Laboratory evidence for co-condensed O- and C-rich meteoritic stardust from nova outbursts

Pierre Haenecour[1*], Jane Y. Howe[1,2], Thomas J. Zega[1,3], Sachiko Amari[4,5], Katharina Lodders[5,6], Jordi José[7], Kazutoshi Kaji[8], Takeshi Sunaoshi[8], and Atsushi Muto[2].

[1.] Lunar and Planetary Laboratory, The University of Arizona, 1629 E. University Blvd, Tucson, AZ 85721-0092, USA.

[2.] Hitachi High-Technologies America Inc., Clarksburg, MD 20871, USA.

[3.] Department of Materials Science and Engineering, The University of Arizona, Tucson, AZ, USA.

[4.] Laboratory for Space Sciences and Physics Department, Washington University in St. Louis, St. Louis, MO, USA.

[5.] McDonnell Center for Space Sciences, Washington University in St. Louis, St. Louis, MO, USA.

[6.] Department of Earth and Planetary Sciences, Washington University in St. Louis, St. Louis, MO, USA.

[7.] Departament de Física, EEBE, Universitat Politècnica de Catalunya, and Institut d'Estudis Espacials de Catalunya, Barcelona, Spain.

[8.] Hitachi High-Technologies Corp., Application Development Department, Ibaraki, Japan.

Revised Manuscript for publication as ***a Letter to Nature Astronomy***

Last edited: June 5, 2019

Although their parent stars no longer exist, the isotopic and chemical compositions and microstructure of individual stardust grains identified in meteorites provide unique constraints on dust formation and thermodynamical conditions in stellar outflows[1–4]. Novae are stellar explosions that take place in the hydrogen-rich envelope accreted onto the surface of a white dwarf in a close binary system[5]. The energy released by a suite of nuclear processes operating in the envelope

powers a thermonuclear runaway, resulting in the ejection of processed material into the interstellar medium. Spectral fitting of features observed in the infrared spectra of dust-forming novae provided the first evidence of the co-condensation of both carbonaceous and silicate dust in stellar outflows within the 50 to 100 days after explosion[6–11]. Here we report the identification of an O-rich inclusion, composed of both silicate and oxide nanoparticles, inside a graphite spherule that originated in the ejecta of a low-mass CO nova[12]. This observation provides laboratory evidence of the co-condensation of O- and C-rich dust in nova outbursts, and is consistent with the transport and mixing of materials between chemically distinct clumps within the nova ejecta.

Dust in stellar and interstellar environments is traditionally studied using space or ground-based telescopes but the laboratory study of circumstellar (presolar) grains identified in extraterrestrial materials (*e.g.,* meteorites) has opened up a new field in astronomy and astrophysics, providing direct ground-truth information on individual stars. Such presolar grains are nanometer- to micrometer-sized minerals, amorphous grains, or aggregates of them, and can include materials such as nanodiamond, SiC, graphites, oxides, and silicates[1,2].

Over the last two decades, about 2000 presolar graphite spherules have been identified and studied exclusively from the acid-resistant residues of the three carbonaceous chondrites: Murchison (CM2), Orgueil (CI1), and Qingzhen (EH3)[13–15]. While most presolar graphite spherules originated from low-metallicity asymptotic-giant-branch stars (~50%) and core-collapse supernovae (~25%), the remaining grains are hypothesized to have also originated from other types of stars, e.g., born-again AGB and J-type stars[14,15]. A few presolar graphite grains also have isotopic compositions suggesting possible origins in the ejecta of novae[16]. Novae are traditionally classified as neon and non-neon, which reflects the composition of the white dwarf that hosts the explosion, i.e., ONe (O- and Ne-rich) and CO (C- and O-rich), respectively[5]. Similarly to other SiC, silicates, and oxides with putative nova origins, these

graphite grains have isotopic compositions that are only qualitatively consistent with direct nova model predictions[17–23].

Recently, a uniquely ^{13}C- and ^{14}N-rich presolar graphite grain (LAP-149) was identified *in-situ* in the primitive CO3.0 carbonaceous chondrite, LaPaz Icefield (LAP) 031117. LAP-149 is about 1µm wide (Fig. 1) with a croissant-like shape (Supplementary Figure 2a) and is characterized by one of the largest ^{13}C enrichments ($^{12}C/^{13}C = 1.41 \pm 0.01$) ever measured in any circumstellar graphite grain[12]. Comparison of its C, N, Si, and S isotopic compositions to stellar nucleosynthesis model calculations and equilibrium dust-condensation models suggest an origin in the ejecta of a low-mass (0.6 solar mass) CO nova[12]. Unlike all other putative nova grains, the isotopic compositions of LAP-149 match both quantitatively and qualitatively CO nova model predictions, providing strong evidence for graphite condensation in nova ejecta[12]. However, the O isotopic composition of LAP-149 ($^{16}O/^{17}O$ and $^{16}O/^{18}O$ are both within solar values) are inconsistent with CO nova models, which predict both large enrichments in ^{17}O and depletions in ^{18}O ($^{16}O/^{17}O$ at least 7.5 times lower than solar and $^{16}O/^{18}O$ at least 60 times higher than solar)[12]. This inconsistency is a recurring and common problem observed for all known putative nova grains, including O-rich grains (silicate and oxide).

A recent study re-examined the isotopic ratios produced by thermonuclear runaways on CO white dwarfs using a Monte Carlo technique that involves a random sampling over the most relevant nova model parameters[24]. It identified, among all the previously reported nova grain candidates, including grain LAP-149 that we report on here, those that are most likely to have formed in CO novae[24]. This study considered a grain to have a CO nova origin if all of its measured isotopic ratios are in quantitative agreement with the model predictions (without assuming any dilution of the ejecta). The new calculations identified six grains deemed most likely to have originated in CO novae. We note that they are all SiC, for which the O isotopic compositions were not measured. In comparison, all the grains with the lowest likelihood of a CO nova origin are those for which O isotopic ratios were measured[24]. In other

words, there is a systematic discrepancy on the O isotopic compositions between all putative nova grains and stellar nucleosynthetic models[12,24]. It was suggested that this discrepancy might reflect equilibration of O isotopic compositions by parent-body alteration or laboratory processing (e.g., acid dissolution), or mixing of the nova ejecta with solar composition material[17,19,20]. However, in the case of grain LAP-149, the NanoSIMS and Auger nanoprobe data did not show any sign of O diffusion into the grain and it was identified *in-situ* in a thin-section of a minimally altered carbonaceous ($CO_{3.0}$) chondrite, thus excluding any possible isotopic dilution due to laboratory processing[12]. Furthermore, new chemical maps of an electron-transparent section of grain LAP-149 obtained by collecting energy-dispersive X-ray spectra (EDS) using scanning transmission electron microscopy (STEM) do not show any sign of O diffusion inside the graphite spherule (Fig. 1). The fact that O diffusion from surrounding matrix material and O isotopic equilibration from laboratory treatments and/or alteration processes can be definitely excluded for grain LAP-149 shows that additional computational models and astronomical observation of nova ejecta are required to reconcile laboratory measurements of all putative nova grains with model predictions for O isotopes.

STEM phase-contrast lattice fringe imaging of LAP-149 and a presolar graphite shows that LAP-149 is composed of nanocrystalline graphite with an interplanar distance (d_{002} spacings = 0.34 nm) consistent with graphite (Fig. 2). This observation is also consistent with the comparison of the electron energy-loss spectroscopy (EELS) C K energy-loss near-edge structure of LAP-149 and two other presolar graphite (Fig. 3).

High-resolution secondary electron, bright-field, and annular dark-field imaging show the presence of an inclusion inside of the LAP-149 graphite grain, measuring approximately 170 × 70 nm (Fig. 1). Both the EELS maps and EDS spectrum images show that the inclusion is composed of O-rich material (Figs. 1 & 4). Furthermore, the heterogeneous distribution of Si and O across the inclusion suggests that it is an aggregate composed of silicate and oxide grains. Electron-nanodiffraction patterns of distinct

regions within the inclusion show that the silicate and oxide grains are nanocrystalline aggregates (Fig. 5). The EDS data reveal that the oxides are Fe- and Al-rich, and the silicates are ferromagnesian in composition (Fig. 4).

Based on spatial relationships, this inclusion must have formed before the host graphite, LAP-149, in the same nova ejecta, as it is completely surrounded by the graphite grain (Figs. 1 & 4). In addition, the inclusion could not have filled a crack or cavity in the graphite spherule because it progressively appeared during the focused-ion beam (FIB) thinning process on both sides of the section, confirming that the inclusion was entirely enclosed inside its host graphite. While many different types of refractory inclusions have previously been reported inside of presolar SiC and graphite grains[25], to our knowledge, this is the first identification of presolar silicate and oxide grains inside a carbonaceous grain. This observation is not consistent with traditional equilibrium thermodynamic calculations of dust condensation in circumstellar environments that predict formation of carbonaceous grains in C-rich stellar envelopes with C/O > 1 and silicate and oxide dust in O-rich envelopes with C/O <1. However, our detection of nanocrystalline silicate and oxide grains inside of a graphite spherule is consistent with astronomical observations over periods of several years in CO nova ejecta that report production of both C-rich (*e.g.,* graphite, SiC and amorphous carbon) and O-rich (*e.g.,* silicates) dust[6–10].

The nanocrystalline silicate and oxide inclusions in LAP-149 provide ground-truth laboratory evidence of the co-condensation of O-rich (silicates and oxides) and C-rich (e.g., graphites) grains in CO nova outbursts. These observations also suggest that either the nova ejecta has temporal or spatial heterogeneities with distinct clumps or layers of C- and O-rich material, or the nova outbursts are significantly asymmetric with different ejecta compositions in different directions[6,8]. Such asymmetries have been previously observed. For example, multi-wavelength spectroscopic observations of several novae, such as observations of the GK Persei nova ejecta with the NASA Hubble space telescope, the NASA Chandra X-ray observatory, and the National Science Foundation's Very Large Array (VLA)

telescope provide evidence of the asymmetric structure of the nova ejecta and show that clumps of material are ejected during nova outbursts[26]. The chemistry of nova outbursts depends on characteristics of the binary star system, such as the nature of the white-dwarf core (CO- or ONe-rich) and the composition of the gas mixture between the white-dwarf surface and the accreted material from the envelope of the companion star[10].

If our observation reflects temporal variations in the nova ejecta, LAP-149 and its silicate-oxide inclusion could have formed within the same clump in which the C/O ratio varied with time. The O-rich inclusion formed first when the C/O ratio was below 1, and the surrounding graphite condensed afterwards in a C-rich environment (C/O ratio above unity). However, this hypothesis is not consistent with astronomical observations of dust condensation in nova ejecta that suggested that C- and O-rich dust formed in different part of the nova outburst, and C-rich grains formed first before the condensation of silicates[6–11]. Grain LAP-149 and its inclusion thus likely formed in distinct clumps of the nova ejecta (with O- and C-rich compositions, respectively). In this case, the observation of the silicate-oxide inclusion inside of LAP-149 indicates dust mixing and transportation between distinct clumps within the nova ejecta. Either way, our data suggest a circumstellar environment that defies conventional wisdom, i.e., co-condensation of both C- and O-rich dust and large-scale transport could have occurred and therefore demands a re-examination of the dynamics of these enigmatic stars.

All the current nucleosynthesis model predictions for novae rely on one-dimensional (1D) simulations. Recent preliminary work on three-dimensional (3D) hydrodynamics simulations of the interaction between the nova ejecta, accretion disk, and stellar companion suggested that, within one hour of the nova outburst (before the condensation of dust grains), a fraction of the ejecta might collide with the accretion disk and/or the envelope of the companion star[27]. Other 3D simulations suggest that turbulent convection might be responsible for the observation of highly fragmented and heterogeneous nova ejecta, and could explain transportation of heterogeneous material across the

ejecta[28,29]. Such processes would only affect a small fraction of the ejecta and could potentially provide an explanation for the transport and mixing material between C- and O-rich clumps in the nova ejecta[28,29]. In this context, our discovery of an O-rich inclusion, composed of nanocrystalline silicate and oxide grains, in a presolar graphite spherule further supports inhomogeneities, mixing and transport of dust within the ejecta and is consistent with a formation model where the nanocrystalline Fe- and Al-rich oxides and ferromagnesian silicates condensed first in an O-rich region of the ejecta, and the silicate-oxide inclusion was then transported to a C-rich region of the ejecta where nanocrystalline graphite condensed around it.

Bright-field (BF) and annular dark field (ADF) images also reveal the presence of two different rims on the top and bottom surface of the grain (Fig. 1). While the top layer reflects Cs^+ ion beam damage due to the NanoSIMS measurements, the rim on the bottom surface of the grain, directly in contact with the surrounding matrix material, was not exposed to either the NanoSIMS measurements or any other laboratory processing. EDS mapping shows that this rim is composed of a mixture of carbonaceous material and O-rich material with an elemental composition consistent with a ferromagnesian silicate (Supplementary Figure S1). EELS measurements of the carbonaceous material in the rim indicate that it is mostly composed of aromatic amorphous carbon (Fig. 3). Because LAP-149 was identified in a pristine (unequilibrated) carbonaceous chondrite that experienced only minimal secondary processing, such as thermal metamorphism and aqueous alteration, the rim does not reflect secondary processing on the meteorite parent-body[30]. Instead, the rim likely reflects processes in the nova ejecta or grain surface processing in the interstellar medium (ISM). While irradiation of the graphite spherule in the ISM could potentially explain the formation of a rim composed of amorphous carbon on the surface of the graphite grain, it would not explain the mixture of both amorphous C and ferromagnesian silicate material. Another possibility is that the rim formed during processing in the parent nova environment. In addition to SiC, graphite and silicates, IR spectroscopic observations of

168 nova ejecta have also suggested the presence of amorphous carbon and aromatic organic
169 nanoparticles[6–11]. The surface coating on grain LAP-149 could then reflect the condensation of
170 amorphous carbon and ferromagnesian silicate nanoparticles on the surface of the grain within the
171 nova outburst.
172
173 **Methods.** An electron-transparent cross section of graphite grain LAP-149 was prepared following
174 previously described focused-ion-beam (FIB) techniques[31], using the dual-beam Helios NanoLab 660
175 FIB-SEM at the University of Arizona (Supplementary Figures S2 and S3). In brief, Pt fiducial markers
176 were first deposited on top of the grain using electron-beam deposition. Using ion-beam deposition, a
177 protective C strap (or capping layer) was then deposited on the top surface of the cross-section to
178 mitigate any potential damage (e.g., Ga^+ ion implantation and amorphization) during the sputtering
179 process. The section was then cut and extracted from the meteorite thin-section using the EasyLift©
180 micromanipulator system. Finally, the section was mounted to the TEM grid and progressively thinned
181 to electron-transparency ($\lesssim$100 nm) using a range of accelerating voltages and current. Final ion
182 polishing was performed at 5 keV and 0.68 nA to remove any potential surface damage caused by
183 higher-voltage milling.
184 We then carried out coordinated high-resolution STEM imaging (secondary electron, bright-field
185 and annular dark-field), energy-dispersive X-ray spectroscopy (EDS) and electron energy-loss
186 spectroscopy (EELS) measurements of the grain using the 30kV Hitachi SU9000 SEM/STEM located at
187 the University of Arizona. The SU9000 is equipped with an Oxford Instruments X-Max 100LE EDS
188 detector and Hitachi EELS system. All measurements were carried out with a 30kV accelerating
189 voltage. Hole Count EDS spectra acquired in the vacuum just above the FIB section show Cu and Al
190 system peaks (Supplementary Figure S4), confirming that all other elements identified in the sample
191 are indigenous to the grain. Based on the EELS low-loss spectrum (zero-loss and plasmon peaks) of

LAP-149, we estimated a t/λ ratio of about 0.55 (where t corresponds to the sample thickness and λ is the local inelastic mean free path), corresponding to a thickness of about 35 nm. This estimate was calculated using the Microscopy & Microanalysis Tool Set[32]. We tested the accuracy of this method by measuring the t/λ ratio of microtome sections of cyanoacrylate standard with a known thickness (30 nm).

Using a Gatan Be double-tilt high-resolution holder, we also acquired additional EDS and electron-nanodiffraction patterns of the O-rich inclusion in LAP-149 using the Hitachi HF5000 200keV STEM/TEM at the University of Arizona to obtain further information on its nanoscale microstructure and to confirm the presence of Al in the inclusion (Supplementary Figure S5).

Finally, we carried out additional NanoSIMS ion raster imaging of C and O isotopes (^{12}C, ^{13}C, ^{16}O, ^{17}O and ^{18}O) of the FIB section of grain LAP-149 at Washington University (WUSTL) using a focused Cs^+ primary beam of ~0.3 pA (~50 nm in diameter) that was rastered over the section (covering a surface of 6×6 μm^2 and 256×256 $pixels^2$) with a and a dwell time of 1 milliseconds/pixel. Unfortunately, despite using a low primary ion beam, the FIB section directly started to fold onto itself as soon as it was exposed to the Cs^+ primary ion beam, so we were not able to obtain isotopic measurements of the inclusion. This was likely due to the extremely low thickness (30 nm) of the section.

Author contributions. P.H. prepared the FIB sample cross-section, carried out the TEM and NanoSIMS measurements and wrote the manuscript; J.Y.H., K.K., T.S., A.M. and T.J.Z. helped with the TEM analyses; T.J.Z., S.A., K.L. and J.J. contributed to the data interpretation; All co-authors contributed to the manuscript.

Materials & Correspondence. P. Haenecour. Lunar and Planetary Laboratory, The University of Arizona, 1629 E. University Blvd., Kuiper Space Science Bldg., Tucson, AZ 85721-0092, USA. Email:

pierre@lpl.arizona.edu. Phone: (520)-626-9810. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Acknowledgment. PH dedicates this paper to his PhD advisor and friend, the late professor Christine Floss (C.F.), for her leading work on coordinated in-situ studies of presolar silicate grains and for fruitful discussions about our discovery of a silicate-oxide inclusion inside grain LAP-149. This research was supported by the NASA NExSS "Earths in Other Solar Systems" (EOS) program (NNX15AD94G), NASA grants NNX15ALJ22G (T.J.Z.), NNX16AD31G (S.A.) and NNX14AG25G (C.F.) from the Emerging Worlds Program, NSF grant AST-1517541 (K.L.), the Spanish MINECO grant AYA2017–86274–P, the E.U. FEDER funds, and the AGAUR/Generalitat de Catalunya grant SGR-661/2017 (J.J.). This article also benefited from discussions within the "ChETEC" COST Action (CA16117). TEM and FIB analyses were carried out at the University of Arizona Kuiper Materials Imaging and Characterization Facility (NSF Grant 1531243 and NASA Grants NNX15AJ22G and NNX12AL47G). The NanoSIMS measurements were also supported by the McDonnell for Space Sciences at Washington University in St. Louis. We thank Paul Wallace for his help with the preparation of the electron-transparent section, and Dr. Josiah Lewis and Tim Smolar for their support with NanoSIMS 50 work.

References

1. Zinner, E. Presolar Grains. in *Treatise on Geochemistry* 181–213 (Elsevier, 2014).
2. Floss, C. & Haenecour, P. Presolar silicate grains: Abundances, isotopic and elemental compositions, and the effects of secondary processing. *Geochem. J.* **50,** (2016).

3. Nguyen, A. N., Keller, L. P. & Messenger, S. Mineralogy of presolar silicate and oxide grains of diverse stellar origins. *Astrophys. J.* **818,** 51 (2016).

4. Hoppe, P., Leitner, J. & Kodolányi, J. The stardust abundance in the local interstellar cloud at the birth of the Solar System. *Nat. Astron.* **1,** 617–620 (2017).

5. José, J. *Stellar Explosions: Hydroynamics and Nucleosynthesis*. (CRC/Taylor & Francis, 2016).

6. Gehrz, R. D. *et al.* The peculiar infrared temporal development of Nova Vulpeculae 1987 (QV Vulpeculae). *Astrophys. J.* **400,** 671 (1992).

7. Sakon, I. *et al.* Concurrent Formation of Carbon and Silicate Dust in Nova V1280 SCO. *Astrophys. J.* **817,** 145 (2016).

8. Smith, C. H., Aitken, D. K. & Roche, P. F. The mid-infrared spectral development of Nova Cen 1986. *Mon. Not. R. Astron. Soc.* **267,** 225–230 (1994).

9. Shore, S. N., Starrffield, S., Gonzalez-Riestrat, R., Hauschildt, P. H. & Sonneborn, G. Dust formation in Nova Cassiopeiae 1993 seen by ultraviolet absorption. *Nature* **369,** 539–541 (1994).

10. Kawakita, H., Ootsubo, T., Arai, A., Shinnaka, Y. & Nagashima, M. Mid-infrared Spectroscopic Observations of the Dust-Forming Classical Nova V2676 OPH. *Astron. J.* **153,** 74 (2017).

11. Gehrz, R. D. *et al.* The Temporal Development of Dust Formation and Destruction in Nova Sagittarii 2015#2 (V5668 SGR): A Panchromatic Study. *Astrophys. J.* **858,** 78 (2018).

12. Haenecour, P. *et al.* Coordinated Analysis of Two Graphite Grains from the CO3.0 LAP 031117 Meteorite: First Identification of a CO Nova Graphite and a Presolar Iron Sulfide Subgrain. *Astrophys. J.* **825,** 88 (2016).

13. Amari, S., Anders, E., Virag, A. & Zinner, E. Interstellar graphite in meteorites. *Nature* **345,** 238–240 (1990).

14. Amari, S., Zinner, E. & Gallino, R. Presolar graphite from the Murchison meteorite: An isotopic study. *Geochim. Cosmochim. Acta* **133,** 479–522 (2014).

15. Jadhav, M. *et al.* Multi-element isotopic analyses of presolar graphite grains from Orgueil. *Geochim. Cosmochim. Acta* **113,** 193–224 (2013).

16. Amari, S., Nittler, L. R., Zinner, E., Lodders, K. & Lewis, R. S. Presolar SiC Grains of Type A and B: Their Isotopic Compositions and Stellar Origins. *Astrophys. J.* **559,** 463–483 (2001).

17. Amari, S. *et al.* Presolar Grains from Novae. *Astrophys. J.* **551,** 1065–1072 (2001).

18. Leitner, J., Kodolányi, J., Hoppe, P. & Floss, C. Laboratory analysis of presolar silicate stardust

from a nova. *Astrophys. J.* **754,** L41 (2012).

19. José, J., Hernanz, M., Amari, S., Lodders, K. & Zinner, E. The Imprint of Nova Nucleosynthesis in Presolar Grains. *Astrophys. J.* **612,** 414–428 (2004).

20. José, J. & Hernanz, M. The origin of presolar nova grains. *Meteorit. Planet. Sci.* **42,** 1135–1143 (2007).

21. Gyngard, F. *et al.* Automated NanoSIMS Measurement of Spinel Stardust from the Murray Meteorite. *Astrophys. J.* **717,** 107–120 (2010).

22. Nguyen, A. N. & Messenger, S. Resolving the stellar sources of isotopically rare presolar silicate grains through Mg and Fe isotopes analyses. *Astrophys. J.* **784,** 149 (2014).

23. Liu, N. *et al.* Stellar Origins of Extremely 13C- AND 15N-Enriched Presolar SIC Grains: Novae or Supernovae? *Astrophys. J.* **820,** 140 (2016).

24. Iliadis, C., Downen, L. N., José, J., Nittler, L. R. & Starrfield, S. On Presolar Stardust Grains from CO Classical Novae. *Astrophys. J.* **855,** 76 (2018).

25. Croat, T. K., Bernatowicz, T. J. & Daulton, T. L. Presolar Graphitic Carbon Spherules: Rocks from Stars. *Elements* **10,** 441–446 (2014).

26. Takei, D. *et al.* X-ray Fading and Expansion in the 'Miniature Supernova Remnant' of GK Persei. *Astrophys. J.* **801,** 92 (2015).

27. Figueira, J. *et al.* Three-dimensional simulations of the interaction between the nova ejecta, accretion disk, and companion star. *Astron. Astrophys.* **613,** A8 (2018).

28. Casanova, J., José, J., García-Berro, E., Shore, S. N. & Calder, A. C. Kelvin–Helmholtz instabilities as the source of inhomogeneous mixing in nova explosions. *Nature* **478,** 490–492 (2011).

29. Li, K.-L. *et al.* A nova outburst powered by shocks. *Nat. Astron.* **1,** 697–702 (2017).

30. Haenecour, P. *et al.* Presolar silicates in the matrix and fine-grained rims around chondrules in primitive CO3.0 chondrites: Evidence for pre-accretionary aqueous alteration of the rims in the solar nebula. *Geochim. Cosmochim. Acta* **221,** 379–405 (2018).

Methods References

31. Zega, T. J., Nittler, L. R., Busemann, H., Hoppe, P. & Stroud, R. M. Coordinated isotopic and mineralogic analyses of planetary materials enabled by in situ lift-out with a focused ion beam scanning electron microscope. *Meteorit. Planet. Sci.* **42,** 1373–1386 (2007).

32. Egerton, R. F. & Cheng, S. C. Measurement of local thickness by electron energy-loss spectroscopy. *Ultramicroscopy* **21,** 231–244 (1987).

FIGURES LEGENDS

Figure 1. Scanning transmission electron microscopy data of grain LAP-149. (a) Bright-field (BF) and (b) annular dark field (ADF) images of grain LAP-149. (c) electron energy-loss spectroscopy 3-window C-K edge map of grain LAP-149 showing the carbon distribution, and (d) false-color composite elemental maps (C = red; O = blue).

Figure 2. Scanning transmission electron microscopy phase-contrast images of presolar graphite grain LAP-149 and a presolar graphite (KFC1b2-gr1) from an acid-residue of the Murchison meteorite. Both grains exhibit lattice fringe with an interlayer graphene distance (002 $d_{spacings}$) of 0.34nm.

Figure 3. Energy-loss near-edge structure of the C, K edge of presolar graphite grains. Comparison of the C K edge energy-loss near-edge structure of LAP-149, the carbonaceous material in its bottom rim and two Murchison presolar graphite grains (KFC1b2-gr1 and KFC1b2-303).

Figure 4. Scanning transmission electron microscopy (STEM) imaging and energy-dispersive spectroscopy (EDS) data of the inclusion inside LAP-149. STEM bright-field (BF), annular dark-field

(ADF) images and EDS false-color elemental maps (O = blue, C = red, Si = green, Fe = pink and Mg = yellow) of the inclusion inside of graphite grain LAP-149.

Figure 5. Transmission electron microscopy (TEM) imaging and electron nanodiffraction data of the inclusion inside LAP-149. Nanodiff 1 & 2 show the diffraction patterns of the nanocrystalline oxides and silicates, respectively.