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RADIATION DEFECTS IN A NEUTRON-IRRADIATED DIAMOND, INVESTIGATED BY PERSISTENT SPECTRAL HOLE BURNING

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Abstract Polarized luminescence and absorption spectra of neutron-irradiated IaB type diamond have been measured in the 700 – 750 nm spectral range. From these measurements the $\langle 111 \rangle$ orientation of electric dipoles for the 723.8 nm line has been concluded. Spectral hole burning (SHB) in the 723.8, 730.9, 731.7, 734.3 nm lines has been performed which allows the analysis of the dynamics of the lattice defects.

Keywords: diamond, neutron-irradiation, hole-burning, lattice defects

INTRODUCTION

Due to attractive industrial applications of diamond a lot of attention has been paid to the investigation of defects and impurities in it.¹ The main impurity in all natural diamonds is nitrogen which exists in the form of single atoms, pairs (A-defects) and larger aggregates (B-defects). A combination of nitrogen with the interstitials and vacancies created by particle irradiation makes the picture quite complicated.² In many spectroscopic investigations defects in the Ib-type diamond, in particular the N-V centre have been studied.^{3,4,5} Recently a thorough experimental work was accomplished, concerning vacancy-related defects in different diamond types.⁶ SHB has been observed in the spectra of N-V, GR1, H4 defects⁷ and photochromic transformation between H2 and H3 defects has been studied.⁸

In our previous investigation⁹ we have performed spectral hole burning in the 649.5, 655 and 681 nm lines in neutron-irradiated IaB-type diamond. In these lines we could create spectral holes thermostable up to 200 K and determine the location of photoproduct for the hole in the 649.5 nm line. It should be mentioned that the thermostability of holes is one of the main preconditions for applications of SHB in information storage.

In this study the polarization spectroscopy and SHB were used to gain information about the defects giving rise to the lines in 700 – 750 nm spectral region.

EXPERIMENTAL RESULTS

A piece of natural IaB-type diamond was subjected to irradiation with a 10^{19} cm^{-2}

dose of high-energy neutrons and annealing at 950°C for half an hour. The measurements were performed in the temperature range of $5 - 100\text{ K}$. Photoluminescence was excited near 704 nm by using an Ar^{+} -ion-laser-pumped linear cw dye laser ($\sim 0.03\text{ nm}$). The emission parallel to the excitation path was recorded by a photon-counting system through a double-pass monochromator. Hole burning was also accomplished with a $\text{Xe} - \text{Cl}$ -excimer-pumped pulsed dye laser.

The emission and absorption spectra of the sample are shown in Figure 1. At 723.8 nm a ZPL could be detected in both spectra. In addition, the spectral region contains lines peaking at 726 , 730.9 , 734.3 and 738.5 nm in luminescence and at 731.7 and 733.1 nm in absorption. From temperature dependence of the latter lines (Figure 3A) a splitting of the ground state levels ($\sim 28\text{ cm}^{-1}$) was concluded.

The asymmetry of the shape of the 731.7 nm line should be noted. A similar asymmetry has been observed for a Si -related defect with ZPL at 738 nm .¹⁰

At LHeT the lines in the spectra are inhomogeneously broadened by 20 to 60 cm^{-1} and are clearly detectable up to temperatures 100 K , being without any significant broadening. None of the lines was observable at room temperature.

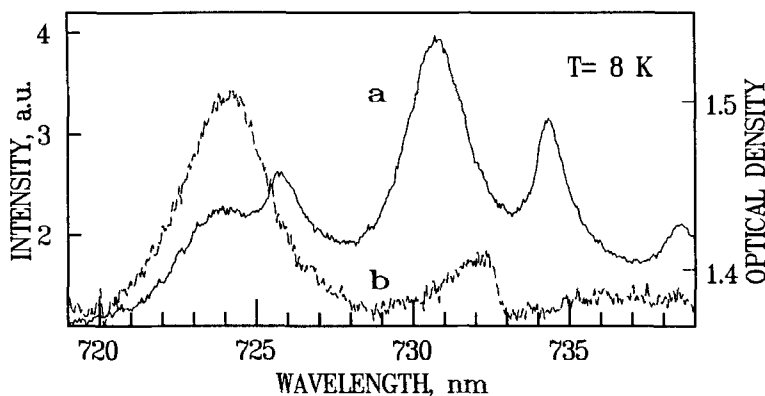


FIGURE 1 The emission (a) and absorption (b) spectra of the neutron-irradiated IaB-type diamond. Photoexcitation at 704 nm , $T = 8\text{ K}$.

In order to determine the orientation of electric dipole moments of the transition polarized luminescence experiments were undertaken. An analysis of the results of these measurements allows the determination of dipole orientation.¹¹

Measurements were carried out in the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions and polarization of luminescence was measured for different orientations of the \vec{E} vector of the exciting beam. Figure 2 shows the polarized spectra for a definite experimental geometry. It was concluded that the electric dipoles of the centre giving rise to the 723.8 nm line are orientated along the $\langle 111 \rangle$ axis of the crystal. As for the other centres, the orientation could not be established reliably.

Persistent HB with a cw laser has been performed in the 723.8 , 730.9 , 731.7 , 734.3 nm lines. At $T = 8\text{ K}$ a 10-minute exposure to 100 mW/mm^2 laser irradiation

yielded holes with the depth of 80 % in the 723.8 nm line. A 50 mW/mm² 3-minute exposure at 734.3 and 730.9 nm caused holes with the depth of 50 % and 30 % respectively. A low-temperature hole width was of the order of 0.1 nm.

As for thermostability, the holes in the 723.8 and 730.9 nm lines are detectable at temperatures up to 70 K, where the intensities of the lines start to decrease essentially.

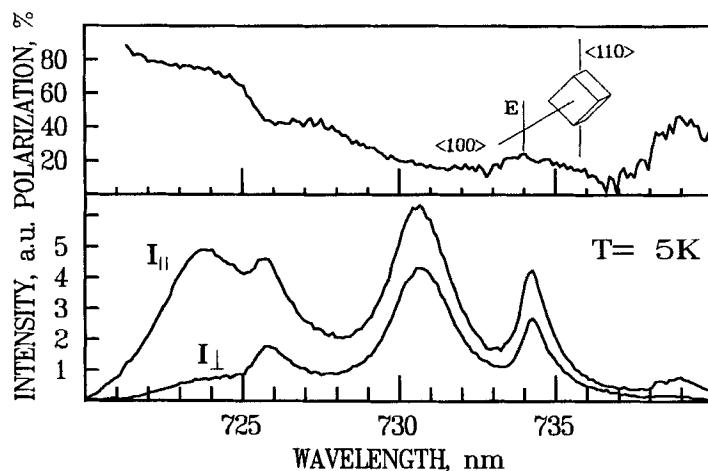


FIGURE 2 Polarized luminescence spectra of the neutron-irradiated diamond. Exciting light beam is parallel to the $\langle 100 \rangle$ direction and plane-polarized with the \vec{E} -vector parallel to the $\langle 110 \rangle$ axis of the crystal.

A strong cross-talk effect of the holes in the 731.7 nm line should be mentioned (Figure 3B). First hole produced at 732 nm (curve a) is almost completely filled as a result of burning at a slightly different site (curve b). This effect is characteristic of a nonphotochemical hole burning process where products are generated dispersed over the whole region of the ZPL.

The burning of a hole at 723.8 nm causes a decrease of the intensity of the 734.3 nm line, which gives evidence of the same origin of the lines. Also, a proportional decrease of the intensity of the 723.8 and 731.7 nm lines has been registered after irradiating the sample at 723.8 nm with a pulsed laser.

None of the lines mentioned above belongs to the well-known centres except the 723.8 nm line which has been observed earlier.¹² The data obtained from the spectroscopic measurements are not sufficient to determine the nature of the defects. Yet it is supposed that they are combinations of nitrogen and radiation defects formed in the course of annealing. It is known that the self-interstitials are mobile at RT⁶ (activation energy 0.15 eV) and that vacancies migrate at temperatures > 600 K (2.3 eV).¹³ In Ia-type crystals vacancies are usually trapped at A and B aggregates, producing H3 and H4 defects (vibronic bands with ZPL-s at 503 and

496 nm). It should be mentioned that the decomposition of the paired nitrogen and the vacancy-enhanced migration of single N have been observed at 800° C.²

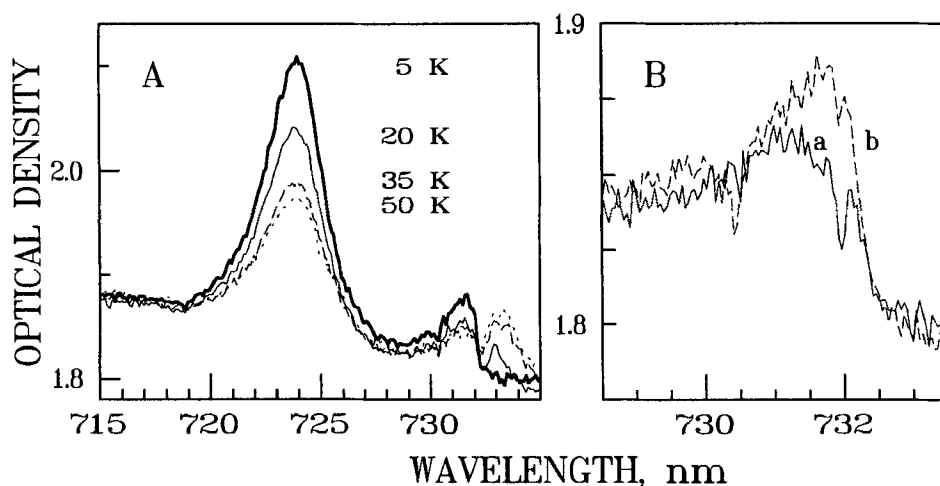


FIGURE 3 A - the temperature dependence of the absorption spectrum of neutron-irradiated diamond; B - cross-talk effect of the holes in the 731 nm line.

CONCLUSION

This study revealed 723.8, 730.9, 731.7, 733, 734.3, 738.5 nm lines in the spectra of a neutron-irradiated IaB-type diamond. These lines belong to the optically active centres created by neutron irradiation and subsequent annealing and possibly formed of nitrogen aggregates and radiation defects. The $\langle 111 \rangle$ orientation of the electric dipoles corresponding to the 723.8 nm line was concluded from the polarized luminescence experiments. Persistent SHB is performable in the 723.8, 730.9, 734.3 and 731.7 nm lines where the holes burnt in at *LHeT* are observable at *LN₂T*. Hole burning revealed that the intensity of the 723.8 nm line is correlated to the intensities of the 734.3 and 731.7 nm lines. Also a nonphotochemical nature of the burning process in the 731.7 nm line was concluded from the cross-talk effect of the holes.

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