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Photoluminescence Spectroscopy of Neutron-Irradiated Cubic SiC Crystals

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Abstract. Photoluminescence (PL) spectroscopy has been used to characterize neutron-irradiated cubic silicon carbide crystals. The effects of thermal annealing (600−1100^{O}C) on the PL bands have been studied. Several PL bands consisting of a sharp line and its phonon replicas have been observed in the 9−80 K temperature range. Certain of them like the D_1 spectrum doublet with 1.975 eV and 1.977 eV zero-phonon lines (ZPL) at 9 K and the *L2* spectrum with ZPL at 1.121 eV were reported previously for ion-implanted and electron irradiated 3C-SiC crystals, respectively. Besides, some new bands with ZPL at 2.027, 1.594, 0.989 and 0.844 eV and a broad band at 1.360 eV have been found. A correlation of PL and EPR spectra intensities of these neutron-irradiated and annealed cubic SiC crystals is briefly discussed.

Introduction

As a sensitive and nondestructive tool for characterization of structural defects in cubic SiC (3C-SiC) the photoluminescence (PL) technique has been extensively used for decades [1-6]. Valuable information about nitrogen-bound excitons [1], donor-acceptor pairs [2], the high-temperature persisted D_1 and D_2 defects [3], the defect-related *W* band near 2.15 eV and *G* band near 1.90−1.92 eV [4], the silicon vacancy-related *L2* center at 1.121 eV [5] and *E* center at 1.913 eV [6] has been gleaned from PL measurements. The radiation damage has been studied by PL spectroscopy in 3C-SiC mainly after electron irradiation and ion implantation. It should be remarked that with the exception of the *L2* center, structures of the vast majority of luminescent defect centers haven't been conclusively elucidated. For example, several suggestions as to the identity of the D_1 center have been made including divacancies, vacancy-nitrogen pairs, antisite pairs and an isolated Si antisite (see [7] and references therein). The possibility for generation of extended defects by neutron irradiation arises considerable interest, however 3C-SiC crystals haven't been actually examined. In this report, the results of our PL study of defects created by relatively high dose neutron irradiation of *n*-type cubic SiC crystals and defect transformation during thermal annealing are presented.

Experiment

Unintentionally doped *n*-type cubic SiC single crystals were grown by thermal decomposition of methyl-trichlorosilane in hydrogen, concentration of nitrogen impurity was about 10^{17} cm^{-3}. The crystals were irradiated by reactor neutrons with a dose of 10^{19} cm^{-2}, they will be labeled as 3C-SiC<n> below. Post-irradiation isochronal thermal annealing procedures were carried out in the 200−1100^{O}C temperature range. Photoluminescence and micro-Raman spectra were studied in the temperature range from 9 to 80 K using a Horiba Jobin Yvon T64000 spectrometer equipped with a confocal microscope and an automated piezo-driven XYZ stage. Discrete lines 488.0 nm and 514.5 nm of an Ar-Kr ion laser were used for excitation. An Olympus BX41 microscope equipped with 100x objective was used to focus the laser light on the sample surface to a spot with a 0.5 μm diameter and to collect the scattered light into the spectrometer. The laser power on the sample surface was always kept below 1 mW in order to obtain acceptable signal to noise ratio and to prevent laser heating. Wavelength analysis was carried by using a thermoelectrically cooled Si-CCD detector for the visible to near-infrared range and an InGaAs detector for the infrared range. The spectral resolution was less than 0.025 meV.

Results and Discussion

Figure 1 gives an overview of typical PL spectra of as-irradiated and thermally annealed 3C-SiC samples in visible and near-infrared ranges at 80 K. Their evolution is a significant evidence of annealing and modification of defects relating to radiative recombination. There are several PL bands consisting of a sharp line and its phonon replicas. Certain of them like the D_I spectrum with no-phonon line at 1.977 eV and a band with ZPL at 1.119 eV were reported previously for ion-implanted and electron irradiated 3C-SiC crystals respectively [3, 5]. Besides, a few new bands with ZPL at 2.027, 1.597, 0.988 and 0.844 eV and a broad band near 1.360 eV have been found. With the exception of the 1.597 eV band, all aforementioned bands are slightly shifted to higher energies at 9 K.

Two components 1.975 eV and 1.977 eV of the D_I no-phonon line are clearly defined at 9 K, its phonon replicas are identical to those reported previously [3]. Moreover, low intensity sharp α_1 (1.962 eV), α_2 (1.958 eV) and δ (1.921 eV) lines [6] are found at the low-energy side of the D_I no-phonon line. Coincident with growth of the D_I spectrum a new band arises with asymmetric ZPL at 1.5970 eV (80 K) and identical to the D_I phonon replica positions. An essential distinction of the 1.5970 eV line is its full width at half maximum being equal to about 15 meV as against 6 meV for the D_I ZPL at 80 K. At 9 K the latter remains as before while the near-infrared peak and its phonon replicas become markedly broader and are shifted to the low-energy side (Fig. 2). We believe that such behavior is associated with an appearance of a new low-temperature form of this spectrum with mostly the same vibrational sidebands similar to the L, M and H forms of the D_I spectrum attributed to the static and the dynamic Jahn-Teller configurations [3]. Since almost the same annealing behavior is observed between the 1.597 eV and the D_I spectra, the former can be assigned to the same defect species.

In contrast to electron-irradiated cubic SiC CVD layers with an intense broad PL band in the 0.88−1.13 eV region [5], the well-resolved *L2* spectrum has been observed for the 3C-SiC<n> samples (Fig. 1b). According to spacing of phonon replicas to the 1.119 eV no-phonon line with 34 meV, 75 meV, 95 meV and 103 meV, they can be attributed to radiative recombination with participation of the TA(L), LA(L), TO(X) and LO(X) phonons, respectively [3].

Two sharp ZPL at 0.844 eV and 0.988 eV emerge in the infrared region after annealing at 900^{O}C and 1100^{O}C respectively. The low-energy broad sidebands of these lines with several sharp peaks decrease intensities with lowering of a temperature, testifying their phonon nature. Nevertheless, no more than one sharp line being at a distance of 47 meV may be attributed to the TA(X) phonon replica. Elucidation of the origin of the rest of the lines as well as the broad structureless 1.360 eV band requires supplementary study.

Similar situation takes place with origin of the sharp 2.027 eV line. It is accompanied by a weak 2.012 eV line and its unabated intensity growth with annealing is continued up to 1000^{O}C. Nevertheless, none of 3C-SiC phonon replicas has been found. The 2.027 eV band, which is dominant in the PL spectrum of the 1000^{O}C annealed sample, is completely absent from the spectrum of the sample annealed at 1100^{O}C where the D_I band becomes principal. This suggests that a defect related to the 2.027 eV band may be a precursor of a defect associated with the D_I band.

Figure 1a shows an enhancement of Raman scattering intensity with increase in annealing temperature. As the position and width of Raman lines are unaltered a rise in their intensities can be concerned with successive improvement of sample structure due to annealing of radiation damage.

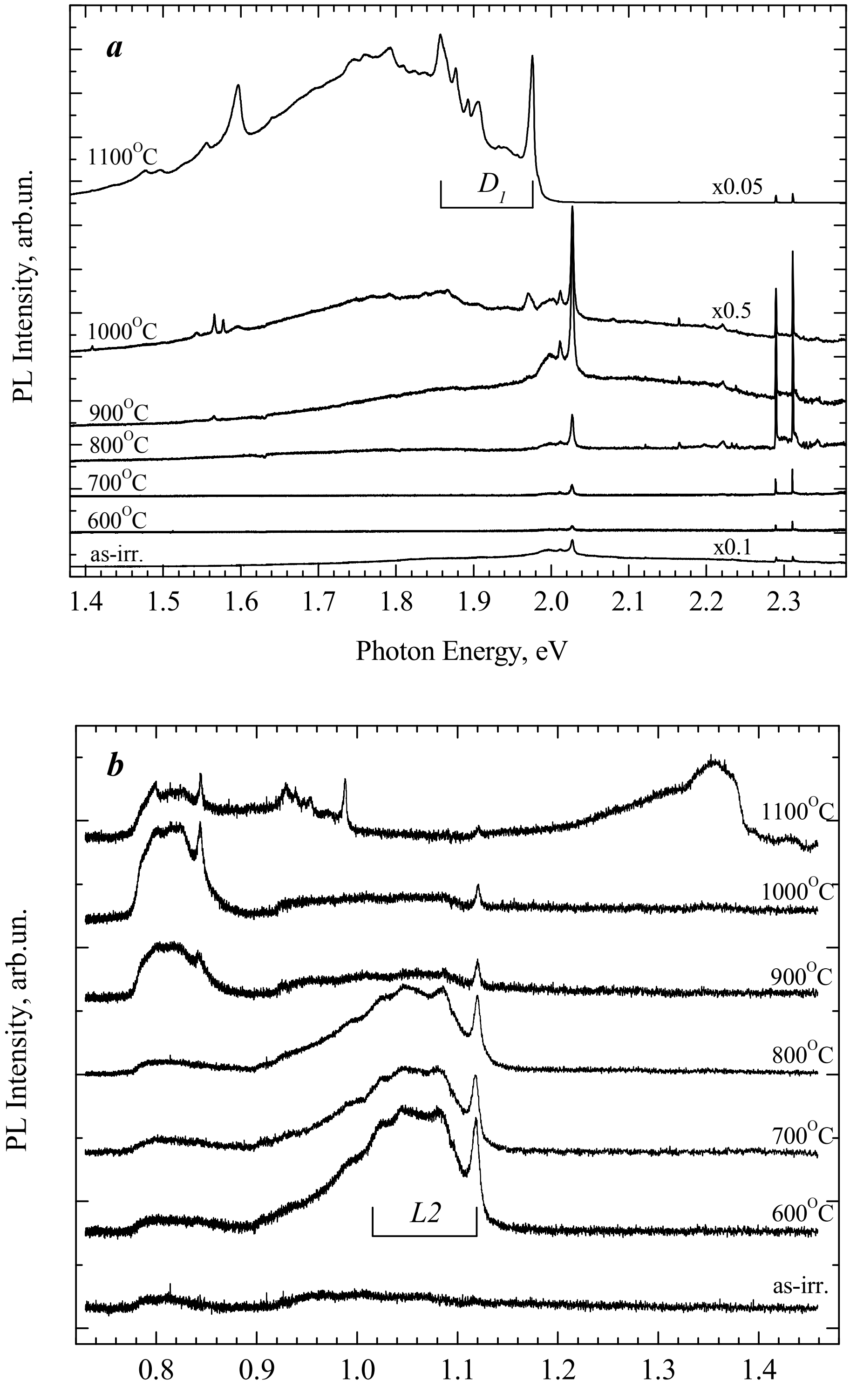

Fig. 1. Typical PL spectra obtained from neutron irradiated 3C-SiC crystals in unannealed and annealed conditions, T=80 K, $\lambda_{exc.}$=514.5 nm. The margins of the D_1 and *L2* bands are marked. A sharp doublet centered around 2.3 eV belongs to the Raman scattering lines.

Several new phonon features are observed in annealed samples in addition to the Raman active 3C-SiC modes by magnifying the intensity 10 times (Fig. 3). According to a theoretical study of localized vibrational modes [9], broad bands around 200 and 520 cm^{-1} and a band at 580 cm^{-1} can be assigned to Si−Si and Si−C vibrations of severely damaged regions respectively.

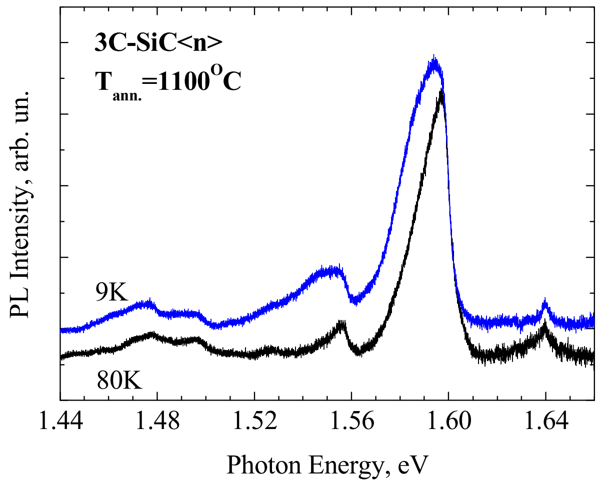

Fig. 2. The 1.597 eV band PL spectra obtained at 80 K and 9 K from annealed at 1100°C 3C-SiC<n> sample.

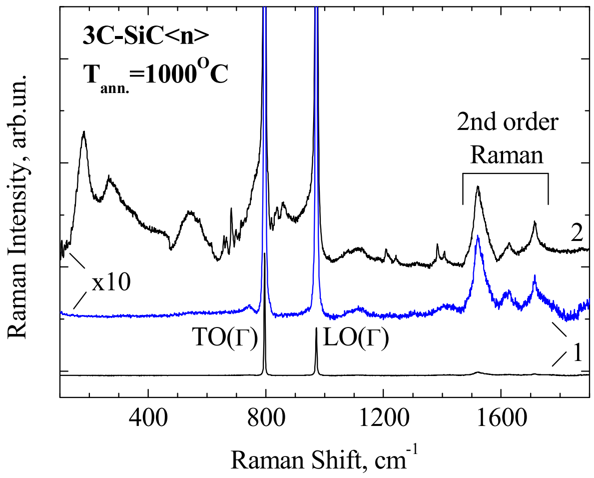

Fig. 3. Room-temperature Raman spectra from virgin 3C-SiC crystal (1) and annealed at 1000°C 3C-SiC<n> sample (2).

Notice, these bands become considerably weaker after 1100°C anneal. Sharp Raman lines in the 640–710 cm^{-1} interval can be attributed to the localized vibrational modes of the antisite Si_C and pair of antisites Si_C–C_{Si} [9]. As regards weak Raman modes in the 1200–1420 cm^{-1} region, they can be associated with different types of bonded carbon interstitial defects [3].

It should be pointed out that the *L2* spectrum attenuation with annealing and its actual disappearance after anneal at 900°C correlates with behavior of the *T1* EPR signal [10] related to negatively charged silicon vacancy. In addition, correlation has been found for intensities of the 0.844 eV PL band and the *T6* paramagnetic defect attributed to a carbon vacancy–interstitial pair.

In summery, the PL spectroscopy has been used to characterize neutron-irradiated and thermally annealed 3C-SiC crystals. Several new PL bands, which are likely to be associated with extended defects, have been observed.

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