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A diamond 14 MeV neutron energy spectrometer with high energy resolution

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A self-standing single-crystal chemical vapor deposited diamond was obtained using lift-off method. It was fabricated into a radiation detector and response function measurements for 14 MeV neutrons were taken at the fusion neutronics source. 1.5% of high energy resolution was obtained by using the $^{12}C(n,\alpha)^{9}Be$ reaction at an angle of 100° with the deuteron beam line. The intrinsic energy resolution, excluding energy spreading caused by neutron scattering, slowing in the target and circuit noises was 0.79%, which was also the best resolution of the diamond detector ever reported. © *2016 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4940929]

I. INTRODUCTION

Diamond radiation detectors have promising properties that include high temperature operation,[1,2] low leakage current, high breakdown voltage, and solar blindness[3] because of the wide bandgap of 5.5 eV. They also have irradiation resistance against fast neutrons,[4] fast response because of high mobility, and high saturation drift velocity.[5,6] Furthermore, they are useful for ion temperature measurements based on the 14 MeV neutron measurement using the $^{12}C(n,\alpha)^{9}Be$ reaction.[7]

Energy resolution of 2.15% for $^{12}C(n,\alpha)^{9}Be$ reactions using natural diamond detector was reported by Krasilnikov *et al.* in 1997.[8] This energy resolution satisfies deuterium-tritium (DT) plasma ion temperature measurements. The method uses direct reactions between 14 MeV neutrons and carbon. No other reaction occurs below 1 MeV of the peak attributable to the $^{12}C(n,\alpha)^{9}Be$ reaction. Therefore, it is possible to measure ion temperatures up to 50 keV.[7] An ideal diamond radiation detector can achieve detection efficiency of 1.0×10^{-5} count/(unit neutron flux) with effective volume of $2.5 \times 2.5 \times 0.13$ mm^3.[9] The detector satisfies DT plasma ion temperature measurements of 100 ms in International Thermonuclear Experiment Reactor (ITER)[7,10] with small size.

Other measurement methods exist for plasma ion temperature based on 14 MeV neutron such as magnetic proton recoil (MPR) spectroscopy[11] and a combination of recoil proton detection and neutron ToF spectroscopy, so-called TANSY.[12] However, these measurements require large instruments of several meters. Particularly, MPR requires several meters of magnetic poles and coils. TANSY requires an approximately 1 m ring scintillator array. In stark contrast, a diamond radiation detector is only several square millimeters. Consequently, it presents benefits for plasma diagnostics, which are usually restricted by spatial limitations. Small diamond detector arrays of the future are expected to be able to supply spatial distribution measurements.

With respect to semiconductor detectors, conventional silicon surface barrier detectors have been widely used in Joint European Tours (JET), but radiation damage appears by 10^{11}–10^{12} neutrons/cm^2.[13] The detectors endure larger neutron fluence in ITER. Moreover, the detector cannot exchange frequently once the experimental period starts. Consequently, silicon surface barrier detectors are difficult to use in such harsh conditions. However, diamond radiation detectors have 100 times radiation resistance compared with silicon because they have small displacement energy per atoms as a result of low atomic number products. Pillon *et al.* reported that diamond radiation detectors show stable energy spectra up to 2×10^{14} neutrons/cm^2 of 14 MeV neutrons.[14] This fluence is greater than the fluence which the detector irradiated during one year of ITER operation. Moreover, a diamond radiation detector is solar blind and is less gamma-ray sensitive than silicon because of its wide bandgap and low atomic number.

As described above, diamond radiation detectors have ideal properties for 14 MeV neutron spectrometry. However, detector grade natural diamond is scarce. Consequently, its application has remained limited. To produce spectroscopic grade diamond routinely, chemical vapor deposition (CVD)

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diamond has been applied to radiation detectors.[14–18] Recent CVD growth technique has enabled routine production of spectroscopic grade diamond. Cazzaniga *et al.* reported 2%–3% energy resolution for $^{12}C(n,\alpha)^{9}Be$ reactions.[17] Nonetheless, CVD diamond has 0.3%–0.4% of energy resolution for 5.486 MeV alpha particles. Energy resolution for $^{12}C(n,\alpha)^{9}Be$ was worse considering neutron energy broadening caused by scattering and slowing in a tritide titanium target. Some room remains for improvement. Sato *et al.* reported that this energy resolution reduction resulted from several percent of electron charge trapping.[18–20] In 14 MeV neutron measurements, charge generation was uniform in the whole sensitive area. The contributions of induced charge were almost identical between holes and electrons. The measured peak was spreading due to electron charge collection process. Thus, energy resolution for $^{12}C(n,\alpha)^{9}Be$ became significantly worse than the energy resolution for alpha particles.

This report describes the achievement of the best energy resolution for 14 MeV neutrons ever reported. A single-crystal CVD diamond radiation detector having excellent charge carrier transportation properties for both holes and electrons was synthesized and fabricated into a radiation detector in our laboratory. A response function for 14 MeV neutrons was measured at the fusion neutronics source (FNS).

II. EXPERIMENTAL

A. Crystal growth and detector preparation

A single-crystal CVD diamond was grown homoepitaxially on a HP/HT type-IIa diamond substrate with an off-angle of three degrees for the ⟨110⟩ direction to suppress abnormal growth. Before CVD growth, the substrate was implanted with carbon ions at energy of 3 MeV and fluence of 2×10^{16} #/cm^2 to fabricate a graphite layer for lift-off processing.[21,22] Then, CVD growth was done on the (001) direction using a microwave plasma reactor (ASTeX 5250; MKS Instruments, Inc.). We prepared two Hokkaido University's samples: HU#1 and HU#2. The gas pressure, methane concentration, and microwave power were, i.e., 110 Torr, 0.25%, and 750 W for HU#1 and 110 Torr, 0.20%, and 1100 W for HU#2, respectively. The substrate temperature was kept 850–900 °C during CVD growth. A self-standing layer was lifted-off from the substrate by electrical chemical etching. The self-standing layer thickness and the growth rate were 70 μm and 0.36 μm/h for HU#1, and 150 μm and 0.86 μm/h for HU#2, respectively.

Then it was oxygen-terminated using dichromic acid. Subsequently, the titanium contact was fabricated on the substrate side of the crystal by evaporation. It was annealed at 400 °C for 30 min to form titanium carbide. Then the gold contact was fabricated on the titanium carbide. The titanium carbide/gold ohmic contact was connected to a Sub Miniature version A (SMA) receptacle using silver paste for bias and read out of signals. The aluminum Schottky contact was fabricated on as-grown side. Then it was connected to the ground.

Alpha-particle-induced charge distribution measurements were conducted for charge collection efficiency evaluation. We used 5.486 MeV alpha particles from an ^{241}Am radioactive source, a charge-sensitive preamplifier (142A; Ortec), a spectroscopy amplifier (672; Ortec), and a multi-channel analyzer (WE7562; Yokogawa Analytical Systems, Inc.). Alpha-particles were injected into aluminum contact side. A silicon surface barrier detector (CU012-050-100; Ortec) was used as the standard for charge collection efficiency. Average energies for electron and hole pair creation of silicon and CVD diamond, i.e., 3.62 eV and 13.1 eV,[23] were used for charge collection efficiency calculations.

TABLE I. Charge transportation properties of the single-crystal CVD diamond detector.

	Charge collection efficiency[a] (%)		Energy resolution (%)		
Sample I.D.	Hole	Electron	Hole	Electron	Thickness (μm)
HU#1	100.1	99.9	0.38	0.38	70
HU#2	100.0	99.7	0.38	0.38	150

[a]Charge collection efficiency was calculated given average hole–electron pair creation energy, i.e., $\varepsilon_{si} = 3.62$ eV and $\varepsilon_{diam} = 13.1$ eV.

In 14 MeV neutron measurements, a charge carrier generates in the whole sensitive area of the diamond detector. Typically, charge transportation properties of electrons were worse than those of holes for scCVD diamond. The full width at half maximum (FWHM) for the $^{12}C(n,\alpha)^{9}Be$ reactions spreads due to worse charge collection efficiency and energy resolution of electrons. The detector with nearly perfect charge collection for both holes and electrons is required to achieve excellent energy resolutions. Table I presents charge collection efficiency and energy resolution for HU#1 and HU#2. High energy resolutions of 0.38% were obtained for both holes and electrons. Electron charge loss was improved from 3% to 0.2% per 100 μm. Details of charge collection properties were reported elsewhere.[24]

B. Response function measurements for 14 MeV neutrons

Response function measurements for 14 MeV neutrons were conducted at the FNS facility of the Japan Atomic Energy Agency (JAEA). The experimental setup is shown in Figure 1. The DT neutron fields of 80° FNS beam line were used. DT neutrons were generated by a 350 kV deuteron beam incident to titanium tritide target. The beam current was 0.5–1 mA. Neutron yields were ca. 10^{11} neutrons/s. A charge-sensitive

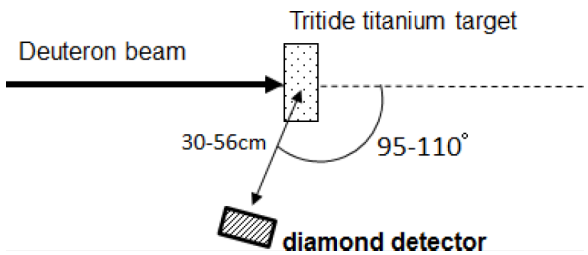

FIG. 1. Experimental setup for response function measurements at FNS 80° beam line. The deuteron beam energy, beam current, neutron yields were 350 kV, 0.5–1 mA and ~10^{11} neutrons/s, respectively. A detector was settled from 30 to 56 cm from the target at an angle of 95°–110° with respect to the deuterium beam for suppressing energy broadening.

preamplifier (142A; Ortec), a spectroscopy amplifier (672; Ortec), a high-voltage power supply (428; Ortec), and a multichannel analyzer (WE7562; Yokogawa Analytical Systems, Inc.) were used for pulse height analysis. An ^{241}Am alpha particle was used for energy calibration. Krasilnikov *et al.* reported that an angle of 95° with respect to the deuterium beam has the smallest neutron energy spreading because of neutron scattering and slowing in the target. Thus, a detector was settled from 30 to 56 cm from the target at an angle of 95°–110° for suppression of energy spreading of DT neutron. The count rate was approximately 700 cps in maximum during the measurements.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 depicts an example of response function for 14 MeV neutron obtained by HU#1. The angle formed with the deuteron beam was 100°. The FWHM of the peak attributable to the ^{12}C(n, α)^{9}Be reaction was 1.5%. The earlier best energy resolution was 2.15% obtained using natural diamond for the same neutron fields that Krasilnikov *et al.* reported in an earlier study.[8] Although CVD diamond growth technology improved, no detector achieved better energy resolution. We achieved the best energy resolution as a 14 MeV neutron energy spectrometer in this study. Figure 3 shows the ^{12}C(n, α)^{9}Be peak shift obtained by HU#1 at an angle of 95°, 100°, and 110° with deuteron beam. The marked ca. 100 keV peak shift and shape variation were observed because of the 5° difference. Table II presents data related to the detector, the measurement angle with the deuteron beam, count rate, and energy resolution. At 95° with the beam line, the HU#1 and HU#2 achieved 1.7% and 1.8% of energy resolution, respectively.

We also evaluated intrinsic energy resolution of diamond detector. The measured 14 MeV neutron spectra energy resolutions σ_{ex} were the following:

$$\sigma_{ex}^2 = \sigma_{diam}^2 + \sigma_{sc}^2 + \sigma_{sl}^2 + \sigma_{noise}^2. \quad (1)$$

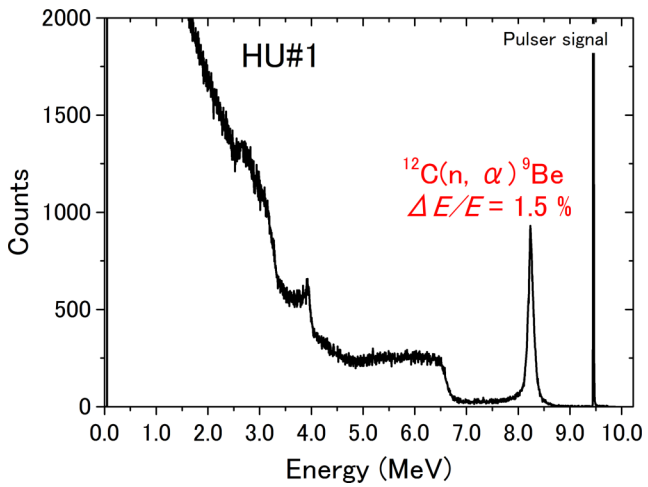

FIG. 2. Example of a response function for DT neutrons obtained using HU#1. The angle with the deuteron beam was 100°. The count rate was approximately 200 cps. The applied electric field was 1.3 V/μm. The energy resolution of the peak attributable to the ^{12}C(n, α)^{9}Be reaction was 1.5%.

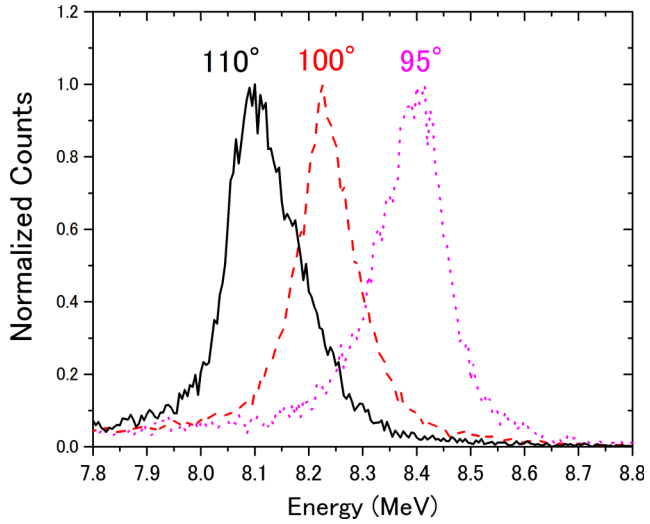

FIG. 3. ^{12}C(n, α)^{9}Be peak shifts obtained by HU#1 at angles of 95°, 100°, and 110° with a deuteron beam. Solid line, dashed line, and dotted line, respectively portray the ^{12}C(n, α)^{9}Be reaction peak at 110°, 100°, and 95°. The marked approximately 100 keV peak shift and shape variation were observed attributable to 5°–10° difference.

In that equation, σ_{diam} is the intrinsic energy resolution of diamond detector. σ_{sc} and σ_{sl} denote energy broadening caused by neutron scattering and slowing in the target. σ_{noise} represents the circuit noise. Krasilnikov *et al.* reported σ_{sc} = 1.48% and σ_{sl} = 0.23% at the angle of 95°.[8] These values were estimated using TRIM code. The circuit noise estimated using a pulser was 0.14% for HU#1 and 0.12% for HU#2 in the present measurements. The intrinsic energy resolution calculated using Equation (1) was σ_{diam} = 0.79% for HU#1 and 0.99% for HU#2. Krasilnikov described intrinsic energy resolution σ_{diam} = 1.95% at an angle of 95° in a report of an earlier study.[8] Cazzaniga reported σ_{diam} = 2.5% obtained at an angle of 90° using CVD diamond.[17] The intrinsic energy resolution obtained in the present work was the best ever reported.

Though the best energy resolutions were obtained, there were some inconsistencies between neutron energy calculations and experimental data. Krasilnikov *et al.* reported that the energy spreading of neutron minimizes at 95°,[8] on the other hand, our experimental data show the smallest energy spreading at 100°.

Provided that energy resolution for 8.4 MeV of ^{12}C(n, α)^{9}Be peak was 0.38% and charge collection efficiency difference between holes and electrons was 0.2%, the calculated

TABLE II. Energy resolution and measurement conditions of DT neutron response function.

Sample I.D.	Angle with the deuterium beam line (deg)	σ_{ex} (%)	$\sigma_{diamond}$ (%)	Count rate (cps)	Electric field (V/μm)
HU#1	110	1.9	...	300	1.3
	100	1.5	...	400	
	95	1.7	0.79	200	
HU#2	110	2.0	...	700	1.3
	100	1.5	...	400	
	95	1.8	0.99	200	

intrinsic energy resolution was 0.47%. Nonetheless, the intrinsic energy resolution calculated by experimental data and neutron energy spreading considering neutron scattering and slowing only in the target was 0.79%. This difference can be caused due to scattering of assembly around the target. This neutron energy spread calculated by law of propagation of errors between calculation and experimental values was 0.6%. To estimate intrinsic energy resolution more precisely, neutron transport calculation which includes detailed assembly around the target will be required as a future work.

Since there has not been a neutron energy spectrometer which has superior to 1.0% of high energy resolution until now, the diamond detector is expected to be applied for detailed comparison of neutron transport calculation with experiment.

IV. CONCLUSION

For this study, self-standing single-crystal CVD diamond radiation detectors were fabricated using lift-off method. Response function measurements were conducted at the FNS 80° beam line. The 1.5% high energy resolution was obtained at an angle of 100°. The intrinsic energy resolution excluding neutron scattering, slowing in the target and circuit noises was 0.79%, which was the best resolution ever reported. We will strive to achieve intrinsic energy resolution to ca. 0.4% for the $^{12}C(n,\alpha)^{9}Be$ reaction in future works by improving the electron charge collection efficiency. Similarly, we will plan to compare neutron transport calculations including detailed assembly around the target with experiment to determine intrinsic energy resolution more precisely.

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