as A and B) appeared in a higher energy region in addition to the peaks seen at 4 K (denoted as C and D). The intensities of peaks A and B become larger as the temperature increases. This fact suggests that the

SnV center has an energy level structure composed of four levels with split ground and excited states (Fig. 2c). The ground and excited state splittings of the SnV center are ~ 850 and ~ 3000 GHz, respectively, being much larger than those of SiV and GeV centers possessing the same four-level structure [5, 9]. Note that the population of the upper branch excited from the lower branch exponentially decreases with the excited state splitting [5,9], as illustrated by the low PL intensities of peaks A and B even at a high temperature of 100 K (compared with SiV [5] and GeV [9]).

In Fig. 2d, we summarize the splittings as a function of the spin-orbit coupling constant [31] of the IV group elements. Both the ground and excited state splittings increase as the spin-orbit coupling constant of the elements increases. The ground state splitting of the SnV center is 17 and 5 times larger than that of SiV and GeV, respectively. The observed splittings should be originated not only from the spin-orbit coupling constant of the IV group elements, but also from the degree of the hybridization of the p orbitals of the elements with carbon dangling bonds (Indeed, minor contribution of valence p states of the group IV impurity to the occupied orbital states was reported in a literature [32], see also Supplemental Materials [24]). In addition, the dynamic Jahn-Teller effect would also contribute to the splittings, as observed in the SiV centers [33]. Its contribution to the splittings will be revealed by observing the fine structures under magnetic fields [33].

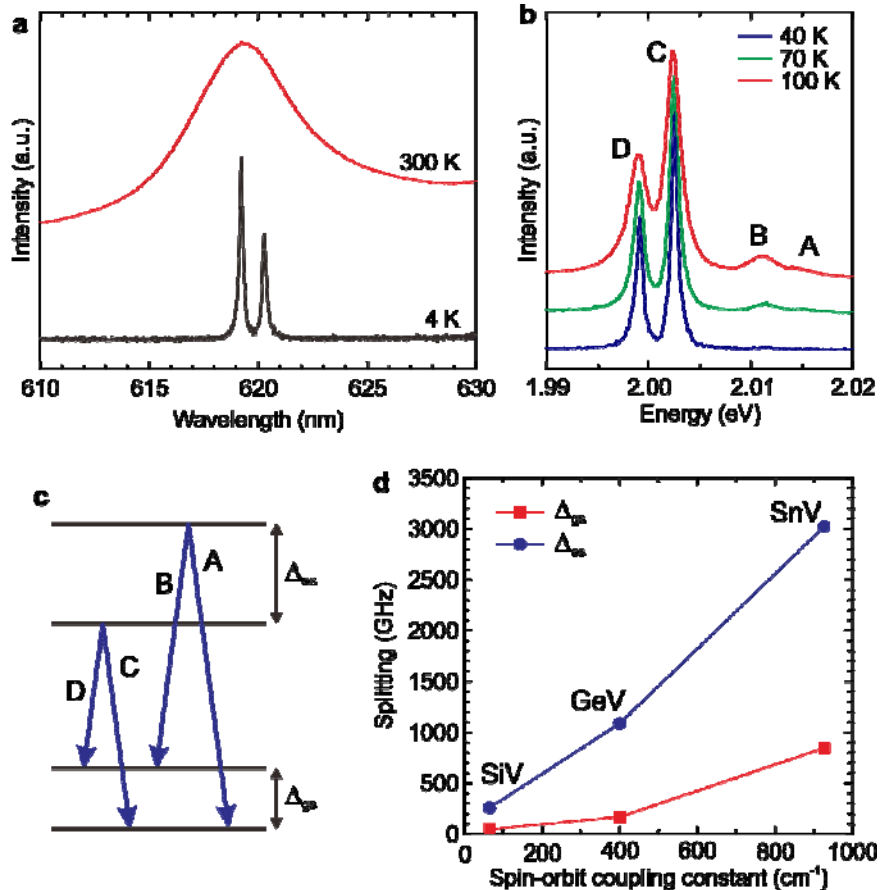


Figure 2. Low-temperature optical characteristics. (a) ZPL of ensemble SnVs at room temperature and at 4 K. The spectrometer resolution is approximately 0.04 nm. Thus, the observed linewidth of 0.2 nm at 4 K is not spectrometer-limited. (b) PL spectra at different measurement temperatures. (c) Fine structure. (d) Ground and excited state splittings as a function of the spin-orbit coupling constant of the IV group element. The values of the SiV and GeV centers were adapted from Refs. [5] and [9], respectively.

We next fabricated single SnV color centers with a lower Sn ion dose of $2 \times 10^8 \text{ cm}^{-2}$. The inset in Fig. 3a shows a confocal fluorescence microscopy image of an isolated SnV center with an FWHM of $\sim 450 \text{ nm}$. The PL spectrum was recorded at the bright spot and background position in Fig. 3a. The bright spot clearly shows a ZPL with a linewidth of 6 nm from the SnV center. The background position also shows multiple peaks in the same region, but they originate from the second-order Raman scattering from the diamond (611 and 620 nm) and probably a surface defect (630 nm) which is not visible in the bulk diamond region. The background-subtracted PL shows the ZPL and PSB from only the SnV center. We performed the Hanbury Brown-Twiss (HBT) interferometry [34] measurements to confirm that the observed spots correspond to a single SnV center. Figure 3b shows the results of a second-order autocorrelation function, $g^2(\tau)$, for different excitation laser powers. Due to the background emission, $g^2(0)$ becomes ~ 0.5 (the right axis in Fig. 3b). Thus, we corrected the background level using the relation $g_{corr}^2(\tau) = [g^2(\tau) - (1 - \rho^2)]/\rho^2$ [35], where $\rho = S/(S + B)$. S is the intensity of the SnV center, and B is the background intensity. ρ takes values of approximately 0.7. As shown along the left axis in Fig. 3b, $g_{corr}^2(0)$ almost reaches zero at a delay time of 0 ns, which is an unambiguous proof of single photon emission [36].

The data were fitted using the equation $g_{corr}^2(\tau) = 1 - (1 + \alpha)e^{-\frac{|\tau|}{\tau_1}} + \alpha e^{-\frac{|\tau|}{\tau_2}}$ [37], where α , τ_1 , and τ_2 are fitting parameters. At low excitation powers, τ_1 yields an estimated excited state lifetime of a fluorescent structure. The two emitters investigated in this study show a τ_1 value of $\sim 5 \text{ ns}$ (another emitter is shown in the Supplemental Materials [24]). For all the laser powers used here, $g^2(\tau)$ exhibits a bunching behavior observed as $g^2(\tau)$ over unity. This means that the SnV center possesses a shelving state in addition to the ground and excited states or that photoionization occurs during the laser excitation [37].

The fluorescence intensity was measured as a function of the laser power (Fig. 3c). We used two kinds of filters: the ZPL intensity alone, which was recorded using a bandpass filter (BPF) with a 20 nm width around the ZPL, and the intensity from the whole SnV spectrum (ZPL+PSB), which was recorded using a 600 nm longpass filter (LPF). Both curves show nonlinear behavior at high laser

powers. The plots were fitted using the equation $I = I_{\infty} \times P / (P + P_{sat})$ [6], where I_{∞} and P_{sat} are the saturation intensity and saturation power, respectively. The saturation intensities of the ZPL and whole spectrum are 280 kcps and 530 kcps, respectively. Note that we used an air objective (numerical aperture (NA): 0.95) here; thus, the utilization of a higher NA oil objective and/or microcavity structures [38-41] will enhance the efficiency of collecting light from the emitter and lead to a further increase in the saturation intensity. Importantly, the SnV center shows a higher fluorescence intensity than that of SiV (56 kcps observed using a 0.95-NA objective [42]) despite the longer excited state lifetime compared with SiV (1~2 ns [6,42]). This implies a high quantum efficiency of the SnV center. We estimated the quantum efficiency based on the rate equations modeled in Ref. [43] and obtained ~ 0.8 (see Supplement Materials). This value is comparable to that of the NV center (0.7-1 [43,44]) and higher than that of the SiV center (~ 0.3 in bulk diamond [45] and < 0.1 in nanodiamonds [46]). It is worth noting that the comparison of the fluorescence intensities of the ensemble with the single SnV center provides a conversion efficiency from Sn ions to SnV centers of approximately 1-2 %. Counting bright spots in confocal microscope images of the single SnV sample, observed with a 620 nm BPF, gives rise to ~ 4 %, which is roughly the same level as that of the estimation made using the ensemble.

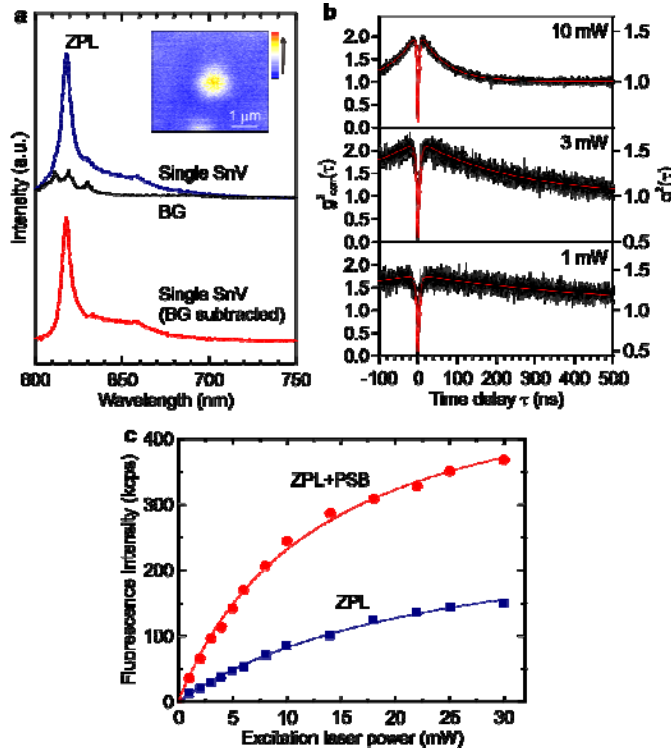


Figure 3. Single SnV color center. (a) PL spectrum for the isolated SnV center shown in inset. Inset: confocal fluorescence microscopy image taken using a BPF with a width of 20 nm around the ZPL.

BG denotes the PL at the background position, where no SnV center exists. The bottom curve is the background-subtracted single SnV spectrum. (b) Background corrected second-order autocorrelation function, $g_{corr}^2(\tau)$, at different excitation laser powers, measured using a BPF. (c) Saturation curves of the ZPL only and of the whole spectrum. The background intensities were subtracted from each curve (see Supplement Materials). The single SnV centers were fabricated via ion implantation (dose: $2 \times 10^8 \text{ cm}^{-2}$, acceleration energy: 150 keV) and annealing at 2100 °C under a high pressure of 7.7 GPa. All measurements were made with an air objective (NA: 0.95) at room temperature.

We performed first-principles calculations of the atomic structure, energy levels, and optical transition energy of the SnV center (see Supplement Materials [24] for calculation details). Figure 4a shows the optimized atomic configuration of a SnV center with a split-vacancy structure. When we start the calculation from a structure with a Sn atom at a substitutional position with a neighboring vacancy (with the same C_{3v} symmetry as that of the NV center), the Sn atom is relaxed to an interstitial site upon structural optimization. Consequently, the Sn atom sits at an interstitial site between two vacancies at carbon lattice sites. This structure possesses the same D_{3d} symmetry as that of the SiV and GeV centers. In accordance with the order of atomic sizes, the nearest C-Si, C-Ge, and C-Sn distances are 1.94, 2.05 and 2.14 Å, respectively. The energy levels of the ground state of the SnV center, assuming a negatively charged state, are shown in Fig. 4b. The transition energy between the negatively charged and neutral states is located below the midgap in the energy gap of the diamond [47], implying that the SnV takes the negatively charged state in the high-purity IIa-type diamond crystals used in this study. The degenerated e_u levels are fully occupied, while one of the degenerated e_g levels is half-occupied. Upon optical excitation, one electron with a down spin in the e_u level is pumped up to the e_g level and then relaxes back while emitting photons with an energy of ~ 2 eV.

Here, we discuss the optical transition energy of the color centers based on the IV group elements. Interestingly, the order of the optical transition energy is not proportional to the atomic size of the element. As shown in Fig. 1b, the SiV centers exhibit a ZPL at 738 nm, while the GeV centers composed of a larger atom exhibit a higher transition energy at 602 nm. However, the ZPL energy of the SnV centers with an even heavier atom is less than that of the GeV center. The transition energy of SnV was calculated using the structural model shown in Fig. 4a and is shown, along with the transition energies for SiV and GeV, in Fig. 4c, which also includes the experimental ZPL transition energies. The experimental order of the optical excitation gap (SiV < SnV < GeV) is consistent with the theoretical results, which was also reported by Goss et al. [47].

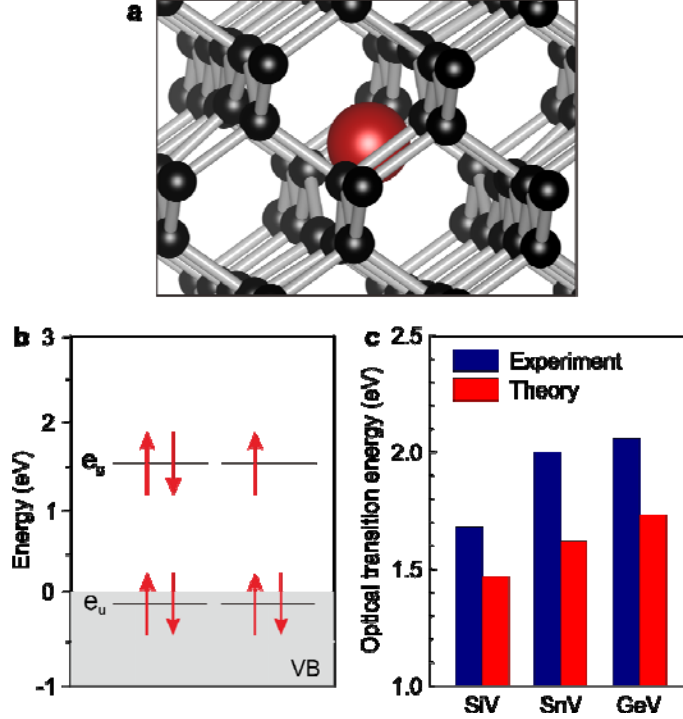


Figure 4. Theoretical calculation of SnV center via first-principles calculation. (a) Atomic structural model. The red sphere and black spheres denote the Sn atom and C atoms, respectively. (b) Energy levels of the ground state. VB denotes the valence band of diamond. (c) Optical transition energies.

Finally, we discuss the expected spin coherence time of the SnV center. The phonon-mediated transition rate is proportional to the occupation of the phonon mode, $n(\Delta_{gs}, T) = 1/(e^{\frac{\hbar\Delta_{gs}}{k_B T}} - 1)$, where \hbar and k_B are the Planck and Boltzmann constants, respectively [21,48]. The occupation decreases exponentially with Δ_{gs} and temperature, leading to the significant reduction in the transition rate in the SnV center with the large ground state splitting of ~ 850 GHz compared with the SiV and GeV centers (Supplemental Materials [24]). Therefore, a long spin coherence time (in the range of milliseconds) can be expected for SnV without cooling down to the subKelvin regime, which is necessary for SiV [23].

We introduced large Sn atoms in the diamond lattice and obtained SnV centers with large level

splitting in the ground state. The incorporation of a heavy atom in diamond will become more important from the viewpoints of materials science [49] and quantum optics. The fabrication of optical centers based on large atoms such as Eu [50], Xe [51], and Tl [52] has thus far been demonstrated. For further development of the heavy atom-related color centers, it is essential to achieve high-quality quantum emitters via an adequate process for each atom, such as high-temperature annealing under high pressure, as demonstrated in this study.

In conclusion, we have demonstrated the fabrication of both ensemble and single SnV centers in diamond via ion implantation and annealing. The high annealing temperature of 2100 °C under a high pressure of 7.7 GPa led to narrow ZPLs from the SnV centers while annealing out undesirable fluorescent structures. Low-temperature optical measurements revealed that the SnV center possesses a four-level structure with large splitting in the ground state, i.e., ~850 GHz. In comparison to the SiV and GeV centers, we showed that the magnitude of the splitting increases as the spin-orbit coupling (atomic number) of the IV group element becomes larger. Our theoretical calculations indicated that the split-vacancy configuration of the SnV center is in agreement with the observed fine structures. The development of a color center with large ground state splitting is expected to provide a way to establish a quantum light-matter interface with a long spin coherence time.

Note: During the preparation of this manuscript, we became aware of another study presenting the room temperature characteristics of Sn-related color centers in diamond [53]. The reported ZPL position is consistent with that of our study. However, because annealing was carried out at a low temperature (950 °C) after the Sn ion implantation, several peaks other than the SnV center remained. Therefore, the high-temperature annealing under high pressure performed in our study is important for obtaining high-quality SnV centers.

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