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Image: Ornamental multiplication of space-time figures of temperature transformation rules (adapted from T. S. Biró and P. Ván 2010 EPL 89 30001; artistic impression by Frédérique Swist).
Fission diamond detectors for fast-neutron ToF spectroscopy

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Abstract – A novel type of fast-neutron (energy $E_n >1 \text{ MeV}$) counter is presented. It is made of a fissionable natural-uranium foil faced to an intrinsic single-crystal diamond that detects the neutron-induced fission fragments escaping the uranium sheet. The fast response of the diamond is a key feature for its use at pulsed spallation neutron sources for applications in beam monitoring and spectrum measurements with mm spatial resolution. This is an important issue to be addressed in the development of beam lines dedicated to the investigation of the so-called single-event effects in electronics, such as the ChipIr instrument designed for the ISIS spallation source in the UK. Tests of the device at the ROTAX beam line at ISIS have shown its potentiality for the proposed application.

Localized neutron flux monitoring is an important issue to be addressed in high-energy neutron irradiation of microelectronics [1–3]. The determination of the cross-section of the so-called single-event effect (SEE) is related to the number of errors of a given type, registered during the irradiation run, and to the neutron fluence onto the chip [4,5]. Neutron detectors featuring mm\textsuperscript{2} sensible surface are desirable for this kind of applications especially at spallation neutron sources, where the intensity of fast neutrons (energy, $E_n >1 \text{ MeV}$) is appreciable. For example, the results obtained at the ISIS spallation source on chip irradiation [2,3] helped the design and future realization of the ChipIr beam line which is dedicated to this kind of applications [6]. An important requirement for this experimental irradiation station is the development of neutron counters capable of measuring the neutron fluence onto the chip, and more desirably the local spectral intensity of the neutron beam. The device should be able to exploit the pulsed nature of the spallation neutron source, allowing the use of the time-of-flight (ToF) technique with fast neutrons over typical distances of 10–15 m, available for ISIS beam lines at Target Station 2 (TS2). To this aim a device was developed that exploits the neutron-induced fission reactions in natural uranium (natU), in turn to detect the fission fragments with a single-crystal diamond detector (SDD) [7–10].

Figure 1 shows the schematic of the device, henceforth named fission diamond detector (FDD). The thickness of the fissionable natU target is 20 µm, while that of the SDD is 25 µm. The natU foil thickness allows only the fission fragments originating on the surface to escape and reach the diamond crystal where these are completely stopped. This is clear from table 1 where the ranges in uranium and diamond are listed for a typical fission fragment (rhodium) produced in natU(n,f) reactions.

The diamond used for the measurements presented here was grown as a small single crystal (few mm\textsuperscript{2} effective area and sub-mm thickness) by microwave chemical vapour deposition [7]. A highly conductive boron-doped diamond film of 20 µm thickness is first grown epitaxially on...
at three different energies in nat the intrinsic layer. After the growth, the intrinsic diamond clean reactor, in order to avoid boron contamination of layer is then grown on the doped surface in a separate Table 1: Nuclear ranges of rhodium nuclide (A = 102, Z = 45) at three different energies in 235U and diamond.

<table>
<thead>
<tr>
<th></th>
<th>Range (µm) at 60 MeV</th>
<th>Range (µm) at 90 MeV</th>
<th>Range (µm) at 300 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>4.0</td>
<td>5.2</td>
<td>10.7</td>
</tr>
<tr>
<td>Diamond</td>
<td>5.8</td>
<td>7.4</td>
<td>16.7</td>
</tr>
</tbody>
</table>

a 4 × 4 mm² commercial high-pressure high-temperature (HPHT) diamond substrate. This B-doped layer is used as a back contact. A thick (e.g., 25 µm) intrinsic diamond layer is then grown on the doped surface in a separate clean reactor, in order to avoid boron contamination of the intrinsic layer. After the growth, the intrinsic diamond layer is oxidized by isothermal annealing at 500 °C for 1 h in air, in order to remove the H₂ surface conductive layer. On top of the layered diamond structure, a 2.5 mm diameter, 100 nm thick aluminium layer is thermally deposited, which is used as top contact. Because of its layered geometry the SDD is only sensitive to interactions occurring within the intrinsic layer (which defines the SDD thickness). Interactions taking place in the much thicker HPHT substrate do not produce charge collected at the electrical contacts. Thus there is no need to remove the substrate, which makes the detector more robust from a mechanical point of view.

The SDD structure acts as a p-type/intrinsic/metal Schottky Barrier Diode. The aluminium contact creates a Schottky junction with the intrinsic diamond that, in turn, is the detector layer sensitive to the ionizing radiation (drift-layer). The SDD’s B-doped layer acts as a p-type layer (hole injector) and determines the unipolarity of the device under direct polarization. The device operates in reverse biased mode, with a positive voltage applied to the top metal contact and a grounded B-doped contact. The electric field in the drift layer is \( E = 2 \cdot 10^4 \text{V cm}^{-1} \). Electron-hole pairs generated in the diamond will be collected at the contacts, provided that the diamond quality is such that recombination and capture (trapping) are negligible. In that case the effective charge carrier lifetime, \( \tau \), is longer than the charge drift time, \( \tau_{\text{drift}} \), across the detector given by

\[
\tau_{\text{drift}} = \frac{D}{\mu_0 E} \left( 1 + \frac{\mu_0 E}{v_{\text{sat}}} \right),
\]

where \( D \) is the depletion layer depth, \( \mu_0 \) is the electron mobility and \( v_{\text{sat}} \) is the saturation velocity. Taking into account that the detector operates in full depletion mode when the applied voltage is high (\( V_0 \sim 1 \text{ V µm}^{-1} \)) and using the values \( D = 25 \mu\text{m}, \mu_0 = 2200 \text{cm}^2\text{V}^{-1}\text{s}^{-1} \) and \( v_{\text{sat}} = 1.35 \cdot 10^7 \text{cm} \text{s}^{-1} \) [11], eq. (1) gives \( \tau_{\text{drift}} \approx 0.24 \text{ns} \).

The FDD was irradiated on the ROTAX test beam line at the ISIS spallation source. Neutrons are produced by a double bunch-structured proton beam, the bunches being about 70 ns wide and separated by about 300 ns. The FDD was placed in the direct beam, exiting a methane moderator at 95 K, at a distance of 15.5 m from the moderator. The neutron energy (\( E_n \)) spectrum is known to feature a peak at about 10 meV and a 1/\( E_n^2 \) tail in the epithermal/fast-neutron region. For each event, the electronics, setup in a biparametric configuration [12,13], allowed to record the time and pulse height (i.e., the energy deposited in the diamond). It has to be stressed that the neutron-induced signals are due to fission fragments generated in the 235U sheet as well as to neutron inelastic reactions [10,14] and neutron capture reactions, such as 12C(n,α)³Be and 12C(n,n)³He, in 12C of SDD. The electronic chain for the data recording consisted of a custom built charge preamplifier (integration time of 500 ns that well matches the sampling time of the digitizer described below and gain of about 0.3 mV IC⁻¹), a timing filter amplifier for additional shaping and gain adjustment and a N1728 CAEN Waveform Digitizer [15]. The timing filter amplifier was used to amplify the signal by a factor of about 4, with an integration constant of 20 ns, in order to reduce the noise and to match the 10 ns sampling rate of the digitizer. The electronic threshold for the acquisition of the events in the detector was set to a value corresponding to \( E_0 \approx 2.5 \text{ MeV} \). The CAEN digitizer implements a trapezoidal filter to evaluate the pulse height from a single waveform [15,16]. The digitizer stores both the waveforms and biparametric data, i.e., the time of arrival of the neutron on the detector and the pulse height, in the memory of the PC. The time of flight (\( t_{\text{ToF}} \)) is then calculated offline as the difference between the time of interaction of the
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Fig. 2: Scatter plot showing the results of biparametric measurements performed with a single-crystal diamond detector without (a) and with (b) nat U fissile target foil.

The uranium fissile target allows for an intrinsic energy calibration of the detector, using the 4.2 MeV energy $\alpha$-particles produced by natural radioactivity that reach the SDD with energy $E_\alpha \leq E_{\alpha,\text{MAX}}$ of about 4 MeV due to the 2 mm air gap between the diamond and the uranium foil.

Figure 2 shows a typical biparametric spectrum of the data recorded without (a) and with (b) the fissionable nat U target foil in front of the 25 $\mu m$ thick SDD. On the vertical axis the pulse height is shown in terms of the energy deposited in the detector, $E_d$; on the horizontal axis the time of flight $t_{\text{ToF}}$ (time of arrival of the pulse). Based on the events measured by the detector, and shown in fig. 2(a), we note that events due to the n-$^{12}\text{C}$ reactions occur before 800 ns and deposit less energy than approximately 20 MeV in the detector. Indeed, the energy threshold of the n-$^{12}\text{C}$ reaction is $E_{\text{TH}} \approx 6$ MeV, that corresponds to a $t_{\text{ToF}}$ of about 460 ns for a neutron produced at $t = 0$ (i.e., the beginning of the first proton bunch) travelling over a flight path of 15.5 m. Considering the proton pulse width and the time separation mentioned before, one expects that the spectrum should extend up to about 830 ns, as can be seen in fig. 2(a). From the comparison of the two plots in fig. 2, three regions are distinguished:

1) The region with a $t_{\text{ToF}} < 800$ ns and $E_d < 20$ MeV, containing events due to both the n-$^{12}\text{C}$ reactions, as in fig. 2(a), and the fission fragments originating from the interaction of fast neutrons with nat U. The fission fragments are mostly originated from $^{238}\text{U}$; the contribution of $^{235}\text{U}$ is comparatively low and can be estimated from the natural abundance of uranium (99.3% of $^{238}\text{U}$ and 0.7% of $^{235}\text{U}$).

2) Events with a $t_{\text{ToF}}$ between 800 ns and 1500 ns are due to fission fragments since n-$^{12}\text{C}$ reactions are below threshold ($E_{\text{TH}} \approx 6$ MeV). Comparing the energy deposited in the diamond in fig. 2(a) and in fig. 2(b) we can infer that all the events with $E_d > 20$ MeV and $t_{\text{ToF}} < 1500$ ns are due to fission events mostly on $^{238}\text{U}$.

3) Events with a $t_{\text{ToF}} > 1500$ ns are attributed to $^{235}\text{U}$ fission events, since the correspondent neutron energies (800 keV) are below the energy threshold for neutron-induced fission on $^{238}\text{U}$.

Figure 3 (dots with statistical error bars) shows the ToF spectrum obtained by projecting the events in the biparametric spectrum of fig. 2(b) onto the $t_{\text{ToF}}$ axis and adopting an offline pulse height cut for events with $E_d \leq 20$ MeV. As discussed before, this cut allows selecting only the events induced by the fission fragments escaping from the nat U target (see fig. 2), thus excluding those due to the direct interaction of neutrons with $^{12}\text{C}$ in the SDD [14]. Indeed, the biparametric data acquisition is very useful to discriminate these two contributions.

The expected ToF spectrum can be calculated by using the nat U fission cross-section, the Energy-ToF Jacobian...
function and the flux at the irradiation position. The latter was determined using the MCNPX Monte Carlo code [14,17]. The calculated ToF spectrum was then broadened by a convolution with the time structure of the ISIS proton beam (two bunches of about 70 ns width and separated by about 300 ns) [14,18].

The solid line in fig. 3 (referred to the right axis scale) represents this calculation, that shows the satisfactory agreement with data (referred to the left-axis scale). This confirms the correct interpretation of the measured spectrum as due to 238U fission events with little or no contribution from other processes or other types of radiation (e.g., γ-rays).

In summary, a fast-neutron counter was designed and developed using a 238U foil as fissile target placed in front of a single-crystal diamond detector. The complete device, named Fission Diamond Detector, can reveal the fission of a single-crystal diamond detector. The complete device, developed using a natural uranium spectrum as due to agreement with data (referred to the left-axis scale). The solid line in fig. 3 (referred to the right axis scale) represents this calculation, that shows the satisfactory separation by about 300 ns) [14,18].

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