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First use of single-crystal diamonds as fission-fragment detector

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ABSTRACT

Single-crystal chemical vapor-deposited diamond (sCVD) was investigated for its ability to act as fission-fragment detector. In particular we investigated timing and energy resolution for application in a simultaneous time-of-flight and energy measurement to determine the mass of the detected fission fragment. Previous tests have shown that poly-crystalline chemical vapor deposited (pCVD) diamonds provide sufficient timing resolution, but their poor energy resolution did not allow complete separation between very low-energy fission fragments, α-particles and noise. Our present investigations prove artificial sCVD diamonds to show similar timing resolution as pCVD diamonds close to 100 ps. Improved pulse-height resolution allows the unequivocal separation of fission fragments, and the detection efficiency reaches 100%, but remains with about a few percent behind requirements for fragment-mass identification. With high-speed digital electronics a timing resolution well below 100 ps is possible. However, the strongly varying quality of the presently available diamond material does not allow application on a sufficiently large scale within reasonable investments.

1. Introduction

The study of neutron-rich nuclei is important to reveal the characteristics of nuclear matter far from stability. One question in this context is the evolution of the shell structure at large deformation and increasing neutron excess. One experimental approach to produce nuclei far from stability at a sufficient rate is by means of neutron-induced fission. In this process two fragments are produced with a ratio between the heavy and the light fragment mass ranging from 1 to about 2.3, at least one of them created with a large neutron excess, whose properties may then be investigated spectroscopically.

A fissioning nucleus undergoes a deformation process, either spontaneously or particle-induced, until it disintegrates at the so-called scission point into two more or less deformed fragments. Both fragments share the excess energy, which is not transferred into kinetic energy. This excitation energy, typically of the order of several tens of MeV, is subsequently released through the emission of neutrons and γ-rays at an early stage after scission. By measuring the multiplicity and total energy of the different particles emitted, in conjunction with the corresponding fragments' mass and kinetic energy, we can learn about the mechanism of the nuclear fission process. Those experimental data enter in fission models, which may allow calculating fission characteristics for isotopes not accessible for experiments because of their short lifetime, high radioactivity or small abundance.

In particular, neutron- and photon-induced reactions on actinides are used to study the fission process and to supply the community with fission fragment data relevant for producing the evaluated nuclear data files (cf. Refs. [1–3]). The continuous improvement of the quality of those evaluated nuclear data files is of utmost importance for the successful design of the next generation nuclear reactors [4], contemplated serving for a cleaner, more sustainable and safe energy supply.

The desired observables to be measured in fission are primarily mass, kinetic energy and nuclear charge of both fission fragments as well as the characteristics of emitted prompt neutrons and γ-rays.

A direct way to determine fission fragment masses is to measure both their velocities and kinetic energies. An experimental setup with a flight path of 50 cm and an energy resolution typical for a silicon detector, i.e. 0.3% FWHM, would require measuring the time-of-flight (TOF) between fission source and fragment detector with a timing resolution of the order of 100 ps (FWHM) in order to achieve a mass resolution of $A/\Delta A \approx 100$. Commonly used parallel-plate

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avalanche counters as fission-event triggers, however, provide an intrinsic timing resolution of the order of 300–400 ps (FWHM). Micro-channel plate based detectors may be faster by a factor of 2 at least, but are very difficult to operate and sensitive to radiation damage.

Another application, where a good timing resolution in fragment detection matters, is the measurement of prompt fission γ-rays. Since those γ-rays are emitted simultaneously with fission neutrons, the time-of-flight technique is applied to separate both. With the recent availability of fast scintillators made of lanthanide halides (see e.g. Refs. [5,6] and references therein) for prompt γ detection, with a timing resolution for a $LaBr_3$:Ce detector of size 5.0 cm × 5.0 cm (diameter × length) as good as 338 ± 18 ps (FWHM) at optimum energy resolution [7], it is important to achieve a comparable or even better intrinsic timing resolution with the fission-fragment detector.

Furthermore, it would be desirable that the fission fragment detector could provide energy information that could be used for identification of the fission fragments to allow studying the dependence of prompt fission γ-ray emission of mass and energy of the fragments.

Another issue is that, due to the very broad fission-fragment mass and energy distributions in fission, a sufficiently high number of events have to be recorded, especially in measurements where coincidence is required with fission γ-rays or neutrons. It is therefore desirable to have an efficiency as high as possible. A simple and economic way of doing so is to maximize the solid angle by placing the detectors as close as possible to the target. Unfortunately, fission experiments involving actinides have often to deal with very high α activity, fragment emission and intense neutron fields. Thus, the detectors must be very resistant to radiations and have as little mass as possible, so that the neutron background is minimized. The relatively small kinetic energy per nucleon and the high charge number of the fragments also implies very short stopping range in matter, requiring minimum energy loss before the fragments enter the active detector volume. Double Frisch-grid ionization chambers can fulfill all these requirements, but they provide only the energy of both fragments. The fragments' mass is obtained afterwards during data analysis with the help of momentum conservation and the knowledge of the prompt neutron emission. Also, the time resolution of ionization chamber is in the range of 1 ns, limiting the use of this signal for time-of-flight measurements. Therefore, a detector material is needed that can be used as ultra-fast fission trigger with a timing resolution equal to or less than 100 ps, mounted very close to the target and providing sufficiently good energy resolution. One candidate is diamond material, which is presently available made by chemical vapor deposition (CVD). This material in its poly-crystalline form has already proven to be radiation resistant to relativistic heavy ions and to posses excellent timing properties [8,9].

The present work focusses on the possibility of measuring fission-fragment masses with high resolution, including measurements of prompt fission γ-rays and neutrons with high efficiency using single-crystal chemical vapor deposited (sCVD) diamonds.

2. Artificial diamonds for fission-fragment detection

Diamond is an insulator with one of the highest electron–hole drift velocity. Electron–hole pairs created by interaction with charge particles can be collected rapidly and, with appropriate pre-amplifiers, the formed pulse has a rise time faster than 1 ns and a width well below 5 ns, depending on the diamond thickness and the strength of the electric field. These features allow diamond to achieve a timing resolution as good as 29 ps (1σ) in experiments involving heavy ions of very high energy [8]. In the late 1990 and early 2000 years, artificial poly-crystalline diamonds produced by means of chemical vapor deposition became available at sufficiently large size. Combined with diamond's very high resistance to light-charged particles, poly-crystalline chemical vapor deposited diamond (pCVDD) has shown to serve as very efficient detector in beam tracking applications. Since a typical fission experiment takes place in a strong (fast) neutron environment, sometimes in combination with a high α-activity of the target material under investigation, a radiation resistant fission detector is needed. The observed features of diamond makes it a very good candidate for detection of fission fragments.

Relatively affordable and available in large dimensions, pCVDD was already used for the detection of fission fragments [9] and an excellent intrinsic time resolution of 106 ± 21 ps was observed. Unfortunately, due to the poor energy response of pCVDD, especially for the heavier low-energy fragments, a part of the fission fragments was not detected, leading to a significant distortion of the TOF spectrum. The poor energy resolution of pCVDD is caused by the recombination of electrons and holes in the vicinity of defects near grain boundaries. It may be expected that the use of single-crystal chemical vapor deposited diamond (sCVDD) will significantly improve the energy resolution and improve the detection efficiency at low particle energies.

3. Single-crystal diamond

Single-crystal diamonds have a defect density that is significantly reduced in comparison to poly-crystalline diamonds. As a consequence, the charge collection efficiency may reach almost 100%, and an energy resolution as good as 0.4% was measured for α-particles in a selected diamond detector [10]. A detector made from sCVD diamond was also used in a TOF setup and a timing resolution as good as 35 ps was measured for 6 MeV protons and 28 ps for 2 AGeV ^{27}Al [11]. If both timing and energy resolution were that good for the detection of fission fragments, diamond detectors would allow unprecedented mass-resolution in TOF measurements. However, the results previously presented were obtained for very light ions such as protons, α-particles or ions of very high energy.

In contrast, fission-fragment mass distributions range roughly from $A=70$ to $A=170$ with kinetic energies between 2 AMeV and 0.5 AMeV. For fission fragments the energy deposition is occurring in the first few micrometers, which leads to a much higher plasma density than in the event of an impinging light ion at ultra-high kinetic energy. Evidently, the above mentioned properties of sCVD diamond detectors may not be the same. In the following we describe the characterization of sCVD diamond detectors with fission fragments.

We characterized two types of diamond detectors with different specifications and designs. The first three diamond detectors, hereafter called detector A(1) to A(3), consist of 4 pixels of 4.6 × 4.6 × 0.3 mm diamonds each, glued in square configuration on a ceramic PCB board [12]. Faces of the detectors were metallized with a 100 nm thick aluminum coating. The rear side is kept at ground potential and each front side pad was connected through a wire bond to a bias voltage $U=+300$ V. The second detector type (B) has one diamond of similar dimensions as above, but a thickness of 0.15 mm [13]. The recommended bias voltage of $U=+100$ V was applied. The diamond is encapsulated in a RF-shield with an opening of 3 mm diameter. Of two such detectors, one has a 200 nm thick Al (B-Al) and the other a Ti–Pt–Au (B-Au) composite metallization, 470 nm thick. The energy resolution of the sCVD diamond detectors was determined with a ^{241}Am source, emitting α particles with an energy $E_\alpha = 5.49$ MeV, and a ^{252}Cf source emitting both α particles and fission fragments.