IDENTIFICATION OF CRYSTALLINE MATERIAL IN TWO INTERSTELLAR DUST CANDIDATES FROM THE STARDUST MISSION. Zack Gainsforth, Alexandre Simionovici, Frank E. Brenker, Sylvia Schmitz, Manfred Burghammer, Peter Cloetens, Laurence Lemelle, Juan-Angel Sans Tresseras, Tom Schoonjans, Geert Silversmit, Vicente A. Solé, Bart Vekemans, Laszlo Vincze, Cheri Achilles, Carlton Allen, Asna Ansari, Sasa Bajt, Nabil Bassim, Ron S. Bastien, H. A. Bechtel, Janet Borg, John Bridges, Donald E. Brownlee, Mark Burchell, Anna L. Butterworth, Hitesh Changela, Andrew M. Davis, Christine Floss, George Flynn, Patrick Fougeray, David Frank, Eberhard Grün, Philipp R. Heck, Jon K. Hillier, Peter Hoppe, Bruce Hudson, Gary Huss, Joachim Huth, Brit Hvide, Anton Kearsley, Ashley J. King, Barry Lai, Jan Leitner, Ariel Leonard, Hugues Leroux, Robert Lettieri, William Marchant, Larry R. Nittler, Ryan Ogliore, Frank Postberg, Mark C. Price, S. A. Sandford, Kate Schreiber, Ralf Srama, Thomas Stephan, Veerle Sterken, Julien Stodolna, Rhonda M. Stroud, Steven Sutton, Mario Trieloff, Peter Tsou, Akira Tsuchiyama, Tolek Tyliszczak, Andrew J. Westphal, Naomi Wordsworth, Daniel Zevin, Michael E. Zolensky, >30,000 Stardust@home dusters , See http://www.ssl.berkeley.edu/~westphal/ISPE for affiliations.

Introduction: NASA's interstellar collector from the Stardust mission captured several particles that are now thought to be of interstellar origin. We analyzed two of these via nanodiffraction at the European Synchrotron Radiation Facility (ESRF) and found them to contain crystalline components. The unit cell of the crystalline material is determined from the diffraction patterns and the most likely mineral components are identified as olivine and spinel.

Experimental: Tracks I1047,1,34 (Hylabrook) and I1043,1,30 (Orion/Sirius) were extracted as aerogel keystones [1] and studied using ESRF beamlines ID13 and ID22 using XRF and nanodiffraction. Hylabrook was rotated through 80º in 0.5º increments to produce a pseudo-powder pattern using 13895 eV photons on ID13. A 2D diffraction image was captured at each angle. Orion/Sirius was rastered through a 13895 eV beam on ID13 and a 17000 eV beam on ID22 with beam waists of about 200 nm to produce 2D spatial maps of the diffractive domains. ID22 used a gray beam about 300 eV wide. Exposure from the ID22 beam apparently amorphized a subset of the crystalline phases seen previously in ID13, allowing us to unambiguously assign several peaks to a specific phase.

Hylabrook: d-spacings for the reflections observed in Hylabrook are shown in Table 1. The set of d-spacings observed allows unambiguous identification of the unit cell geometry assuming we select the highest symmetry and lowest cell size possible. We assumed one crystalline phase is responsible for the pattern, and excluded the cubic, hexagonal, trigonal, and tetragonal crystal systems with McMaille [2]. An orthorhombic cell is the next highest symmetry cell and is compatible with the diffraction pattern, which was then indexed on the assumption of orthorhombic symmetry. The unit cell was computed with error bars using a Monte Carlo simulation written in MATLAB, and found to be a=4.85 ± 0.08 Å, b = 10.34 ± 0.16 Å, c = 6.08 ± 0.13 Å, $\alpha = \beta = \gamma = 90º$ where errors are 2σ. The only naturally occurring compatible mineral found in the PDF-4+ mineral database is the olivine family. We place no constraints on the chemical composition. Comparison of the observed peaks seen in Hylabrook's diffraction pattern with a modeled fit for olivine shows that all modeled peaks are present in the measured data except for three very low intensity reflections. All measured peaks are explained by modeled peaks as would be expected for a correct mineral identification. It should be noted that the unit cell dimensions are an experimentally measured quantity independent of the choice of olivine as the most likely crystalline phase.

The diffraction pattern shows that multiple domains are present through polygonalization of the diffraction pattern. Peak broadening is also present and was analyzed using the double-Voigt approach [3]. The diffracting domain sizes are found to be larger than 15 nm (2σ) with an expectation value of several tens of nm. We set an upper limit on the strain fields present at ≤ 0.3% (2σ) with an expectation value of 0.2%. According to Abramson et al. [4], this corresponds to stresses ≤ 2 GPa in forsterite, which is consistent with aerogel capture, but could also be native to the original grain. This pressure regime may be inconsistent with an origin as secondary ejecta from an impact on the spacecraft, but needs further investigation.

Orion/Sirius: The set of d-spacings seen in Orion/Sirius is also consistent with olivine plus at least one other phase. The most likely secondary phase is spinel where four peaks are identified from the scans on ID13 and ID22 and four are expected from the model. The ID22 scan observed primarily the four spinel peaks. This is consistent with the radiation hardness of spinel, so that it was the primary crystalline phase to survive the scan [5]. Two additional peaks exist in the ID13 data which cannot be ascribed to olivine or spinel, yet are also absent from the ID 22 data indicating they belong to a third unidentified phase. A rigorous determination of the unit cells without mineralic assumptions and a size-strain analysis of the phases is in progress.

Orion/Sirius was mapped spatially in 2D by rastering the sample with a 200 nm X-ray beam. By analyzing the presence or absence of specific diffraction reflections in the 2D dataset, we obtain a spatial image of the locations of these crystalline phases. This is called an X-ray topograph, akin to a TEM darkfield image. Figure 1 shows topographs produced from both beamlines alongside a scanning transmission X-ray microscopy (STXM) image. The leftmost topograph is from ID13 and shows a white pixel where any diffracting material exists. This does not contain information about orientation or grain size since it contains a brightness contribution from all reflections measured in the diffraction pattern. Outlined in red is a topograph produced from one specific reflection, which indicates that the entire region is diffracting as a single diffractive domain corresponding to olivine. In this way, we see that the grain was approximately a micron long by a few hundred nm thick. The middle frame shows the sum topograph from IDs22 which is the location of the spinel material. None of these crystallites are larger than a single pixel, and therefore ≤ 200 nm across.

Mass fraction: While we unambiguously show the presence of crystalline material in these particles, we do not place a strong constraint on the fraction of material that could be crystalline. Because the amorphous component of these grains is invisible to diffraction, additional work is in progress to place a useful number on the crystalline fraction by combining our datasets with STXM XANES, and through complete analysis of the topographs.

Implications: Several lines of evidence point independently to an interstellar origin for these particles [6]. The presence of diffracting crystalline material is not inconsistent with astronomical observations [7] indicating a large fraction of amorphous material in the ISM: the fraction of crystalline material in these particles is not yet quantified, and may be in fact be small. Also, these particles, which are >1000 larger in mass than most interstellar dust particles, may not be mineralogically representative of the IS dust. And finally, infrared spectroscopic techniques may not accurately determine crystallinity for nanograins. Nevertheless, the presence of some crystalline material indicates that at least some small fraction of the sample is of circumstellar origin and can survive transport through the ISM or is generated in the ISM through radiation induced crystallization or other mechanisms.

References: [1] Westphal A. J. et al. (2004) MAPS 39, 1375. [2] Le Bail A., (2004) Powder Diffr. 19, 249. [3] Balzar D. and Ledbetter H. (1993) J. Appl. Cryst. 26, 97. [4] Abramson E. et al (1997) J. Geophys. Res. 102, 12253. [5] Hobbs L. et al. (1994) J. Nucl. Mater. 216, 291. [6] Westphal, A. J. et al (2012) 43rd LPSC submitted. [7] Kemper, F. et al. (2004) ApJ, 609, 826.

Hylabrook d-spacings (Å)	(hkl) indexed to olivine
5.18	020
4.39	110
3.94	021
3.80	101
3.55	111 and 120
3.06	121 and 002
2.81	130
2.55	131
2.50	112
2.38	200
2.36	041 and 210
2.30	122 and 140
2.20	211 and 220
2.06	132 and 221

Table 1: Measured d-spacings for Hylabrook alongside the most likely reflections assuming olivine. Three peaks were not observed corresponding to the 022, 040 and 141 reflections.

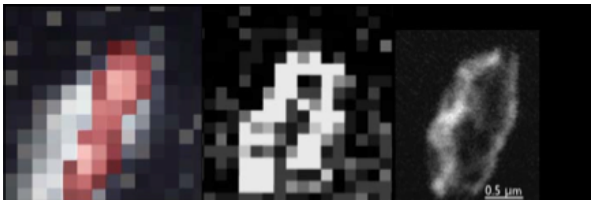

Figure 1: Left image shows the sum of all topographs from ID 13, showing the location of all diffracting material in the particle. Overlay in red is a single topograph from a reflection at 2.28 Å (olivine) showing one grain about 1 µm x 200-600 nm wide. Middle image shows the sum of topographs from ID 22 where most reflections map to spinel. Right image shows a STXM optical density map at 1568 eV for reference.