Title
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Journal
Proceedings of the National Academy of Sciences of the United States of America, 117(14)

ISSN
0027-8424

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Publication Date
2020-04-07

DOI
10.1073/pnas.1918056117

Peer reviewed

Superconductivity found in meteorites

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Contributed by Mark Thiemens, January 16, 2020 (sent for review October 18, 2019; reviewed by Zachary Fisk, Laura H. Greene, and Munir Humayun)

Meteorites can contain a wide range of material phases due to the extreme environments found in space and are ideal candidates to search for natural superconductivity. However, meteorites are chemically inhomogeneous, and superconducting phases in them could potentially be minute, rendering detection of these phases difficult. To alleviate this difficulty, we have studied meteorite samples with the ultrasensitive magnetic field modulated microwave spectroscopy (MFMMS) technique [J. G. Ramírez, A. C. Basaran, J. de la Venta, J. Pereiro, I. K. Schuller, *Rep. Prog. Phys.* 77, 093902 (2014)]. Here, we report the identification of superconducting phases in two meteorites, Mundrabilla, a group IAB iron meteorite [R. Wilson, A. Cooney, *Nature* 213, 274–275 (1967)] and GRA 95205, a ureilite [J. N. Grossman, *Meteorit. Planet. Sci.* 33, A221–A239 (1998)]. MFMMS measurements detected superconducting transitions in samples from each, above 5 K. By subdividing and remeasuring individual samples, grains containing the largest superconducting fraction were isolated. The superconducting grains were then characterized with a series of complementary techniques, including vibrating-sample magnetometry (VSM), energy-dispersive X-ray spectroscopy (EDX), and numerical methods. These measurements and analysis identified the likely phases as alloys of lead, indium, and tin.

superconductivity | meteorites | extraterrestrial

Meteorites can preserve the oldest phases in the solar system (1), which can form under hundreds of gigapascals of pressure (2), with typical crystallization temperatures of 500 to 700 °C (3). In addition, they can have cooling rates of 100 to 10,000 °C per million years. Because of this, meteorites (particularly those with extreme formation conditions) can contain material phases such as quasicrystals, which are not found in terrestrial environments (4). Past studies of extraterrestrial materials have led to new, previously unpredicted insights (5–7). In this report, we investigated a diverse population of meteorites for superconductivity, including 15 meteorites, spanning the range of meteoritic classes (for more details on methods, including meteorite selection criteria, see *SI Appendix*). Of these, we detected superconductivity in two meteorites: Mundrabilla and GRA 95205. Both Mundrabilla and GRA 95205 are nonchondritic meteorites (they do not possess glassy chondrules). Nonchondritic meteorites have been melted and recrystallized in their history and do not preserve an original record of the presolar interstellar medium (1). Mundrabilla is an iron meteorite, a class of metal meteorites formed largely from melting in asteroidal cores. It is an FeS-rich meteorite with extremely slow cooling times, estimated to be 3 °C/y (8, 9). GRA 95205 is a ureilite meteorite that was heavily shocked during formation (10). Ureilites are primitive (meaning that they are nearly chondritic chemical composition), and most contain large grains of olivine, pigeonite, and pyroxene. The interstitial material consists of a unique mineralogical mixture of silicates, carbides, sulfides, and metals.

Shocked ureilites also contain diamond, graphite, lonsdalite, and other forms of carbon (11). Meteorites with extreme formation conditions are ideal for observing exotic chemical species, such as superconductors.

Magnetic field modulated microwave spectroscopy (MFMMS) is an ultrasensitive technique that can measure 10^{-12} cm^3 of superconducting material. This sensitivity is critical in measuring possible minute phases within inhomogeneous materials (12). MFMMS has been used to search for novel superconductivity in several types of inhomogeneous samples, such as phase spread alloys (13), bulk samples (14), and even natural samples, including meteorites (15, 16). However, previous searches for superconductivity in meteorites have not identified any superconducting compounds.

Results

MFMMS measurements were made on a powder sample extracted from Mundrabilla (MUND-1). At low direct current (DC) field, $H_{DC} = 15$ Oe, there were sharp transitions at $T_{c1} = 5$ K and $T_{c2} = 6$ K that indicated superconducting transitions (Fig. 1*A*). By applying increasing DC fields, these peaks were suppressed in both temperature and in magnitude and were barely visible at $H_{DC} = 1{,}000$ Oe. This peak shape and field evolution are characteristic of a superconducting transition (12). Peaks in the MFMMS signal were observed in five of the 10 samples collected from this meteorite.

Similar MFMMS measurements were made of a sample extracted from a piece of GRA 95205 (GRA-1). When the applied DC field (H_{DC}) was 15 Oe, there was a peak at $T_{c3} = 5.5$ K, and increasing H_{DC} suppressed this transition in temperature and magnitude (Fig. 1*B*), just like Mundrabilla. Only one of six samples taken from this meteorite exhibited such a peak.

In order to confirm that the peaks observed in MFMMS indicated superconductivity, vibrating-sample magnetometry (VSM) measurements were performed on samples from Mundrabilla (Fig. 2). Zero-field-cooled (ZFC) measurements of sample MUND-1 showed a strong diamagnetic response, characteristic of a superconducting transition (Fig. 2*A*). This diamagnetic response was suppressed in onset temperature and in magnitude at increased DC fields

Significance

In this paper, we report the presence of superconducting material in two meteorites. We further characterize these phases as alloys of lead, tin, and indium. These findings could impact our understanding of several astronomical environments. Superconducting particles in cold environments could affect planetary formation, shape and origin of magnetic fields, dynamo effects, motion of charged particles, and other processes.

Author contributions: J.W., M.T., S.C., Y.Z., and I.K.S. designed research; J.W. and S.C. performed research; M.T. contributed new reagents/analytic tools; J.W., M.T., S.C., Y.Z., and I.K.S. analyzed data; and J.W., M.T., S.C., Y.Z., and I.K.S. wrote the paper.

Reviewers: Z.F., University of California, Irvine; L.H.G., Florida State University; and M.H., Florida State University.

The authors declare no competing interest.

Data deposition: Data from Superconductivity Found in Meteorites have been deposited in University of California San Diego Library Digital Collections (https://library.ucsd.edu/dc/object/bb24282657).

See online for related content such as Commentaries.

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This article contains supporting information online at https://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1918056117/-/DCSupplemental.

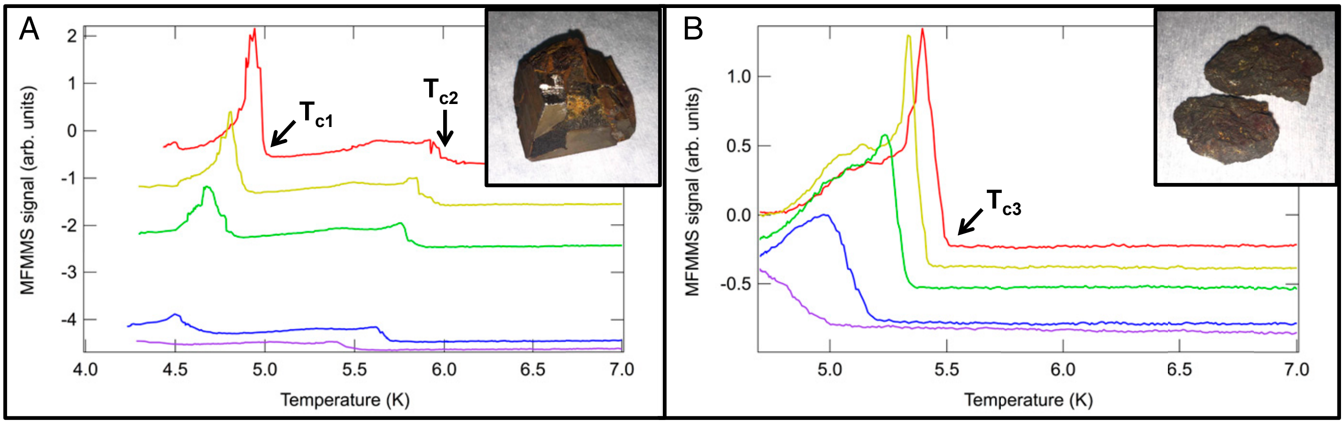

Fig. 1. MFMMS temperature sweeps of samples from Mundrabilla sample MUND-1 (*A*) and GRA 95205 sample GRA-1 (*B*). These sweeps were performed with a DC field, H_{DC} set to 15 Oe (red), 100 Oe (yellow), 200 Oe (green), 500 Oe (blue), and 1,000 Oe. An alternating current (AC) field, H_{AC} = 15 Oe is applied in parallel to the DC field. *A* and *B*, *Insets* show meteorite fragments from which all samples (e.g., MUND-1 and -2) were obtained (17). Arb., arbitrary.

(Fig. 2*B*). In addition, the curves showed multiple inflection points, which were likely the result of the multiple superconducting transitions observed in MFMMS. Low-magnetic-field ZFC and field-cooled (FC) measurements (5 and 10 Oe; Fig. 2*C*) showed similar behavior. The weaker response observed in FC measurements is consistent with superconductivity. The onset temperature for the superconducting transition observed with the VSM was ∼1 K higher than the onset temperature observed by using MFMMS, which is within the temperature uncertainty for the flow cryostat used in MFMMS and is likely a thermal lag typical of that technique.

In order to calculate the average magnetic susceptibility, the volume of the samples was estimated from two-dimensional images by using image-processing software, $V_{\mathrm{MUND\text{-}1}} = 9.10 \times 10^{-5} \pm 7.99 \times 10^{-5}$ cm^3 (SD) and $V_{\mathrm{MUND\text{-}2}} = 8.06 \times 10^{-4} \pm 5.40 \times 10^{-4}$ cm^3. Using this volume, low-magnetic-field measurements (Fig. 2*C*) gave a superconducting volume fraction across the whole sample of ∼5% (17). Measurements on another sample from Mundrabilla (MUND-2) showed a similar transition (Fig. 2*D*), in addition to other magnetic behavior observed at higher temperatures.

Since samples from both meteorites contained superconducting phases, we performed a "divide-and-conquer" process to isolate individual grains that contain the largest superconducting fractions. This isolation allowed us to determine their chemical composition. We examined the samples (denoted as "parent samples") with an optical microscope and completely divided them based on their visual morphology (i.e., what the samples looked like) into different subsamples. If the strength of the superconducting response substantially depends on the visual morphology, then these subsamples could be subjected to a battery of further tests.

MFMMS data were taken from a parent sample collected from Mundrabilla (sample MUND-2; Fig. 3*A*), and the subsamples were derived from the divide-and-conquer process. Under the microscope, there were three visually distinct morphologies that were separated into subsamples A, B, and C. Subsample A contained grains that were apparently metallic and homogenous. Subsample B contained samples that appeared nonreflective, with colors ranging from oranges to dark browns. Subsample C contained samples that appeared inhomogeneous and partially metallic.

Subsample A showed the strongest superconducting response in MFMMS; subsample B showed a slight superconducting response;

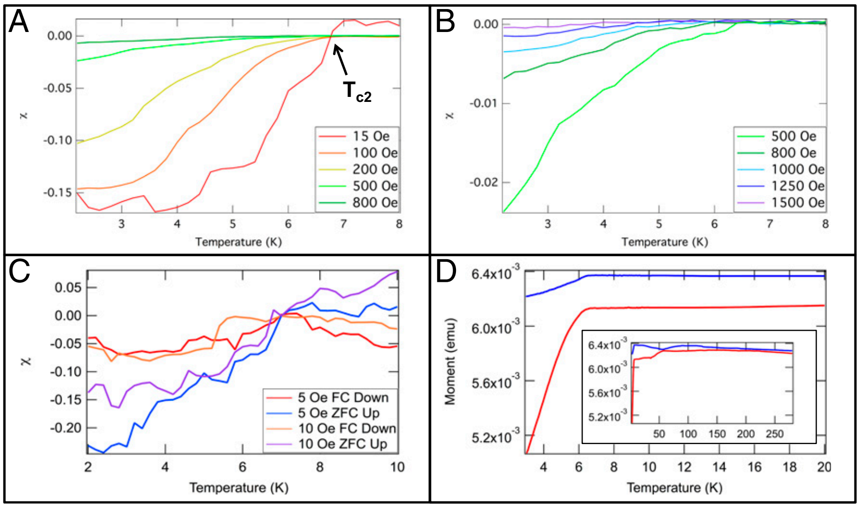

Fig. 2. VSM measurements of samples from Mundrabilla. ZFC measurements on sample MUND-1 were performed with applied magnetic field ranging from 15 to 1,500 Oe. (*A* and *B*) Measurements from 15 to 800 Oe (*A*) and 500 to 1,500 Oe (*B*) shown separately for visual clarity. (*C*) FC and ZFC measurements at 5 and 10 Oe were taken with multiple consecutive data taken at each temperature. All measurements offset for visual clarity. (*D*) ZFC measurements (red) and FC measurements (blue) were performed on a second sample, MUND-2, with an applied magnetic field of 500 Oe. *D*, *Inset* shows the full temperature range (17).

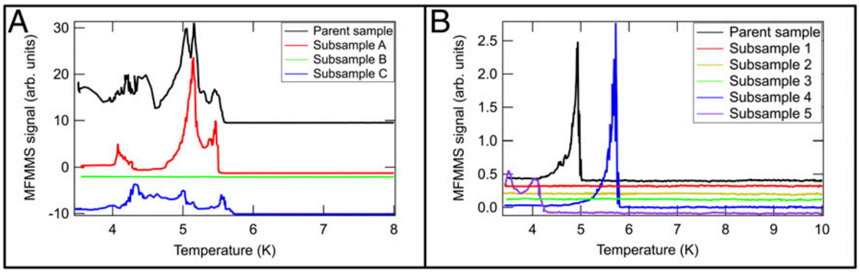

Fig. 3. MFMMS temperature sweeps of divide and conquer samples. These measurements are performed with $H_{DC} = H_{AC} = 15$ Oe for parent samples and subsamples from the Mundrabilla meteorite (*A*) and GRA 95205 (*B*). The data are vertically offset for visual clarity. Arb., arbitrary (17).

and subsample C showed a moderate response. Thus, the more metallic the sample appeared, the stronger the superconducting response. Further, the small response from subsample B indicated that almost all of the superconducting material was in grains that have metallic luster. The phases responsible for the superconductivity are then likely to be the macroscopic phases responsible for the luster of the grains.

Similar divide-and-conquer data measurements were performed on samples from GRA 95205 (Fig. 3*B*). The parent sample (taken from sample GRA-1) was divided into subsamples 1 through 5. Subsample 1 contained black grains, subsample 2 contained semitranslucent grains, and subsample 3 was brown and stony. Subsample 4 was white and crumbly with apparently metallic inclusions, and subsample 5 appeared semimetallic. These apparently metallic subsamples were the only two to exhibit a superconducting response. To determine the superconducting phase within these apparently metallic subsamples, it was necessary to measure the elemental composition of all of the subsamples and observe the differences.

Each of the subsamples measured in the divide-and-conquer process were measured by using the energy-dispersive X-ray spectroscopy (EDX) detector on a scanning electron microscope

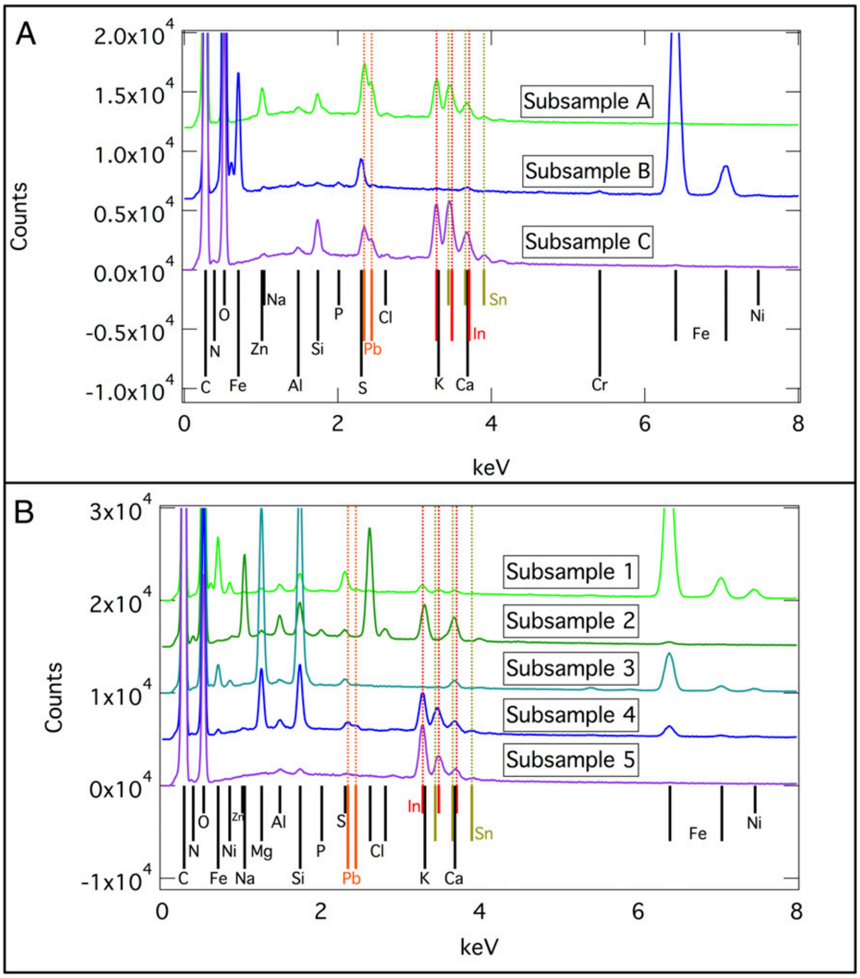

Fig. 4. EDX data measured from grains taken from subsamples of Mundrabilla (*A*) and GRA 95205 (*B*). Measurements are normalized and vertically offset for clarity. Black lines show ideal energies for EDX peaks of elements. Pb, In, and Sn have colored labels to highlight them for visual clarity (17).

(SEM). The EDX spectra for the subsamples from Mundrabilla (Fig. 4*A*) showed that the superconducting subsamples, A and C, contained lead, indium, and tin. The nonsuperconducting subsample, B, did not contain lead, indium, or tin. Similarly, the EDX spectra for the subsamples from GRA 95205 (Fig. 4*B*) showed that only the spectra from the superconducting subsamples (4, 5) had significant indium and tin peaks. Subsample 1 showed very small indium and tin peaks, while subsamples 2 and 3 did not show any evidence of indium or tin. None of the GRA 95205 samples showed a significant lead presence. This suggested that the superconducting phase within the samples was likely an alloy of lead, indium, and tin in Mundrabilla and an alloy of indium and tin in GRA 95205.

The multiple peaks observed in the MFMMS signal for the various subsamples in Fig. 3 are consistent with this conclusion. Lead and lead alloys are superconducting at these temperatures (18). Indium and lead alloys have a range of critical temperatures, decreasing with increasing indium concentration (19). Indium and tin alloys have critical temperatures below 4 K at high concentrations of either metal, but alloys with moderate concentrations of each have critical temperatures above 6 K (20). Therefore, multiple grains with varying ratios of metals would produce multiple superconducting critical temperatures.

To support this explanation, we created an algorithm to compare each set of EDX data with every superconducting phase in the Superconducting Material Database (SuperCon) (21) with T_c between 5 and 8 K. This method created a list of every superconducting candidate in SuperCon, for each subsample from Mundrabilla and GRA 95205 based on the elements detected by using EDX. These lists were sorted by the maximum volume percentage that each superconducting candidate could have in the sample, based on the availability of elements in the meteorite subsamples (*SI Appendix*, Table S1) (17). This analysis assumed that the EDX analysis was representative of the bulk content. The calculation showed that, out of all superconductors in SuperCon, those with the highest possible volume percentage in the superconducting subsamples were alloys of indium, tin, and lead.

To verify this, a state-of-the-art Super EDX detector on a transmission electron microscope (TEM) was used to measure one of the superconducting grains from Mundrabilla subsample A. The surface layers of the sample were removed with focused ion beam (FIB) and TEM images were taken of the interior (*SI Appendix*, Fig. S1), along with EDX analysis of nanoscale regions of the sample (*SI Appendix*, Table S2). All of the regions imaged were predominantly indium, lead, and tin, with indium making up the majority of each region (17). Trace quantities of aluminum, silicon, and magnesium were also present in some regions. Because alloys of lead, indium, and tin have been shown to be superconducting at these temperatures, and these elements were present with high purity in the grains that are superconducting, these alloys are likely the phases responsible for the MFMMS response.

Note that while these techniques provide strong evidence that the metallic species observed here is a superconducting metallic alloy, a direct measurement of the resistivity and/or the magnetization of the superconducting species cannot be made in these minute phases embedded in heterogeneous samples.

Discussion

Superconductivity in natural samples (i.e., those formed by natural processes, without further treatment) is extremely unusual (22). Naturally collected minerals are not phase-pure materials. Even the simplest superconducting mineral, lead, is only rarely found naturally in its native form, and, to our knowledge, there are no previous reports of natural lead samples superconducting (though it is still possible that sufficiently pure natural lead exists). In fact, we are only aware of one previous report of superconductivity in natural materials, in the mineral covellite, with a T_c of 1.6 K (22). However, the superconducting phases reported here likely exist in other meteorites, since such similar phases have been found in two dissimilar meteorites. This assumption is consistent with the abundance of these elements in other meteorites and the solar system at large. Lead and tin have moderate abundance, whereas indium is another order of magnitude less abundant, but still more abundant than the rare earths (1). Within ureilites, irons, and other meteorites, their abundances vary significantly (23–26). In an extensive review of siderophile elements in ureilites, it is observed that the abundance of bulk indium in particular is quite low (it had an abundance of 1 ng/g, while lead was about 500 ng/g) (26).

Given that these superconducting species have not been reported in the literature, determining the mechanism of formation of the species reported here is beyond the scope of this paper. Shock must be considered as a likely contributing factor, but deeper insight into speciation and siting in the meteorites will require further study. Notably, exotic mineral species enriched in metals (termed "mysterite") have been reported, including in ordinary chondrites. Though there have not been reports of superconductivity in mysterite, it has been used to explain the overabundance of several transition metals in chondrites, and a similar mechanism could explain the formation of the metal alloys reported in this study (27, 28).

When reporting new material phases in meteorites, it is important to show that the phase is not an impurity phase and is native to the meteorite. During sample processing, extreme care was taken to ensure that no outside contaminants were introduced (*SI Appendix*). In addition, the MFMMS and EDX analyses indicated that the superconducting phases were certainly natural in origin. The several peaks observed in the MFMMS samples were consistent with multiple, different superconducting phases with significantly differing stoichiometry (19, 20). This would not be expected from a laboratory contaminant. SEM EDX analysis indicated that the superconducting phases in the two meteorites differed, something that would be expected from natural phases, but not from contaminants that originated in a laboratory. Finally, TEM EDX measurements were performed on a sample cut with an FIB to allow analysis of the interior of the sample. This analysis showed that within a single superconducting fragment taken from Mundrabilla subsample A, the stoichiometry at different locations differed significantly, and there were at least six elements with higher than 1% concentration in one or more images (*SI Appendix*, Fig. S1 and Table S2). It is not plausible that such a sample could be formed from laboratory contaminants, so this implies that it must be natural.

While these nonchondritic meteorites do not carry a history of the interstellar medium (1), the fact that these phases can form naturally in macroscopic grains shows that there exists a natural process that can create these phases. This suggests the possibility that superconducting material phases could also be found in interstellar grains deep within the coldest regions of space, where these phases would be in a superconducting state. For example, cold, dense molecular clouds have typical temperatures as low as 10 K (29), but in regions with particularly low ultraviolet stellar radiation, temperatures are estimated to be as low as 5 K (30) or even colder (31). In unusual parts of space, temperatures can be even lower (32). The superconducting phases detected in these meteorites would be superconducting in these regions. Furthermore, other metallic alloys have been shown to have critical temperatures above 10 K (33, 34), which could exist naturally if they could be created in similar conditions to the phases detected in this manuscript.

Superconducting particles within cold regions of space could have implications on the structure of stellar objects. Specifically, superconducting particles could sustain microscopic current loops generated by transient fields and contribute to nearby magnetic fields. The origins of some magnetic fields, such as those observed in giant molecular clouds (35), are not well understood. Whether or not naturally superconducting phases would have any significant effect would depend on the critical temperature and quantity of such phases, and therefore deserves further study. There is an aggressive expansion of research and new missions to space, searching for new materials in extraterrestrial objects. This study suggests that the search for naturally occurring superconducting grains and their roles in astronomical objects should be a new component of such research.

Materials and Methods

A diverse population of meteorites was selected, and powder samples were dislodged or scraped from the meteorite, for measurement in MFMMS (12). Extreme care was taken to avoid contamination in handling the samples to assure that the results reflected the properties of the meteorites. Superconducting candidate samples were identified from peaks in the MFMMS response. The grains with the most superconducting material were isolated by subdividing these samples and measuring the subsamples in the MFMMS. These subsamples were subsequently measured with EDX in both an SEM and a TEM to characterize the superconducting grains. VSM ZFC and FC measurements were also taken to confirm superconductivity. For a full description of these materials and methods, see *SI Appendix*.

Data and Material Availability. Data are available from Superconductivity Found in Meteorites, University of California San Diego Library Digital Collections (https://library.ucsd.edu/dc/object/bb24282657). Materials are available from NASA (GRA 95205) and the Smithsonian (Mundrabilla).

ACKNOWLEDGMENTS. We thank Ilya Valmianski for help with SEM-EDX measurements; Alex Hojem for help editing the manuscript; and James Day for taking complementary measurements of the meteorites reported on in this paper. Harold Weinstock's original suggestion for the search for superconductivity in extraterrestrial materials is gratefully acknowledged. Insightful comments by three reviewers are gratefully acknowledged. We thank the Field Museum of Chicago and the Johnson Space Center for providing samples of Mundrabilla and GRA 95205 meteorites, respectively. This work was supported by Air Force Office of Scientific Research Grant FA9550-14-1-0202; and the University of California San Diego Chancellors Associates (M.T.). TEM work at Brookhaven National Laboratory was supported by the US Department of Energy, Office of Basic Energy Science, Division of Materials Science and Engineering Contract DE-SC0012704.

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