SPECIAL TOPIC — Ion beam technology

In situ luminescence measurement of 6H-SiC at low temperature[*]

Meng-Lin Qiu(仇猛淋)[1], Peng Yin(殷鹏)[1], Guang-Fu Wang(王广甫)[1,2,†], Ji-Gao Song(宋纪高)[1],
Chang-Wei Luo(罗长维)[1], Ting-Shun Wang(王庭顺)[1], Guo-Qiang Zhao(赵国强)[1],
Sha-Sha Lv(吕沙沙)[1], Feng-Shou Zhang(张丰收)[1,2], and Bin Liao(廖斌)[1]

[1] *Key Laboratory of Beam Technology of Ministry of Education, College of Nuclear Science and Technology, Beijing Normal University, Beijing* 100875, *China*
[2] *Beijing Radiation Center, Beijing* 100875, *China*

To understand the evolution of defects in SiC during irradiation and the influence of temperature, *in situ* luminescence measurements of 6H-SiC crystal samples were carried out by ion beam induced luminescence (IBIL) measurement under 2 MeV H^+ at 100 K, 150 K, 200 K, 250 K, and 300 K. A wide band (400–1000 nm) was found in the spectra at all temperatures, and the intensity of the IBIL spectra was highest at 150 K among the five temperatures. A small peak from 400 nm to 500 nm was only observed at 100 K, related with the D1 defect as a donor–acceptor pair (D–A) recombination. For further understanding the luminescent centers and their evolution, the orange band (1.79 eV) and the green band (2.14 eV) in the energy spectrum were analyzed by Gaussian decomposition, maybe due to the donor–deep defect/conduction band–deep defect transitions and Ti related bound excition, respectively. Finally, a single exponential fit showed that when the temperature exceeded 150 K, the two luminescence centers' resistance to radiation was reduced.

1. Introduction

Silicon carbide (SiC), a third generation semiconductor material, has shown excellent physical properties: wide band gap, high intrinsic temperature, high critical breakdown field strength, high thermal conductivity, and resistance to irradiation. Hence, it has been applied in fabricating high-frequency and radiation resistant electron devices, which are expected to be operated in the radiation environments of the military and astronomy.[1,2]

Ion implantation could be helpful for controlling the carrier life due to defects that are created during irradiation.[3] However, heavy radiation damage during the irradiation up to amorphization could influence the properties of SiC. In previous works, luminescence measurements have been applied to study the defects induced by irradiation, such as photoluminescence (PL),[4–6] cathodoluminescence (CL),[7] and thermoluminescence (TL).[8] Studies of defects in SiC mainly focused on important defects such as the D_1 center, E_1/E_2, and Z_1/Z_2.[9,10] There are three no-phonon lines contained in the D_1 center in 6H-SiC: L_1 = 472.3 nm (2.625 eV), L_2 = 478.8 nm (2.590 eV), and L_3 = 482.5 nm (2.570 eV). All these lines were observed in low temperature photoluminescence measurement. However, up to now, there is no consensus regarding the micro-structure around the D_1 center. The Z_1/Z_2 defect and E_1/E_2 defect could be observed by deep level

transient spectroscopy in n-type 6H-SiC after being irradiated by electrons.

However, all the mentioned studies of SiC were conducted after irradiation or annealing and therefore the formation and quenching of defects with the ion beam can not be observed during the irradiation or annealing. During the irradiation, point and structural defects would be produced mainly by nuclear interactions as well as the related luminescence centers (photons emitted from outer shell transitions of materials) excited by intensive ionization. Hence, ion beam induced luminescence (IBIL) is a very sensitive technique to monitor the formation and quenching of the optically active defects directly (with the best detection limit of part per billion), through the luminescence evolution during the irradiation with the ion beam.[11,12] However, IBIL has rarely been used in the analysis of defects in SiC.

In this work, the IBIL spectra of SiC were acquired under 2 MeV H^+ at various low temperatures, helping to understand the evolution of defects in SiC during irradiation, and the dependence on temperature.

2. Experiment details

The single crystal 6H-SiC (10 mm×10 mm×1 mm, ⟨0001⟩) used in this work was supplied by MTI Corporation (KJ Group, China). The doping element was nitrogen to make

it into an n-type semiconductor. The samples were irradiated by 2 MeV H$^+$ at the GIC4117 2×1.7 MV Tandem accelerator in Beijing Normal University. The IBIL setup has previously been introduced in detail in Ref. [12]. The beam current was approximately 20 nA with a diameter of 6.7 mm. The integration time was 0.5 s for each spectrum. The total fluence during the irradiation was approximately 1.27×10^{14} ions/cm^2. The

spectra were obtained at five different temperatures (100 K, 150 K, 200 K, 250 K, 300 K).

3. Result and discussion

Figure 1 shows the evolutions of IBIL spectra of n-type 6H-SiC crystals under 2 MeV H$^+$ bombardment with fluence at five different temperatures.
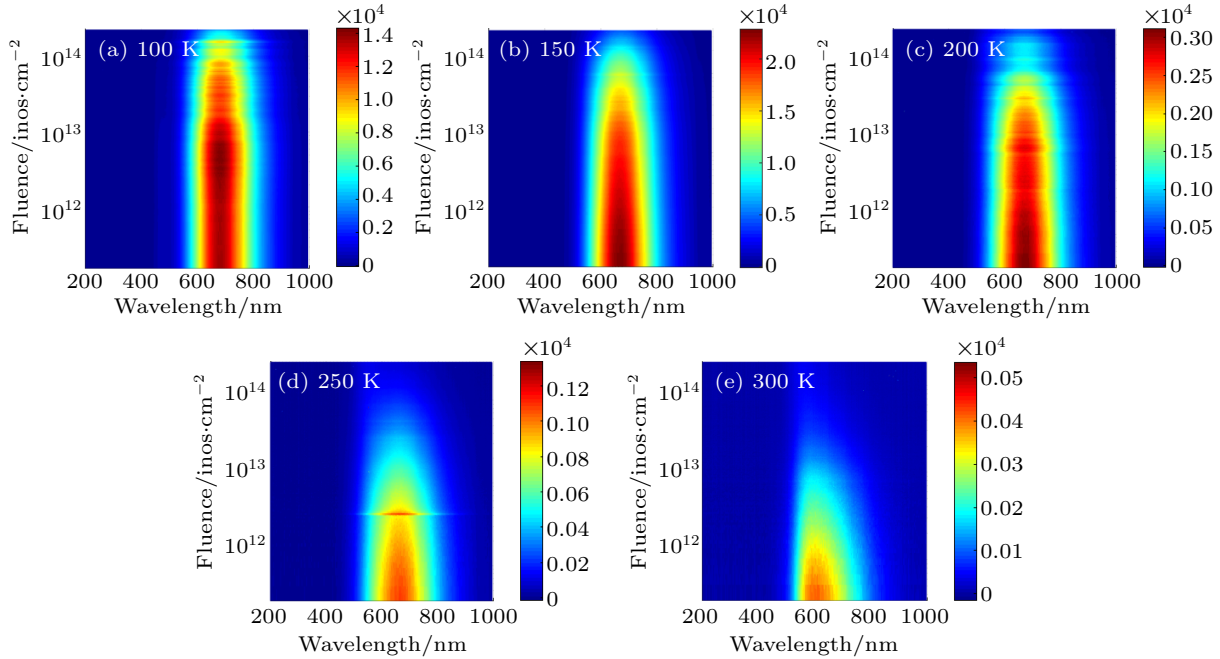

Fig. 1. Evolution of IBIL spectra of n-type 6H-SiC crystals under 2 MeV H$^+$ bombardment with fluence at five different temperatures: (a) 100 K, (b) 150 K, (c) 200 K, (d) 250 K, (e) 300 K.

The main wide bands in the IBIL spectra at five temperatures all appeared from 400 nm to 1000 nm. The intensities monotonically decreased with fluence and the decay rate slowed down with the dropping of temperature. The probable causes of these phenomena were the quenching of the optically active defects damaged by the incident ions, as well as the slowing defect migration rate and the increasing free carrier lifetime with the decreasing temperature. In addition, it should be pointed out that the spike at 250 K was due to the sudden beam current fluctuation, even though the synchronous counting of the back scattering ions was acquired to correct the luminescence intensity. The band at 300 K exhibited the significant asymmetry, which indicates that the band has several components, and the intensities of different components decreased with fluence at different rates.

To compare the evolution of luminescence intensities with temperature, the typical spectra of 6H-SiC at different temperatures with the same fluence of 5×10^{11} ions/cm^2 are shown in Fig. 2(a). All the spectra had similar shapes. The maximum intensity of the IBIL spectra of 6H-SiC first increased with increasing temperature, then decreased with increasing temperature above 150 K. Additionally, a small peak around 400–500 nm was observed when the temperature was

100 K. The D$_1$ defect could be the origin of this peak. [13–15] According to the references, [4,5] the D$_1$ defect was assigned to a donor–acceptor pair (D–A) recombination, and the D$_1$ defect was extremely temperature stable. However, when the temperature exceeded 130 K, its intensity in the PL spectrum of 6H-SiC was extremely weak, matching well with the IBIL results. Figure 2(b) shows the intensities at different temperatures that were normalized to the maximum. The obvious shifts between peak centers were observed, but presenting a non-monotone variation with temperature (683 nm at 100 K, 665 nm at 150 K, 670 nm at 200 K, 664 nm at 250 K, 617 nm at 300 K). Different types of luminescence centers showed particular sensitivities to the temperature. Hence, the different intensity ratios and specific evolution rates of the components in a band at different temperatures could be the possible reason of this phenomenon.

The overlapping between the closed components of a wide band was common in luminescence spectra, which would have a negative effect on the fitting model of the spectral data. The parameters in a suitable Gaussian function could have each correlated actual physical meanings. Therefore, the Gaussian function was often used to fit the spectra to further study the evolution of the spectra. [16] The Gaussian decom-

position shown in Fig. 3 for the IBIL spectrum of n-type 6H-SiC irradiated by 2 MeV H$^+$ at 150 K suggested that the peak around 670 nm was actually composed of two peaks at 1.79 eV (orange band) and 2.14 eV (green band).
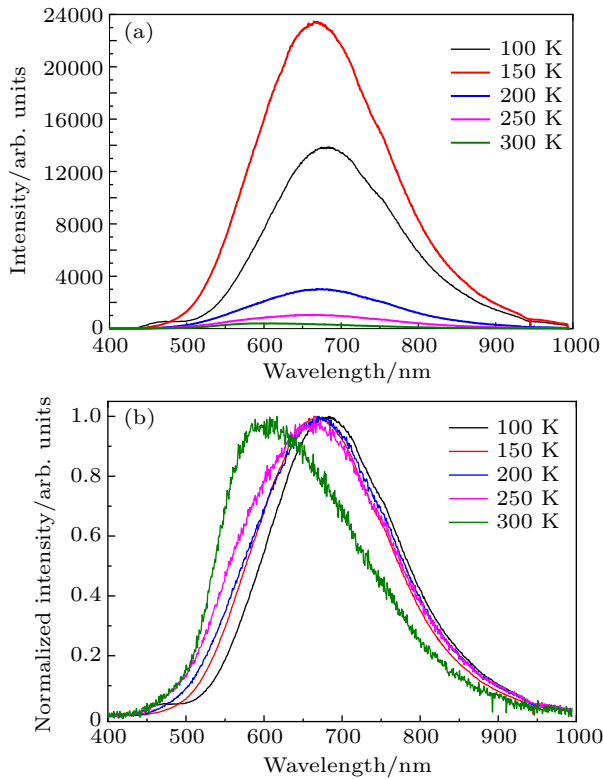

Fig. 2. The IBIL spectrum of n-type 6H-SiC irradiated by 2 MeV H$^+$ with the same fluence (5×10^{11} ions/cm^2) at different temperatures (a) and normalized to the maximum (b).
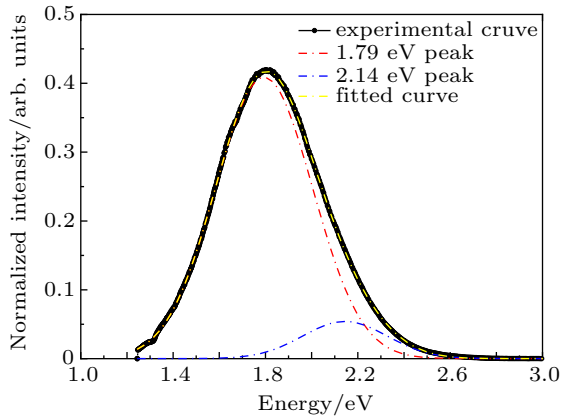

Fig. 3. A typical Gaussian decomposition of IBIL spectrum of n-type 6H-SiC irradiated by 2 MeV H$^+$ with a fluence of 5×10^{11} ion/cm^2 at 150 K.

As shown in Table 1, the band center positions gained by the Gaussian decomposition of the IBIL spectrum were relatively stable. Figure 4 shows that the full widths at half maximum (FWHM) of the fitted Gaussian peaks at 1.79 eV and 2.14 eV were stable with fluence.

Table 1. The band center positions of two bands with the same fluence.

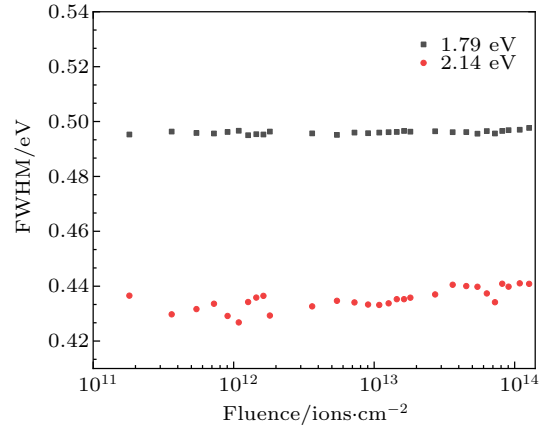| Temperature/K	100	150	200	250	300
Orange band/eV	1.771	1.793	1.771	1.758	1.807
Green band/eV	2.136	2.139	2.137	2.102	2.136

Fig. 4. The FWHM evolution of the fitted peaks at 1.79 eV and 2.14 eV with fluence at 150 K.

According to Ref. [5], the orange band combined with the blue band can be explained in a model, assuming a donor–acceptor pair (DAP) for the blue band emission and the donor–deep defect and conduction band–deep defect transitions for the orange band. According to the results of PL, the emission intensity of the blue band decreased quickly when the temperature increased and the emission was restrained when the temperature exceeded 130 K. To the contrary, the emission intensity of the orange band increased with increasing temperature up to 140 K, and then decreased.[5] This matched with the IBIL result shown in Fig. 2(a). For the green band, it may be caused by the Ti related bound exciton.[6] In the formation of the 6H-SiC crystal, the Ti would be unintentionally doped. The Ti related bound exciton is an effective luminescence center and its recombination could cover a board range from 1.79 eV to 2.86 eV at a low temperature. However, the process of bound exciton recombination would be suppressed as temperature increased.

The Gaussian decomposition of the IBIL spectrum of the n-type 6H-SiC irradiated by 2 MeV H$^+$ at other temperatures also found the same band, but with the peak position shifted. Table 1 gives the positions of the two bands' peaks at different temperatures. The peak position of the orange band was strongly influenced by the temperature, but the peak position of the green band was stable with temperature. This may be due to the ionization of the subsequent donor level.[5]

Figures 5 and 6 show the evolution of the 1.79 eV peak and 2.14 eV peak with fluence. The intensities of the two luminescence centers monotonously decreased as the fluence increased. The quenching of the luminescence centers could be due to the crystal structure damage induced by ion beam and the accompanying trapping of electrons and holes by different types of defects.[12] Furthermore, a single exponential formula fitting was used to study the fluence evolution of the 1.79 eV peak and 2.14 eV peak gained from the Gaussian decomposition,

$$I = a + b\mathrm{e}^{-F/f}, \qquad (1)$$

where I is the luminescence intensity, F is the fluence, and a, b, f are the fitting parameters.[17] The f is the parameter of the decay rate. Its value could represent the resistance of the luminescence center to radiation damage, which is usually applied to express the anti-radiation performance or self-repairing capability in the spectral analysis of scintillator materials under different conditions. The larger f would mean the slower evolution rate and indicate the better stability or the stronger radiation resistance.[17,18] Table 2 lists the values of f at different temperatures, f_a belonging to the 1.79 eV peak and f_b belonging to the 2.14 eV peak. When the temperature exceeded 150 K to 300 K, there was a negative impact on the resistance to radiation for both luminescence centers. The higher temperature would result in the higher defect migration rate and shorter free carrier lifetime. Hence, the quenching ef-

fect would be enhanced and the resistance of the luminescence center gradually dropped from 150 K to 300 K. However, a positive impact was shown with temperature below 150 K. The main cause may be due to the competition of the non-radiative recombination centers, as well as the decrease of the effective damage cross section, with the rising temperature up to around 150 K.[19] In addition, the existence of the D_1 defect at 100 K may be another influence factor. Finally, to a certain extent, the varied evolution rates deduced from the values of f_a and f_b matched the discussion of Figs. 2(a) and 2(b).

Table 2. The f calculated using Eq. (1) to fit the fluence evolution of the two peaks at different temperatures, with f_a belonging to the 1.79 eV peak and f_b belonging to the 2.14 eV peak.

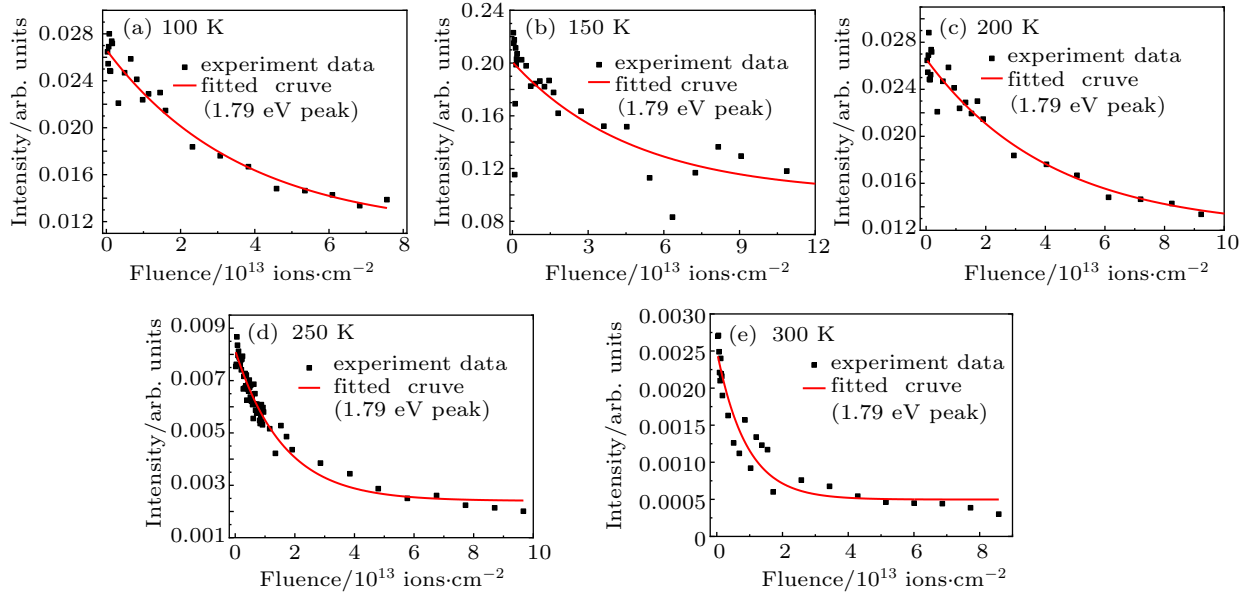| Temperature/K	100	150	200	250	300
$f_a/10^{13}$ ions/cm^{-2}	3.65	4.81	4.18	1.61	0.9
$f_b/10^{13}$ ions/cm^{-2}	3.49	5.18	3.85	1.33	0.623

Fig. 5. The evolution of the 1.79 eV peak with fluence at five different temperatures: (a) 100 K, (b) 150 K, (c) 200 K, (d) 250 K, (e) 300 K.
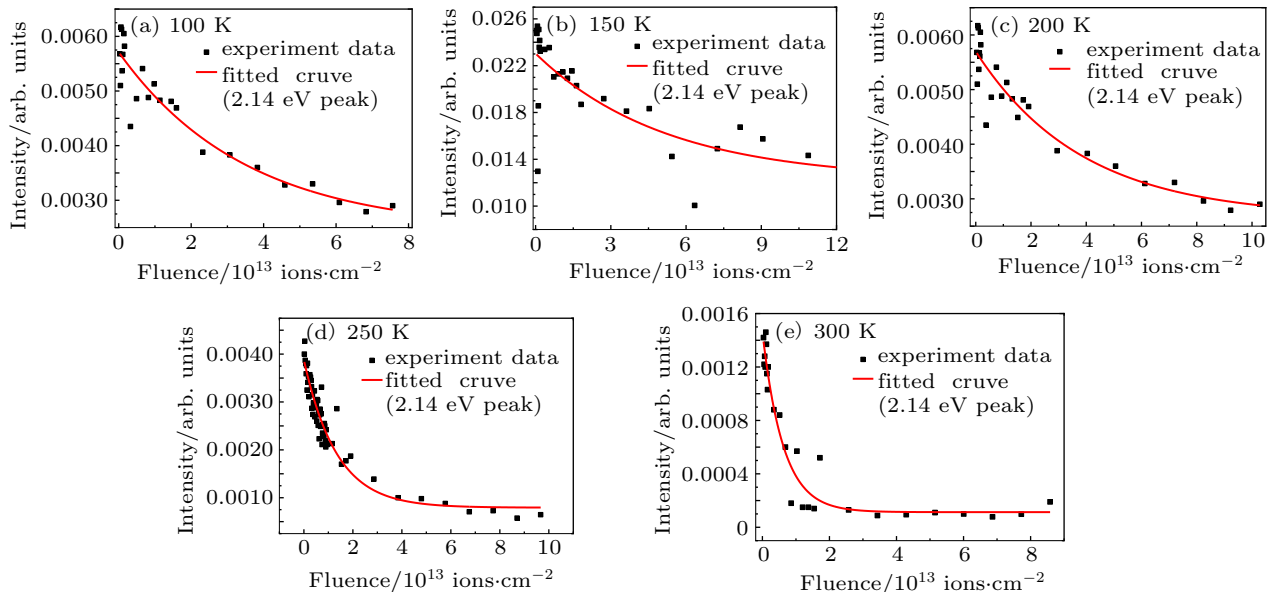

Fig. 6. The evolutions of the 2.14 eV peak with fluence at five different temperatures: (a) 100 K, (b) 150 K, (c) 200 K, (d) 250 K, (e) 300 K.

4. Conclusion

In summary, IBIL could be used to analyze the evolutions of defects during irradiation. Three bands were found in the IBIL spectra of 6H-SiC. The blue band, due to the D_1 defect, only appeared at 100 K. The other two bands calculated by Gaussian decomposition existed at all temperatures. The orange band was caused by the donor–deep defect and conduction band–deep defect transitions. For the blue and orange bands, the results of IBIL matched the PL results well. The green band could be explained by the Ti related bound excition. The whole luminescence intensity was the highest at 150 K among the five temperatures. Additionally, the intensity increased as the temperature increased up to 150 K. Finally, according to decay rate parameter f from the single exponential fitting, the temperature had an important impact on the resistance to radiation of the two luminescence centers. When the temperature exceeded 150 K to 300 K, the resistance to radiation of the two luminescence centers decreased. The phenomenon appeared at 100 K needs further study.

References

[1] Neudeck P G 1995 *J. Electron. Mater.* **24** 283
[2] Casady J B and Johnson R W 1996 *Solid. State. Electron.* **39** 1409
[3] Fissel A, Richter W, Furthmüller J and Bechstedt F 2001 *Appl. Phys. Lett.* **78** 2512
[4] Storasta L, Carlsson F H C, Sridhara S G, Bergman J P, Henry A, Egilsso T, Hallén A and Janzén E 2001 *Appl. Phys. Lett.* **78** 46
[5] Wysmołek A, Mroziński P, Dwiliński R, Vlaskina S and Kamińska M 1995 *Acta Phys. Pol. A* **87** 437
[6] Huang W, Chen Z Z, Chang S H, Li Z Z and Shi E W 2010 *Mater. Sci. Eng. B* **170** 139
[7] Young A P, Jones J and Brillson L J 1999 *J. Vac. Sci. Technol. A.* **17** 2692
[8] Wei Y, Künecke U, Wellmann P and Ou H 2017 *2017 International Conference on Silicon Carbide and Related Materials*, September 17–22, 2017, Washington DC, USA
[9] Eberlein T A G, Jones R, Öberg S and Briddon P R 2006 *Phys. Rev. B* **74** 144106
[10] Klettke O, Reshanov S A, Pensl G, Devaty R P and Choyke W J 2001 *Physica B* **308–310** 687
[11] Peres M, Alves L C, Rocha F, Catarino N, Cruz C, Alves E, Silva A G, Víllora E G, Shimamura K and Lorenz K 2018 *Phys. Status. Solidi. A* **215** 1800190
[12] Qiu M L, Yin P, Luo C W, Zheng L, Chu Y J, Xu M and Wang G F 2019 *Nucl. Instrum. Meth. Phys. Res. B* **450** 69
[13] Pensl G, Schmid F, Ciobanu F, Laube M, Reshanov S A, Schulze N, Semmelroth K, Schöner A, Wagner G and Nagasawa H 2003 *Mater. Sci. Forum* **433–436** 365
[14] Devaty R P and Choyke W J 1997 *Phys. Status Solidi. A* **162** 5
[15] Wang O, Ding Y L, Zhong, Z Q, Yuan J, Gong M, Chen X D, Fung S and Beling C D 2004 *Chinese. J. Light Scattering* **16** 66 (in Chinese)
[16] Chu Y J, Wang G F, Qiu M L, Xu M and Zheng L 2016 *Chin. Phys. Lett.* **33** 106101
[17] Zheng L, Wang G F, Qiu M L, Chu Y J, Xu M and Yin P 2017 *Chin. Phys. Lett.* **34** 087801
[18] Umeda N, Vasilets V N, Bandourko V V and Kishimotoc N 2002 *Nucl. Instrum. Methods Phys. Res. Sect. B* **191** 708
[19] Sullivan P A and Baragiola R A 1994 *J. Appl. Phys.* **76** 4847