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Supporting information for article:

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Supplemental Material

Neutron Scattering “Halo” Observed in Highly Oriented Pyrolytic Graphite

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Materials

Three HOPG samples with different grades were purchased from Union Carbide Corp. (TX, USA) and they arrive in strip shape. The ZYA HOPG (Mosaic $\sim 0.4^\circ$ FWHM) has dimension of $10\text{mm} \times 70\text{mm} \times 2\text{mm}$, the ZYB HOPG (Mosaic $\sim 0.8^\circ$ FWHM) has dimension of $20\text{mm} \times 100\text{mm} \times 2\text{mm}$ and the ZYH HOPG (Mosaic $\sim 1\text{-}2^\circ$ FWHM) has dimension of $50\text{mm} \times 40\text{mm} \times 5\text{mm}$. Five additional HOPG samples were supplied by Panasonic Corp. (Osaka, Japan), labeled as PGC $\times 04$, PGC $\times 05$, PGC $\times 07$, PGC $\times 10$ and PGC $\times 20$ with mosaic spread of $0.4^\circ \leq \theta < 0.5^\circ$, $0.5^\circ \leq \theta < 0.6^\circ$, $0.6^\circ \leq \theta < 1.0^\circ$, $1.0^\circ \leq \theta < 2.0^\circ$ and $2.0^\circ \leq \theta < 3.0^\circ$, respectively. These square samples have the same dimensions of $20\text{mm} \times 20\text{mm} \times 2\text{mm}$. All samples were examined as received.

Elastic and Inelastic Neutron Scattering Measurements

All measurements were carried out at ambient conditions. SANS measurements were performed at all three SANS instruments at ORNL, including EQ-SANS, Bio-SANS and GP-SANS. The sample-to-detector distance is about 2m and a diameter of 10mm pinhole sample aperture was used. Inelastic neutron scattering measurements were carried out with the cold neutron triple-axis spectrometer (CTAX) at the high flux isotope reactor (HFIR), ORNL. The incident and final neutron wavelengths were selected by a PG (002) monochromator and analyzer, respectively. Contamination from higher-order reflections was removed by a cooled Be filter placed between the sample and the monochromator. Like the geometry in elastic scattering measurements, the sample was oriented in a way so that the crystallographic c axis was parallel to the incident neutron beam. Inelastic neutron data were collected using the 3-axis mode with the incident neutron wavelength fixed at 4.0 Å, 4.5 Å and 5 Å, respectively. Elastic scattering data were collected without the use of the analyzer, with incident neutron wavelength fixed at 4.0 Å and 4.5 Å, respectively. Data was collected as a function of sample tilting angle and temperature.

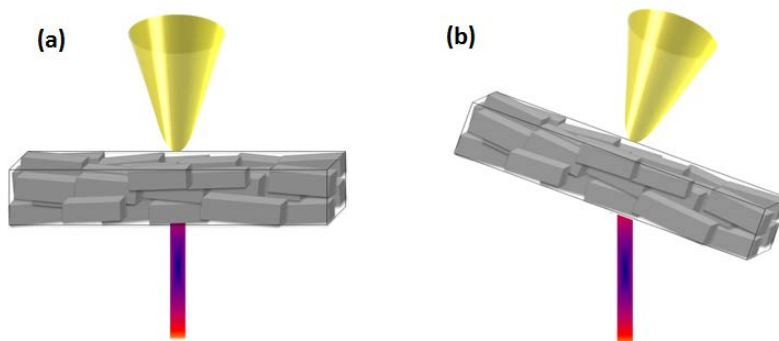


Figure S1 Schematic view of the experimental geometry for both SANS and cold-neutron triple-axis spectrometer measurements, the cone indicates neutron trajectory of the ring feature. (a) with no sample tilting, the centers of the incident beam and the ring overlap; (b) After tilting the sample by 30°, the ring shifts towards the same direction as the sample tilts.

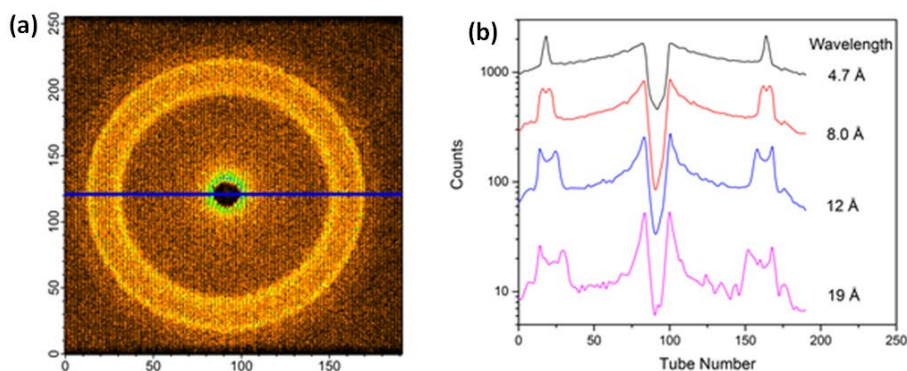


Figure S2 Peak analysis of the ring at different wavelengths. (a) the location of crossing lines in the analysis. (b) the peak shape against wavelength.

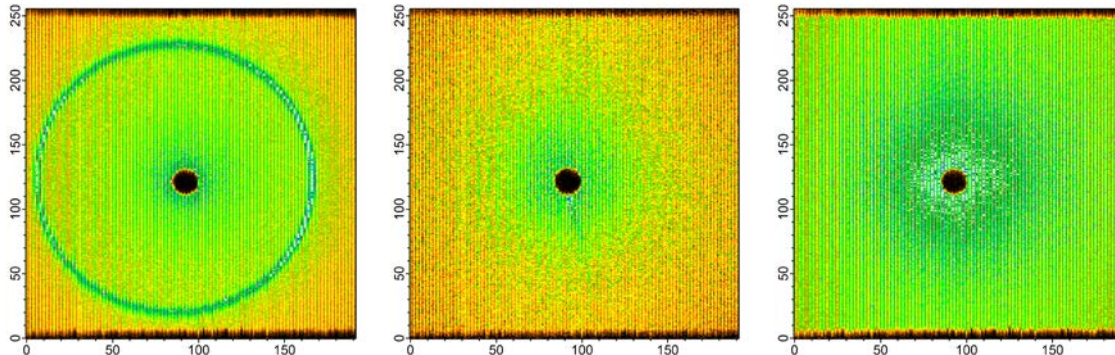


Figure S3 2D images of different materials: ZYA grade HOPG (left), Mica (middle) and single crystal Germanium (right).

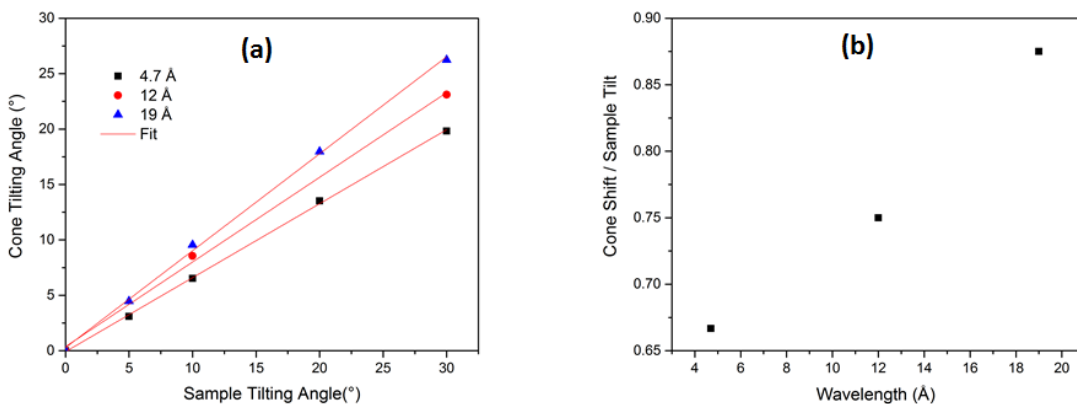


Figure S4 We analyzed the shift of the cone as we tilted the sample and changed the wavelength (a) Cone tilting angle vs sample tilting angle at different incident wavelengths; (b) The ratio of cone shift and sample tilting angle as a function of wavelength

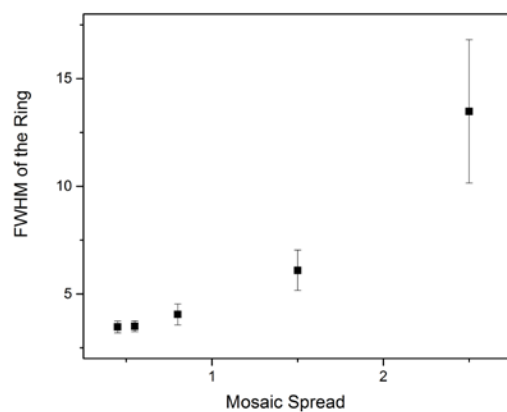


Figure S5 The ring peak as a function of mosaic spread. The FWHM was obtained by fitting the peak using a Gaussian function.

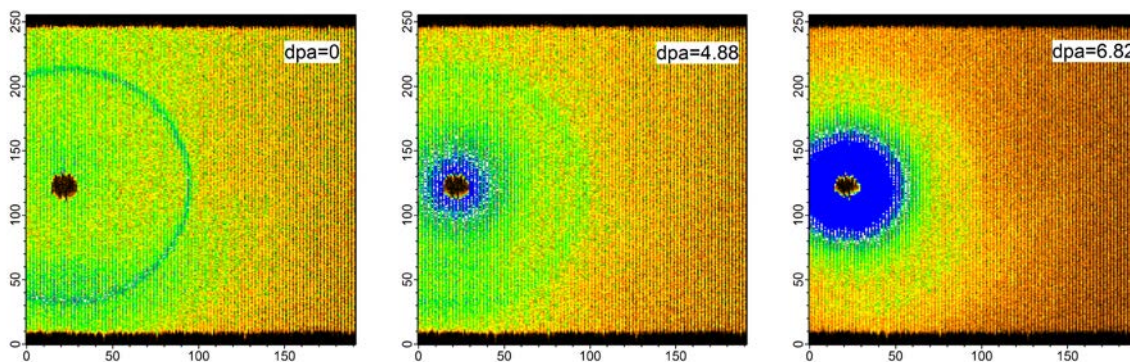


Figure S6 2D images of HOPG samples with different neutron irradiation doses. The ring fades as the quality of the sample decreases.