

## A FLUORESCENT AEROGEL FOR CAPTURE AND IDENTIFICATION OF INTERPLANETARY AND INTERSTELLAR DUST

GERARDO DOMÍNGUEZ<sup>1</sup> AND ANDREW J. WESTPHAL  
Space Sciences Laboratory, University of California, Berkeley, CA 94720

MARK L. F. PHILLIPS  
Pleasanton Ridge Research Corporation, Hayward, CA 94542

AND

STEVEN M. JONES  
Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109  
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### ABSTRACT

Contemporary interstellar dust has never been analyzed in the laboratory, despite its obvious astronomical importance and its potential as a probe of stellar nucleosynthesis and Galactic chemical evolution. Here we report the discovery of a novel fluorescent aerogel that is capable of capturing hypervelocity dust grains and passively recording their kinetic energies. An array of these “calorimetric” aerogel collectors in low-Earth orbit would lead to the capture and identification of large numbers of interstellar dust grains.

*Subject headings:* astrochemistry — dust, extinction — instrumentation: detectors — interplanetary medium — meteors, meteoroids — techniques: image processing

### 1. INTRODUCTION

Interstellar dust is an important component of the interstellar medium. Dust dominates the opacity from the far-ultraviolet through the far-infrared and hence controls the spectral appearance of most interstellar objects. Because of dust shielding against dissociating FUV radiation, molecules can form in dense clouds, which allows cooling to low temperatures and thus, eventually, allows gravity to overwhelm pressure support and the formation of new stars. Small dust grains also dominate the heating of the interstellar gas through the photoelectric effect and hence controls the structure of the interstellar medium. Despite some 50+ years of active research, the composition of interstellar dust is still largely guessed at. In essence, our ignorance reflects the difficulty of inferring dust composition from remote astronomical observations. Here we propose a novel collection agent that allows the discriminatory collection of interstellar grains and separation from solar system debris. This promises to open up a new window on the solid component of the interstellar medium.

Although it is known that interstellar (IS) dust penetrates into the inner solar system (Grün et al. 2000; Taylor, Baggaley, & Steel 1996), to date not even a single contemporary IS grain has been captured and analyzed in the laboratory. Using sophisticated chemical separation techniques, certain types of refractory ancient IS particles (so-called presolar grains) have been isolated from chondritic meteorites (e.g., Amari & Zinner 1997). Isotopic abundance patterns within these individual grains often differ wildly from solar system values, and point to the formation of these grains in specific astrophysical environments such as supernova ejecta and the winds of asymptotic giant branch (AGB) stars. However, because only the most chemically robust particles (e.g., graphite, SiC, Al<sub>2</sub>O<sub>3</sub>) survive the harsh

chemical separation, this sample is extremely biased, and it is unlikely that these particles are typical of those found in the interstellar medium. A sample of IS dust collected by spacecraft in the inner solar system would be less biased, and could lead to the first laboratory characterization of the “typical” IS dust particle. Furthermore, such a sample would allow us to detect isotopic, elemental, and mineralogical differences between dust in the protosolar cloud and dust currently residing in the local ISM, and to probe Galactic chemical evolution over the 4.6 Gyr since the formation of the solar system. Presolar grains are already proving to be a valuable probe of Galactic chemical evolution and stellar nucleosynthesis (Amari et al. 2001a; Nittler et al. 1997).

The vast majority (~84%) of large (>1 μm) ancient presolar grains appear to be of one type, so-called mainstream SiC grains. These grains are enriched in <sup>22</sup>Ne, show *s*-process signatures in Kr and Xe, and probably originate in the outflows of AGB stars. Grains from other astrophysical sites have been identified but are relatively rare (e.g., Type A, B SiC, tentatively identified with J-type carbon stars [Amari et al. 2001b], 3%–4%; Type X SiC from supernovae [Amari & Zinner 1997], 1%; and alumina from high-metallicity red giants, <0.5%.) A few presolar grains show isotopic patterns that are unique among the thousands that have been studied so far (Nittler et al. 1997). If dust in the local ISM shows a similar pattern of diversity, with a dominant common type and relatively rare populations of exotic grains, a large-statistics collection technique will be required to capture, identify, and study contemporary IS dust grains from a wide variety of astrophysical sources.

Aerogels are extremely low density solids whose superiority as capturing media for hypervelocity ( $v > 0.5 \text{ km s}^{-1}$ ) grains has been well established (Barrett et al. 1992; Hörz et al. 2000; Kitazawa et al. 1999). A prominent example is the use of silica aerogel as the collecting medium for cometary and interstellar grains on NASA’s *Stardust* mission (Brownlee et al. 1997). Aerogel collectors have been deployed in low-Earth orbit, but severe background from

<sup>1</sup> Department of Physics, University of California, Berkeley, CA 94720.

anthropogenic orbital debris has so far prevented the identification of more than a handful of interplanetary particles (Hörz et al. 2000). No interstellar particles have been identified so far. Since they are on hyperbolic orbits, extraterrestrial particles are faster than orbital debris, so could in principle be identified on that basis, but existing aerogels give little information on impact velocity. With this in mind, we have developed a novel calorimetric aerogel that passively records the kinetic energy of captured hypervelocity particles.

The capture of a hypervelocity dust particle in aerogel produces a shock wave that deforms, heats, and vaporizes the aerogel material in the vicinity of the projectile's trajectory, resulting in the formation of a permanent track. The correlation between captured projectile velocity and track characteristics (e.g., track length, track volume, etc.) is poor (Kitazawa et al. 1999). This behavior is expected theoretically (Anderson & Ahrens 1994; Westphal, Phillips, & Keller 1998; G. Domínguez 2003, in preparation). The amount of local heating, however, is nearly linearly proportional to the projectile kinetic energy (Anderson & Ahrens 1994). If this local heating alters some property of the aerogel in the vicinity of the track, then this property could be used as a calorimeter. We chose to focus on inducing a fluorescence signal.

## 2. OBSERVATION OF FLUORESCENCE FROM CAPTURE EVENTS

We have observed fluorescence resulting from the thermal alteration of aerogels previously in various doped aerogel systems, which fluoresce weakly in their amorphous state and strongly when baked at high temperatures ( $\approx 1000^\circ\text{C}$ ) for extended periods of time ( $\sim 1$  hr). A simple example of such a system is alumina aerogel doped with chromium (III). The amorphous, unheated phase is only very weakly fluorescent under UV illumination (254 or 365 nm). Heating the aerogel to  $1450^\circ\text{C}$  causes it to crystallize to the well-known luminescent phase  $\alpha\text{-Al}_2\text{O}_3\text{:Cr}$ , known in nature as ruby, which glows red ( $\lambda_{\text{max}} \approx 700$  nm) under UV illumination. More complex systems include alumina gels co-doped with Gd and Tb. Gd acts as a sensitizer by absorbing UV light at certain wavelengths and nonradiatively transferring energy to Tb, which emits at several wavelengths, principally in the green.

Local heating that results from the capture of hypervelocity projectiles is rapid and confined to small regions in the aerogel. However, the production of a fluorescent state as a result of rapid ( $t < 200 \mu\text{s}$ ), local heating (within  $< 100 \mu\text{m}$  of the particle track) in an aerogel has previously not been reported. To test whether local heating in an aerogel could induce an irreversible phase transformation into a fluorescent phase, the effects of hypervelocity projectile capture were first simulated by exposing samples of Cr-doped and (Gd, Tb)-doped alumina aerogels ( $\rho \sim 170 \text{ mg cm}^{-3}$ ) with a pulsed  $\text{CO}_2$  laser (300 Hz,  $50 \mu\text{m}$  spot size, pulse width =  $50 \mu\text{s}$ , power = 0.25–0.50 W). The energy per pulse is approximately the energetic equivalent of a glass sphere  $10 \mu\text{m}$  in diameter impacting at  $10 \text{ km s}^{-1}$ . Some of these aerogels displayed brilliant green fluorescence in the regions of local heating. This was encouraging evidence that the capture of hypervelocity dust particles could induce a fluorescent phase in alumina aerogels. These alumina aerogel samples were selected for shots with hypervelocity projectiles (a mix of

powdered meteorite and glass beads) at the Advanced Vertical Gun Range at NASA Ames Research Center. Two of these samples showed intense green fluorescence in the heated material surrounding the particle tracks, thus establishing that the phase transformation occurs in alumina aerogels. Quantitative measurements with these shots were precluded because of the large spread in particle sizes and the unknown effect of particle ablation. These shots were followed more recently, again at Ames, with projectiles consisting of a mixture of monodisperse glass spheres. This allowed us to do quantitative measurements of the fluorescence yield as a function of particle size and velocity.

## 3. ANALYSIS OF FLUORESCENCE OBSERVATIONS

We measured the fluorescence yield using a standard fluorescence microscope with a cooled color CCD video camera. The fluorescence was excited at 365 nm using a standard bandpass filter cube at the excitation side and imaged using a long pass filter ( $\lambda \geq 395$  nm). The samples were imaged within 2 hours of each other to minimize the effects of UV lamp intensity variations. High-resolution images of the aerogel surface where tracks entered were taken and the background fluorescence (weak and mostly blue) was subtracted as follows. A local blank region of aerogel was sampled, and the average ratio of green to blue,  $f_{\text{gb}}$  was determined; for each pixel we defined the net fluorescence in the green as:

$$I_{\text{green}}^{\text{net}} = I_{\text{green}} - \overline{f_{\text{gb}}} I_{\text{blue}}, \quad (1)$$

where  $I_{\text{blue}}$  is the blue pixel value. We chose this background subtraction method because a linear increase in both the green and blue channels would be expected, even in the absence of a phase transformation, because of the increased density of aerogel in the vicinity of the track mouth. We define the fluorescence yield as the sum of  $I_{\text{green}}^{\text{net}}$  for  $I_{\text{green}}^{\text{net}} > 2.5 \sigma$  above the pixel noise in the region surrounding the track mouth. The yield increases dramatically with increasing velocity within each particle population (Fig. 1). In Figure 2, we show the fluorescence yield as a function of kinetic energy. Over the range from 2 to  $20 \mu\text{m}$  (3 orders of magnitude in mass), the fluorescence yield appears to be consistent with being a single-valued function of the particle kinetic energy,  $I_g \propto E_k^{0.69}$ . We found that the exponent is insensitive to the choice of fluorescence signal-to-noise ratio threshold.

A reasonable model for the energetics of grain capture can be used to explain, at least qualitatively, the calorimetric aspects of the aerogel. In this model, we treat the aerogel as a fluid. In the limit of large Reynolds number, the energy deposited per unit path length by a grain of radius  $r$ , density  $\rho_g$ , and kinetic energy  $E$  is

$$\frac{dE}{dx} \sim \frac{3}{2} \frac{1}{r} \frac{\rho_a}{\rho_g} E = \frac{E}{\lambda}, \quad (2)$$

where  $\rho_a$  is the aerogel density, and

$$\lambda = \frac{2}{3} r \frac{\rho_g}{\rho_a}. \quad (3)$$

This stopping length scale agrees to within 10% of the value obtained following the more detailed treatment by Anderson & Ahrens (1994). The range of the particle in its

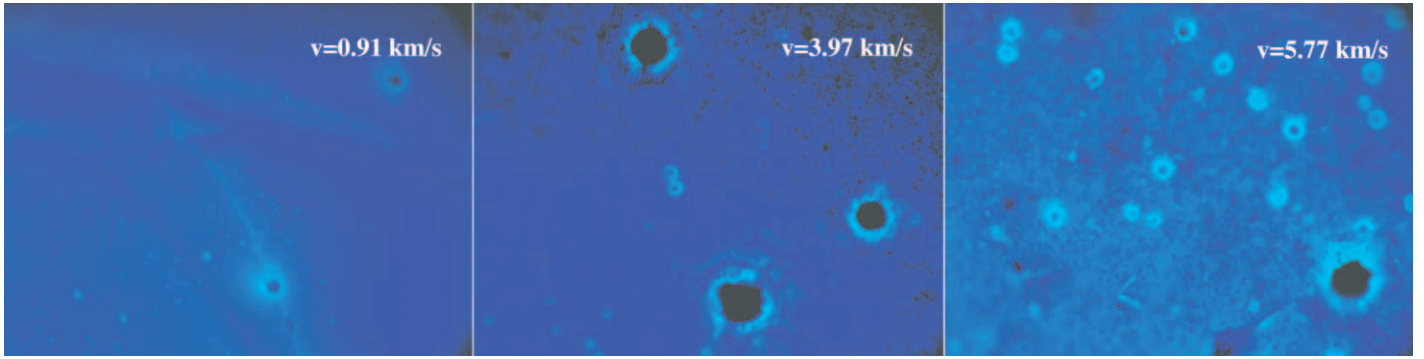


FIG. 1.—Fluorescence images of the surfaces of a Gd, Tb-doped alumina aerogel shot with monodisperse glass (diameters = 2, 5, and 20  $\mu\text{m}$ ) beads at various velocities at NASA Ames. Images were taken with a Zeiss Axiophot fluorescence microscope with an attached Optronics DEI-750 three-chip analog camera. Image contrast was enhanced to improve visibility of fluorescence.

supersonic slowing phase  $R$  is

$$R_{\text{super}} \sim 2\lambda \ln\left(\frac{v}{v_{\text{sonic}}}\right), \quad (4)$$

where  $v_{\text{sonic}}$  is the speed of sound in the aerogel. The logarithmic dependence of the supersonic range on velocity is consistent with the weak dependence observed experimentally (Kitazawa et al. 1999). If some fraction of the energy loss contributes to the local heating of the aerogel, we should expect the amount of aerogel crystallized to increase as the projectile kinetic energy increases. Assuming that the luminescence we observe is dominated by one fluorescent phase, the mass per unit track length that is converted into this fluorescent phase is expected to be

$$\frac{dm_{\text{fl}}}{dx} \propto v^2 r^2. \quad (5)$$

The dependence of  $I_{\text{green}}$  on the amount of crystallized aerogel is not necessarily straightforward, as it depends on the optical properties (ultraviolet and visible) of the aerogel as well as the track length. For events with large track lengths, such as those due to 20  $\mu\text{m}$  diameter grains, the

fluorescence yield may be dominated by the fluorescence at or near the surface of the aerogel. If so, then  $I_{\text{green}}$  should be proportional to the amount of aerogel crystallized near the track entrance. For constant density, therefore

$$I_{\text{green}} \propto m^{2/3} v_0^2, \quad (6)$$

where  $v_0$  is the initial impact velocity. Conversely, for shallow events, the fluorescence yield is expected to be more sensitive to the total amount of aerogel crystallized; hence, we would expect  $I_{\text{green}} \propto E_0$ . Interestingly, a fit of  $I_{\text{green}}$  versus  $m^{2/3} v_0^2$  appears to be a better fit to the data than  $I_{\text{green}}$  versus  $E$ . Regardless of which functional form ( $E_0$  or  $E_0 m^{-1/3}$ ) turns out to be more accurate when additional studies are done, the main implication of the results reported here is the same. Together with an independent measure of the mass, for example, using in situ optical imaging or X-ray fluorescence, the velocity of the embedded projectile can be determined (Flynn et al. 1996; Flynn, Sutton, & Horz 2000).

The expectation that the amount of crystallized aerogel scales with the amount of heat deposited is not obvious. The dependence of emission intensity on grain kinetic energy is complicated by the fact that several phase transitions can occur as alumina aerogels are heated (amorphous  $\rightarrow \gamma\text{-Al}_2\text{O}_3 \rightarrow \theta\text{-Al}_2\text{O}_3 \rightarrow \alpha\text{-Al}_2\text{O}_3$  [corundum]) (Mizushima & Hori 1995). Fluorescence efficiencies of dopant ions typically depend strongly on their local crystal field, and thus it is likely that even if higher grain kinetic energy does not crystallize a larger mass of aerogel, the higher temperatures produced within the track will yield phases of different luminescence. For example, the heating of  $\text{Al}_2\text{O}_3\text{:Gd, Tb}$  doped aerogels to even moderate temperatures ( $\sim 1100^\circ\text{C}$ ) can precipitate phases such as the perovskite phase  $\text{GdAlO}_3\text{:Tb}$  and the metastable garnet phase  $\text{Gd}_3\text{Al}_5\text{O}_{12}\text{:Tb}$ , which yield brilliant green luminescence under UV illumination.

The kinetics of crystallization in doped alumina aerogels are not known but a more detailed understanding should be helpful in maximizing their usefulness. The density of the aerogels used in this study are higher than optimal for capture of small hypervelocity particles. Developing larger samples with lower density is a major focus of our current work. This will allow us to characterize the optical properties of our calorimetric aerogel, which will in turn help us understand the effects that oblique impacts and particle fragmentation may have on integrated fluorescence. Studies of the crystallographic phase and fluorescence dependence on temperature could be used as an in situ temperature

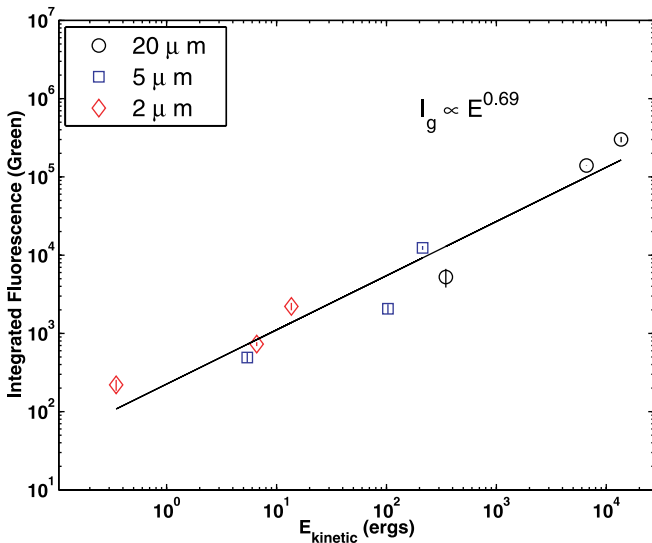


FIG. 2.— $I_g$  vs. kinetic energy of hypervelocity projectiles. The error bars are statistical only.

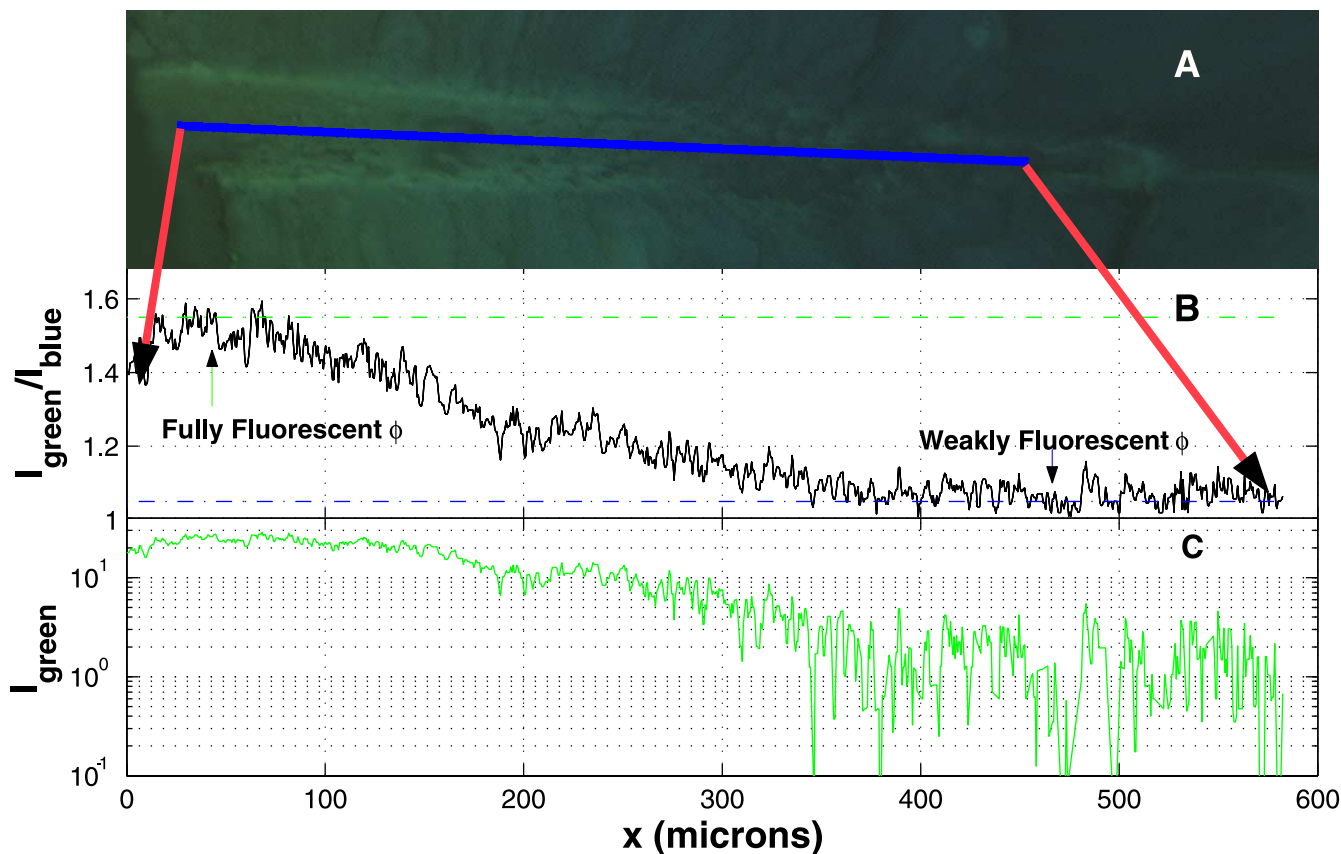


FIG. 3.—(a) Fluorescence image of a cleaved track produced by a particle  $\approx 30 \mu\text{m}$  in diameter and initial velocity equal to  $4.72 \text{ km s}^{-1}$ . (b) The ratio of green to blue, indicating the degree to which the sampled track region has been transformed into the fluorescent phase. (c) Fluorescence signal in the green channel sampled along the track. Notice that the fluorescence drops off significantly at the end of the track. Image contrast was not enhanced.

gauge along tracks and could improve our understanding of the kinetics of grain capture in aerogels. Detailed, systematic studies of *fluorescence along a track* may provide information on the instantaneous energy loss that a captured dust particle experiences (see Fig. 3).

#### 4. DISCUSSION

The *Stardust* spacecraft, whose primary mission is to return samples of cometary dust to Earth for laboratory study, has exposed aerogel collectors to the interstellar dust stream during two periods of its cruise phase. The *Stardust* collectors will be returned in 2006. Models of the IS dust flux in the inner solar system indicate that the *Stardust* collectors will capture  $\sim 10$   $1 \mu\text{m}$  particles, and perhaps one  $2 \mu\text{m}$  particle. An array of calorimetric aerogel, with collecting area of  $3 \text{ m}^2$  deployed in low-Earth orbit for 2 yr, would have enough collecting power to collect several hundred  $1 \mu\text{m}$  IS particles (Landgraf et al. 2000). A collector deployed on the wake side of a spacecraft in low-Earth orbit could collect IS dust at moderate velocities ( $< 10 \text{ km s}^{-1}$ ) during periods of the year when the Earth's motion is most parallel to that of the IS dust stream (Grün et al. 2000). Furthermore, the largest particle expected to be captured by such an array would be  $\sim 30$  times more massive than the largest particle expected to be collected by *Stardust* (Landgraf et al. 2000) (see Fig. 4). These particles would be large enough to apply multiple chemical, mineralogical, and

isotopic analysis techniques to each particle (Zolensky et al. 2000).

The main point of this paper is that we have discovered a method of distinguishing between copious anthropogenic

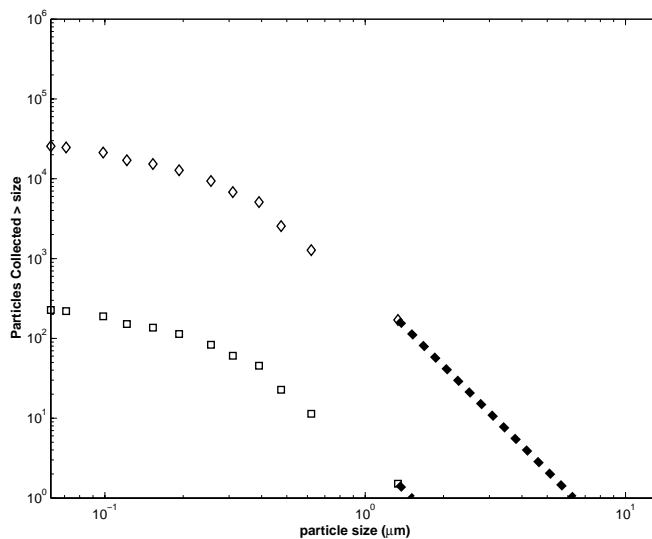


FIG. 4.—Cumulative number of interstellar dust grains vs. size expected to be collector in low-Earth orbit (diamonds) compared to the *Stardust* mission (squares). The collecting area of the low-Earth orbit collector is assumed to be  $3 \text{ m}^2$ , exposed for 3 yr (30% duty cycle). The extrapolated portion of the calculation assumes an IS flux that falls off as  $m^{-1.1}$ .

debris and relatively rare extraterrestrial particles captured in a collector in low-Earth orbit. Although we have chosen to emphasize the importance of capturing large numbers of contemporary interstellar grains for the first time, it is inevitable that interplanetary dust particles would also be collected. Both of these populations would be of scientific interest, and separating these two populations is a complex problem, beyond the scope of this paper. A large-statistics collection of interplanetary dust collected in space would be a valuable resource for the meteoritics and planetary science community. So-called interplanetary dust particles (IDPs) have been collected in the stratosphere for many years (Brownlee et al. 2002). Micrometeorites have also been collected terrestrially in Antarctica—these are the so-called Antarctic micrometeorites (AMMs) (Engrand & Maurette 1998). Each of these collection techniques has its own biases. Both are biased toward particles that can survive atmospheric entry. The effects of atmospheric contamination are poorly understood (Flynn et al. 1995). Terrestrially collected micrometeorites are selected toward particles that survive weathering and that are readily recognized as extraterrestrial.

A few chondritic particles have been extracted from ordinary silica aerogel collectors flown in space and ana-

lyzed (Hörz et al. 2000). These chondritic grains were selected from a large background of anthropogenic particles. The relationship between IDPs, AMMs, micrometeorites, and ordinary chondrites is not clear (Brownlee et al. 2002; Flynn 2002), but it appears that AMMs constitute a different population than IDPs, and may have a different origin. A single collector that is large enough to capture, in space, several approximately 100  $\mu\text{m}$  particles—characteristic of AMMs—along with IDPs could clarify the relationship between them. The origin of AMMs in particular is important, since they constitute the greatest contemporary mass input to the Earth (Maurette 2000) and could have contributed a significant amount of water and organics to the early Earth.

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