Origin of isolated olivine grains in carbonaceous chondrites

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Abstract—We report microscopic, cathodoluminescence, chemical, and O isotopic measurements of FeO-poor isolated olivine grains (IOG) in the carbonaceous chondrites Allende (CV3), Northwest Africa 5958 (C2-ung), Northwest Africa 11086 (CM2-an), and Allan Hills 77307 (CO3.0). The general petrographic, chemical, and isotopic similarity with bona fide type I chondrules confirms that the IOG derived from them. The concentric CL zoning, reflecting a decrease in refractory elements toward the margins, and frequent rimming by enstatite are taken as evidence of interaction of the IOG with the gas as stand-alone objects. This indicates that they were splashed out of chondrules when these were still partially molten. CaO-rich refractory forsterites, which are restricted to $\Delta^{17}O < -4_{/00}$, likely escaped equilibration at lower temperatures because of their large size and possibly quicker quenching. The IOG thus bear witness to frequent collisions in the chondrule-forming regions.

INTRODUCTION

In their seminal description of Murchison, Fuchs et al. (1973) reported isolated olivine grains (henceforth IOG), one of which earned the honor of their frontispiece. With the condensation models in full swing since the fall of Allende (e.g., Marvin et al. 1970; Grossman 1972), it was but natural to interpret these grains, soon found in other carbonaceous chondrite clasts, as nebular condensation products, for which Olsen and Grossman (1974, 1978) added morphological evidence such as large size, euhedral shapes, and patterned crystal surfaces. Yet chemical analogies of the IOG with chondrule olivine, as well as the presence of glass inclusions chemically comparable to chondrule mesostases, suggested early that they were chondrule fragments, possibly liberated after alteration of the mesostasis (e.g., McSween 1977; Richardson and McSween 1978; Desnoyers 1980; Nagahara and Kushiro 1982; Jones 1992).

Still, Steele (1986) drew attention to a subset of IOG, both in carbonaceous and ordinary chondrites, with particularly forsteritic and refractory mineral chemistries and bright blue cathodoluminescence (CL), which led him to entertain again a condensate origin (e.g., Steele 1988, 1989), for which the SIMS (secondary ion mass spectrometer) measurements of Weinbruch et al. (1993, 2000) found supporting evidence in their $^{16}O$ enrichment (i.e., in the direction of refractory inclusion compositions) and flat rare earth element (REE) patterns in olivine, inconsistent with igneous partitioning. Yet, as further data confirmed, the oxygen isotopic composition of the IOG largely overlaps with chondrules in carbonaceous chondrites (e.g., Jones et al. 2000; Russell et al. 2010; Ushikubo et al. 2012). Also, in situ laser ablation inductively coupled mass spectrometer (LA-ICP-MS) analyses have revealed that IOG olivine has, in fact, quite fractionated REE, in accordance with near-equilibrium igneous partitioning (e.g., Pack et al. 2005; Jacquet et al. 2012; Jacquet and Marrocchi 2017). While the balance of evidence thus remains in favor of a genetic link between IOG and chondrules, some systematic differences, at least among the forsteritic ones, with the general population of chondrules remain to be understood so as to decide whether they sample the same heating events. For
instance, the relative \(^{16}\text{O}\) enrichment of Tagish Lake (C2-ung) IOG indicated “more primitive material than the forsterite-rich chondrules” to Russell et al. (2010) while Jacquet and Marrocchi (2017) attempted to relate their coarse grain size and incompatible element enrichment of IOG in Northwest Africa (NWA) 5958 (C2-ung) to longer thermal processing. The IOG would be obviously important (olivine-rich) pieces of the puzzle of chondrule origin, if only their exact formation context vis-à-vis mainstream chondrules could be ascertained.

Our recent SIMS/electron microprobe (EMP) work on type I chondrules (i.e., reduced, with \(F_{90-100}\) olivine) with porphyritic olivine (PO) textures in carbonaceous chondrites has increased our understanding of the formation of olivine in these objects (Marrocchi et al. 2018, 2019). We have identified Al-Ti-poor cores in central olivine grains whose oxygen isotopic deviation from the host revealed their relic nature and likely derivation from amoeboid olivine aggregate (AOA)-like precursors. These are overgrown by olivine enriched in incompatible elements and crystallized from melt produced during initial heating. Finally, palisadic olivine grains near the chondrule margins show evidence for gas-assisted outward growth, as also beautifully depicted by the CL maps of Libourel and Portail (2018). These different generations of olivine multiply our opportunities to find suitable analogs for IOG, at least for the magnesian population which dominates in carbonaceous chondrites. We have thus set out to extend our work from type I chondrules to their isolated olivine counterparts (which we will also refer to as “type I IOG”) in carbonaceous chondrites, where such reduced compositions dominate their high-temperature fraction. Specifically, we present in this paper combined petrographic (CL), chemical (EMP), oxygen isotopic (SIMS) data for magnesian IOG and chondrules in CM, CO, and CV chondrites. The comparison will afford new insights on the genesis of these IOG.

### SAMPLES AND METHODS

We surveyed the polished sections NWA 5958-1 and NWA 5958-4 of NWA 5958, the thin section 3181lm4 (or 3181-4) of Allende (all three from the Muséum national d’Histoire naturelle, Paris, France), the thin section ALHA 77307,96 of Allan Hills (ALH) 77307 (from the NASA Antarctic Search for Meteorites program), and a thick section of NWA 11086 prepared at CRPG. Allende is an oxidized Allende-like CV subgroup classified as a CV3.6 by Bonal et al. (2006). ALHA 77307 is a CO3.0 chondrite (Grossman and Brearley 2005; Bonal et al. 2007; Busemann et al. 2007). Opaque assemblages, chondrule mesostases, olivine textures, and compositions of chondrules show no indication of alteration and diffusional exchange. NWA 5958 corresponds to a C2-ung CM-like chondrite with type II chondrule olivine Cr content and opaque petrography indicating minimal thermal metamorphism (<300 °C) but chondrule mesostases bearing witness to significant aqueous alteration (Jacquet et al. 2016; Jacquet and Marrocchi 2017). NWA 11086 has been classified as a CM-an due to \(^{16}\text{O}\) enrichment and the absence of phyllosilicates according to X-ray diffraction (Gattacceca et al. 2019), but significant aqueous alteration is indicated by the presence of PCP (poorly characterized phases, also known as tochilinite cronstedtite intergrowths; see e.g., Lentfort et al. 2020), conversion of chondrule mesostasis to “spinach” (Fuchs et al. 1973), and paucity of Fe-Ni metal, so it might be a plain, if somewhat \(^{16}\text{O}\)-rich, CM2 chondrite. In the plots and the discussion, data from these latter two chondrites and ALH 77307 will be subsumed in the CM/CO clan.

Secondary electron microscope (SEM) imaging was performed on a JEOL JSM-6510 SEM equipped with a Genesis EDX detector at the Centre de Recherches Pétrographiques et Géochimiques (CRPG-CNRS, Nancy, France) using 3 nA electron beam accelerated at 15 kV. Modal abundances of high-temperature components were also estimated by manual point counting on backscattered electron (BSE) maps using the JMicrovision software (N. Roduit; https://jmicrovision.github.io) with 4000 points (randomly chosen by the software) per section (except for ALHA 77307, 2000 points). An object was considered an IOG if its interior was dominated by one olivine grain. During closer BSE observation, only IOG large enough for SIMS analyses (~15 μm spot; this is also roughly the scale over which the BSE image allowed assignment to IOG in the point counting) were selected for further study. The effective radius of the IOG or the coarsest olivine of each studied chondrule was calculated as the radius of the equal area disk, from images. Cathodoluminescence (CL) imaging of chondrules was performed using (i) an RGB CL detection unit attached to a field emission gun secondary electron microscope JEOL J7600F at the Service Commun de Microscopie Electronique (SCMEM, Nancy, France) and (ii) a field-gun ZEISS Supra 55 VP equipped with an OPEA cathodoluminescence device (imaging and spectral) with a high-tech parabolic mirror. Quantitative chemical compositions of olivine grains were obtained using a Cameca SX Five EMP at the Université Pierre et Marie Curie (UPMC, Camparis, Paris, France) using a 150 nA focused beam (about ~2 μm in diameter) accelerated to 15 kV. We analyzed Na, Mg, Si, Al, K, Ca, Fe, Ti, Cr,
and Mn in olivine grains. The high beam current allowed detection limits for silicates of 100 ppm for Al, Ca, and Ti; 150 ppm for Mn and Si; and 200 ppm for Na, K, Cr, Fe, and Mg. The PAP software was used for matrix corrections.

We measured the oxygen isotopic compositions of chondrule olivine and isolated olivine grains, where CL and EMP had shown fairly homogeneous compositions (typically near the center), with a Cameca IMS 1270 E7 at CRPG-CNRS (Nancy, France). $^{16}$O, $^{17}$O, and $^{18}$O ions produced by a Cs$^+$ primary ion beam (~15 µm, ~4 nA) were measured in multi-collection mode with two off-axis Faraday cups (FC) for $^{16}$O, $^{17}$O, and the axial FC for $^{17}$O. The FC had $10^{11}$ Ω amplifiers. To remove $^{16}$OH$^-$ interference on the $^{17}$O peak and to maximize flatness atop the $^{16}$O and $^{18}$O peaks, the entrance and exit slits of the central FC were adjusted to obtain mass resolution power of ~7000 for $^{17}$O. As an additional safeguard against $^{16}$OH$^-$ interference, an N$_2$ trap was used to reduce the pressure in the analysis chamber to $<$5 x $10^{-6}$ mbar. The multicollection FCs were set on exit slit 1 (MRP = 2500). Total measurement times were 240 s (180 s measurement + 60 s pre-sputtering). We used three terrestrial standard materials (San Carlos olivine, magnetite, and diopside) to define the instrumental mass fractionation line for the three oxygen isotopes and correct for instrumental mass fractionation for olivine. To obtain good precision on analytical measurements, we analyzed, in order, four standards, eight chondrule forsterites (recognized by CL) had CaO > 0.4 wt% and such compositions represent two-fifths of our IOG analyses, and since Pack et al. (2004) only counted the luminescing part of their olivine, this would indicate IOG modal fractions of order 1 vol%. More recently, manual identifications on X-ray maps by Ebel et al. (2016) resulted in an average of 1.16 vol% of “isolated olivine grains or aggregates in matrix” in CO chondrites and 1.37 vol% in Allende. Part of the differences between studies may lie in the ambiguity in discriminating IOG from chondrules. Despite this systematic uncertainty, an order of magnitude of a few percent for IOG modes in carbonaceous chondrites seems overall sound.

RESULTS

Petrography

Type I chondrules present the usual mineralogical zoning (Fig. 1) of olivine phenocrysts dominating in the interior and enstatite laths, poikilitically enclosing olivine chadacrysts, concentrating near the outside (e.g., Libourel et al. 2006; Friend et al. 2016). The mesostasis is altered in the CM chondrites. When viewed in CL, interior olivine grains often display dark cores (Fig. 1) that correspond to relict grains for those chondrules investigated by Marrocchi et al. (2018, 2019), surrounded by brighter overgrowths (identified to the in situ crystallized host). The “palisadic” olivine grains near the outside of PO chondrules (Libourel and Portail 2018; Marrocchi et al. 2018, 2019) have brightest CL in their inner edge and darken toward the exterior (Fig. 1D).

Point counting reveals 1.7–4.2 vol% of IOG in the studied carbonaceous chondrites (Table 1). Fuchs et al. (1973) reported 8 vol% of isolated mineral grains in Murchison, about two-thirds of which appeared to be olivine; Browning et al. (1996) reported 6 vol% of IOG in Murchison, and down to 1.5 vol% in more altered CM chondrites. The average 22 vol% of “single grain and grain fragments” of Olsen and Grossman (1974) for C2 chondrites must be in error as it equals the total high-temperature fraction normally seen in CM chondrites (e.g., Table 1; Howard et al. 2011). As for CO3 chondrites, McSween (1977) quoted 8 vol% for IOG. While our numbers thus tend to be systematically lower than past literature estimates based on optical microscopy, we note that Pack et al. (2004) never found more than 0.35 vol% of “refractory forsterites” in carbonaceous chondrites. Since their refractory forsterites (recognized by CL) had CaO > 0.4 wt% and such compositions represent two-fifths of our IOG analyses, and since Pack et al. (2004) only counted the luminescing part of their olivine, this would indicate IOG modal fractions of order 1 vol%. More recently, manual identifications on X-ray maps by Ebel et al. (2016) resulted in an average of 1.16 vol% of “isolated olivine grains or aggregates in matrix” in CO chondrites and 1.37 vol% in Allende. Part of the differences between studies may lie in the ambiguity in discriminating IOG from chondrules. Despite this systematic uncertainty, an order of magnitude of a few percent for IOG modes in carbonaceous chondrites seems overall sound.

While most IOG are of type I, it is noteworthy that the type II/type I modal ratio is significantly higher for IOG (0.1–0.6) than for bona fide chondrules (0.03–0.11), by a factor of 4–7. This is qualitatively in line with the histograms of McSween (1977), which indicate values of 2 and 0.8 for this ratio in CM and CO chondrites, respectively, with that of Desnoyers (1980) indicating a ratio of 1.2 for Niger (C2), although those histograms are based on microprobe analyses and not areas (perhaps accounting for the systematic difference). Seven of the 12 Tagish Lake IOG of Russell et al. (2010) were of type II; same for 25 of 101 CI chondrite
IOG compiled by Piralla et al. (2020). This point being made, we henceforth exclusively focus on type I IOG.

As noted by Olsen and Grossman (1978), the IOG tend to be big compared to chondrule phenocrysts (Fig. 2, even though the first bin is cut off by our selection biases). In fact, in CV chondrites, their size distribution is comparable to that of the coarsest chondrule phenocrysts; in CM/CO chondrites, more than 90% of the IOG studied here are even bigger than the mode of the coarsest chondrule phenocrysts (near 30 µm effective radius). About half (21 of 41) of the IOG examined here by BSE/CL and EMP have equidimensional, often euhedral/subhedral shapes, although the others are evidently fragments of larger objects (e.g., Figs. 3C, 3D and 4G, 4H). The CL of the former, generally blue in the core, is concentrically zoned, with intensity decreasing toward the margin, which may have a redder tint. Discrete darker CL streaks, partially or entirely surrounding the center, are often superimposed on the background trend, generally paralleling the grain edges. An oscillatory sector zoning is spectacularly visible in NWA 11086 IO12 (Fig. 5); some may be discernible in NWA 5958-I 107 as well (Fig. 4F). Despite their name, the IOG are not pure olivine. The olivine may enclose metal grains and glass inclusions, and, when whole, is nearly always surrounded, partly or entirely, by enstatite (from a few microns to tens of microns in thickness), sometimes with mesostasis (although we did not find as large a mesostasis patch as in IOG N6-5 in fig. 1e of Jacquet and Marrocchi 2017) and even further olivine phenocrysts. Enstatite rims, partial or total, are sometimes seen even around fragments (e.g., Figs. 3C and 3D). We note that Jones (1992) called large olivine grains surrounded by pyroxene in ALHA 77307 “macroporphyritic chondrules,” in contradistinction to “bare” isolated olivine grains, but given the continuum between them, we maintain the name IOG for all those objects. The continuum, in fact, extends to bona fide chondrules (e.g., Jacquet and Marrocchi 2017), with some chondrules exhibiting disproportionately large olivine crystals (e.g., Figs. 1A and 1B; chondrule N5-21 with three aligned coarse olivine grains in fig. 2b of Jacquet et al. 2016), not to mention chondrule-IOG or IOG-IOG compounds (see e.g., Figs. 1G and 3E, 3F). Overall, this continuum makes the assignment of objects to IOG rather than chondrules relatively subjective and must have contributed to the scatter in IOG modes in the literature discussed above. We recall that we have considered here an object to be IOG when its interior was dominated by one olivine crystal.

Table 1. Modal abundances of high-temperature components in the studied chondrites; 4000 points were counted per section except for ALHA 77307 (2000 points).

| Classification | NWA 5958 | NWA 11086 | ALHA 77307 | Allende |
|----------------|---------|-----------|------------|--------|
| Type I chondrules |         |           |            |        |
| Bona fide | 20.3 | 23.2 | 36.3 | 31.3 |
| IOG | 2.3 | 3 | 2.6 | 1.5 |
| Type II chondrules |         |           |            |        |
| Bona fide | 1.2 | 1.4 | 3.8 | 1 |
| IOG | 0.9 | 1.1 | 1.6 | 0.2 |
| Refractory inclusions |         |           |            |        |
| AOA | 1.2 | 0.8 | 1.6 | 5.7 |
| CAI | 1 | 0.5 | 0.6 | 3.8 |

1 Fragments may be numerically more numerous for smaller sizes which we have not selected. Still, it may not affect the modal ratio much (here the studied “whole” IOG outweigh fragments also in this respect, with a total area of 0.74 mm² versus 0.59 mm²).
Individual profile location and analyses are shown in the supporting information and some representative ones are illustrated in Fig. 6. The CL zoning in IOG olivine is reflected by EMPA traverses, with Ca, Al, Ti decreasing from core to rim, whereas Fe, Mn, Cr symmetrically increase outward. Thus, the zoning is basically of decreasing “refractoriness” toward the margin (Fig. 6; Jones 1992; Jacquet and Marrocchi 2017). This is seen in the coarsest chondrule olivine grains as well (more asymmetrically for the palisadic grains, with 0.2–0.5 wt% CaO; Marrocchi et al. 2018) although many have flatter minor element profiles (Figs. 6A–D). In general, excluding obvious fragments, flat profiles are associated with uniformly low CaO (<0.4 wt%) abundances. While Al and Ti roughly parallel Ca, they may suffer abrupt drops or peaks (e.g., Figs. 6A and 6D) which correspond to oscillations seen in CL (whose lengthscales may be shorter than the profile steps) or occasional relict grains (mostly for chondrules; Marrocchi et al. 2018). This is consistent with the CL profiles shown in Fig. 6; Jones 1992; Jacquet and Marrocchi 2017)
with the contention of Libourel and Portail (2018) that CL intensity is largely controlled by Al (in the absence of sufficient amounts of the CL quencher Fe). NWA 11086 IO12 (Fig. 6H) alone shows Ca oscillations unattenuated relative to Al and Ti.

In the biplots of this paper (Figs. 7 and 8), the IOG will be represented by their apparent core composition. CaO spans 0.16–0.89 wt% in IOG and 0.05–0.66 wt% in chondrule coarsest olivine (with averages of 0.44 versus 0.30 wt%); Al₂O₃ spans 0.06–0.42 wt% in IOG and chondrule coarsest olivine 0.02–0.53 wt% (with averages of 0.20 versus 0.12 wt%). Crystal size seems to correlate negatively with Fe, Mn, but the positive correlation with Ca and Al noted by Jacquet and Marrocchi (2017) is very rough (Fig. 7). This may be to some extent a 2-D sectioning artifact as more equatorial sectioning of the grains (providing the widest areas) would pass closest to the actual refractory core. Excluding Allende, whose olivine FeO contents up to 5 wt% are obviously secondary, the IOG have low FeO contents (0.35–1.4 wt%) anticorrelated with CaO, similar to chondrule phenocrysts in this compositional range (Fig. 8).

The oxygen isotopic compositions of chondrule olivine and IOG plot along the primitive chondrule mineral (PCM; Fig. 9) line of Ushikubo et al. (2012). When analyzed multiple times, the IOG appear generally isotopically homogeneous, with the exception of NWA 11086 IO22 (with Δ¹⁷O of −0.18 ± 0.60‰ and −5.39 ± 0.60‰), but the number of analyses per object is usually small. Δ¹⁷O ranges from −12.65 to −0.2‰ for chondrules and −7.46 to 0.84‰ for IOG. The distribution (Fig. 10) is fairly similar to the chondrule host (i.e., non-relict) olivine grains of Marrocchi et al. (2019), with a minor ¹⁶O-poor population above the ~4‰ hiatus noted by Ushikubo et al. (2012). The four IOG in this range are not more fayalitic than the others, unlike the trend shown by type I chondrules in several studies (e.g., Ushikubo et al. 2012; Schrader et al. 2013; Tenner et al. 2013, 2015) but similar to the Jacquet and Marrocchi (2017; see their fig. 8a) data for NWA 5958. Nevertheless, high refractory element
Fig. 6. Minor element profiles in chondrule olivine (A–D) and IOG (E–H), with profile positions in the indicated figures. Recall that NWA 5958-4 IOG21 is a compound object where the profile transects two successive (formerly isolated) olivine grains (Figs. 3E and 3F).
Isolated olivine grains

Genetic Link Between Isolated Olivine Grains and Chondrules

The general overlap in $\Delta^{17}O$ (Fig. 10) between IOG and chondrule host olivine, as well as in minor elements, indicates, as in previous work (McSween 1977; Desnoyers 1980; Jones 1992; Jones et al. 2000), that IOG are closely linked to chondrules rather than early nebular condensates such as AOAs. The presence of mesostasis is also evidence for an igneous origin incompatible with gas–solid condensation (e.g., McSween 1977; Richardson and McSween 1978). Other phases associated with olivine (metal, occasional companion olivine crystals, pyroxene) are also most analogous to chondrule petrography; with IOG CL being comparable to that of the coarsest olivine in chondrules (Fig. 1). We may also recall the REE fractionation measured in IOG olivine (Pack et al. 2005; Jacquet et al. 2012; Jacquet and Marrocchi 2017) which is at variance with the flatter patterns in AOA olivine which formed by condensation (Ruzicka et al. 2012; Jacquet and Marrocchi 2017). The oscillatory sector zoning shown by the exceptional NWA 11086 IO12 (Fig. 5) also suggests departure from equilibrium, but the analogy with fast diffusion-controlled grown

Fig. 7. Olivine size versus minor element content (A: FeO; B: MnO; C: CaO; D: Al$_2$O$_3$) in IOG and chondrule coarsest olivine. Data for all analyzed chondrule phenocrysts in Marrocchi et al. (2018, 2019) are also shown. (Color figure can be viewed at wileyonlinelibrary.com.)

The isotopic, chemical, and geometrical data for all analyzed objects are shown in supporting information.

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planetary igneous olivine (Welsch et al. 2014) is consistent with an igneous character. Adding to this the textural continuum between IOG and bona fide chondrules, we contend, in agreement with the latest literature (e.g., Jones et al. 2000; Pack et al. 2005; Russell et al. 2010; Ushikubo et al. 2012), that IOG are genetically related to chondrules.

Isolated Olivine Grains Formed Hot

It does not, however, follow that IOG result from the cold fragmentation of chondrules. Olsen and Grossman (1978) had already voiced skepticism about the feasibility to cleanly separate olivine phenocrysts from the mesostasis. While Richardson and McSween (1978) had invoked the friability of the altered mesostasis at least for CM chondrite chondrules, that alteration is now widely believed to have occurred on the parent body (Brearley 2014), too late for chondrule–chondrule collisions, and would not be relevant for the least altered chondrites such as ALHA 77307. The large size of IOG compared to chondrule phenocrysts is also problematic if the former are fragments of the latter (e.g., Olsen and Grossman 1978). The larger IOG/chondrule ratio for type II compared to type I objects in carbonaceous chondrites would not be understandable if the chondrules were fragmented after being mixed together in the general protoplanetary disk.

The euhedral or equidimensional morphology of many IOG and their frequent concentric CL zoning patterns indicates that those feature unbroken olivine. Although, again, this study focuses on type I IOG, we recall that the ferroan IOG N6-7 in NWA 5958 exhibited oscillatory zoning visible in BSE (Jacquet et al. 2016; Jacquet and Marrocchi 2017). The frequent presence of near-complete enstatite margins (e.g.,

Fig. 8. Minor element biplots of IOG, chondrule coarsest olivine of this study, and chondrule olivine (keyed by chondrule) from Marrocchi et al. (2018, 2019). A) CaO versus FeO. The correlation is negative for FeO below ~1.5 wt% and positive afterward. Individual chondrules plot in only one of these trends. IOG all belong to the former. B) Al₂O₃ versus TiO₂. C) MnO versus FeO. D) MnO versus Cr₂O₃. (Color figure can be viewed at wileyonlinelibrary.com.)
Figs. 3G, 3H and 4C, 4D) in type I IOG is comparable to those shown by whole chondrules. These are ascribed to the influx of SiO into residual melts, promoting pyroxene crystallization at the expense of olivine (e.g., Libourel et al. 2006; Chaussidon et al. 2008; Friend et al. 2016; Barosch et al. 2019). This indicates that the IOG experienced such influx as independent objects. In fact, prior to enstatite precipitation, the decrease of refractory incompatible elements toward IOG olivine margins reflected by the CL zoning also suggests condensation of Mg and SiO into the parent melt, inducing dilution of these elements, as advocated by Jacquet and Marrocchi (2017), as well as Marrocchi et al. (2018, 2019) and Libourel and Portail (2018) for the palisadic olivine. More volatile Fe, Mn, Cr would have recondensed contributing to their outward increase in the olivine. Perhaps in some cases—for those IOG which really were bare euhedral olivine grains—the late olivine condensed directly, without any intermediate liquid, which may account for the patterned surfaces which Olsen and Grossman (1974) compared to terrestrial sublimate olivine. In such cases, glass inclusions would of course have to date back to the time the olivine was still hosted in the partially molten chondrule. Thus, IOG experienced gas–melt (or gas–solid) interaction as isolated objects. In that sense, condensation played a role, after all, in IOG formation.

Further evidence for a hot formation of IOG is afforded by compound objects, either between IOG (e.g., Figs. 3E and 3F) or with chondrules (e.g., Fig. 1G), which require collisions in a plastic state (Gooding and Keil 1981; Akaki and Nakamura 2005). Some bona fide chondrules with coarse olivine phenocrysts may have engulfed them during cooling—that is, be “enveloping” compound chondrules in the parlance of Wasson et al. (1995). To be sure, many IOG are mere misshapen olivine (±mesostasis, pyroxene) debris, but even those may sometimes be fragments of hot-formed IOGs (e.g., the IOG “half” NWA 5958-4 IO13; Figs. 3C and 3D), or have been fragmented hot, if enstatite now surrounds them (e.g., NWA 5958-4 IO1 in supporting information). We note that Jones (1992) did envision isolated olivine grain formation from fragmentation of “macroporphyritic chondrules,” although the formation of the latter was left unexplained.

We conclude from enstatite rimming and concentric CL zoning in many IOG (Figs. 3 and 4) that they formed as stand-alone high-temperature objects. In that sense, we deem it warranted to also call them “chondrules” (sensu lato, e.g., Jacquet et al. 2012; Jacquet and Marrocchi 2017), whenever the distinction is not of interest, as they are part of the same continuum of objects (with the word “chondrule” being already applied to a diverse suite of objects: ferromagnesian, aluminum-rich, chromite-rich, etc., see e.g., Connolly and Desch 2004). This does not exclude the possibility that some isolated olivine grains in chondrites have different origins, for example, as AOA or a forsterite-bearing CAI fragment, although no evidence (e.g., isotopic) to that effect has been found for those of this study. To avoid any confusion, we remind the reader that the expression “bona fide chondrule” in this paper continues to refer to a non-IOG chondrule.

**Isolated Olivine Grains as Chondrule Splashes**

Since IOG are (by definition) olivine-dominated, they cannot be of chondritic composition, if only because of the Mg/Si (atomic) ratio (2 in endmember forsterite versus 1.02 for solar abundances, Lodders 2003). This undercuts the Jacquet and Marrocchi (2017) effort to explain the IOG olivine incompatible element enrichment relative to “mainstream” type I chondrules in terms of varying olivine subtraction from a given parental melt. Most likely, the IOG precursors oversampled olivine. We must thus attempt to identify their nature.

Given the general analogy of chondrule relict grains with AOA olivine (Marrocchi et al. 2018, 2019), a first candidate would be simply AOAIs. However, melting experiments of AOA analogs by Whatdam et al. (2010)
produced porphyritic textures. Also bulk AOA composition, although deficient in silica relative to type I chondrules, is closer to the latter than IOG (because of their CAI-like inclusions). Perhaps, though, a fragment of the olivine portion of a compact AOA (the dominant AOA texture in CM and related chondrites; Krot et al. 2004; Jacquet et al. 2016) would do. However, the (polycrystalline) olivine parts rarely extend beyond a few tens of µm in extent (e.g., Krot et al. 2004; Komatsu et al. 2015), at variance with the large IOG sizes. Furthermore, the abundance of AOAs overall in the studied chondrites is too low (~1 vol%) to account for the abundances of magnesian IOG (2–3 vol%). Indeed, the substantial matrix fraction in carbonaceous chondrites indicates that only a minority of their matter, and in particular of their AOAs, has been converted in chondrules (Marrocchi et al. 2019).

A second precursor candidate that comes to mind would be olivine phenocrysts from previous generations of chondrules. Now, if the chondrule/matrix ratio of carbonaceous chondrites, in particular CM-related chondrites, can be again taken as a measure of the extent of chondrule formation, chondrule-to-chondrule recycling must have been limited. Assuming chondrule-forming events were independent, Marrocchi et al. (2019) calculated that a fraction of 12% of chondrule material was inherited from earlier chondrules (sensu lato) in NWA 5958 (the same formula would yield 19% for Allende and 26% for ALHA 77307). At face value, this would, when multiplied by the chondrule abundance, approximate the modal abundance of IOG. However, it is unlikely that the recycled chondrules were preferentially in the form of free-floating olivine phenocrysts when they were remelted. This is because, again, fragmentation would not necessarily liberate bare olivine phenocryst (Olsen and Grossman 1978), and presumably, before the purported second chondrule-forming event, such olivine grains would have mixed

Fig. 10. Probability density function of Δ^{17}O for IOG and coarsest chondrule olivine, compared to host and relict data compiled by Marrocchi et al. (2019). Parts (A and B) compare data within chondrite clans (CM/CO and CV, respectively) and (C and D) compare chondrule and IOG data across the clans. (Color figure can be viewed at wileyonlinelibrary.com.)
with chondritic dust, such as rim high-temperature objects in CM chondrites (Metzler et al. 1992), erasing their olivine-dominated character most of the time.

Separating olivine crystals from the mesostasis would be conceivable if the mesostasis was liquid, that is during chondrule formation. The crystals and liquid could separate (to some extent) owing to buoyancy during sudden accelerations. Such could be caused by disruptions under strong headwind (Kato et al. 2006) or more generically by chondrule-chondrule collisions, producing splashes of melt and crystals, with varying proportions thereof, the IOG representing the olivine-dominated end of the spectrum. Some crystals may have been fragmented, either during initial splashing or subsequent collisions, and the latest ones not rounded up by further olivine crystallization may account for the misshapen IOG (devoid of euhedral/subhedral shape and concentric CL). Collisions among hot chondrules must have been relatively frequent as a few percent of chondrules are compound (Gooding and Keil 1981; Wasson et al. 1995; Ciesla et al. 2004; Akaki and Nakamura 2005). A collisional origin during chondrule formation may explain the higher proportion of IOG among type II chondrules as the higher concentrations of solids inferred for their formation regions (e.g., Schrader et al. 2013; Tenner et al. 2015, 2017) would promote collisions, although their rate also depends on relative velocities and the time in plastic state (which may have been shorter; Jacquet et al. 2015). Incidentally, the overrepresentation of type II chondrules among IOG might explain the dominance of ferroan olivine in Wild 2 terminal particles (13 versus 8; non-AOA-...
like) forsterites in the compilation of Defouilloy et al. (2017), at variance with type I chondrule dominance in carbonaceous chondrites. Indeed, if chondrule-forming regions did not extend to the accretion reservoir of this comet, the latter may have received some of their products by aerodynamic transport, but these would be biased toward the smaller fragments, less easily decoupled from the gas (Jacquet 2014).

Although the chemical and isotopic properties of IOG overlap with bona fide chondrules, their large size compared to the phenocrysts of the latter might lead us to question whether they derive from the same chondrules rather than some largely lost population. However, IOG represent 5–13% of type I chondrules sensu lato (Table 1), and if correctly interpreted as chondrule splashes, are only the tip of the iceberg: One would need to add isolated pyroxene grains, some cryptocrystalline chondrules (melt-dominated splashes), and larger fragments with more representative silicate/melt ratios, which, if subsequently relaxed to some extent to spherical shapes, would be indistinguishable from unsplashed chondrules. Hence, the IOG source chondrules have to represent a few tenths at least of the whole type I chondrule inventory in carbonaceous chondrites. If we are to allow a significant population of non-IOG-related chondrules, we need to assume near-complete destruction of the IOG source chondrules, and very little splashing of the others, that is, very different collisional histories despite the thermal history similarity suggested by petrography. It seems simpler to assume that they formed in the same regions and that collisions induced about a tenth of the chondrule matter to be splashed out in the form of IOG.

If so, how can we then explain the large size of IOG? The PO chondrules studied by Marrocchi et al. (2018, 2019) and Libourel and Portail (2018) may offer a clue. Indeed, they are surrounded by thick (∼100 μm) palisades of olivine that asymmetrically grew toward the exterior, presumably as a result of Mg and Si addition from the gas (Figs. 1C and 1D; Marrocchi et al. 2018, 2019). Now, an isolated olivine expelled from its parent chondrule in its inventory from the parent chondrule would have interacted with the gas on all sides (as inferred in the Isolated Olivine Grains Formed Hot section), with no competing crystal around, so may well have hereby attained a diameter double of that of the palisadic olivine grains, about as observed (Fig. 2). Roughly speaking, the refractory (bright CL) core of the IOG might correspond to the olivine crystallized in the parental chondrule and the margin (if concentric) to that formed after splashing, but diffusion (for Ca) after splashing as well as recondensation onto the chondrule before would make the actual boundary between these two stages uncertain.

Later, as mentioned in the previous subsection, the olivine would have reacted with SiO and have been replaced by enstatite to varying extent. Some objects may hence have lost their olivine-dominated nature: This may explain the largest isolated pyroxene grains seen by Jones (1999) in ALHA 77307 even though some isolated pyroxene grains may be merely surficial sections of pyroxene-armored IOG. Yet others could of course have been expelled as pyroxene grains, after pyroxene crystallization had commenced in chondrules. At any rate, Jones (1999) deemed derivation of isolated pyroxene grains from chondrules likely from textural and mineral chemical similarities with them, but a dedicated study beyond her work only published in abstract form would be desirable.

The emerging scenario of IOG formation is sketched in Fig. 12.

**Refractory Forsterites and Diversity among Type I Chondrules in Carbonaceous Chondrites**

We have yet to understand refractory forsterites, that is, those CaO-rich IOG studied by, for example, Steele (1988), Weinbruch et al. (2000), and Pack et al. (2005). CaO enrichment is also seen in the above-mentioned palisadic olivine grains in PO chondrules, which certainly would not have been the least prone to expulsion upon collisions. Marrocchi et al. (2018, 2019) explained the Ca, Al, Ti-enrichment in those by precipitation out of a Ca-Al-Ti-rich melt. Indeed, in a reservoir with dust/gas ratio below unity, the melt upon olivine crystallization should be fairly refractory because of evaporation (Ebel and Grossman 2000; Ruzicka et al. 2008). The palisadic olivine grains would have escaped diffusional resetting of their Ca after recondensation because of their size, the same would hold a fortiori for the large isolated refractory forsterites. This incidentally constrains the diffusion length of Ca to a few tens of microns, hence the timescale of days inferred by Marrocchi et al. (2018, 2019).

It remains to be understood why high refractory element contents are restricted to the $^{16}$O-rich ($\Delta^{17}$O < $-4\%$) population of IOG or chondrules at large, as observed by Jacquet and Marrocchi (2017). These authors inferred protracted cooling to allow equilibration of the olivine with a late, incompatible element-enriched melt, but their closed-system olivine crystallization reasoning failed to recognize, as above, that the initial melt was incompatible element rich. Ushikubo et al. (2012), Schrader et al. (2013), and Tenner et al. (2015, 2017) noted that $^{16}$O-poor chondrules tended to have more ferroan olivine than the $^{16}$O-rich counterparts, which they ascribed to greater solid/gas ratios in the chondrule-forming regions (with the solids [dust and ice] being $^{16}$O-depleted). $^{16}$O-poor type I IOG do not, however, present such a FeO enrichment, but their large size may have prevented diffusion of Fe$^{2+}$ into their core
during cooling. A higher solid/gas ratio would lead to lower refractory element concentrations in olivine at the onset of its crystallization and an upturn in the CaO versus FeO trend (e.g., Ebel and Grossman 2000), as observed (Fig. 8A).

In type I porphyritic chondrules, moderately high fayalite contents (above ~ 2 mol%) seem correlated with pyroxene proportion: The three PP chondrules in CR3 chondrites studied by Tenner et al. (2015) have Mg# \(\approx 100 \times \frac{Mg}{(Mg + Fe)}\) of 94.2–97.8, with no PO in this range although it comprises a majority of their type I chondrules; the four PP chondrules of Lewis Cliff 85332 (C3-ung, CR-related) analyzed by Wasson et al. (2000) have Fs4.2-9. Outside carbonaceous chondrites, type IAB and IB chondrules in Semarkona (LL3.0) span F\(_s\) 1.8–6.7 in the study of Jones (1994). No Mg# below 98.4 (nor \(\Delta^{17}O = /C0_{3.55}\)) is reported in any type I chondrule from the CV3 Kaba and NWA 8613 analyzed by Hertwig et al. (2018, 2019), although they include POP and PP chondrules, same for the Murchison (CM2) chondrules studied by Chaumard et al. (2018), save for one granular olivine pyroxene (GOP) chondrule with Mg# = 96 (\(\Delta^{17}O = −3.55\%_{oo}\)) is reported in any type I chondrule from the CV3 Kaba and NWA 8613 analyzed by Hertwig et al. (2018, 2019), although they include POP and PP chondrules, same for the Murchison (CM2) chondrules studied by Chaumard et al. (2018), save for one granular olivine pyroxene (GOP) chondrule with Mg# = 96 (\(\Delta^{17}O = −3.55\%_{oo}\)). The situation of the Yamato 81020 (CO3.0) study by Tenner et al. (2013) is more marginal, with the 3 PP having Mg# of 97.8–99 (all \(^{16}O\)-rich), while all three type I chondrules with Mg# <97 are POP and \(^{16}O\)-poor. Of course, the estimate of the true proportions of pyroxene is hampered by sectioning artifacts (Hezel and Kießwetter 2010). Still, if there is indeed some link between “ferroan forsterite” and pyroxene abundance, this may indicate that the chondrules in question cooled more slowly, since pyroxene should appear at lower temperature than olivine (e.g., Ebel and Grossman 2000). Indeed, in Vigarano, the most pyroxene-rich chondrules seem to have the lowest proportion of clinoenstatite in low-Ca pyroxene, indicative of slower cooling at least around 1000°C (Brechtley et al. 1993; Soulé 2014).

Thus, the \(^{16}O\)-rich type I chondrules and the associated refractory forsterites may have cooled more rapidly, under lower solid/gas ratios, than their generally more ferroan \(^{16}O\)-poor counterparts. Are these two subtypes of type I chondrules derived from different sources? Or were two styles of type I chondrule formation events overlapping in individual reservoirs? The \(^{16}O\)-poor variety is more prevalent in CR chondrites than in other carbonaceous chondrites: Keeping \(\Delta^{17}O = −4\%_{oo}\) as our arbitrary boundary between the two, Tenner et al. (2015) found a 10:35 ratio between \(^{16}O\)-rich and \(^{16}O\)-poor type I chondrules in CR3 chondrites; the same research group obtained a ratio of 19:4 in the CO3.0 chondrite Y 81020 (Tenner et al. 2013), 23:2 in the CM2 chondrite Murchison (Chaumard et al. 2018), and 71:7 in CV chondrites (Rudraswami et al. 2011; Hertwig et al. 2018, 2019). One could envision that \(^{16}O\)-poor type I chondrule formation was merely more frequent in the CR chondrite reservoir without denying \(^{16}O\)-poor type I chondrule formation elsewhere. However, \(^{16}O\)-poor chondrules appear systematically more \(^{54}Cr\)-rich than their \(^{16}O\)-rich counterparts in both CR and CV chondrites (e.g., Williams et al., 2020; Schneider et al. 2020). This indicates a different reservoir of origin for the \(^{16}O\)-poorer type I chondrule, which presumably was spatially and/or temporally closer to the accretion event of CR chondrites. We note that CR chondrules exhibit initial \(^{26}Al\) abundances lower than their counterparts in CO and CV chondrules (Nagashima et al. 2014;
Schrader et al. 2017; Tenner et al. 2019), but possibly comparable to cometary samples (Ogliore et al. 2012; Nakashima et al. 2015). The Fe/Mn ratio of olivine in type II chondrules, lower in CR chondrites than in other carbonaceous chondrites (e.g., Berlin et al. 2011; Jones 2012; Frank et al. 2014), also sets the CR chondrule population apart among carbonaceous chondrites.

Hence, the emerging picture is that of a CR-like chondrule population, with apparently higher solid/gas ratios and slower cooling than the others (for the type I chondrules), which contributed to non-CR reservoirs dominated by $^{16}$O-rich chondrules. Such a limited mixing of chondrules beyond their formation region could be reproduced in the simulations of Goldberg et al. (2015). This limited mixing would also have involved refractory inclusions, as suggested by Al-Mg isotopic evidence (Larsen et al. 2020). While the $^{16}$O-poor type I chondrules of different carbonaceous chondrite groups (CO, CV, CM) may thus come from a single source, the $^{16}$O-rich ones, although O isotopically similar, may still have diverse origins, as suggested by distinctive petrographic features in the different chemical groups (e.g., size, prevalence of fine- or coarse-grained rims, occurrence of primary feldspar, and so on; see Jones 2012).

**CONCLUSION**

We have carried out microscopic, cathodoluminescence, chemical, and O isotopic measurements of type I isolated olivine grains (IOG) in the carbonaceous chondrites Allende (CV3), Northwest Africa 5958 (C2-ung), Northwest Africa 11086 (CM-an), and Allan Hills 77307 (CO3.0).

The IOG typically represent a few percent of the studied carbonaceous chondrites, and are dominated by type I IOG but with an overrepresentation of type II compositions. The type I IOG present O isotopic signatures similar to bona fide chondrules, although they are generally coarser than chondrule phenocrysts. CaO, which may reach up to 0.9 wt% in $^{16}$O-rich IOG, as well as Al$_2$O$_3$ and TiO$_2$ decrease from core to rim, while FeO, MnO, Cr$_2$O$_3$ increase. EMP traverses and CL imaging often reveal concentric zoning, with enstatite frequently rimming the olivine's outer edge.

The general isotopic and chemical similarities, indeed the textural continuum, with bona fide chondrules indicate that the IOG were derived from them. Their not uncommonly unbroken morphology and evidence of interaction with the gas, with recondensation on all sides possibly accounting for their large size, indicate that rather than being cold fragments of chondrules, they were expelled from chondrules while these were molten, likely during chondrule-chondrule collisions. Among IOG, the refractory forsterites retained their high-temperature composition presumably established by equilibration with a refractory melt, as some palisadic olivine grains in PO chondrules. These $^{16}$O-rich type I chondrules may have undergone quicker quenching than their $^{16}$O-poorer (type I) counterparts, possibly derived from a CR chondrite-like reservoir.

Thus, IOG were likely derived from chondrules, as favored by most recent authors (e.g., Jones 1992; Jones et al. 2000; Pack et al. 2005; Russell et al. 2010), but were affected by significant gas-solid interactions (before and after expulsion from the parent chondrules), as inferred by their earliest students (e.g., Fuchs et al. 1973; Olsen and Grossman 1974, 1978; Steele 1986, 1988, 1989), hereby reconciling the two endmembers of the isolated olivine literature.

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**Editorial Handling**—Dr. Timothy Fagan

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**SUPPORTING INFORMATION**

Additional supporting information may be found in the online version of this article.

**Table EA.1** Oxygen isotope compositions of chondrule coarsest olivine and isolated olivine grains in this study.

**Table EA.2** Size of chondrule coarsest olivine and isolated olivine grains in this study.

**Table EA.3** Chemical composition of chondrule coarsest olivine and isolated olivine grains in this study.