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Response function measurement of layered type CVD single crystal diamond radiation detectors for 14 MeV neutrons

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Response function measurement of layered-type chemical vapor deposition single crystal diamond radiation detectors for 14 MeV neutrons was carried out. The detector had a layered structure that was composed of a boron-doped diamond layer of $0.5\ \mu\text{m}$ in thickness and a nondoped diamond layer of $20\ \mu\text{m}$ on an inexpensive high pressure and high temperature-type Ib diamond substrate. The detector had energy resolution of 2.6% for 5.5 MeV α particles. This experiment was mainly carried out in order to understand the present status of the detector as a 14 MeV neutron spectrometer and an extent of charge trapping. As result, a peak caused by the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ reactions was clearly observed; the best energy resolution of 6% as for a synthetic diamond radiation detector was achieved. Detection efficiency was 3.2×10^{-7} counts/unit neutron fluence. However, taking the energy resolution for α particles, etc., into account, the energy resolution for 14 MeV neutrons was not so high. Further improvement based on better crystal growth is indispensable.

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I. INTRODUCTION

A diamond radiation detector has several merits in terms of high radiation resistance,¹ high temperature operation,² high chemical resistance,³ etc. Moreover, a diamond radiation detector can be applied to 14 MeV neutron energy spectroscopy using the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ reactions. This method corresponds to neutron energy spectroscopy using the $^{28}\text{Si}(n, \alpha)^{25}\text{Mg}$ and $^{28}\text{Si}(n, p)^{28}\text{Al}$ reactions in a silicon surface barrier detector.⁴ This capability of a diamond radiation detector was reported by Kovalchuk in 1994 in Ref. 5. The energy resolution of 2% for 14 MeV neutrons was the best result reported in the previous studies using a natural diamond radiation detector.⁶ This energy resolution satisfied a required capability of a 14 MeV neutron energy spectrometer for ion temperature measurements for DT plasmas. A very compact size of a diamond radiation detector has an advantage in plasma diagnostics that are usually forced strict spatial limitation. Moreover, a diamond radiation detector is at least 100 times tougher than a silicon surface barrier detector in radiation resistance.¹ As mentioned above, a diamond ra-

diation detector has ideal properties for a 14 MeV neutron energy spectrometer for plasma ion temperature measurements. However, it was very difficult to routinely produce diamond radiation detectors that had enough performance, because natural diamond crystals for this purpose were very scarce.

On the other hand, a synthetic technique of diamond has made remarkable progress. The authors reported the first trial of 14 MeV neutron energy spectroscopy using a radiation detector made of a synthetic diamond grown by a high pressure and high temperature (HP/HT) method in Ref. 7. Although the detector succeeded to work as an energy spectrometer, there was severe trapping on electrons⁸ and its reduction was indispensable for fruition of a practical detector. Accordingly, characterization of impurities,⁹ applying higher purity diamond crystals¹⁰ and estimation of charge trapping mechanism into the HP/HT-type IIa diamond crystal¹¹ were carried out. At the same time, the application of chemical vapor deposition (CVD) diamond single crystal was carried out successfully.¹² However, it was revealed that yield rate of CVD single diamond for energy spectrometer was extremely poor. Recently, development of a layered-type CVD single crystal diamond radiation detector was

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FIG. 1. Schematic drawing of a cross section of a layered-type CVD single crystal diamond radiation detector. The detector was fabricated on an inexpensive HP/HT-type Ib diamond single crystal substrate. The boron-doped CVD diamond layer worked as a contact. The nondoped CVD diamond layer of 20 μm in thickness was a sensitive layer.

succeeded,¹³ one main motivation of this development was improvement of yield rate. In this article, response function measurement of a layered-type CVD single crystal diamond radiation detector for 14 MeV neutrons was described. This experiment was carried out in order to obtain a guide to the next improvement, namely fabrication of a thick sensitive layer.

II. DETECTOR AND EXPERIMENTAL SETUP

A. Layered-type CVD single crystal diamond radiation detectors

Figure 1 shows a schematic drawing of a cross section of a layered-type CVD single crystal diamond radiation detector. This detector was developed aiming at improvement of poor yield rate of CVD single crystal diamond radiation detectors and fabrication of a thin sensitive layer enough for α particle energy spectroscopy.¹³ This detector was fabricated on an inexpensive HP/HT-type Ib diamond single crystal substrate, i.e., yellow diamond. The detector had a layered structure composed of a boron-doped single crystal diamond layer of 0.5 μm thickness and a nondoped single crystal diamond layer of 20 μm on the substrate. These layers were homoepitaxially grown on (100) surface of the substrate by plasma assisted CVD method in order of boron-doped and then nondoped diamond layers. After crystal growth, a part of the nondoped diamond layer was removed by oxygen plasma etching with a metallic mask; at this part, a Ti/Au electrode was fabricated by evaporation, and the boron-doped diamond layer worked as a back contact. Moreover, an aluminum Schottky contact was fabricated on nondoped diamond layer by evaporation, too.

Owing to the boron-doped diamond contact, the detectors had strong rectification properties. The detectors had energy resolution of 2.6% and 2.8% for 5.5 MeV α particles.¹³ These values were far from higher energy resolution of 0.4% achieved by radiation detectors made of the HP/HT-type IIa single crystal diamond¹² and the CVD single crystal diamond.¹⁴ It had already been known that the quality of the nondoped diamond layers was not very high; it was obvious from their cathode luminescence spectra in which the strong band A luminescence around 420 nm was observed with the free exciton recombination luminescence at 235 nm, indicat-

ing high quality. It was probably caused mainly by poor crystallinity of the boron-doped layer owing to growth instrumentation; the boron-doped layer gave a bad influence on crystallinity of the nondoped layer on it. On the other hand, the detector had a very thin 20 μm layer that was very difficult to fabricate by mechanical polishing and a self-standing diamond substrate, thus there was the possibility of improvement on suppression of charge trapping on electrons that was a big issue with the HP/HT-type IIa diamond. Compared with self-standing CVD single crystal diamonds, the yield rate of the layered-type detector was overwhelmingly high. In this experiment, two layered-type CVD single crystal diamond radiation detectors were used.

B. Experimental setup and measurement electronics

Response function measurement experiment for 14 MeV neutrons was carried out at the fusion neutronics source (FNS) facility of the Japan Atomic Energy Research Institute. The diamond radiation detector was set at an exit of a 14 MeV neutron collimator. At the position of the detector, peak energy of neutrons was calculated to be 14.2 MeV, and it had energy broadening of 1.0% in full width at half maximum. The detector was connected to a CANBERRA 2003T charge sensitive preamplifier by a cable whose length was 15 cm. A CANBERRA 2021 spectroscopy amplifier and Genie-ESP analysis system were used in this measurement.

III. EXPERIMENTAL RESULT AND DISCUSSIONS

Figure 2 shows examples of response functions for 14 MeV neutrons obtained by one of the layered-type CVD single crystal diamond radiation detectors. Bias voltage of +15 V (7.5 kV/cm) was applied to the aluminum contact, and gain and shaping time of the spectroscopy amplifier were 0.6 $\times$ 1 k and 10 μs , respectively. Continuous measurement of 120 min was carried out with counting rate of approximately 20 counts/s. Figure 2(a) shows the pulse height spectrum obtained during the first 40 min, and Fig. 2(b) shows the pulse height spectrum obtained during the third 40 min, namely from 80 to 120 min. In Fig. 2(a), basic structure caused by the $^{12}\text{C}(n,n')^{12}\text{C}$, $^{12}\text{C}(n,n')^{12}\text{C}^*$, $^{12}\text{C}(n,n')3\alpha$ and $^{12}\text{C}(n,\alpha_0)^9\text{Be}$ reactions¹⁵ was clearly observed. In contrast, the structure became dull with increase of measurement time as shown in Fig. 2. It was obvious we had to get the influence of accumulated charge in the detector.

Figure 3 shows a response function measured by another detector. In this measurement, bias voltage of +25 V (12.5 kV/cm) was applied to the aluminum contact. Gain and pulse shaping time constant of the spectroscopy amplifier were 0.6 $\times$ 500 and 6 μs , respectively. To obtain this spectrum, six times of a 20 min measurement were summed up. Between each measurement, some bias voltage of opposite polarity was applied to the detector in order to remove space charge. As result, the fine spectrum compared with Fig. 2 was obtained. Moreover, energy resolution of 6% at the peak caused by the $^{12}\text{C}(n,\alpha_0)^9\text{Be}$ reactions was achieved; it was the best result for synthetic diamond radiation detectors.

Detection efficiency was 2.8×10^{-7} counts/unit neutron flux for this detector. Figure 4 shows dependence of detec-

FIG. 2. Change of pulse height distribution spectra obtained by the layered-type CVD single crystal diamond radiation detector for 14 MeV neutrons due to measurement time. (a) the first (0–40 min) measurement of series three times measurement in total of 120 min, (b) the third (80–120 min) measurement. Bias voltage of +15 V was applied to the aluminum contact. Gain and shaping time of a spectroscopy amplifier was 0.6×1 k and 10 μ s, respectively. It was obvious that spectra were distorted according to the measurement terms.

tion loss on the thickness of a sensitive layer calculated by analytical approximation. Range of α particles created by the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ reactions was approximately 15 μm in diamond. Thus, for 20 μm in detector thickness, approximately 18% of counts caused by the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ reactions was not

FIG. 3. An example of response function of the layered-type CVD single crystal diamond radiation detector for 14 MeV neutrons. Bias voltage of +25 V (12.5 kV/cm) was applied to the aluminum contact. Gain and shaping time of linear amplifier were 0.6×500 and 6 μ s, respectively. This spectrum was made by summing up of six times of a 20 min measurement. Between each measurement, bias voltage of opposite polarity was applied to the detector in order to remove space charge in the detector.

FIG. 4. Dependence of detection efficiency of the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ reactions on thickness of sensitive layer of diamond. This figure was calculated using an analytical approximation. The range of α particle produced by the $^{12}\text{C}(n, \alpha_0)^9\text{Be}$ was approximately 15 μm . In this calculation, influence of ^9Be was ignored, because the range was at most 2 μm .

able to deposit all the energy to the detector. However, taking the bad energy resolution of approximately 3% for α particles and the broadening of neutron energy of 1% into account, calculated detection efficiency of 3.2×10^{-7} was almost equal to the experimental result of 2.8×10^{-7} .

For the detectors used in this study, it was difficult to observe transfer properties of electrons and holes separately, because the thickness of the sensitive layers was almost the same length as a range of a 5.5 MeV α particle, i.e., 14 μm . If there was no charge collection loss in the detector, approximately 65% of the pulse height of a signal was composed by motion of electrons for 5.5 MeV α particles.¹⁶ Moreover, the detectors had strong rectification properties, thus it was impossible to change drift direction of electrons or holes by applying opposite bias polarity. As reported in Ref. 13, in the experimental results obtained using α particles, the detectors had low charge collection efficiency and slow drift velocity of both charges, i.e., $700\text{--}2 \times 10^4$ cm/s. On the other hand, for 14 MeV neutrons, all reactions occurred in the detector homogeneously. Therefore, average contribution to a pulse height of a signal was equal for both electrons and holes. Taking all the information described above into account, there were charge losses not only on electrons but also on holes. Probably, there were trapping centers related to bad crystallinity.

In order to improve detector performance, improvement on the boron-doped diamond layer and optimization for growth condition of nondoped diamond layer are indispensable. For the latter issue, a growth condition achieving higher growth rate of 4 $\mu\text{m}/\text{h}$ with very high crystal quality was observed. If this growth condition applied to fabrication of the detector, a 100- μm -thick detector can be fabricated in 25 h. Therefore, the authors believe that a practical synthetic diamond radiation detector as a 14 MeV neutron energy spectrometer will come about in the near future.

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