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Magneto-Luminescence Study of Silicon-Vacancy in 6H–SiC

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The magneto-spectroscopy studies of luminescence related to silicon-vacancy, in high quality 6H–SiC crystals grown by the seeded physical vapor transport method, are presented. The superior optical quality of these crystals allowed us to resolve a doublet structure of the 1.398 eV emission line (V_2 line), commonly assigned to the transitions involving two singlet states of the silicon-vacancy. Experiments performed in magnetic fields up to 20 T showed that each doublet constituent of the V_2 line splits into four components for the magnetic field parallel to the *c*-axis of the 6H–SiC crystals. This result could be hardly explained in terms of a singlet to singlet transition. The analysis of the angle-resolved luminescence experiments in high magnetic fields serves us to discuss the symmetry of the defect states responsible for the V_2-line in silicon carbide.

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1. Introduction

Silicon carbide (SiC) is a very promising wide band-gap semiconductor for application in high-power and high-frequency devices. In contrast to silicon, many

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intrinsic defects in SiC are stable at room temperature. The detailed knowledge about these defects is essential for the device performance. In spite of many efforts, information about intrinsic defects in SiC is still very limited. An important example of the defect in SiC is the silicon-vacancy. Since early 1970s, this defect has been claimed to be responsible for characteristic photoluminescence (PL) band, with zero-phonon lines at 1.433, 1.398, and 1.368 eV in 6H–SiC polytype, referred as V_1, V_2, and V_3 line, respectively [1, 2]. The number of the observed zero-phonon lines was attributed to the number of inequivalent lattice sites characteristic of 6H–SiC: V_1 and V_2 are proposed to be related with two inequivalent sites of cubic symmetry, whereas the V_2 line is expected to be connected with the hexagonal site. The assignment of V_1, V_2, and V_3 lines to the silicon-vacancy has been eventually confirmed by optically detected magnetic resonance (ODMR) studies [3]. The observed ODMR signals revealed angle behavior typical of a spin-triplet configuration. However, no evidence has been presented that those states are indeed involved in the radiative recombination. Later photoluminescence and photoluminescence excitation experiments performed in magnetic fields up to 5 T did not show any observable Zeeman effects, which leads to the conclusion that the observed zero-phonon lines correspond to transitions between spin singlet states [4].

In this report magneto-spectroscopy studies of silicon- vacancy related luminescence in high quality 6H–SiC crystals grown by the seeded physical vapor transport (PVT) method are presented.

2. Experimental details

The silicon carbide crystals used in the PL experiments were grown in the Institute of Electronic Materials Technology in Warsaw by the seeded PVT method using a graphite resistance heater. The single crystals were grown in argon atmosphere on c-face (0001) of 6H–SiC seeds. The temperature measured on the back side of the crystal holder was in the range of $2100-2300^\circ$C.

Photoluminescence measurements were performed at low temperature (4.2 K) in a magnetic field up to 20 T using a two-fiber optical system allowing PL excitation and detection. He–Cd laser operating at 3.813 eV line was used as an excitation source. PL spectra were collected for different angles between the c-axis of the 6H–SiC crystals and the magnetic field direction. The spectra were analyzed with a single 0.5 m monochromator equipped with a CCD camera. The spectral resolution provided by the experimental setup was better than 0.1 meV.

3. Results and discussion

The low temperature spectrum of high-quality 6H–SiC crystal is presented in Fig. 1. The observed energy positions of the V_1, V_2, and V_3 lines correspond very well to those attributed to the silicon-vacancy emission [1–4]. In this paper we concentrate only on the V_2 line assigned to the silicon-vacancy at the hexagonal site. As it is clearly seen in the inset of Fig. 1, the V_2 line splits into two narrow components characterized by FWHM of about 0.2 meV. In order to verify whether the initial or final recombination state is responsible for this splitting variable temperature experiments were performed. In Fig. 2 several PL spectra measured in the temperature range between 4.2 and 40 K are presented. It is observed that the relative intensities of the V_2 components remain constant in this temperature range, which strongly suggests that the observed doublet structure arises from the splitting in the final recombination state.

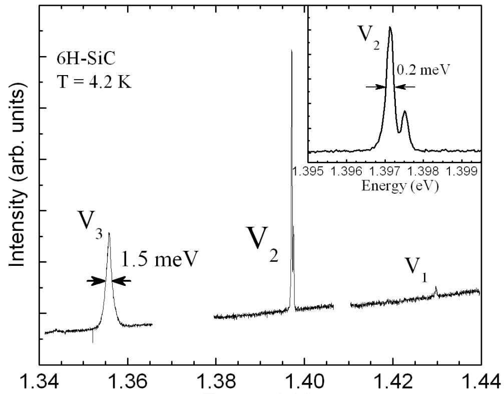

Fig. 1

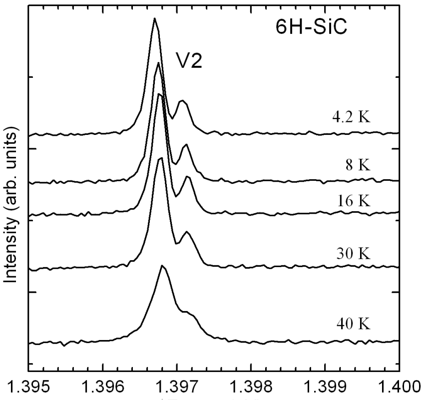

Fig. 2

Fig. 1. The low temperature (4.2 K) photoluminescence assigned to the silicon-vacancy, measured for high-quality 6H–SiC crystal. Zero field splitting of the V_2 line is presented in the inset.

Fig. 2. PL spectra of the V_2 line measured at $B = 0$ T for different temperatures between 4.2 and 40 K.

In Fig. 3, PL spectra measured in the magnetic field up to 20 T aligned along *c*-axis of the 6H–SiC crystal are presented. It is observed that each doublet component of the V_2 line splits into four components. This result clearly shows that the energetic structure involved in this recombination is more complicated than the singlet–singlet transition proposed in Ref. [4].

Taking into account that the zero field splitting of the V_2 line arises from the splitting in the final recombination state the eight-component splitting pattern observed in the magnetic field could be interpreted as a result of the recombination involving two Zeeman components of the initial state (labeled as "u" and "d" in Fig. 3) and two pairs of Zeeman doublets in the final state (labeled as "A", "B", "C", and "D"). In order to determine magnetic field behavior of the states taking part in the recombination process a phenomenological analysis similar to the procedure performed for the luminescence due to neutral acceptor in GaN [5] was performed. The emission lines observed in a magnetic field were separated

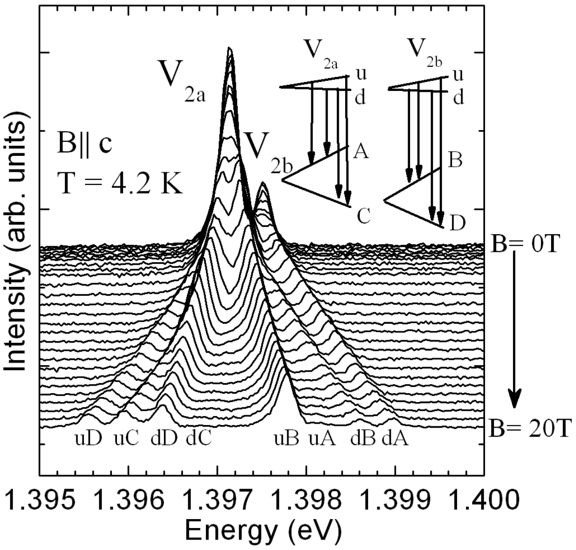

Fig. 3. PL spectra measured in the magnetic field between 0 and 20 T, applied along c-axis of the 6H–SiC crystal.

into pairs that correspond to the same final Zeeman component (eg. uD and dD in Fig. 3). The energy difference between such lines corresponds to the splitting of the initial recombination state (see diagrams in Fig. 3). The energy differences, corresponding to the splitting of the initial recombination state, calculated for all components of the split final states, as a function of the magnetic field applied parallel to the c-axis are shown in Fig. 4. It was found that splitting of the initial recombination state shows a linear dependence versus magnetic field which can be parameterized using the effective g-factor. For the magnetic field parallel to the c-axis the value $g_i^0 = 0.726 \pm .004$ was obtained.

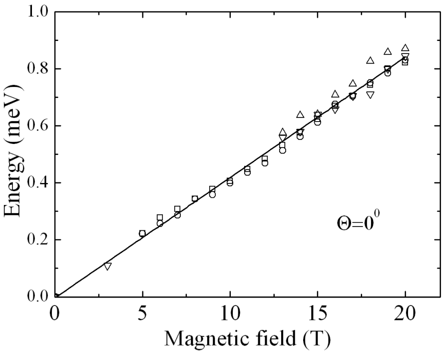

Fig. 4. Magnetic field dependence of the splitting of the initial recombination state for the magnetic field applied along c-axis ($\theta = 0°$). Different symbols represent the splitting extracted from the transitions involving different components of the final recombination state (see the text).

The same procedure performed for the magnetic field applied along a direction tilted from the c-axis by the angle θ shows that g_i decreases with increasing θ, and in the first approximation, could be described as $g_i(\theta) = g_i^0 \cos(\theta)$. In the next step of the analysis, subtracting from the transition energies, the experimentally term $E_i = \pm(1/2) g_i \mu_\mathrm{B} B$ responsible for the magnetic field dependence of the initial state, the components of the final state can be obtained. The result of such procedure is shown in Figs. 5 and 6 for $\theta = 0°$ and $\theta = 30°$, respectively.

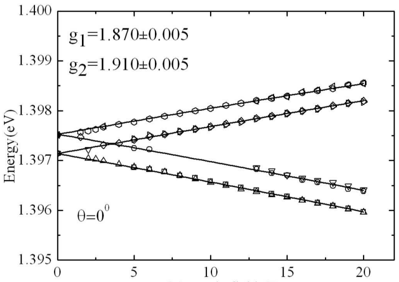

Fig. 5

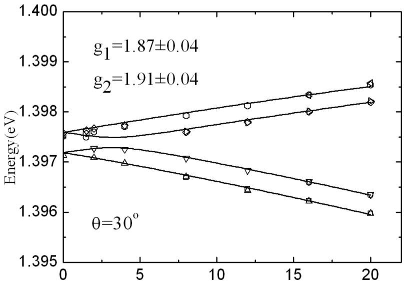

Fig. 6

Fig. 5. The splitting pattern of the final recombination state for the magnetic field applied parallel to the c-axis ($\theta = 0°$). Symbols: experimental data, solid lines — calculations (see the text).

Fig. 6. The splitting pattern of the final recombination state for the field applied along a direction tilted from the c-axis by angle $\theta = 30°$. Symbols: experimental data, solid lines — calculations (see the text).

As it is seen in Fig. 5, the magnetic field behavior of the final recombination state for $\theta = 0°$ can be successfully described assuming linear splitting of the two Zeeman doublets. The situation observed for the $\theta = 30°$ configuration is more complicated. The splittings are different from those observed for $\theta = 0°$ and show some signatures of the interaction between the split components. Such behavior can be well reproduced using an effective Hamiltonian in the following form:

$$H = \begin{bmatrix} +\frac{1}{2} g_1 \mu_\mathrm{B} B \cos(\theta) & , & 0 & , & 0 & , & +\frac{1}{2}\sqrt{g_1 g_2}\mu_\mathrm{B} B \sin(\theta) \\ 0 & , & -\frac{1}{2} g_1 \mu_\mathrm{B} B \cos(\theta) & , & +\frac{1}{2}\sqrt{g_1 g_2}\mu_\mathrm{B} B \sin(\theta) & , & 0 \\ 0 & , & +\frac{1}{2}\sqrt{g_1 g_2}\mu_\mathrm{B} B \sin(\theta) & , & -\Delta + \frac{1}{2} g_2 \mu_\mathrm{B} B \cos(\theta) & , & 0 \\ +\frac{1}{2}\sqrt{g_1 g_2}\mu_\mathrm{B} B \sin(\theta) & , & 0 & , & 0 & , & -\Delta - \frac{1}{2} g_2 \mu_\mathrm{B} B \cos(\theta) \end{bmatrix},$$

where Δ — zero field splitting of the V_2 components.

The results of calculations performed for $\theta = 0°$ and 30° are shown in Figs. 5 and 6, respectively. They reproduce experimental data very well. In both cases the same effective g-factors $g_1 = 1.87$ and $g_2 = 1.91$ were used for two interacting Zeeman doublets in the final recombination state. This simple model can be successfully used for $\theta < 60°$, while for bigger values of θ, the proper reconstruction of the fine structure of the final states involved in the V_2 emission requires a more advanced description.

4. Conclusion

Experiments performed in magnetic fields up to 20 T showed that each doublet component of the V_2 line splits into four components for the magnetic field parallel to the c-axis of the 6H–SiC crystals. This result cannot be explained in terms of the singlet to singlet transition proposed in Ref. [4]. The analysis of variable temperature and angle-resolved luminescence experiments in high magnetic fields allowed us to separate the magnetic field behavior of the initial and final recombination state corresponding to the V_2 recombination. This analysis implies that the initial recombination state consists of a single doublet, which shows a strong anisotropy, typical of the Γ_9 symmetry in the wurtzite structure, which can be described by an effective g-factor $g_i(\theta) = g_i^0 \cos(\theta)$. The final recombination state of the V_2 luminescence is formed from two interacting Zeeman doublets. The simple description presented in the paper accounts well for the experimental results obtained for the magnetic field applied along a direction tilted from the c-axis by angle $\theta < 60°$, while for larger angles a more advanced model, that properly accounts for the actual symmetry of the initial and final states involved in the V_2 recombination, is required.

Acknowledgments

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