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Magnetically Enhanced Coagulation of Very Small Iron Grains

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Laboratory experiments, in which very small (~20 nm) grains are produced in the presence of a magnetic field on the order of 100 Gauss in a low-pressure hydrogen atmosphere, have demonstrated that such smokes can become permanently magnetized. We show that magnetization results in an enormous enhancement in the coagulation efficiency of such materials even in the absence of external magnetic fields. Small iron grains should have been produced in the solar nebula by thermal processing of preexisting interstellar grains. If such processing occurred via high-energy electromagnetic events then the resultant magnetized grains could have triggered the formation of centimeter- to meter-sized protoplanetessimals by acting as "nets" capable of sweeping up nonconductive silicates suspended in the gas. It is possible that the presence of conductive fractal aggregates observed in modern-day protostellar disks could be explained by the enhanced coagulation efficiency of very small magnetized iron particles. © 1994 Academic Press, Inc.

I. INTRODUCTION

Chondrules most likely formed in the primitive solar nebula (Grossman 1988), the result of an energetic, transient process (Levy 1988) which transformed a loosely bound aggregate of refractory grains into a molten—or semimolten—spheroidal droplet on the order of 1 mm in size. Solar furnace experiments on likely chondrule precursors (King 1983), studies of meteor ablation spherules (Brownlee *et al.* 1983), and speculations on the fate of heated interstellar grain aggregates (Clayton 1980, Nuth 1989) all demonstrate the likelihood that such an aggregate will undergo an internal oxidation/reduction reaction between iron oxides and carbonaceous grains or underoxidized silicon, producing a glassy silicate component plus immiscible iron metal. These small iron grains could be brought to the grain surface through centrifugal forces in a spinning chondrule (Grossman 1988) or possibly be excluded from the interior of the forming droplet as the surface tension of the molten silicate forced the irregular, partially molten aggregate to contract into a sphere.

Iron grains which were produced *in situ* by such oxidation/reduction processes but which had no means of escape to the chondrule surface are thought to be responsible for the "dusty" olivine or pyroxene grains (Fredrikson *et al.* 1969, Nagahara 1981, Rambaldi 1981) observed in some chondrules. Individual ~0.1 μm interstellar grains subjected to high temperatures should undergo similar degrees of reaction, possibly resulting in the production of seperate populations of silicate and nanometer-sized iron grains. For this reason the formation of small iron grains via the same energetic processes which formed chondrules should be considered firmly established, and because of the widespread prevalence of the chondrule formation process in the solar nebula (Grossman 1988) one can assume that the nebula also contained a significant number of such small iron grains.

The high number density of submicrometer-sized iron grains is not significant in and of itself, but can become extremely important if large, transient magnetic fields were present to a significant degree in the nebula, as suggested by Grossman (1988). Whipple (1966) and Cameron (1966) have suggested that lightning discharges could have provided the energy source necessary to form chondrules. Similarly, Sonett (1979) has suggested that large

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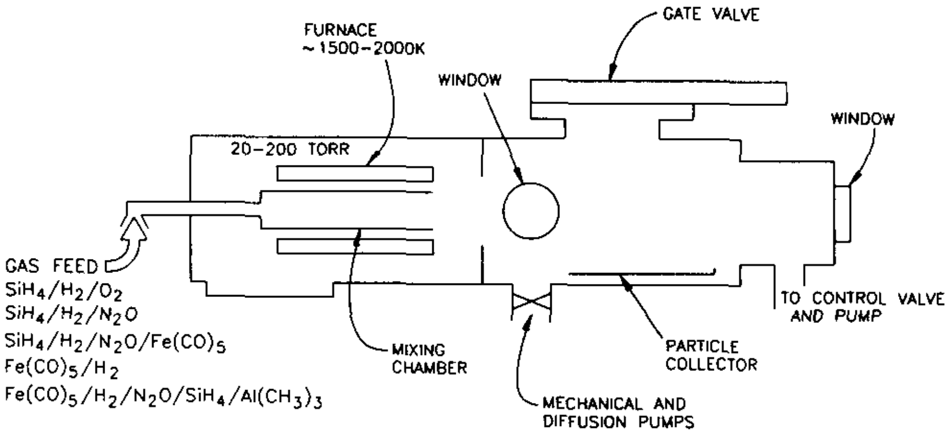

FIG. 1. A schematic diagram of the experimental apparatus showing the location of the furnace, collector, and windows, which are 4 inches in diameter.

fluxes of relativistic electrons could have served the same purpose and would have been generated by magnetic field reconnection events in the nebula. More recent evaluations of these scenarios (Levy 1988) suggest that both are possible to some degree—lightning near the disk plane and magnetic flare events high in the disk corona. Levy (personal communication) has calculated that lightning discharges in the solar nebula would yield currents of 5 ($\pm$2) $\times$ 10^6 amperes in a cylinder approximately 1 km across. This yields a transient field at the edge of the cylinder of from 5 to 15 Gauss. In addition, it is possible that a magnetohydrodynamic dynamo field of between 1 and 10 Gauss might have been generated within the disk itself (Levy 1988). No matter the exact mechanism, studies of the remanent magnetization observed in chondritic meteorites attest to the presence of some level of magnetic field in the nebula (Sugiura and Strangway 1988) at the epoch of chondrule formation. It is the combination of large numbers of submicrometer iron grains with a magnetic field—possibly of considerable strength—at the time such grains formed, which may have significant consequences for the formation of planetesimals in the primitive solar nebula (Nuth *et al.* 1992). Exposure of submicrometer iron grains to this magnetic field, even if the field is transient, will result in the formation of permanent magnetic dipoles.

Grain coagulation in the primitive nebula is one of the most important and least understood processes leading to the formation of planets (Meakin and Donn 1988, Weidenschilling *et al.* 1989, Weidenschilling and Cuzzi 1993). This same process is also important on a smaller scale in the formation of chondrule precursors—the fluffy aggregates processed via the unknown energy source must have formed somewhere and the efficiency of aggregate formation must have been high since the efficiency of chondrule formation in some regions of the nebula must also have been quite high (Grossman 1988). Finally, grain coagulation must occur in protostellar systems in order to convert the small (0.1-μm) grains or grain aggregates found in typical dark clouds (Cassen and Boss 1988) to the much larger grains or aggregates observed around T-Tauri stars (Beckwith and Sargent 1991, Miyake and Nakagawa 1992). We report here on a series of experiments which may shed some light on these processes.

II. EXPERIMENTAL PROCEDURE

Refractory metal and metal-oxide grains can be made by passing volatile precursors such as silane, trimethyl aluminum, titanium tetrachloride, or iron pentacarbonyl, diluted in a hydrogen carrier gas and mixed with an oxidizer such as oxygen or nitrous oxide, through a tube furnace at elevated temperatures (e.g., Rietmeijer and Nuth 1991, Nelson *et al.* 1989). The tube furnace consists of an aluminum oxide cylinder that passes through a 5.6-cm-diameter graphite bar through which numerous holes have been drilled to increase the electrical resistance of the graphite. The graphite bar is heated by passing a low-voltage AC current through it at a high amperage, usually between 500 and 1200 amperes. The particles produced via chemical decomposition of the $Fe(CO)_5$, $TiCl_4$, $Al(CH_3)_3$ or the subsequent reaction of SiH_4 with the oxidizer, followed by vapor-phase condensation, are collected on copper plates placed downstream from the furnace at temperatures on the order of 300 K. A schematic drawing of the apparatus is shown in Fig. 1.

In these experiments pure iron grains on the order of 20–50 nm in diameter were produced by flowing iron pentacarbonyl diluted in hydrogen through a furnace at temperatures in excess of 500 K at total pressures around 100 torr. However, unlike our previous experiences with this apparatus, the iron particles did not slowly settle out from the gas phase. After several seconds of producing iron

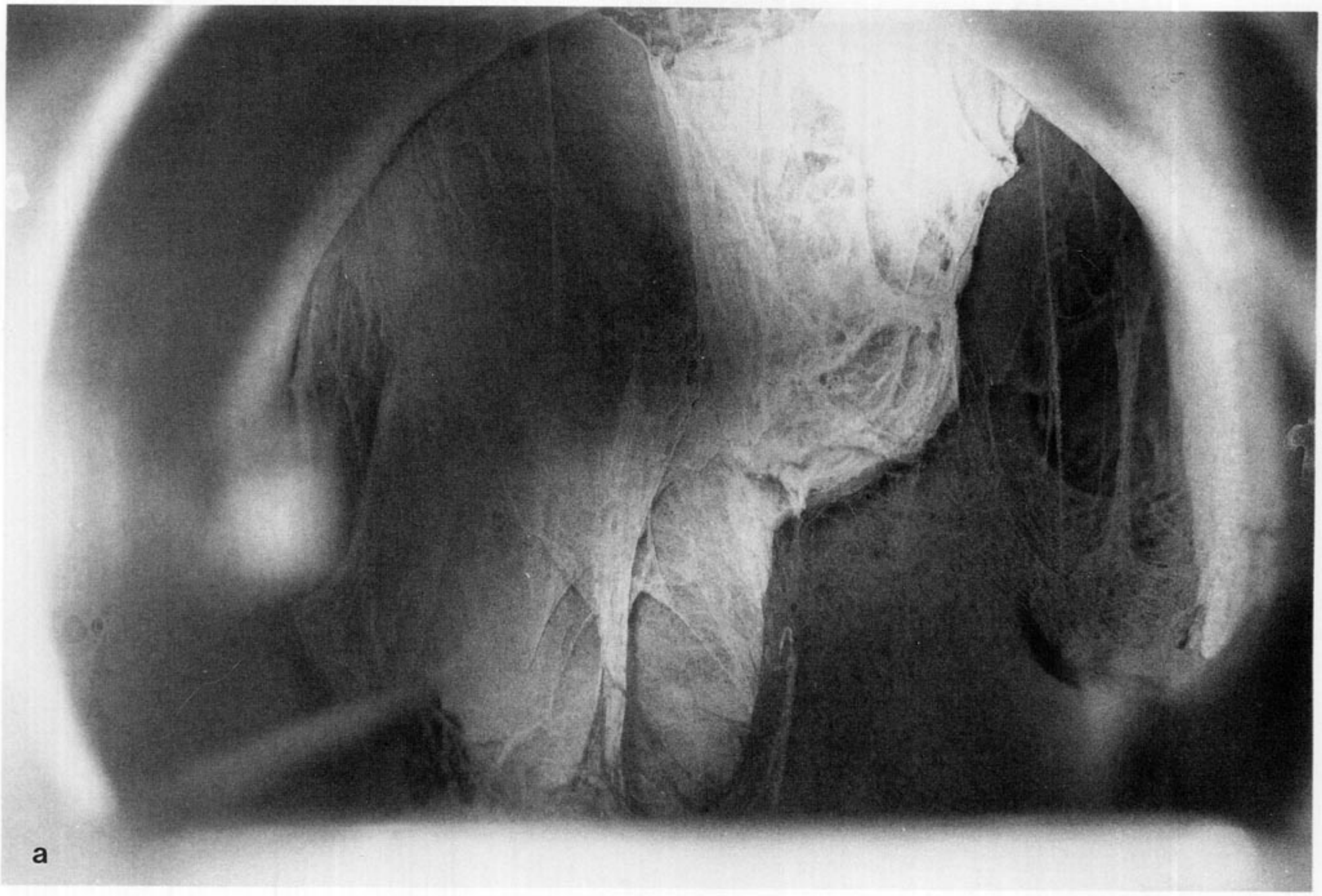

FIG. 2. Two views into the experimental chamber. (a) View of webs from the side port, illuminated through the side port. (b) View of webs from the rear window looking along the axis toward the furnace while illuminated via the side port.

"smoke," the particles suddenly gelled into a dense mass of "spiderwebs" (see Fig. 2a,2b). Dilution of the iron carbonyl-hydrogen mixture from roughly 10 mol% iron carbonyl to approximately 0.1 mol% resulted in the formation of (3–4-inch) "streamers" of fluffy iron aggregates 20–30 sec after initiation of the experiments. Further dilution—by roughly a factor of 10—resulted in the formation of small fluffy (~1 mm) dustballs after several minutes.

We have run several hundred experiments in this flow system using a wide variety of reactant precursors to produce grains of varying compositions. Only pure iron grains form aggregates while suspended in the hydrogen flow. No other material has ever shown the slightest evidence for growth via coagulation in our experimental apparatus beyond aggregates of a few tens of grains: such growth is only apparent using electron microscopy. No other material but iron has ever produced a morphology characterizable as "dustballs," "streamers," or "spiderwebs." We hypothesize that these aggregates formed as the result of magnetic effects. To test this hypothesis we ran a series of experiments in which the furnace was heated to ~1000 K in a pure hydrogen flow. Power to the furnace was then turned off and a typical hydrogen–iron carbonyl mixture was passed through the furnace. Plenty of iron smoke was observed to form as the temperature of the furnace decreased at a rate of approximately 100 K per minute. No dustballs, streamers, or spiderwebs were observed in these experiments; the grains were collected on copper plates after settling out of the hydrogen flow and were found to be normal "smokes."

We performed several experiments in which iron spiderwebs were collected on glass slides covered in nonmagnetic epoxy. The strands appeared to sink evenly into the epoxy which was then allowed to dry *in vacuo;* these iron strands were collected without exposure to the atmosphere and were the subject of later magnetic analyses. We attempted to perform a series of experiments in which the iron particles condensed above the Curie Point by bringing the furnace to ~1300 K prior to starting the flow of iron carbonyl. Unfortunately, the temperature gradient downstream of the furnace is extremely high (>500 K/inch) as is the electrical current used to heat the furnace

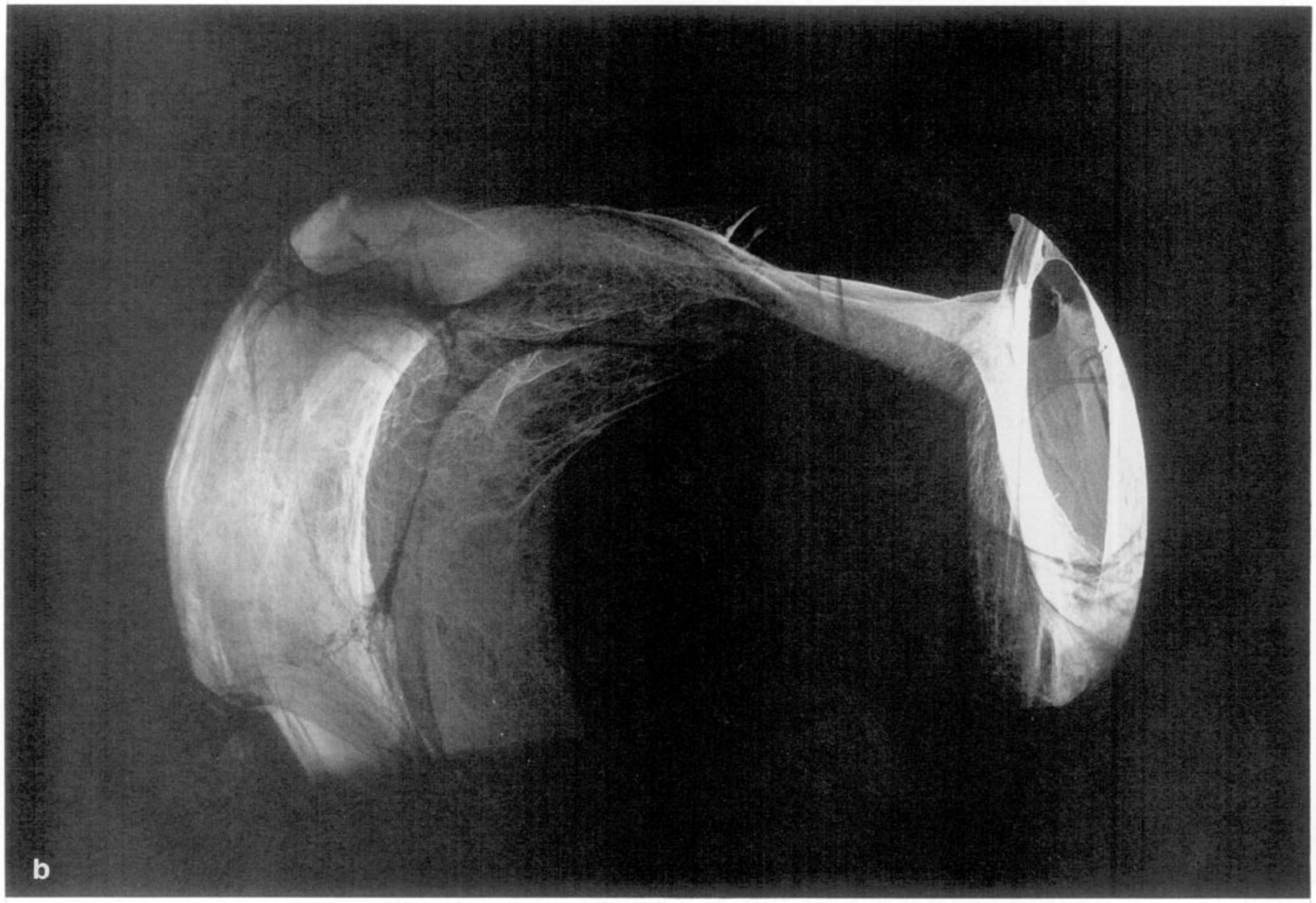

FIG. 2—*Continued*

to this temperature (~1200 Amps). We were therefore never able to observe grains formed at high temperature but which passed through the Curie Point at very low magnetic field strength. In each of these experiments we formed copious quantities of webs quite rapidly.

III. ANALYSES

The magnetic remanance (RM) of the iron webs collected in epoxy and dried *in vacuo* was measured using a liquid helium cooled superconducting magnetometer. The samples were then subjected to a 10,000-Gauss (1.0 Tesla) magnetic field and the saturation remanant magnetization (SIRM) measured on the same superconducting magnetometer. The ratio (REM) of the remanant magnetization in the web samples produced in the flow system to the saturation magnetic remanance for each of the three samples is given in Table I. REM values of a few percent suggest that the remanance acquisition mechanism is extremely efficient. The magnetic remanence in the web samples was acquired as the result of exposure to a magnetic field estimated to be, at most, between 35 and 90 Gauss just outside the furnace, although no field measurements were made. This estimate was based on the geometry of the system and the ~500- to 1200-ampere AC current typically used to heat the furnace.

Alternating field demagnetization studies were also performed on the web samples; the results were identical for

TABLE I
Magnetic Remanence

Sample	RM (10^{-4} emu)	SIRM (10^{-4} emu)	REM
C	0.834	35.87	0.023
D	0.782	25.60	0.030
E	0.814	25.95	0.031

Note. RM, remanence magnetization in the web material measured after deposition on a glass slide and set permanently in epoxy; SIRM, saturation remanent magnetization acquired upon application and removal of a magnetic field of 10,000 Gauss (0.1 Tesla); REM, ratio RM/SIRM.

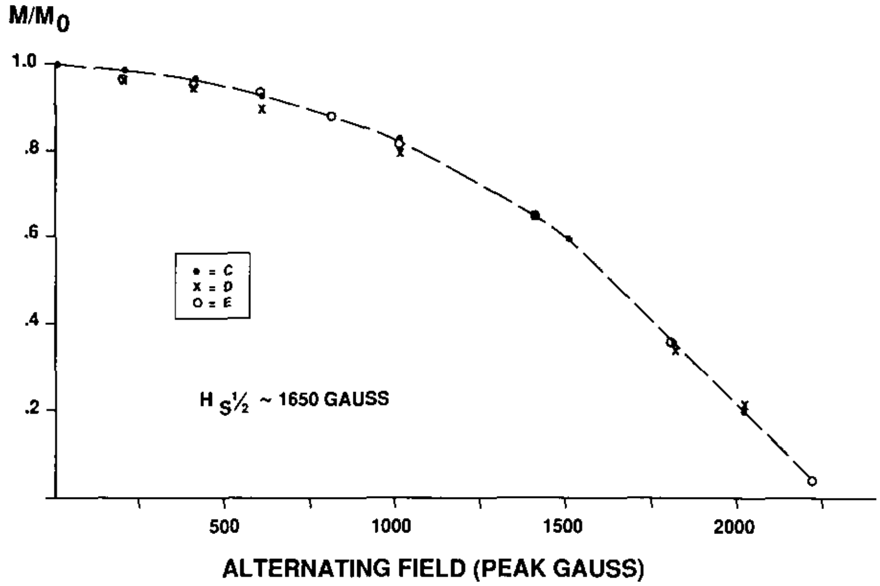

FIG. 3. Alternating field demagnetization curves were identical for the three samples studied: The 1650-Gauss field required to demagnetize one-half of the saturation remanance (M_0) indicates that the web material is magnetically hard. M is the instantaneous remanence remaining in the sample after exposure to a magnetic field of the indicated strength.

three samples and are presented in Fig. 3. A field of 1650 Gauss (165 mT) was required to demagnetize one-half of the saturation remanance in the samples, indicating that the material making up the web is magnetically hard. These chains appear to be magnetically harder than a noninteracting dispersion of iron spheres in a copper matrix with a size distribution between ~20 and 35 nm (Wasilewski 1981). It may be that the coagulation of the initial distribution of 20- to 50-nm-diameter spheres into sintered chains (see below) allows an approximation to stable single domain behavior in strings containing tens of iron particles. It is also likely that "pole to pole" coagulation would tend to eliminate much of the net remanent magnetic field from individual grains in such sintered chains. This would imply that a net remanence of a few percent for the chain probably resulted from the incomplete cancellation of nearly completely saturated single domain particles in the irregular chains.

The morphology of the web was studied using optical microscopy, scanning electron microscopy, and transmission electron microscopy. The web consisted of intersecting continuous strands a fraction of a millimeter thick and 4–5 inches long (the collection chamber is 4 inches in diameter). Individual strands consist of many kinked smaller strands of iron particles ~20–50 nm in diameter and many hundreds of particles long (Fig. 4). Individual particles were connected to their neighbors in head-to-tail chains at angles usually between about 90° and 180°. Very few triplets were observed at angles of either 90° or 180°; the vast majority appear to form at angles on the order of 130°–150°. A dipole in the presence of a nonuniform magnetic field is subject to the greatest force at an angle which maximizes the divergence of the field (Purcell 1965). The strongest attraction between two single-domain particles is therefore likely to result in grains which stick together at some angle to their magnetic poles. This would tend to explain the prevalence of 130°–150° bond angles.

Individual particle boundaries show evidence for much more than simple "sticking"; some evidence is observed for martensitic transformations at the joints, some joints have deformed into contact planes, and the chains show evidence for a continuous overgrowth of a metal oxide layer. We suggest that these grains underwent some degree of contact welding due to the interaction of both surface tension and magnetic dipole forces shortly after or during coagulation.

IV. DISCUSSION

If grains falling into the solar nebula from the interstellar medium are processed via magnetic reconnection events (Sonett 1979), lighting discharges (Whipple 1966), shock heating (Wood 1984) or simple release of chemical potential energy (Clayton 1980) and such processing leads to the production of a significant number of very small iron grains and if such grains are then exposed to even a transient magnetic field, then magnetically enhanced coagulation could have been an important step in the formation of planetessimals. Magnetic dipoles in a relatively quiescent,

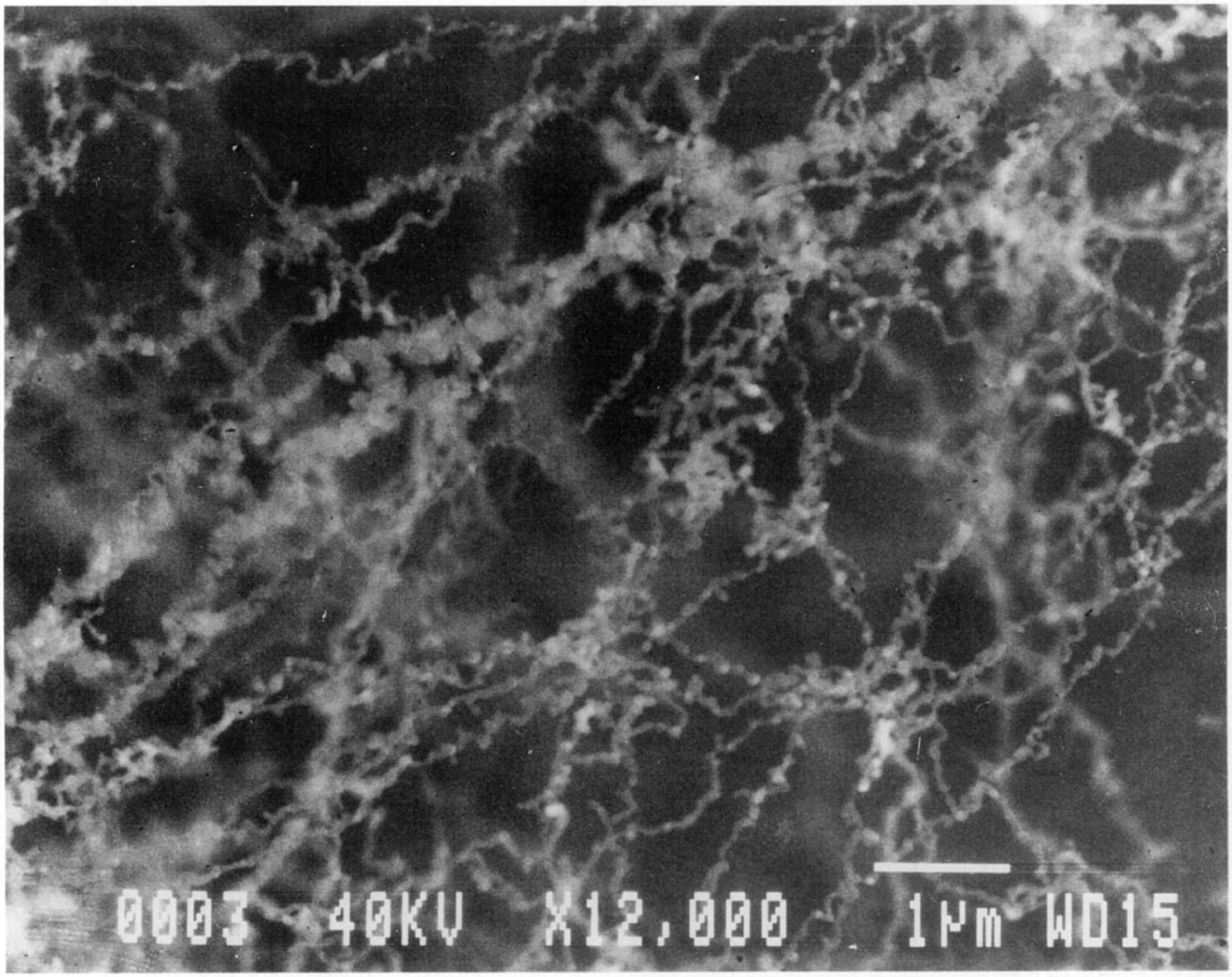

FIG. 4. Scanning electron microscope image of a small portion of web material (12,000 × magnification) taken at 40 KV. Scale bar is 1 μm long.

low-pressure environment interact over spatial scales hundreds to thousands of times larger than the grain radius (KenKnight 1992). No external field is necessary for this interaction to occur. Once "nets" of iron particles form they will be extremely effective at sweeping up additional particles which may include unprocessed interstellar grains, nebular condensates, previously annealed crystalline grains or residues from previously processed fluffy aggregates.

Grains in a molecular cloud core are likely to be relatively well oxidized. Therefore it is likely that very few—if any—metallic iron grains will be present in such a population (Nuth and Hecht 1990). However, as solids are processed in the nebula an increasing fraction of the grain population will consist of small iron grains. It should be emphasized that iron formation can occur when an *individual* interstellar grain (possibly coated with carbonaceous material) is heated. It is not necessary to form large aggregates for this process to take place. Such grains could have been magnetized by nebular fields or by transient fields generated by nebular lightning. Grain–grain interactions among magnetized grains will be very efficient in a relatively quiescent or low-pressure environment (KenKnight 1992).

As a check on this hypothesis one can calculate the approximate increase in the effective radius of two single-domain magnetic dipoles interacting in the nebula using the magnetic analogy to gravitational focusing. The force (F) between two magnetic dipoles is given by

$$F = \frac{m_1 m_2}{\mu r^2}, \tag{1}$$

where m_1 and m_2 are the magnetic pole strengths, r is the particle separation, and μ is the magnetic permeability, equal to unity in a vacuum. By analogy with gravitational focusing (Öpik 1974), the effective radius (R) of one dipole interacting with another will be given by

$$R = a(1 + V_e^2/V_0^2)^{1/2}, \tag{2}$$

where a is the radius of the larger particle, V_e is the escape velocity, and V_0 is the initial velocity of the particle. The "escape velocity" for two magnetically interacting dipoles can be derived by substituting the magnetic potential in place of the gravitational potential to yield

$$V_e = \left[\frac{2m_1m_2}{M\mu r}\right]^{1/2}, \tag{3}$$

where M is the reduced mass of the particles. The average interparticle distance will depend on the initial number density (n_0) and Eq. (2) therefore yields

$$R = a\left[1 + 2\left(\frac{4\pi}{3}\right)^{1/3} \frac{m_1m_2}{M\mu}\frac{n_0^{1/3}}{V_0^2}\right]^{1/2}. \tag{4}$$

If we assume that single-domain iron particles have the bulk saturation magnetization for iron of 1720 emu/cm^3 (Butler and Banerjee 1975), a density of 7.86 g/cm^3, and a geometric mean particle size equivalent to the grains in our experiments, we obtain a value for m of $\sim 5 \times 10^{-4}$ esu and for M of $\sim 7 \times 10^{-17}$ g. If we further assume an initial number density (n_0) of 10^6 cm^{-3} and V_0 of ~ 1 cm/sec (Weidenschilling and Cuzzi 1993), equation (4) yields an enhancement in the geometric radius (a) by a factor of $\sim 10^6$.

One can easily see from Eq. (4) that the enhancement is relatively insensitive to the initial number density—varying as $n_0^{1/6}$ and only somewhat sensitive to particle mass—varying as $M^{-1/2}$. However, the enhancement is directly proportional to the magnetic pole strength and inversely proportional to particle velocity. Because single domain particles will be completely saturated after exposure to a strong magnetic field, m will vary as the particle radius (a) cubed. Due to the limited size stability range of single domain iron particles (Butler and Banerjee 1975) between ~ 15 and 60 nm, uncertainty in the particle size we assumed for our calculation (~ 30 nm) introduces a factor of ~ 8 variation in the enhancement factor. This is partially compensated for by the dependence of M on a, which reduces the variation to a factor of ~ 3. Of course, particles exposed to a weaker field—possible because they were located at some significant distance from a transient event such as lightning—will not become as strongly magnetized.

A much larger variation in the enhancement factor arises from the likely natural distribution in V_0. In a near vacuum V_0 could become quite large and would severely reduce R. However, in the solar nebula at a pressure between 10^{-3} and 10^{-7} atmospheres very small grains will be closely coupled to the gas and their relative velocities are likely to be quite small (Weidenschilling and Cuzzi 1993). For velocities as low as 10^{-2} cm sec^{-1} the effective radius of single domain particles could be increased by as much as a factor of 10^8.

Finally, since it is unlikely that iron grains would form in isolation we can ask what effect the presence of dielectric silicate grains might have on the coagulation of the less abundant magnetic dipoles. If we assume that silicate grains have a radius of ~ 20 nm and an initial n_0 of $\sim 10^6$ but that iron grains have an initial n_0 of only 1 cm^{-3}, then two magnetic dipoles must traverse a distance of approximately 1 cm in order to collide and stick. Since each silicate grain has a cross-sectional area of $\sim 10^{-11}$ cm^2, one iron grain in $\sim 10^5$ will collide with a silicate grain before coagulating with another iron particle. If the silicate grain sticks to the iron particle it will increase the effective mass of the composite grain by less than 50% due to the much lower density of the silicate. This will result in a negligible decrease in the effective collision cross section due to the inverse square-root dependence of R on M.

One can envision the formation of iron nets that quickly sweep up dielectric grains into a fluffy, composite dust ball which awaits processing via the next transient nebular heating episode. Such a scenario might suggest a relationship between the size and number density of chondrules in a particular meteorite class and the degree of nebular processing that the initial interstellar grain population underwent in order to produce the individual constituents which came together to form that particular meteorite parent body. As an example, CM chondrites contain only a few, relatively small chondrules whereas the ordinary chondrites contain many, larger ones: the components of the latter obviously saw a considerably higher degree of nebular processing than did the components of the former. This would not necessarily imply that ordinary chondrites were processed at higher nebular temperature than CM or CO chondrites, only that the interstellar matter which became the ordinary chondrites saw a larger number of transient heating events (i.e., lightning strokes) either because they were formed in a more "active" region of the nebula or in a region of "normal" activity but over a more extended time span. Another way to view this scenario is to consider the average number of transient heating events experienced per unit meteorite parent body. Material in CM parent bodies experienced many fewer transient heating events prior to their aggregation into planetessimals than did the material which became the ordinary chondrites.

It is interesting to note that Wright's (1987) calculation of the optical properties of fractal iron grains are consistent with observations of the spectral properties of modern protostellar systems (Beckwith and Sargent 1991). These observations suggest that the opacities of particles in these disks follow a power law dependence on frequency whose spectral index lies between 0 and 1: the index for crystalline materials is 2, whereas the index for amorphous materials may be as small as 1 (Tielens and Allamandola 1987) but only under special circumstances. The index for *conducting* fractal aggregates lies within the range from 0 to 1: calculations of the optical properties of nonconducting fractal aggregates indicate that the spectral index for such particles is greater than unity. Therefore, if the millimeter observations of protostellar disks are dominated by conducting fractal aggregates (e.g., of iron) as the observations suggest, rather than by the equally (or more) abundant silicates, it may be because of the greatly increased coagulation efficiency of magnetically interacting grains.

V. CONCLUSIONS

Magnetically hard, open nets of very small iron metal grains rapidly form via coagulation in a laboratory system in which large-scale coagulation has never before occurred. We hypothesize that magnetically induced dipole–dipole interactions between the magnetized iron grains increase the grain coagulation efficiency by several orders of magnitude and probably also increase the strength of the interparticle bonds in the aggregate. We infer that very small iron grains formed by high temperature processing of individual interstellar grains in energetic events in the protosolar nebula and may have been magnetized by exposure to even transient nebular fields. These magnetic grains could then have acted as catalysts for the coagulation of chondrule precursor "dustballs." Processing of such a "dustball" in a subsequent event would result in the formation of a chondrule, some additional silicate condensate and more iron grains. Unprocessed dustballs would slowly settle to the nebular midplane and aggregate into solid bodies.

The presence of magnetic iron dust (20–30 nm diameter) in the solar nebula as the result of energetic processing of interstellar materials could lead to a very efficient mechanism for the initial coagulation of centimeter-sized aggregates from the initial interstellar grain population. Long-range dipole–dipole magnetic interactions in the low-pressure solar nebula could increase the effective grain radius of these particles by several orders of magnitude and lead to the formation of fractal iron aggregates which may have swept up surrounding nonmagnetic material. These fractal iron aggregates may have already been observed in the long-wavelength spectra of modern protostellar nebulae.

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