



HAL
open science

Analogs for Unlayered-Graphene Droplet-Formation in Stellar Atmospheres

P Fraundorf, Melanie Lipp, Taylor Savage

► **To cite this version:**

P Fraundorf, Melanie Lipp, Taylor Savage. Analogs for Unlayered-Graphene Droplet-Formation in Stellar Atmospheres. *Microscopy and Microanalysis (M&M 2016)*, Jul 2016, Columbus, OH, United States. pp.1816-1817. hal-01356394

HAL Id: hal-01356394

<https://hal.science/hal-01356394>

Submitted on 25 Aug 2016

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Analogues for Unlayered-Graphene Droplet-Formation in Stellar Atmospheres.

P. Fraundorf,* Melanie Lipp, and Taylor Savage

Physics & Astronomy/Center for NanoScience, U. Missouri-StL (63121)

(Dated: August 25, 2016)

The unlayered spherical graphene cores, found in a subset of micron-sized presolar graphite onions extracted from primitive meteorites, show graphene but not graphite inter-sheet spacings, sheet “coherence widths” of about 4nm, and (in electron-phase-contrast images) evidence of edge-on obtuse-angle graphene-sheet junctions. The literature on solidification of metallic liquids predicts supercooling by as much as 30% below the melt-temperature, suggesting that at low pressures carbon vapor will predictably condense first as a liquid. Density measurements, possible TEM evidence of faceted-pentagones, and preliminary studies of 5 and 6 atom loop formation in a solidifying melt, all suggest that these unlayered graphene cores (which bear isotopic signatures of formation in asymptotic-giant-branch star atmospheres after third dredge-up) may have resulted from dendritic solidification of “slow-cooled” carbon droplets. Preliminary notes on a strategy for slow-cooling carbon vapor-condensation in the laboratory (with help from an evaporating oven) are also provided.

CONTENTS

I. Introduction	1
II. Open questions	1
III. Density	1
IV. Droplets and solidification	1
V. Synthesis in the laboratory	2
Acknowledgments	2
References	2

I. INTRODUCTION

Micron-sized presolar carbon-spheres condensed before the origin of our solar system, in the atmosphere of asymptotic-giant-branch stars from carbon atoms dredged up following nucleosynthesis in the stellar interior, have been extracted by dissolution from primitive carbonaceous chondrite meteorites, identified by noble-gas and major-element isotopic analysis, and available to electron microscopists on earth for over two decades¹⁻⁴. Some of these spheres appear to consist of graphite condensed on carbide grains, but others instead show graphite-onion rims that surround a spherical-core of unlayered-graphene i.e. carbon that in diffraction shows graphite (hk0) spacings only. The high spatial-frequency tail of these graphene lines⁵ suggests a graphene-sheet coherence-width in the 4-nm size range¹, consistent with electron-phase-contrast images of single (sometimes segmented) edge-on sheets^{2,4}.

II. OPEN QUESTIONS

Three key questions are: What process in a low-pressure atmosphere can form condensed spheres of graphene with densities approaching that of graphite, but with graphitic-layering absent? How might we synthesize such material in the laboratory? What interesting properties might it have as a result?

Our group has been focusing on three aspects of this problem: (i) Measurements of core-material density (relative to the graphite rim) in ultra-microtomed spheres using zero-loss/deflection analysis⁶ via energy-filtered transmission electron microscopy or TEM, (ii) modeling of graphene-sheet nucleation and growth from a carbon melt to get atom-positions for TEM image simulation and estimate the likelihood of pentagonal-loop nucleation as explanation of apparent edge-on facets² using the Tersoff potential⁷, and (iii) the laboratory synthesis of unlayered-graphene via slow-cooling of carbon vapor.

III. DENSITY

Carbon plasmon-peak energies¹ suggest an unlayered graphene-density near the 2.3[g/cc] of graphitic carbon. Zero-loss/deflection analysis [6] shows promise for inelastic mean-free-path mapping in a wide range of disordered materials, but specular reflection by diffraction can complicate life (e.g. via Pendullosung or thickness-fringe contrast) by putting electrons into Bragg storage if the crystals are thick enough for multiple scattering within the same crystal. Although our first analyses suggest a density closer to 1.8[g/cc] for presolar unlayered-graphene, this may not be confirmed without further calibration studies on the presolar specimens themselves.

IV. DROPLETS AND SOLIDIFICATION

Condensation of particles in a low-pressure stellar-atmosphere will require local density increases¹ e.g. asso-

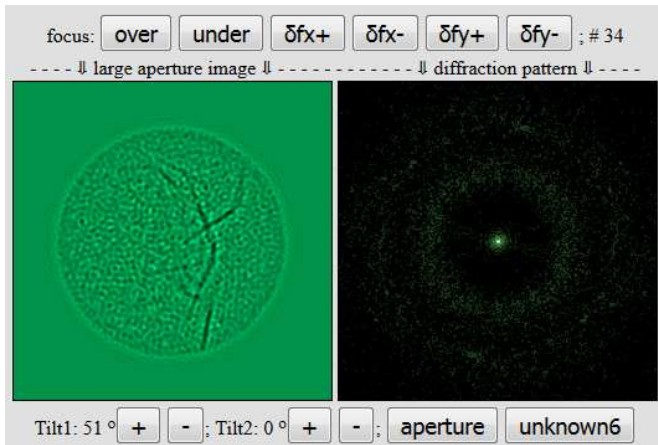


FIG. 1. On-line strong-phase-object simulation of 15 faceted-nanocones in an 8-nm sphere with 26,846 “non-touching” C atoms. The projected-potential power-spectrum at right shows graphene {100} and {110} rings, as well as rel-rods associated with some of the edge-on graphene-sheets.

ciated with jets of material around sunspots, and might take place one C atom at a time, by clustering of polycyclic aromatic hydrocarbons, or by condensation of droplets from a carbon vapor. The latter has the advantage that it will give us the spherical core-shape and density. Although thermodynamics predicts that below pressures of 10.8[MPa], carbon on warming sublimates to a vapor at around 3915K, containerless metals on cooling generally supercool in the liquid state until as much as 30% below the liquid condensation temperature⁸, de-

pending on the amount of rearrangement needed between liquid and solid state configurations.

A liquid condensation temperature for carbon around 4600K, in this context, along with the major rearrangement between 12-neighbor coordination in an fcc liquid and the graphene-sheet structure, thus suggest that supercooling to temperatures as low as 3220K (2950C) might be expected in the absence of condensation seeds (like the carbide grains seen at the center of other particles). This synthesis path might also explain the possible TEM evidence^{2,4} that the graphene sheets are flat, except where they are seen edge-on to connect at sharp bends whose geometry is suggestive of nucleation on pentagonal (rather than hexagonal) carbon-loops. So far, we are assembling graphene-sheet and graphene-pentacene structures for TEM image simulation (Fig. 1), but only beginning to look at simulated annealing models of pentagonal versus hexagonal loop nucleation.

V. SYNTHESIS IN THE LABORATORY

Finally, slow-cooled carbon-vapor synthesis in the inside of a resistively-evaporating carbon-cylinder now looks possible. However we are only beginning to examine carbon spheres created as a result.

ACKNOWLEDGMENTS

Thanks to Roy Lewis at U. Chicago and T. J. Bernatowicz at Washington U. for the presolar specimens used here.

* pfraudorf@umsl.edu; also Physics, Washington University (63110), St. Louis, MO, USA

¹ T. Bernatowicz, R. Cowsik, P. C. Gibbons, K. Lodders, S. Amari, and R. S. Lewis, “Constraints on stellar grain formation from presolar graphite in the murchison meteorite,” *Astrophysical Journal* **472**, 760 (1996).

² P. Fraundorf and Martin Wackenhut, “The core structure of pre-solar graphite onions,” *Astrophysical Journal Letters* **578**, 153–156 (2002), arXiv:astro-ph/0110585.

³ Thomas K. Croat, Frank J. Stadermann, and Thomas J. Bernatowicz, “Presolar graphite from AGB stars: Microstructure and s-process enrichment,” *Astrophysical Journal* **631**, 976–987 (2005).

⁴ Eric Mandell, *Electron beam characterization of carbon nanostructures*, Ph.D. thesis, University of Missouri - St.

Louis and Rolla (2007).

⁵ B. E. Warren, “X-ray diffraction in random layer lattices,” *Phys. Rev.* **59**, 963–698 (1941).

⁶ P. Fraundorf, K. Pisane, E. Mandell, and R. Collins, “Zero-loss/deflection map analysis,” *Microscopy and Microanalysis* **16:S2**, 1534–1535 (2010).

⁷ J. Tersoff, “Empirical interatomic potential for carbon, with applications to amorphous carbon,” *Physical Review Letters* **61**, 2879–2882 (1988).

⁸ K. F. Kelton and A. L. Greer, *Nucleation in condensed matter* (Elsevier, 2010).