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Silicon carbide light-emitting diode as a prospective room temperature source for single photons

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Generation of single photons has been demonstrated in several systems. However, none of them satisfies all the conditions, e.g. room temperature functionality, telecom wavelength operation, high efficiency, as required for practical applications. Here, we report the fabrication of light-emitting diodes (LEDs) based on intrinsic defects in silicon carbide (SiC). To fabricate our devices we used a standard semiconductor manufacturing technology in combination with high-energy electron irradiation. The room temperature electroluminescence (EL) of our LEDs reveals two strong emission bands in the visible and near infrared (NIR) spectral ranges, associated with two different intrinsic defects. As these defects can potentially be generated at a low or even single defect level, our approach can be used to realize electrically driven single photon source for quantum telecommunication and information processing.

Robust and cheap light sources emitting single photons on demand are at the heart of many demanding optical technologies[1,2]. Single photon emission has been demonstrated in a variety of systems, including atoms[3], ions[4], molecules[5–7], quantum dots (QDs)[8,9] and color centers in diamond[10,11]. The most significant progress has been achieved for QDs[12–14], however, the necessity to use cryogenic temperatures and high inhomogeneity (the emission wavelength is individual for each QD) make this system impractical. Electrically driven single photon sources in the visible spectral range have also been demonstrated using nitrogen-vacancy (NV) centers in diamond[15,16], but the compatibility of this system with the present-day integrated circuits manufacturing is not obvious.

The operation principle of single photon sources is based on the quantum mechanical properties of a single two-level system. When a single photon is desired, this system is put into the excited state by an external stimulus, and a single photon is emitted upon relaxation into the ground state. A perspective approach to fabricate an efficient, room temperature single photon source based on this principle is to use color centers in semiconductors. In our work, we exploit two defect centers in SiC, the so-called D_1 defect[17] and the silicon vacancy (V_{Si}) defect[18], making two-color LED [Fig. 1(a)].

Remarkably, the V_{Si} defects in SiC comprise the technological advantages of semiconductor quantum dots and the unique quantum properties of the NV defects in diamond[19]. In particular, V_{Si} spin qubits can be optically initialized and read out[18,19], and, therefore, our demonstration of room temperature EL from V_{Si} defects is an important step towards realization of all-electrical control of V_{Si} spins. Further, the V_{Si} EL reveals a broad-band emission spectrum in NIR (850 – 1050 nm), where the absorption of silica glass optical fibers is relatively weak. While this spectrum range is still below the telecom window (1.3 μm), it can be changed in the direction of longer wavelengths by proper choosing over family of deep defect centers in different SiC polytypes[20]. Alternatively, the frequency conversion of NIR photons to a telecom wavelength can be applied[21,22]. Therefore, the integration of defect-based SiC LEDs with existing telecommunication infrastructure seems feasible.

SiC with highly developed device technologies (e.g. MOSFETS, MEMS, sensors) is a very attractive material for practical applications. SiC is also known as the material on which the first LED has been created[23]. Until the 90's, SiC was used for commercial yellow and blue LEDs, but later it was replaced with GaN. One of the disadvantages of SiC for opto-electronics is its indirect bandgap. As a consequence, direct band-to-band (BB) radiative recombination is inefficient, compared to recombination via subgap states [Fig. 1(b)]. Engineering and isolating single defects with proper transition energy on demand can open a route for an efficient electrical single photons source.

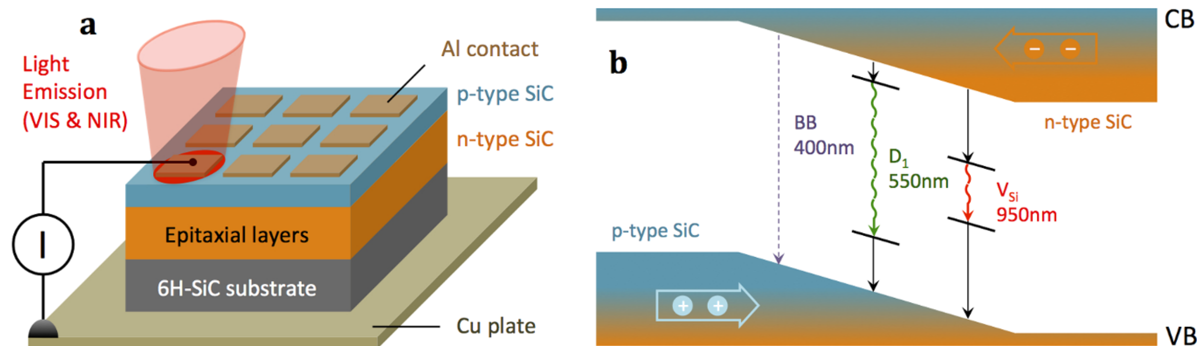

Figure 1 | SiC LED with intrinsic defects. (a) A scheme of the SiC LED. (b) Electron-hole recombination through the D_1 and V_{Si} defects results in the 550 nm and 950 nm emission bands, respectively. The radiative band-to-band recombination (BB) at 400 nm is inefficient because SiC is an indirect bandgap semiconductor.

Results

A scheme of the SiC LED structure, consisting of a single p-n junction, is presented in Fig. 1(a). Intrinsic defects in these structures were generated by electron irradiation. We mount LED samples on a Cu plate serving as the back electrode. Upon applying voltage between one of the Al contacts and the Cu plate the luminescence glow is seen by the naked eye [Fig. 2(a)]. The room temperature EL spectrum of one of our LEDs is presented in Fig. 2(b). It consists of two broad emission bands, labeled as D_1 and V_{Si}. The corresponding recombination processes at the p-n junction are schematically shown in Fig. 1(b).

We now discuss the EL bands of Fig. 2(b) in details. The emission energies are seen to be significantly smaller than the bandgap of 6H-SiC (3.05 eV). We, therefore, ascribe them to the defects in SiC. The emission in the spectral range 450 – 650 nm is characteristic of the D_1 zero-phonon lines (ZPLs) and their phonon replicas[24], merging together at room temperature. The nature of this defect is still not clear – several models have been proposed, including a bound-exciton-like center[24] and a first-neighbor antisite pair Si_C – C_{Si}[25]. The second emission band in the NIR spectral range 850 – 1050 nm coincides with the photoluminescence (PL) spectrum of the silicon vacancy defects V_{Si}[26] in the reference 6H-SiC bulk sample.

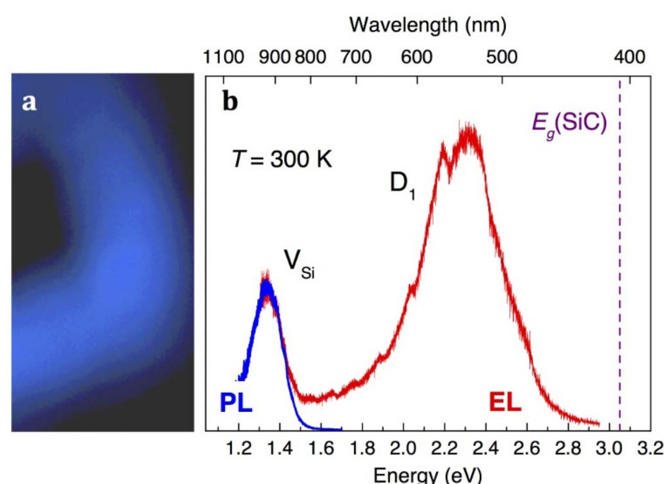

Figure 2 | Room-temperature electroluminescence of intrinsic defects in SiC. (a) An image of the luminous LED around an Al contact. (b) Electroluminescence (EL) spectrum of the SiC LED and photoluminescence (PL) spectrum of the reference SiC sample recorded at room temperature. The PL spectrum is excited by a He-Ne laser with E_{exc} = 1.96 eV (633 nm). The bandgap of 6H-SiC is E_g(SiC) = 3.05 eV.

To prove this interpretation we repeat the experiment of Fig. 2(b) at a temperature of 77 K [see Fig. 3(a)], when the spectroscopic features, individual for each defect, can be resolved. The results are summarized in Fig. 3 and below we discuss them in detail.

Discussion

First, we demonstrate the presence of V_{Si} defects in our LED structures. Figure 3(b) shows photoluminescence (PL) spectrum recorded under excitation with the energy E_{exc} = 1.96 eV, which is below the D_1 emission energy. Three ZPLs at 1.368 eV, 1.398 eV and 1.434 eV are the well known fingerprint of the V_{Si} defects in 6H-SiC[27]. These three ZPLs originate from three nonequivalent crystallographic sites

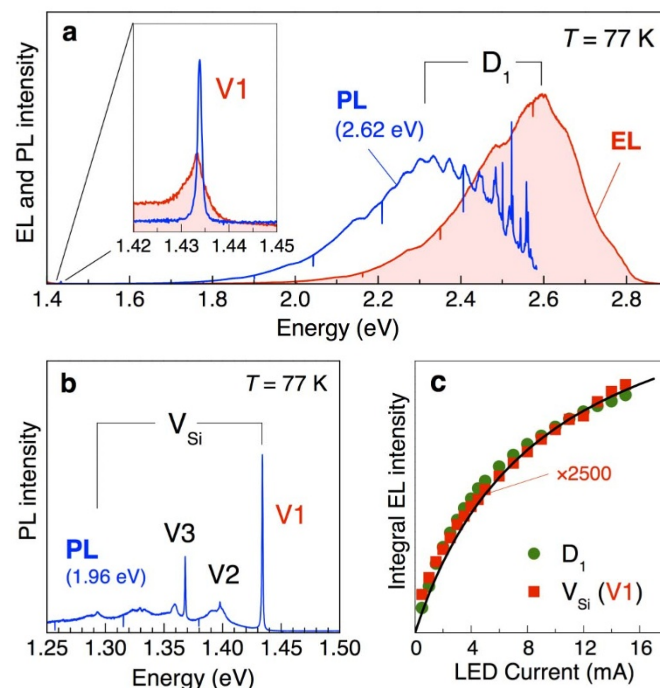

Figure 3 | EL and PL spectra of SiC LED recorded at 77 K. (a) Comparison of the EL (shaded area) and PL spectra under excitation with an energy E_{exc} = 2.62 eV (473 nm). Inset: The same, but shown in the spectral range where the strongest V_{Si} ZPL (V1) is expected. (b) PL spectrum obtained under excitation with a He-Ne laser with E_{exc} = 1.96 eV (633 nm). The V1, V2 and V3 ZPLs characteristic for the V_{Si} defects in SiC are clearly seen. (c) Integral intensity of the V_{Si} and D_1 emission bands [the shaded areas in (a)] as a function of LED current. The solid line is a fit (see text for details).

in this SiC polytype and are frequently labeled as V3, V2, and V1, respectively. The highest ZPL intensity is observed for V_{Si}(V1).

Second, we demonstrate that the V_{Si}(V1) defect can be electrically driven. Figure 3(a) shows EL spectrum recorded at $T = 77$ K. In contrast to room temperature [Fig. 2(b)], the D_1 emission dominates in the spectrum. The reason is the much higher concentration of D_1 defects than of V_{Si} defects. However, at room temperature most of the D_1 defects are ionized due to the thermal activation of the defect-bound electrons in the conduction band. This is consistent with a small activation energy (about 60 meV[28,29]) of the D_1 defect in a different polytype 4H. With lowering temperature, the activation process becomes inefficient and EL (PL) intensity increases. On the other hand, the activation energy of the V_{Si} defects is much larger and their intensity weakly depends on temperature. Indeed, we observe the characteristic V_{Si}(V1) ZPL at 1.434 eV in the EL spectrum [the inset of Fig. 3(a)]. The larger ZPL spectral width in EL compared to PL is most probably caused by current-induced charge fluctuations in the vicinity of V_{Si} defects, leading to an increase of inhomogeneous broadening.

Third, we verify that the electrical excitation of the V_{Si}(V1) defect shown in the inset of Fig. 3(a) is not due to the re-emission process via D_1. We excite into the maximum of the D_1 band (2.62 eV), leading to the Stokes shift of the D_1 emission spectrum [Fig. 3(a)]. The laser intensity per area is several orders of magnitude higher than that of the D_1 emission, but no significant enhancement of the V_{Si}(V1) PL is observed. This means that while the reemission may potentially take place, it is inefficient as compared to electrical excitation. Therefore, we conclude, the recombination of electrically injected electrons and holes is responsible for the V_{Si} EL, as schematically shown in Fig. 1(b).

Finally, we present an input-output characteristic of one of our LED devices [Fig. 3(c)]. A clear tendency to saturation of the emission intensity P with injection current I is seen. This behavior can be reasonably well described by the equation $P \propto I/(I + I_0)$, corresponding to the simple model when the carrier capturing rate by the defects is proportional to the injection current. From this fit we estimate the characteristic saturation current $I_0 = 10$ mA. It is higher than that in QD-based single photon LEDs[30] and comparable to that in NV-based single photon LEDs[16].

In conclusion, we generated intrinsic defects in SiC devices and demonstrated that these defects can be electrically driven, resulting in the efficient EL with emission energies well below the SiC bandgap. Our LEDs are two-color in a sense that they show two spectrally different emission bands associated with different defects. The D_1 defects show EL in visible, which is intense at low temperatures but quenches with rising temperature. The V_{Si} defects emit in NIR even at room temperature. By varying the irradiation dose one can control defect concentration, which should allow to isolate single defects, similar to single NV centers in diamond or single semiconductor QDs. Because isolated defects are ideal single photon emitters, our findings open a new way to fabricate cheap and robust LEDs emitting single photons on demand.

Methods

The LED structures used in our experiments were grown on a n-type 6H-polytype SiC substrate. First, an epitaxial 15-μm-thick SiC layer was grown by the sublimation method. It is n-type and contains N (3×10^{18} cm^{-3}) and Ga (2×10^{18} cm^{-3}). The layer is followed by a p-type SiC layer of thickness 5 μm grown at a temperature of 2300°C in Ar atmosphere in the presence of Al vapors (pressure 100 Pa). This results in the concentration of Al acceptors of ca. 10^{20} cm^{-3}. In order to generate intrinsic defects at the p-n junction the samples were irradiated with 0.9 MeV electrons to a dose of 10^{18} cm^{-2}. After irradiation, the samples were annealed for 1 minute in Ar atmosphere at a temperature of 1700°C. At the final stage, 0.4×0.4 mm^2 Al contacts were deposited on the top of the p-type SiC layer.

EL and PL were recorded using LabRAM system for microscopy (Horiba Scientific) equipped with a CCD camera. In low temperature experiments, the samples were inserted in liquid nitrogen and the Cu back electrode plate was also used as a cold finger.

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Author contributions

F.F. conducted the experiments and analyzed the experimental data; V.A.S. and S.V. conducted the experiments; E.N.M. fabricated the samples; P.G.B., G.V.A. and V.D. conceived the experiments; G.V.A. wrote the main manuscript text; F.F. and V.D. critically reviewed and corrected the manuscript; all authors discussed the results.

Additional information

Competing financial interests: The authors declare no competing financial interests.

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