Spectroscopy Letters
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/lstl20

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Available online: 17 Oct 2011

To cite this article: Henner Busemann, Nicole H. Spring, Conel M. O'D. Alexander & Larry R. Nittler (2011): Raman Spectroscopy on Cometary and Meteoritic Organic Matter, Spectroscopy Letters, 44:7-8, 554-559

To link to this article: http://dx.doi.org/10.1080/00387010.2011.610425

Spectroscopy Letters, 44:554–559, 2011

ISSN: 0038-7010 print/1532-2289 online
DOI: 10.1080/00387010.2011.610425

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Raman Spectroscopy on Cometary and Meteoritic Organic Matter

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ABSTRACT Extraterrestrial organic matter (OM) is an important component of meteorites that originate from asteroids and from cometary dust collected from comet Wild 2 and the Earth's atmosphere. Some of this OM was incorporated into the first planetary bodies during the formation of the solar system, 4.56 Ga ago, and did not experience significant alteration. Here, we summarize our efforts in the comparative C Raman spectroscopy of this OM. We aim for the identification of the most primitive OM that might be of solar nebula origin and a better understanding of the modifying processes on parent bodies, during transport in space and sample collection. We (i) determine the degree of order of meteoritic OM and hence the metamorphism experienced on their parent bodies; (ii) assess potential effects during sample collection of comet Wild 2 dust, and (iii) compare possibly cometary interplanetary dust particles (IDPs) with primitive meteorites.

KEYWORDS asteroids, carbon Raman spectroscopy, comets, interplanetary dust particles, meteorites, organic matter, Raman imaging, Stardust, thermal metamorphism

INTRODUCTION

The identification and characterization of the most pristine extraterrestrial materials available are essential for a better understanding of the origin and evolution of our solar system. This material has survived since the formation of the first solids that accreted 4.56 Ga ago. Comets and undifferentiated asteroids in the outer asteroid belt, sampled by meteorites, may best preserve such primitive matter that possibly escaped significant parent body processing. The effects due to parent body alteration, transport in space, entry into the Earth's atmosphere, terrestrial weathering, and sample collection must be identified to assess the primitiveness of any extraterrestrial material and to understand its evolution through the history of the solar system. The effects can be best studied by comparing meteoritic and cometary materials that experienced different degrees of alteration on their parent bodies and reached the Earth through different pathways.

Organic matter is an abundant primordial component in the protosolar cloud. It can be traced spectroscopically in the interstellar medium and in

This submission was presented during the CORALS-2 Meeting on Micro-Raman Spectroscopy and Luminescence Studies in the Earth and Planetary Sciences, which was held between May 19 and 21, 2011, in Madrid, Spain. This is an invited paper for a special CORALS-2 GEO-SPECTROSCOPY issue of *Spectroscopy Letters*.

Received 1 July 2011; accepted 25 July 2011.

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comets and can be found in the laboratory in essentially all pristine extraterrestrial samples.

In this contribution, we summarize our efforts in the comparative C Raman spectroscopy of extraterrestrial organic matter (OM).[1–5] Our samples comprise (i) insoluble OM (IOM) extracted from meteorites originating from asteroids;[6] (ii) interplanetary dust particles (IDPs),[7] typically ≤100 μm–sized particles collected by NASA aircraft in the stratosphere, >80% of which may sample Jupiter-family comets (JFCs);[8] and (iii) JFC Wild 2 dust returned with NASA's Stardust mission.[9]

MATERIALS AND METHODS

1. Meteoritic OM (typically a few weight-percent) is found in the most primitive members of all chondrite classes. Fewer than 25% are soluble in water or organic solvents.[10] IOM is separated by demineralization of the main chondritic silicates and the removal of solvent–soluble OM.[11] IOM is not completely characterized yet. It consists of macromolecular networks of aromatic molecules interlinked with short aliphatic chains and shows variable abundances of heteroatoms N, O, and S.[6,12] IOM in the most primitive meteorites (and IDPs) carries isotope anomalies in H, C, and N that could be a record of chemical processing in the interstellar medium or cold protoplanetary disk.[13–15] All meteorite parent bodies may have incorporated the same mixture of OM.[6] Parent-body aqueous alteration and thermal metamorphism may then have altered this OM, causing, for example, carbonate formation, graphitization, and the (partial) loss of aliphatic chains and isotope anomalies.[6,16,17] Particle irradiation in space or radioactivity may have induced some and amorphization of the OM.[15,18,19]
2. Primitive IDPs (of the "chondritic-porous" type) often show C abundances (typically ~10%[7]) larger than do meteorites and extreme bulk and localized micro-meter-sized isotope anomalies.[2] OM in IDPs is in general similar to but less altered than meteoritic OM.[2,20]
3. Organics in comet Wild 2 "Stardust" is—despite the harsh conditions during collection—remarkably chemically complex[21] and similar to OM in IDPs.[22] Wild 2 OM collected in aerogel shows less pronounced isotopic anomalies and presolar grain abundances than that found in meteoritic IOM and IDPs.[5]

In addition to macromolecular OM, primitive meteorites, IDPs, and Stardust contain (at particles-per-meter level) submicron-sized amorphous objects called "nanoglobules" of possibly interstellar origin[23,24]; graphitic whiskers[25]; and diamonds, SiC, and graphite that formed around previous generations of stars and carry isotopic signatures of stellar nucleosynthesis.[e.g., 26, and references therein] Presolar diamond separates, and single graphite and SiC grains have been analyzed by Raman spectroscopy[e.g., 27–29] and micro-Raman imaging of IOM is capable of detecting embedded graphite (Fig. 1) as well as comparably amorphous grains.[1,30]

Here we discuss general C Raman characteristics of disordered to microcrystalline macromolecular OM. We will focus on averaged Raman results of meteorites, IDPs, and Stardust and correlate them with other observations, such as the presence of isotope anomalies or thermal metamorphism experienced on meteorite parent bodies.

Most data were obtained in image mode with a scanning micro-Raman WiTec α-SNOM with 532 nm exciting laser.[e.g.,31] Experiments on meteoritic IOM with other laser wavelengths (514 and 632 nm)[30,32] show significant dispersion of the D and G bands, which can be accounted for by comparing the results and shifting the band positions in the data sets accordingly by a few wave numbers.[e.g., 4,32] Here

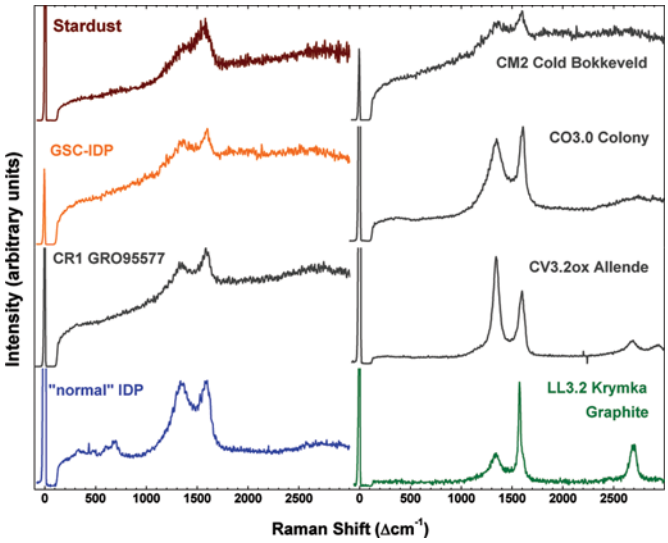

FIGURE 1 Raman spectra of meteoritic IOM, IDPs, and comet Wild 2 "Stardust."[1–5]. (color figure available online.)

we focus on data obtained with a 532 nm laser alone. Typical spatial resolution of the Raman mapping was ~0.4 μm, integration times were 3–4 s, and the laser power was kept at ≤55 μW.[1,2,4] Tests with the WiTec instrument with various laser powers applied to meteoritic IOM of different degrees of disorder showed no significant systematic modification in the C Raman parameters.[1] However, our most recent experiments indicate that C Raman parameters depend to a small degree but systematically on the chosen laser power.[32] These laboratory-induced effects are small compared to the observed natural trends (discussed below).[32] D and G bands were fitted to 2 Lorentzian profiles and a free floating linear background. Spectra with indication of fluorescence (maximum/minimum background variation between 600 and 3000 $cm^{-1} > 15\%$) were discarded. Details are described elsewhere.[1]

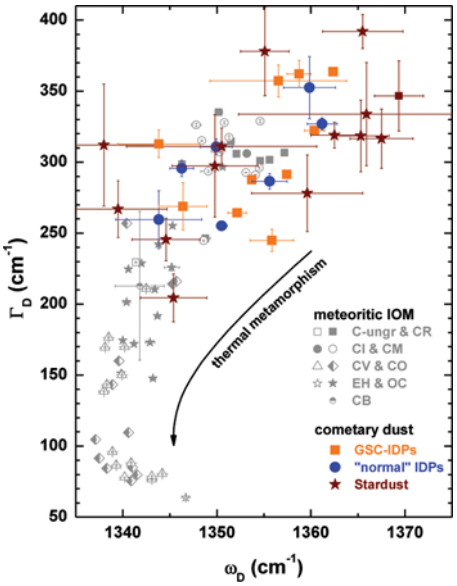

FIGURE 2 D band width and positions of meteoritic IOM, IDPs, and comet Wild 2 "Stardust."[1–5]. (color figure available online.)

RESULTS

Typical spectra of meteoritic IOM extracts, IDPs, and comet Wild 2 dust OM are shown in Fig. 1. Most spectra only show the carbon D and G bands. IOM obviously only occasionally contains other, non-C Raman active phases, such as oxides that survive demineralization.[1] However, for IDPs and Stardust, the frequent lack of non-C Raman features is surprising, as cometary phases such as sulfides and silicates are present.[7,9] This probably reflects the non-C grains being coated and held together with OM[33] and C being a strong opaque adsorbent with a particularly large Raman response.[34,35] Amorphous and very disordered C shows broad overlapping bands, while increasing order and graphitization cause the bands at first order to sharpen. This is visible in Fig. 1, showing spectra in the order of increasing order. D and G band parameters will be used here to assess the degree of order of the OM and hence the metamorphism experienced, much as in studies on terrestrial materials.[e.g., 36–39]

Trends in Meteorites

D band parameters for averaged meteoritic IOM of various classes from distinct parent bodies are shown in Fig. 2. Meteorites are classified based on their chemical, isotopic, and mineralogical compositions,[40] which reflect, for example, thermal and aqueous alteration, impact heating, the survival of primordial nebula materials, the prevailing O fugacity, and temperature or heliocentric distance of the region where the parent body accreted. CR, CI, and CM chondrites as well as the most primitive ordinary chondrite (Semarkona) show very similar C Raman parameters. These are considered the least thermally altered meteorites. In contrast, CO, CV, and OC chondrites that experienced thermal metamorphism, with assigned petrologic types from 3.0 to 3.7,[40] show a much more ordered OM. These trends are also correlated with the N/C, H/C, and less pronounced isotopic compositions of the IOM (not shown).[1] All these properties are sensitive to thermal metamorphism.[1,6] Hence, as applied first systematically in studies by Bonal et al.,[41,42] Raman band parameters can be used to assess the petrologic subclassification and the experienced peak metamorphic temperatures (not shown)[1] that have traditionally been determined by other means, for example, noble gas concentrations or mineral compositions.[43] Figure 3 shows the D band width as a function of petrologic type for three meteorite classes.[1] However, as demonstrated in Fig. 3, the petrologic subclassification cannot be directly compared between meteorites from different chemical classes,[41,42] as conditions—such as O fugacity, presence of water, closed/open system alteration, chemical environment, and depth within the parent body

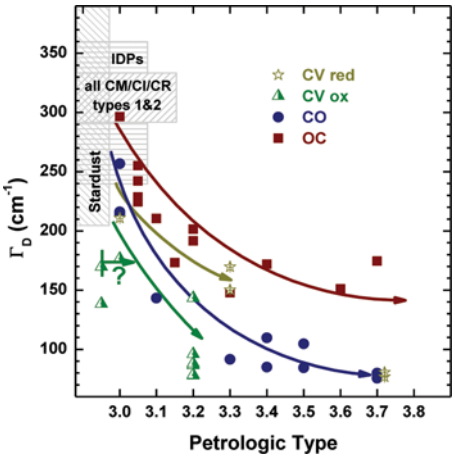

FIGURE 3 D band widths as a function of petrologic type for type 3 meteorites and of the range in IDPs and comet Wild 2 "Stardust."[1–5]. (color figure available online.)

and heating timescales—will be different for CV, CO, and ordinary chondrites. Aqueous alteration (increasing from types 3.0 to 1) does not affect the C Raman characteristics.[1]

Trends in IDPs

Figure 2 also shows data for several IDPs compared to our averaged meteoritic IOM data. A more detailed discussion of our results on a larger set of IDPs is in preparation. IDPs and meteoritic IOM cover the same space, supporting the idea that both asteroidal bodies in the inner solar system and comets in the outer solar system acquired similar OM, albeit in different amounts.[e.g., 6] OM in some IDPs shows a particularly disordered character, as observed already earlier, possibly due to irradiation in space.[e.g., 15,34,44] A subset of IDPs was recovered in a timed collection during Earth's passage through comet Grigg-Skjellerup's dust stream (GSC), increasing the probability of having sampled fresh dust from a known cometary source.[45] Data from GSC-IDPs and IDPs not associated with a specific dust stream are similarly distributed. This shows that exposure to irradiation during transport in space (typically 1–100 kyr) does not systematically alter C Raman characteristics. On average (not shown), C Raman properties of IDP OM are similar to the averages for primitive meteoritic IOM, supporting the idea of a common reservoir of OM for both asteroids and comets. However, certain particularly primitive GSC-IDPs with features such as extremely high D enrichments and presolar grain abundances[2] or the presence of the previously unknown mineral Brownleeite[46] also show the most disordered C Raman parameters.

Trends in Cometary Stardust

Wild 2 material was decelerated from an impact velocity of 6.1 km/s within a few millimeters to centimeters of aerogel and could have suffered flash heating, evaporation, ablation, and complete to partial melting. OM might have experienced peak temperatures of >1000 K for a short period (~1 μs).[47] Tests on carbonaceous Stardust analogues shot into aerogel show that the capturing process causes significant changes on the surfaces of the grains.[48] Both crystallization of disordered C and amorphization of graphitic C occur. However, the presence of isotope anomalies[49] and a large variety of organic species in the actual Stardust [e.g., 5,21,22] indicates that some pristine cometary material survived the capture. Raman spectroscopy is capable of assessing the degree of alteration this cometary OM experienced.[4,5] Figure 2 shows the D band parameters of Wild 2 OM. The data points cover the same range defined by primitive chondrite IOM and IDPs, indicating that even severe heating during capture, albeit for only extremely short periods, did not alter Wild 2 OM to the same extent as prolonged thermal metamorphism on the CV, CO, and OC chondrite parent bodies (Fig. 2). Similarly, the Stardust G band parameters (not shown) exhibit a large spread over the whole parameter space determined by meteoritic IOM, indicating both particularly (hydrogenated) amorphous material,[50] but also relatively graphitic OM,[4] similar to thermally metamorphosed OM in chondrites.

DISCUSSION

Comet Wild 2 OM collected by the Stardust mission shows pristine characteristics comparable to IDPs, despite having been collected under less harsh conditions. While the collection process inevitably altered the material, cometary signatures, such as particularly disordered C correlated with other primitive characteristics, are preserved, rendering Stardust samples the most important cometary materials available as their provenance is known. The Raman study

of meteoritic IOM can be used to assess petrologic type and rough peak metamorphic temperatures. Raman characteristics of OM from asteroids and comets are on average very similar. However, individual cometary IDPs showing particularly primitive Raman signatures, indicating highly disordered macromolecular or amorphous material, often show the most extremely primitive characteristics. Highly disordered C Raman features in OM are consistent with irradiation in space.

ACKNOWLEDGMENTS

The authors are grateful for support by STFC (H.B. and N.S.) and NASA (L.N. and C.A.). The authors also thank J. Garcia Guinea and A. Guscik, organizers of the Madrid CORALS II meeting.

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