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Isotopic Composition and Origin of Meteoritic Nanodiamonds studied by Atom-Probe Tomography and Complementary Techniques

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Presolar grains, which formed in the outflows of distant stars, are the only source of information from other star systems that is accessible to direct laboratory microanalysis [1]. Meteoritic nanodiamonds (NDs), separated from carbonaceous chondrite meteorites by acid dissolution [2], are the smallest suspected type of presolar grain, 3 nm in median diameter [3]. Bulk analyses of billions of NDs in aggregate, accompanied by disordered C and other acid residue, have been conducted by stepped heating. They reveal trace amounts of isotopically anomalous Xe-HL, which originates from core-collapse supernovae [4,5], but bulk measurements of the C isotopic ratios are within the limits of variation in our solar system [6, and references therein]. Isotopic analysis of individual NDs is necessary to solve the mystery of their origins and to connect our knowledge of NDs to our knowledge of the stars.

Atom-probe tomography (APT) has uniquely allowed us to probe the $^{12}C/^{13}C$ isotopic ratio of individual and small groups of meteoritic NDs and disordered carbon [7–9]. Pt-ND-Pt layer deposition and FIB liftout techniques were developed for targeting isolated NDs by APT (Fig. 1 and 2) [8], assisted by correlated and complementary transmission electron microscopy (Fig. 2) [10]. Laboratory detonation synthesized NDs were used as a standard to detect and normalize for artifacts in isotopic measurements, and statistical techniques were employed to correct for multihit signal loss [9,11]. Using density maps and trace element analysis we assessed the possible loss of NDs during field-evaporation from the surfaces of nanotips [12].

We find no extreme carbon isotopic anomalies in roughly 100 NDs analyzed using a LEAP 4000X Si. Taken together with statistical analysis of many clusters of thousands of NDs collected using nanoscale secondary ion mass spectrometry [13], we find this indicative that the fraction of NDs with a supernova origin has an upper bound of 1 in 10^3 NDs. This would require that the supernova fraction of the NDs carry a fairly high concentration of supernova Xe-HL, 1 Xe-HL atom per 10^3 supernova NDs. Presolar graphite and silicon carbide grains from supernovae, though much larger than NDs, carry no Xe-HL, so our results suggest that supernova NDs condensed with Xe-HL in different supernovae or different regions within expanding supernovae remnants, compared to other supernovae grains. As for the remaining majority of NDs, they may have formed in the solar system or around asymptotic giant branch stars.

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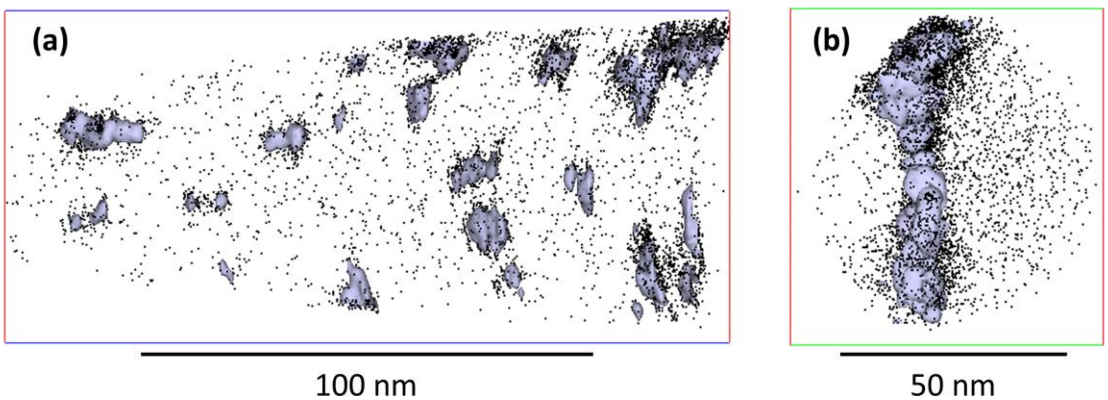

Figure 1: 2D projections of a 3D atom-probe reconstruction of a nanotip containing several nanodiamonds. Black dots are C atoms, gray contours are 2% C concentration isosurfaces, which highlight (a) probable nanodiamonds in the perspective transverse to the nanotip axis, and (b) the carbon deposition layer in the axial perspective. Not shown are Pt from the sandwich layers and trace elements.

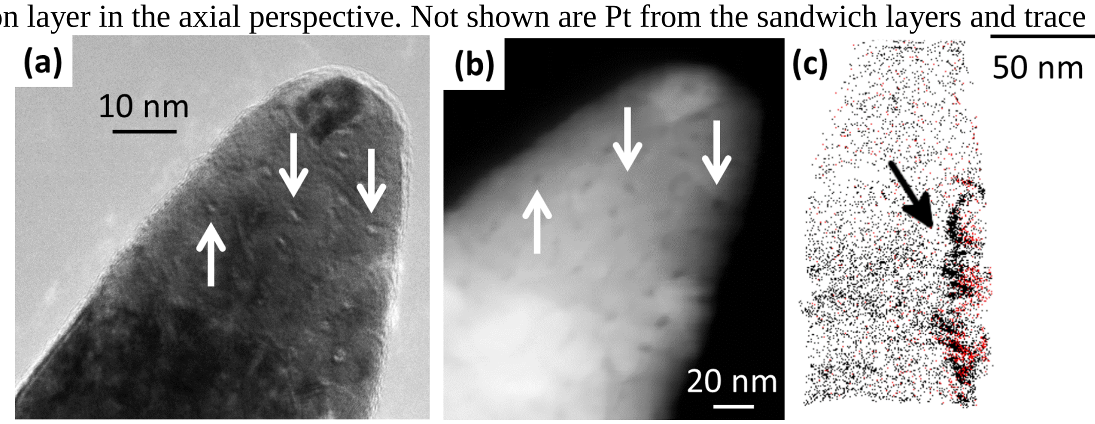

Figure 2: Correlated APT-transmission electron microscopy of a nanotip. (a) Bright-field and (b) dark-field transmission electron images with low-Z features consistent with ~3 nm-nanodiamonds. (c) Atom-probe reconstruction, showing carbon atoms (black) surrounding low-density features filled with NaO (red) from the acid dissolution process, suggesting loss of nanodiamonds during APT of this nanotip.

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