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FIRST LABORATORY OBSERVATION OF SILICA GRAINS FROM CORE COLLAPSE SUPERNOVAE

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ABSTRACT

We report the discovery of two supernova silica (SiO_2) grains in the primitive carbonaceous chondrites LaPaZ 031117 and Grove Mountains 021710. Only five presolar silica grains have been previously reported from laboratory measurements but they all exhibit enrichments in ^{17}O relative to solar, indicating origins in the envelopes of asymptotic giant branch stars. The two SiO_2 grains identified in this study are characterized by moderate enrichments in ^{18}O relative to solar, indicating that they originated in Type II supernova ejecta. If compared to theoretical models, the oxygen isotopic compositions of these grains can be reproduced by mixing of different supernova zones. While both theoretical models of grain condensation and recent NASA *Spitzer Space Telescope* observations have suggested the presence of silica in supernova ejecta, no such grains had been identified, until now, in meteorites. The discovery of these two silica grains provides definitive evidence of the condensation of silica dust in supernova ejecta.

Key words: circumstellar matter – ISM: supernova remnants – meteorites, meteors, meteoroids – nuclear reactions, nucleosynthesis, abundances – stars: winds, outflows – supernovae: general

Online-only material: color figures

1. INTRODUCTION

Presolar grains are tiny mineral and/or amorphous grains (from a few nanometers to several micrometers in diameter) that formed in stellar outflows or supernova ejecta, survived the collapse of the molecular cloud from which our solar system formed, and were incorporated into primitive extraterrestrial materials (meteorites, interplanetary dust particles, Antarctic micrometeorites, and cometary samples) in which they are found today (e.g., Zinner 2007, 2013). Presolar grains are identified by their highly unusual and anomalous isotopic compositions, which cannot be explained by any known processes acting in the solar system (e.g., radioactive decay, mass-dependent fractionation); their isotopic compositions can only be explained by nuclear reactions occurring in stellar environments (outside the solar system), indicating that these grains have stellar origins (Lodders & Amari 2005; Meyer & Zinner 2006).

Since the first identification of presolar silicates in interplanetary dust particles and primitive meteorites (Messenger et al. 2003; Nagashima et al. 2004; Nguyen & Zinner 2004), new presolar phases, such as FeO, MgO, and SiO_2 have been identified thanks to the use of Auger spectroscopy to characterize the elemental compositions of presolar silicate and oxide grains (Bose et al. 2012; Floss & Stadermann 2009; Floss et al. 2008; Nguyen et al. 2010).

Dust grains from core-collapse (Type II) supernovae constitute the second most abundant presolar grain population after grains from asymptotic giant branch (AGB) and red giants stars. Supernovae thus constitute an important source of dust during the time of solar system formation. A portion of this dust could have originated from a nearby supernova explosion, which might have triggered the collapse of the molecular cloud from which the solar system formed (Cameron & Truran 1977; Lattimer et al. 1977). A number of different presolar phases from supernovae have been identified to date in extraterrestrial materials, including graphite, SiC, Si_3N_4, and various oxides and silicates (Amari et al. 1992, 1993; Messenger et al. 2005; Nittler et al. 1998, 1995). Here, we report the discovery of two supernova silica (SiO_2) grains in primitive meteorites.

2. EXPERIMENTAL METHODS

Thin sections of the two meteorites LaPaZ (LAP) 031117 (CO3.0) and Grove Mountains (GRV) 021710 (CR2) were obtained from the NASA meteorite curatorial facility and the Polar Research Institute of China, respectively. The sections were initially carbon-coated and examined in a petrographic microscope to locate areas rich in dark fine-grained material for presolar grain searches.

The NanoSIMS 50 isotopic maps were acquired by rastering a focused Cs^+ primary beam of ∼1 pA, with a diameter of ∼100 nm over preselected matrix areas. Secondary ions of $^{12,13}C^-$ and $^{16,17,18}O^-$, as well as secondary electrons (SE), were simultaneously acquired in multicollection mode. Each measurement consisted of 5–10 scans of $10 \times 10\,\mu m^2$ (256×256 pixels) areas rastered within $12 \times 12\,\mu m^2$ regions that were pre-sputtered to remove the carbon coat. Images of individual layers were added to produce a single image for each isotope analyzed. Isotopic ratio images were used to detect isotopically anomalous grains. A grain was considered presolar if it fulfilled two conditions: (1) its isotopic composition deviated from that of the average surrounding material by more than 5σ, and (2) the anomaly was present in at least three consecutive layers.

The elemental compositions of O-anomalous grains identified by NanoSIMS analysis were determined with the PHI 700 Auger Nanoprobe. The areas of interest were initially sputter cleaned by scanning a defocused 2 kV, 1 μm Ar^+ ion beam over a broad area (∼2 mm) to remove atmospheric surface contamination and the grains were located on high-resolution SE images. Auger electron energy spectra of each grain were obtained with

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Table 1
Isotopic and Elemental Compositions of the Two SN Silica Grains of this Study and of Other Presolar Silica Grains

Meteorite	Size (nm^2)	$^{17}O/^{16}O$ ($\times 10^{-4}$)	$^{18}O/^{16}O$ ($\times 10^{-3}$)	O	Si	O/Si	Reference
LAP 031117 (CO3.0)	195 × 195	2.85 ± 0.4	2.82 ± 0.2	73.8	26.2	2.8	This study
GRV 021710 (CR2)	300 × 260	4.26 ± 0.2	2.73 ± 0.08	66.9	33.1	2.0	This study
ALHA 77307 (CO3.0)	175 × 175	7.1 ± 0.4	1.98 ± 0.08	56	25	2.2	1
	360 × 280	10.5 ± 0.4	1.61 ± 0.05	65.4	34.6	1.9	2
	280 × 160	5.9 ± 0.1	1.93 ± 0.02	69.1	30.9	2.2	2
	300 × 300	8.40 ± 0.03	1.99 ± 0.03	69.1	30.9	2.2	2
QUE 99177 (CR3)	150 × 150	17.9 ± 0.4	1.99 ± 0.05	67.9	32.1	2.1	3
Solar system reference (SMOW)		3.84	2.01				

Notes. Errors on isotopic compositions are 1σ. Elemental abundances are expressed in at.%.
References. (1) Nguyen et al. 2010; (2) Bose et al. 2012; (3) Floss & Stadermann 2009.

a 10 kV 0.25 nA primary electron beam in the energy range of 30–1730 eV. The compositions of the grains are calculated from the peak-to-peak heights of the differentiated spectra and are corrected for elemental sensitivities determined from standards. Errors are based on uncertainties in the measurements of the elemental compositions of a series of silicate standards, including SiO_2 (Stadermann et al. 2009).

3. RESULTS

Based on their oxygen isotopic compositions, Nittler et al. (1997) proposed a classification of presolar oxide–silicate grains into four groups, reflecting different stellar origins and/or nucleosynthetic processes. As shown in Figure 1(A), two silica grains identified in the carbonaceous chondrites LAP 031117 and GRV 021710 exhibit moderate enrichments in ^{18}O relative to solar and are classified as group 4 presolar grains (Nittler et al. 1997). The grain identified in LAP 031117 also has a slightly subsolar $^{17}O/^{16}O$ ratio, while the one identified in GRV 021710 has an $^{17}O/^{16}O$ ratio close to solar (Figure 1(A)). Both LAP 031117 and GRV 021710 are characterized by relatively high abundances of presolar silicates grains (Zhao et al. 2013; Haenecour et al. 2013), indicating that they are primitive meteorites that did not experience significant thermal processing and/or aqueous alteration.

The Auger spectra of these two presolar SiO_2 grains show that they consist only of silicon and oxygen (Figure 2); all other elements (e.g., Mg, Fe) are below detection limits (typically ∼3 at.%, depending on the quality of the spectrum, e.g., signal-to-noise). Quantification of the Auger spectrum of the grain found in GRV 021710 gives an O/Si ratio of 2.0 ± 0.6, consistent with SiO_2. The O/Si ratio of the grain identified in LAP 031117 is somewhat higher (O/Si = 2.8 ± 0.6); this could reflect residual surface oxygen contamination, although it is likely that the grain does indeed have a higher O/Si ratio than SiO_2, as many presolar silicates are also non-stoichiometric (Bose et al. 2012). If this is the case, this grain is likely to be amorphous. Both grains are relatively small with sizes of 190 × 190 nm^2 (LAP) and 300 × 260 nm^2 (GRV), respectively (Table 1).

4. DISCUSSION

Although five other presolar silica grains have been previously found in the CR3 meteorite QUE 99177 (Floss & Stadermann 2009), and the CO3.0 meteorite ALHA77307

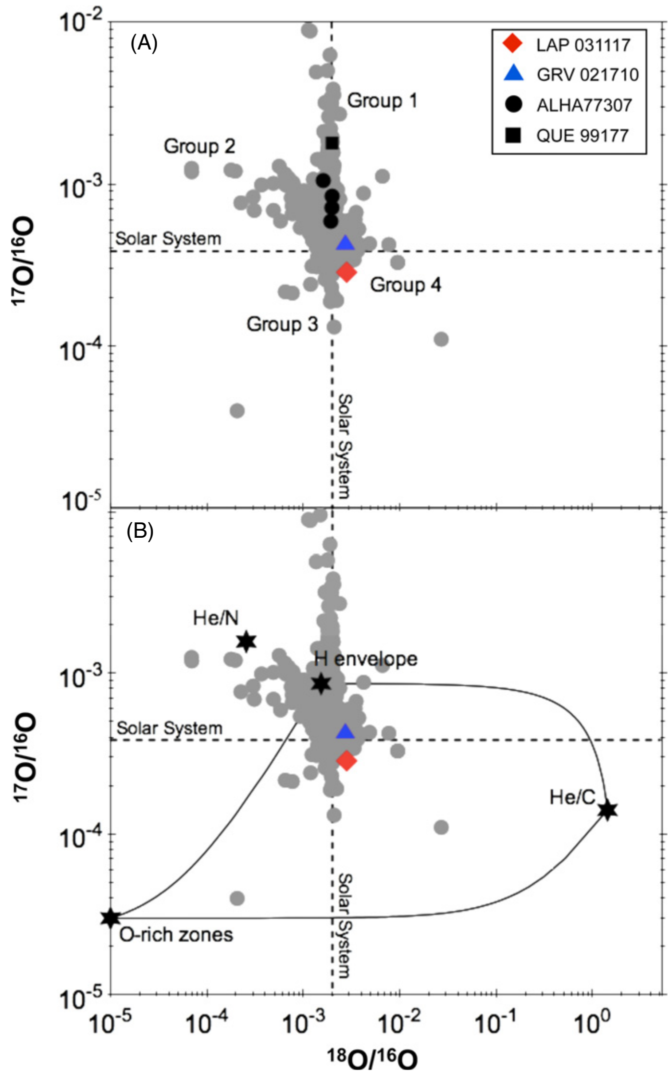

Figure 1. Oxygen three-isotope plot. (A) Oxygen isotopic compositions of the silica grains identified in LAP 031117 and GRV 021710 (this study), as well as those from QUE 99177 (Floss & Stadermann 2009), and ALHA 77307 (Bose et al. 2012; Nguyen et al. 2010). Other data (gray) are silicate and oxide grains from the Presolar Grain Database (Hynes & Gyngard 2009). Group classifications are from Nittler et al. (1997). (B) Supernova mixing calculations (lines) between different zones (stars) of a 15 $M_{\odot}$ supernova model from Rauscher et al. (2002).

(A color version of this figure is available in the online journal.)

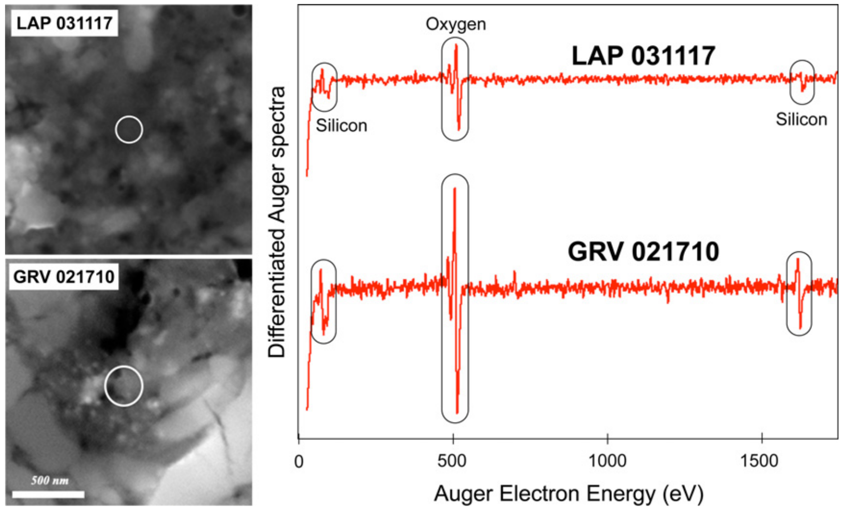

Figure 2. Auger nanoprobe secondary electron images and differentiated Auger elemental spectra of the silica grains identified in LAP 031117 and GRV 021710. The spectra show that the grains are composed only of silicon and oxygen; other elements are below detection limits (within background).

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(Bose et al. 2012; Nguyen et al. 2010), to our knowledge the two presolar silica grains identified in this study are the first Group 4 presolar silica grains. Below we discuss models of silica dust formation in circumstellar environments and compare them to astronomical observations.

4.1. Presolar Silica Grains from Low- to Intermediate-mass Red Giant or AGB Stars

Unlike the two presolar silica grains identified in this study, the five SiO_2 grains previously identified have larger than solar $^{17}O/^{16}O$ ratios and close to solar $^{18}O/^{16}O$ ratios (Table 1, Figure 1(A)), and are classified as Group 1 presolar grains (Nittler et al. 1997). Group 1 grains likely originated in the envelopes of low- to intermediate-mass red giant or AGB stars. Within errors, these grains all exhibit O/Si ratios consistent with SiO_2 (O/Si = 2). Although Auger spectroscopy provides compositional information, it does not provide the structural information needed to determine whether the grains are amorphous or crystalline. A recent study of one presolar silica grain by transmission electron microscopy (TEM) indicated that it was amorphous (Bose et al. 2012). However, it is not known whether the grain condensed as an amorphous phase, or was initially crystalline (e.g., quartz, tridymite, or cristobalite) and was later amorphized. Amorphous silica grains with large enrichments in both ^{17}O and ^{18}O ($^{17}O/^{16}O$ and $^{18}O/^{16}O$ ratios of about 0.01) have been discovered within insoluble organic matter isolated from the Murchison CM meteorite (Aléon et al. 2005). However, it has been argued that these grains have origins in the solar system, with the oxygen isotopic anomalies being produced through irradiation by high-energy particles accelerated during an active phase of the proto-Sun (Aléon et al. 2005).

Infrared spectroscopy studies of AGB stars indicate that about half of these stars, mainly semi-regular and Mira variable stars, exhibit a feature at 13 μm (Sloan et al. 1996). It has been suggested that SiO_2 is the carrier of the 13 μm feature observed in about half of these stars (Speck et al. 2000); however, other species, such as corundum (α-Al_2O_3) and spinel ($MgAl_2O_4$), have also been proposed as carriers of this feature (DePew et al. 2006).

Silica has been described as a "mythical condensate" because it is not expected to condense in stellar environments according to thermodynamic calculations for equilibrium condensation from a solar composition gas (Lodders & Fegley 1999). Silicon is expected to be completely consumed in circumstellar environments by the formation of Mg- and/or Fe-rich silicates, mainly olivine and pyroxene (Lodders & Fegley 1999). However, other thermodynamic studies (Ferrarotti & Gail 2001; Gail & Sedlmayr 1999) suggest that $MgSiO_3$ and SiO_2 constitute significant dust components in stellar atmospheres characterized by Mg/Si ratios of less than 1. Astronomical observations indicate that only about 10% of all stars have Mg/Si ratios favorable for the condensation of SiO_2 grains (Gail & Sedlmayr 1999), which would explain the low number of silica grains identified so far; close to 600 oxygen-anomalous presolar grains have been analyzed with the Auger Nanoprobe, but only seven (∼1%) are silica grains.

4.2. Presolar Silica Grains from Supernovae

The two silica grains identified in our study belong to Group 4, with moderate excesses in ^{18}O relative to solar. Group 4 presolar grains represent about 10% of the presolar oxide–silicate population in primitive meteorites (Haenecour et al. 2012) and their origins have remained unclear for many years. While both low-mass/high-metallicity red giant or AGB stars and Type II supernova have been proposed as possible sources of these grains, Group 4 presolar grains are now thought to originate in Type II supernova ejecta (e.g., Choi et al. 1998).

Before its explosion as a supernova, the interior of a massive star ($>8\,M_{\odot}$) consists of individual layers, with an onion-type structure, that contain the products of nuclear burning at increasing temperatures toward the core. The zones are named after the dominant elements present in each and consist, from the core to the surface, of the Ni zone, the Si/S zone, the oxygen-rich zones (O/Si, O/Ne, and O/C), and the He/C and He/N zones, surrounded by the H envelope (Meyer et al. 1995, Figure 3). Supernova mixing calculations indicate that the O isotopic compositions of many Group 4 oxide and silicate grains fall along a single mixing line of ^{16}O-rich material from the supernova interior with material from the He/C zone and the H envelope (Nittler et al. 2008).

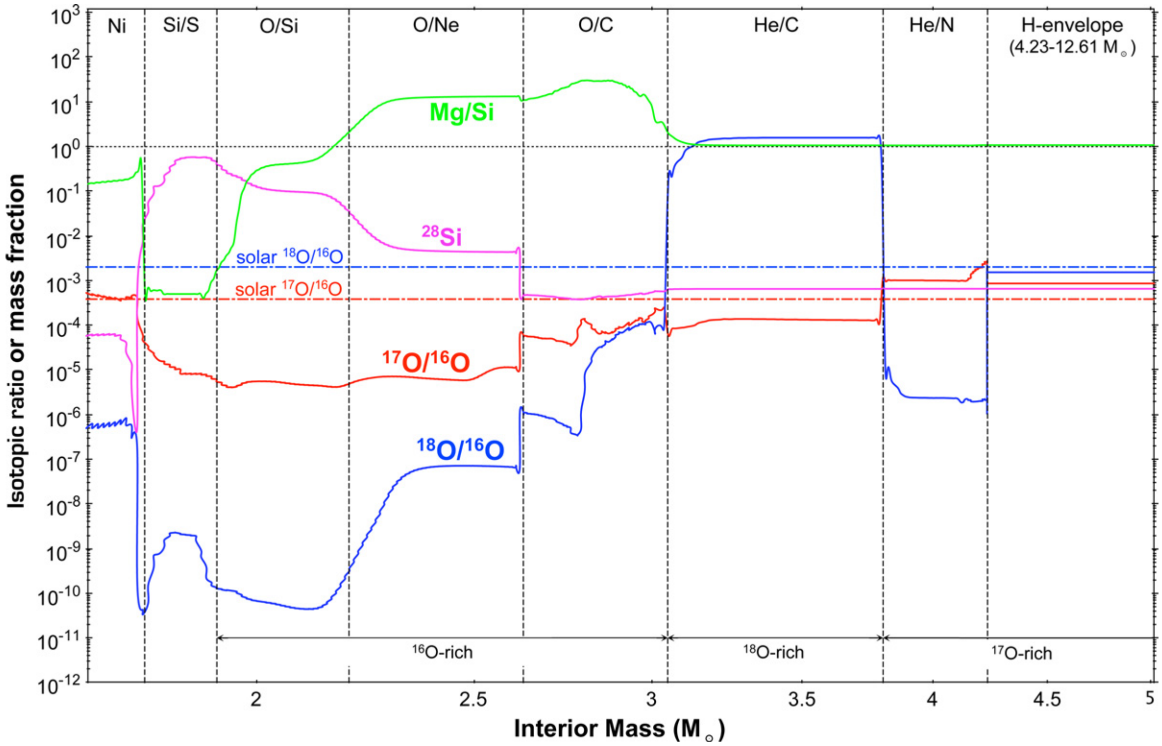

Figure 3. Oxygen isotopic ratios in the interior of a 15 $M_\odot$ Type II supernova. The diagram shows the $^{17}O/^{16}O$ and $^{18}O/^{16}O$ ratios, as well as the ^{28}Si mass fraction and Mg/Si elemental ratio as function of the interior mass of the 15 $M_\odot$ supernova model by Rauscher et al. (2002). The different SN layers are labeled according to the most abundant elements present.

(A color version of this figure is available in the online journal.)

Both equilibrium and non-equilibrium condensation models have predicted the condensation of silica grains in supernova ejecta, but no supernova presolar silica has been identified so far in primitive extraterrestrial materials. Equilibrium condensation calculations indicate that SiO_2 should be produced in abundance at high temperature in the massive O-dominated supernova zones, in the absence of polyatomic gaseous molecules (e.g., CO, SiO, H_2O, etc.; Ebel & Grossman 2001). However, it is not clear how likely such a scenario is. Another model, also based on equilibrium condensation for different layers of a 21 $M_\odot$ supernova, indicates that tridymite, a high-temperature polymorph of quartz, is a possible condensate in the O/Si zone (Fedkin et al. 2010). Non-equilibrium condensation calculations, based on non-steady-state nucleation and grain growth, also suggest the formation of SiO_2, through condensation from SiO molecules left over after the formation of Mg-silicates (Nozawa et al. 2003). Furthermore, recent modeling of the formation of supernova dust precursor molecules indicates that silica constitutes a prominent condensate in supernova ejecta (Cherchneff & Dwek 2010). However, supernova condensation models generally do not discuss the presence of SiO_2 because of the absence, until now, of this phase from the supernova presolar grain inventory in meteorites (Fedkin et al. 2010).

Recent astronomical observations by the NASA *Spitzer Space Telescope* indicate the possible presence of silica dust in several supernova remnants. Spectral fitting of the feature observed at 21 μm in *Spitzer* infrared spectrograph spectra of supernova remnants, including Cassiopeia A (Rho et al. 2008), and 1E0102−7219 in the Small Magellanic Cloud (Rho et al. 2009), suggests the presence of SiO_2, Mg-protosilicates, Al_2O_3, and FeO grains. It was suggested that SiO_2 might have originated from the inner O and Si/S layers of the supernova (Rho et al. 2008).

If silica grains can condense in O-rich layers of supernovae, as suggested by both thermodynamic calculations and astronomical observations, these grains should exhibit large excesses in ^{16}O relative to solar (Figure 3). However, the two silica grains identified in LAP 031117 and GRV 021710 exhibit enrichments in ^{18}O. Following the mixing calculations used in previous studies to reproduce the isotopic compositions of supernova oxide and silicate grains (Choi et al. 1998; Nittler et al. 2008; Qin et al. 2011), we carried out partial mixing calculations, using the O isotope yields for a 15 $M_\odot$ supernova model from Rauscher et al. (2002). We calculated the average oxygen isotopic compositions for the H envelope, the He/C zone, and the three O-rich zones and found that the oxygen isotopic compositions of the two silica grains may be reproduced by two somewhat different mixtures of small amounts (1%–2%) of ^{16}O-rich material from the O-rich inner zones and of ^{18}O-rich material from the He/C zone, with large amounts of material (about 97%) from the H-envelope (Figure 1(B)). In agreement with Nittler et al. (2008) we used the 15 $M_\odot$ model, but we emphasize that similar results are obtained from supernova models spanning a large range of masses, including the 21 $M_\odot$ one used by Fedkin et al. (2010). As suggested for silica grains from AGB stars, an Mg/Si ratio of less than 1 is likely to constitute a condition for the condensation of SiO_2 grains in supernovae. However, the mixing calculations that we carried out for the two silica grains result in Mg/Si ratios slightly higher than 1. As shown in Figure 3, only the O/Si and Si/S zones are characterized by an Mg/Si ratio of less than 1. There is thus a discrepancy between the theoretical models which predict condensation of silica grains in the inner O-rich layers of a supernova and our calculations based on the oxygen isotopic compositions of the grains, which suggest formation of the grains mainly from H-envelope material. More generally, other oxygen-rich silicate and oxide grains are also expected to condense in the inner ^{16}O-rich layers of supernovae and should, thus, exhibit enrichments in ^{16}O relative of solar (Figure 3). However, only two ^{16}O-rich presolar grains have been found to date (Nittler et al. 1998; Gyngard et al. 2010) and most presolar silicate and oxide grains are thought to come from supernovae instead show excesses in ^{18}O (Nittler et al. 2008), like those observed in the two presolar silica grains from our study (Figure 1(B)). The reason for this discrepancy remains unclear.

5. SUMMARY AND CONCLUSION

We report the first identification of two supernova silica (SiO_2) grains in the primitive meteorites LAP 031117 and GRV 021710. The enrichments in ^{18}O relative to solar observed in those two grains can be reproduced by mixing small amounts of material from the O-rich inner zones and of ^{18}O-rich material from the He/C zone with large amounts of material from the H-envelope of a supernova.

Theoretical models of grains condensation have predicted the presence of SiO_2 in supernova ejecta and silica has also been invoked to model the IR spectra of supernova remnants; however, no supernova SiO_2 grains had been identified in extraterrestrial materials. With the in situ identification of two supernova SiO_2 grains in primitive meteorites, our study provides definitive evidence of the presence of SiO_2 dust in supernova ejecta and reconciles supernova condensation model predictions and the presolar grain inventory in meteorites.

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