(NASA-CR-141169) TH
CARBONACEOUS METEORITES Ph.D. Thesis
(Texas A&M Univ.) 124 p HC $5.25
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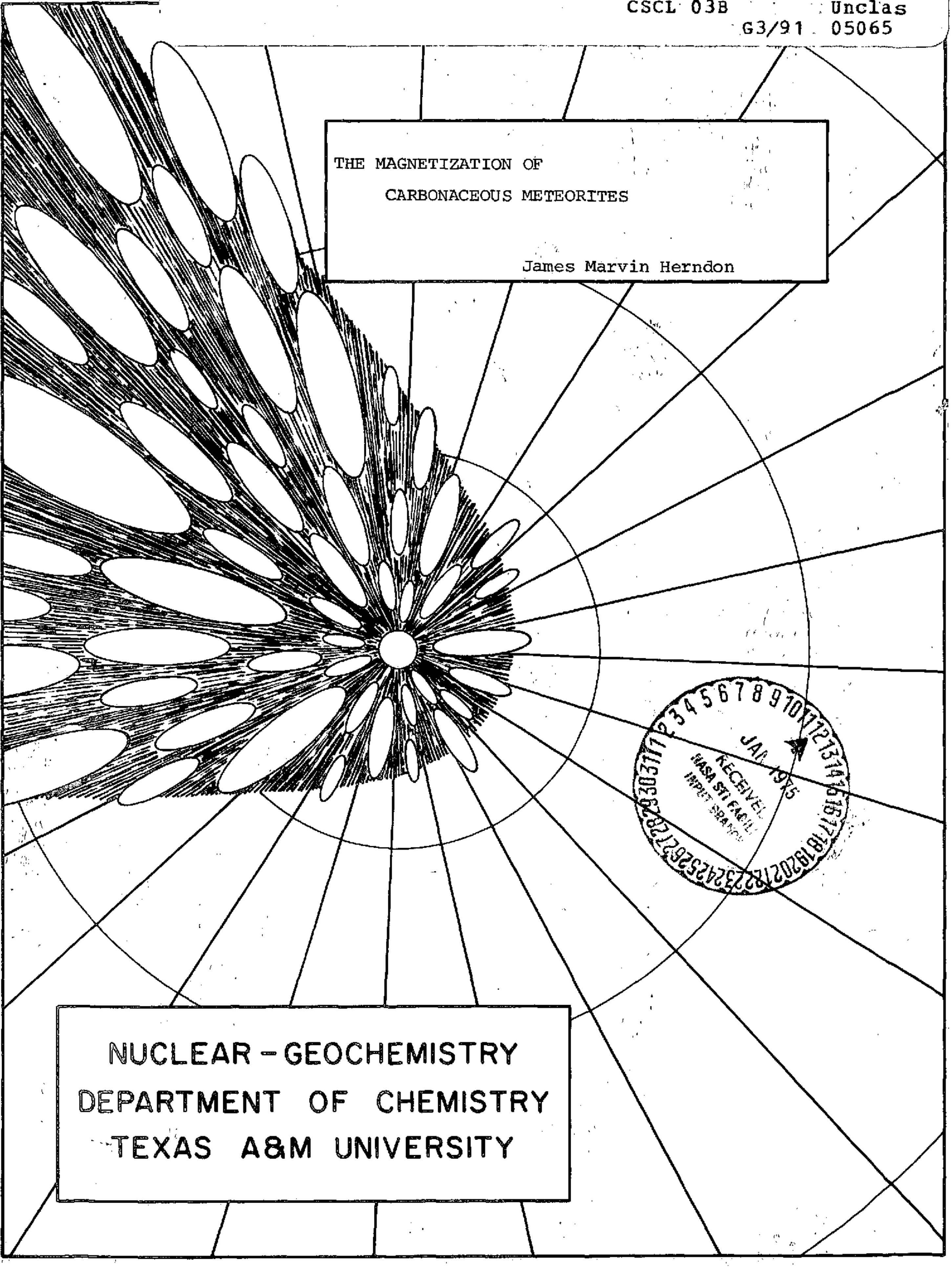

THE MAGNETIZATION OF CARBONACEOUS METEORITES

James Marvin Herndon

NUCLEAR - GEOCHEMISTRY
DEPARTMENT OF CHEMISTRY
TEXAS A&M UNIVERSITY

THE MAGNETIZATION OF CARBONACEOUS METEORITES

A Dissertation

by

James Marvin Herndon

Submitted to the Graduate College of
Texas A&M University in
partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

August 1974

Major Subject: Chemistry

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Approved as to style and content by:

(Chairman of Committee)

(Head of Department)

(Member)

(Member)

(Member)

(Member)

(Member)

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ABSTRACT

The Magnetization of Carbonaceous Meteorites. (August, 1974)
James Marvin Herndon, B.A., University of California, San Diego
Directed by: Dr. Marvin W. Rowe

Alternating field demagnetization experiments have been conducted on representative samples of the carbonaceous meteorites (carbonaceous chondrites and ureilites). The results indicate that many, if not all, of these meteorites possess an intense and stable magnetic moment of extraterrestrial origin. Thermomagnetic analyses have been conducted on samples of all known carbonaceous meteorites. In addition to yielding quantitative magnetite estimates, these studies indicate the presence of a thermally unstable component, troilite, which reacts with gaseous oxygen to form magnetite. It is proposed that the magnetite found in some carbonaceous chondrites resulted from the oxidation of troilite during the early history of the solar system. The formation of pyrrhotite is expected as a natural consequence of magnetite formation via this reaction. Consideration is given to the implications of magnetite formation on paleointensity studies.

ACKNOWLEDGEMENTS

I am greatly indebted to Professor E. E. Larson, University of Colorado, and to D. E. Watson, National Oceanic and Atmospheric Administration (NOAA), for their many helpful discussions, advice, and the courtesies extended me in using their laboratories. I am further grateful to Professor M. W. Rowe, my research advisor, for his continued interest and constant encouragement.

For generously providing meteorite samples, I thank Dr. D. D. Bogard, NASA, Houston; Dr. H. Carstens, Norges Geologiske Undersokelse, Trondheim, Norway; Dr. R. S. Clark, Jr., U. S. National Museum, Smithsonian Institution, Washington, D. C.; Dr. J. Fabries, Museum National d' History Naturelle, Paris; Dr. R. E. Folinsbee, University of Alberta, Edmonton; Dr. S. V. P. Iyengar, Geological Survey of India, Calcutta; Dr. E. A. King, Jr., University of Houston; Dr. E. L. Krinkov, Academy of Sciences, U.S.S.R., Moscow; Dr. D. V. Manson, American Museum of Natural History, New York; Mr. O. Monnig, Fort Worth, Texas; Dr. C. B. Moore, Director, Arizona State University Center for meteorite studies, Tempe; Dr. E. Olsen, Museum of National History, Chicago; Dr. H. Papunen, Institute of Geology and Mineralogy, University of Turku, Finland; and Dr. J. H. Reynolds, University of California, Berkeley.

This research was supported by the National Science Foundation and by the National Aeronautics and Space Administration.

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INTRODUCTION

The physical conditions which existed during the formation of our solar system have been of concern to scientists for centuries. Much attention has been directed to the meteorites for they have been shown to hold a remarkably intact record of some of the very early processes occurring during and shortly after the birth of the solar system. Meteorites have long been known to be very ancient (Patterson, 1955), even having retained the residue from the decay of the short-lived extinct radioactivities I^{129} (Reynolds, 1960) and Pu^{244} (Rowe and Kuroda, 1965). As John Wood (1968) put it, "This is why we study the meteorites: not primarily because they come to us from outer space, but rather because of their ancientness." Studies of meteorites are of interest for providing clues on the origin, history, and composition of the solar system.

Meteorites are broadly classified according to the relative amounts of nickel-iron and stony matter present. Iron meteorites consist mainly of nickel-iron, whereas stone meteorites are composed primarily of silicate material. Stony-iron mateorites contain nickel-iron and silicate material in roughly equal proportion. Stone meteorites constitute about 93% of the observed falls and are broadly subdivided as either chondrites or achondrites (Hey, 1966). The most abundant stone meteorites are chondrites which are

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The citations on the following pages follow the style of <u>Geochimica et Cosmochimica Acta.</u>

characterized by the presence of chondrules - small, predominately spherical silicate bodies ranging in size from microscopic to, in rare instances, about 1 cm, with the vast majority being less than 2 mm in diameter. Achondrites are stone meteorites characterized by their lack of chondrule structures. The least differentiated members of each subdivision are the carbonaceous chondrites and the ureilites (carbonaceous achondrites), respectively. Table 1 sets forth all known carbonaceous meteorites, their data of fall (or find) and the total mass recovered.

Carbonaceous chondrites, while numbering only 36 and comprising about 2% of all known meteorites, are extremely interesting for scientific investigation. These meteorites are considered to be quite primitive - their chemical composition and unusual mineralogy attesting to minimum chemical fractionation and thermal alteration (Mason, 1960; Ringwood, 1961; DuFresne and Anders, 1961, 1962; Wood, 1967). Particular significance is attached to this group of meteorites because of the identification of their bulk chemical content with solar abundance for condensable elements (Anders, 1971; Kerridge, 1972). C1 chondrites are considered to represent the closest approximation to elemental solar abundance, while C2, C3, and C4 chondrites show an increasing degree of fractionation with increasing petrologic type (Anders, 1968). Attempts have been made to determine genetic relationships among the various petrologic types of carbonaceous chondrites (Mason, 1962) and the ureilites (Anders, 1964; Vdovykin, 1970). Recent studies, however, point to a very

Table 1. The carbonaceous meteorites

Meteorites	Date of Fall	Recovered Mass(Kg)
C1 CHONDRITES		
ALAIS, France	1806	~6
IVUNA, Tanganyika	1938	0.7
ORGUEIL, France	1864	~12
REVELSTOKE, Canada	1965	~0.001
TONK, India	1911	<0.01
C2 CHONDRITES		
AL RAIS, Saudi Arabia	1957	0.162
BELLS, Texas, U.S.A.	1961	0.3
BORISKINO, U.S.S.R.	1930	1.34
COLD BOKKEVELD, South Africa	1838	~4
CRESCENT, Oklahoma, U.S.A	1936	0.0884
ERAKOT, India	1940	0.113
ESSEBI, Congo	1957	0.3
HARIPURA, India	1921	0.3
KABA, Hungary	1857	~3
MIGHEI, U.S.S.R.	1889	~8
MOKOIA, New Zealand	1908	~4
MURCHISON, Australia	1969	~105
MURRAY, Kentucky, U.S.A.	1950	7

Table 1. The carbonaceous meteorites (cont'd)

Meteorites	Date of Fall	Recovered Mass(Kg)
NAWAPALI, India	1890	~0.06
NOGOYA, Argentina	1879	~4
POLLEN, Norway	1942	0.2536
RENAZZO, Italy	1824	~5
SANTA CRUZ, Mexico	1939	~0.05
C3 CHONDRITES		
ALLENDE, Mexico	1969	~2000
BALI, Cameroun	1907	<0.011
EFREMOVKA, U.S.S.R.	1962*	21
FELIX, Alabama, U.S.A.	1900	~3
GROSNAJA, U.S.S.R.	1861	~3.5
KAINSAZ, U.S.S.R.	1937	200
LANCE', France	1872	~52
LEOVILLE, Kansas, U.S.A.	1962*	8.1
ORNANS, France	1868	~6
VIGARANO, Italy	1910	16.0
WARRENTON, Missouri, U.S.A.	1877	~45
C4 CHONDRITES		
COOLIDGE, Kansas, U.S.A.	1937*	4.5
KAROONDA, Australia	1930	42

Table 1. The carbonaceous meteorites (cont'd)

Meteorite	Date of Fall	Recovered Mass (Kg)
UREILITES		
DINGO PUP DONGA, Australia	1965*	0.120
DYLAPUR, India	1872	~1.4
GOALPARA, India	1868*	~2.7
HAVERO, Finland	1971	1.544
NORTH HAIG, Australia	1961*	0.964
NOVO UREI, U.S.S.R.	1886	1.9

* Indicates date found

complex picture which probably precludes establishment of any such simple relationship among these meteorites. The controversiality of this subject is typified by the proposed genesis of material constituting a single Cl chondrite, Orgueil (Anders, 1972a, 1972b; Kerridge, 1971).

Obviously there is a multiplicity of reasons for the existing divergent theories on the origin of carbonaceous meteorites. Certainly one significant factor involved is the difficulty of obtaining experimental data on these complex meteorites with the limited amount of material available for study. For example, prior to the work reported here, measurements of magnetite content had been made on only two meteorites, Orgueil and Mokoia.

Cloetz (1864) first determined that magnetite was present in Orgueil on the basis of magnetic susceptibility measurements, metallic iron being absent. His application of magnetic techniques is of particular interest historically, since the magnetite analyses presented in this work were obtained on the same meteorite (and others) by application of a similar, although more sophisticated thermomagnetic analysis technique. Early applications of this technique clearly demonstrated its potential as a method for cosmological investigation. The comprehensive work of Smith (1908) laid a solid foundation for understanding the temperature dependent magnetic transformations in the Ni-Fe system. Upon this foundation Smith and Young (1939) utilized thermomagnetic analysis to measure the nickel content of taenite _in situ_ in iron meteorites. Lovering, _et al_.

(1960) employed this technique in a novel application to determine the thickness of reaction rims resulting from ablation heating in two nickel-rich ataxities. These data coupled with their theoretical calculations permitted estimation of ablation mass loss in these meteorites. Lovering and Parry (1962) first demonstrated the utility of the thermomagnetic technique for the identification of the magnetic phases present in meteorites. Stacey, et al. (1961) determined the magnetic content of the Mokoia meteorite with this technique. Later meteorite magnetism studies incorporated thermomagnetic analysis solely for the qualitative identification of the magnetic phases present (Banerjee and Hargraves, 1972; Butler, 1972; Gus'kova, 1963, 1965a,b; Weaving, 1962a).

Prior to 1959 virtually nothing was known about the magnetic properties of meteorites and their magnetic record. Substantial progress has been made, however, following the earliest reported investigations of Lovering (1959) and Stacey and Lovering (1959). It was established in these studies and those which followed that many meteorites possess a stable magnetic moment of extraterrestrial origin. The intensity of the magnetic field under which the remanent magnetization (NRM) formed was estimated to be only slightly less that the present terrestrial field (Stacey, et al., 1961; Weaving, 1962a,b; DuBois, 1965). During the past decade, Russian scientists (primarily Gus'kova and Pochtarev) conducted an impressive series of magnetic studies in which they measured the NRM and magnetic susceptibility in over 900 meteorite samples located in museums throughout the Soviet Union (Gorshkov, et al., 1972; Gus'kova, 1963, 1965a, b,c, 1969a,b, 1970; Gus'kova and Pochtarev, 1967, 1969; Kashkai, et al., 1961; Pochtarev, 1967; Pochtarev and Gus'kova, 1962). The Russian scientists, in the papers referred to above , also reported more sophisticated studies involving both thermal and alternating field demagnetization on about 20 meteorites. Herndon, et al. (1972) recently reviewed the Russian work on meteorite magnetism.

As with most meteorite studies, almost all of the research efforts cited so far were concentrated on ordinary chondrites and iron meteorites - the two most abundant classes of meteorites. Only Stacey, et al. (1961) performed thermal demagnetization experiments on a carbonaceous chondrite, Mokoia. Unlike the ordinary chondrites studied, Stacey, et al. (1961) reported that Mokoia gave no indication of a magnetic moment of extraterrestrial origin and suggested that the small observed moment which disappeared after heating to only 200°C was the result of isothermal induction in the earth's magnetic field. Their interpretation of the Mokoia data admitted two possibilities. Either Mokoia was never heated above 200°C, or if it was, then it cooled in the absence of any significant magnetic field.

The apparent lack of emphasis on the magnetization of carbonaceous chondrites during the 1960's has been more than counterbalanced by the magnetic studies during the early 1970's. Virtually every study during the past three years has focused on this important class of meteorites.

Banerjee and Hargraves (1971) were the first of the recent investigators to concentrate their interest on carbonaceous chondrites. In contrast to Stacey, et al.'s (1961) observation on Mokoia, Banerjee and Hargraves found that the four carbonaceous chondrites they examined held evidence of a stable magnetic moment of extraterrestrial origin. More recent studies from other laboratories (Banerjee and Hargraves, 1972; Brecher, 1972; Butler, (1972) as well as from our own (Larson, et al. 1973) confirm Banerjee and Hargraves' observation that most, if not all, carbonaceous meteorites have recorded evidence of a magnetic field(s) of extraterrestrial origin.

Presently all techniques employed in meteorite magnetism studies have been adopted virtually unchanged from terrestrial paleomagnetism. In terrestrial magnetic studies, however, the primary emphasis is placed on determining the direction, rather than the intensity, of the earth's magnetic field in the geological past. As directions in meteorites are necessarily arbitrary (except within a single meteorite specimen) the primary goal of meteorite magnetism studies is to determined the intensity of magnetic fields operating in the early solar system. With due regard for the divergent formative processes experienced by meteorites and earth rocks (and for the observed chemical and petrological differences, as well), it might be expected that at some point terrestrial magnetism techniques and/or interpretations may prove inadequate in meteorite magnetism studies.

The proposed intent of this dissertation research was to investigate the stability of the natural remanent magnetization observed in carbonaceous meteorites and to investigate the thermal stability of their magnetic components. The latter aspect is important as all presently known techniques employed to estimate the intensity of magnetic fields in the early solar system involve heating of the meteorite samples. This research is necessary in order to properly access previous work and to provide a firm foundation upon which to base future studies. While fulfilling the proposed intents, the results of this investigation have yielded previously unattained information on the occurrence of magnetite (Fe_3O_4) in carbonaceous chondrites. Furthermore, the results of the thermal stability studies have led this author (Herndon, 1973) to propose that meteorite magnetite formed from the oxidation of troilite [(Fe, Ni)S] rather than from the reaction of water with metallic iron as suggested by Urey (1952). The formation of high nickel pyrrhotite [$(Fe, Ni)_{1-x}S$] is seen as a direct consequence of magnetite formation from the mild oxidation of troilite. No adequate genesis has previously been proposed for meteorite pyrrhotite (Kerridge, 1972).

GLOSSARY OF MAGNETISM TERMINOLOGY

The magnetization observed in meteorites is a complex phenomenon which depends upon (1) the intensity of the magnetic field(s) to which the meteorite or its mineral phases was exposed, (2) the temperatures to which the meteorite was subjected, particularly during and after exposure to magnetic fields, (3) the magnetic minerals in the meteorite, and (4) the processes by which these minerals became magnetized. Following the initial recognition of the potential of studies of terrestrial rock magnetism [see, for example, Chevallier (1925); Mercanton (1926); Matuyama (1929), notable progress has been made in understanding the processes by which rocks (and later on, partially by analogy, meteorites) may become magnetized. To foster more complete understanding of the processes, numerous laboratory techniques have been developed through the years to help resolve the complex magnetization record stored in the sample and thereby release new information concerning physical processes operating in the early solar system.

Virtually all the magnetism in meteorites is carried by magnetite and nickel-iron alloys, although other minerals such as schreibersite and magnetite sulfide phases may also contain significant information. The identification and study of minor magnetic mineral components in meteorites is an area which has not been investigated to any extent.

Any grain of a magnetic mineral is said to consist of one or more magnetic domains. Sufficiently small grains (~ 10^2Å) can be regarded as single domains. Separating the domains are domain walls which may be considered as magnetic energy barriers. For a given magnetic mineral within a particular sample, the intensity of the magnetization per unit volume is the same in all domains although the directions of the magnetization are not necessarily the same.

In the absence of an applied external magnetic field, the direction of magnetization of each domain of a magnetic mineral will be preferentially oriented along one of several axes which is determined by the shape of the grain and/or by the crystalline anisotropy of the mineral. In an applied magnetic field the direction of the magnetization of a domain will be changed in such a way that it will tend toward alignment with the direction of the applied field. If the magnetic field is relatively weak, the direction of magnetization of the domain will reversibly return to its initial preferred direction after removal of the external field. Such reversible magnetization is termed induced magnetization. If, however, the energy supplied by the applied field is greater than the magnetic energy barrier of the domain, the direction of magnetization of the domain will not return to its preferred position when the applied field is removed. This is remanent magnetization, (NRM). The coercive force is defined as the value of the applied field when the irreversible transformation takes place.

If a meteorite specimen is allowed to remain at room temperature in a sufficiently strong magnetic field, it may acquire a remanent magnetization in the direction of the applied field provided that the field is larger than the lowest coercive force of its magnetic minerals. This is called isothermal remanent magnetization (IRM).

If the magnetic field is weaker than the lowest coercive force of the magnetic minerals present and if the meteorite specimen remains for a long period of time in some fixed orientation with respect to the direction of the field, the weak field will effect a bias on the normally random thermal agitation which may allow the magnetization of the domains to cross over energy barriers otherwise unexpected in such a weak field. This magnetization is acquired slowly and, when the applied field is removed, the magnetization of the domain will reversibly return to its initial preferred direction on a time scale comparable to its acquisition. This is viscous remanent magnetization (VRM).

Aside from the effects caused by an externally applied magnetic field on the magnetization of a mineral such as described above, one must consider the effect of temperature on the magnetization. If a magnetic mineral, carrying a remanent magnetization is heated, the thermal energy increases until it equals the magnetic barrier energy at which point the magnetic moments become randomized. The temperature at which this occurs is characterized of the particular mineral involved and is called the Curie point or Curie temperature. If a magnetic mineral cools through its Curie point in the presence of a magnetic field, at the Curie temperature it begins to develope a magnetization in the direction of the applied field. This magnetization, for weak fields comparable in magnitude to the earth's field, is proportional to the intensity of the external field. This is an important characteristic which allows estimation of the magnetic field intensities in the early solar system. A magnetization acquired as the mineral cools through its Curie temperature is called thermal remanent magnetization (TRM).

If a magnetic mineral is heated to some temperature below the Curie temperature, then cools in a magnetic field, a partial thermal remanent magnetization (PTRM) forms. Reheating the mineral in the absence of an external field will cause the magnetization to disappear in the same temperature range that it was acquired. Thus a PTRM preserves a faithful memory of the temperature and external field which intially caused the acquisition of the PTRM. This is an important characteristic of thermal demagnetization analysis which may allow estimation of paleotemperatures in some meteorites.

Chemical changes taking place in the presence of an external magnetic field can cause newly created magnetic mineral grains to grow in alignment with the direction of the applied field and acquire a magnetization. This is termed chemical remanent magnetization (CRM) and may take place for either non-magnetic or magnetic minerals undergoing reactions at low temperatures which change them to different magnetic minerals. The character of this magnetization is virtually indistinguishable from a TRM. If sufficient heating is involved a combined TRM and CRM may result. This is thermochemical *remanent magnetization* (TCRM).

If the low temperature compaction of a meteorite such as a carbonaceous chondrite were to involve very small grains which had previously acquired a TRM, it is possible for these grains to become aligned with any external magnetic field operating at the time. This could lead to a net magnetization of the compact body, called *depositional remanent magnetization* (DRM). Since the grains originally had a TRM, the DRM described above would have the characteristics of a TRM. The relative stabilities of the different types of a remanent magnetization discussed above are VRM < IRM < TRM ≈ CRM ≈ DRM.

All the above types of remanent magnetization are generally acquired in weak magnetic fields (~0.5 Oe). This should be distinguished from laboratory induction in very large fields (~1000 Oe or greater) which results in *saturation magnetization* (J_s). The temperature dependent behavior of the force of attraction between the magnetic components in the meteorite sample and a very strong magnetic field (displayed graphically in a J_s-T plot) forms the basis for thermomagnetic analysis (discussed below).

For the convenience of the reader, abbreviations frequently used in this text are set forth in Table 2.

Table 2. Abbreviations used in this dissertation

Abbreviation	Terminology
CRM	Chemical remanent magnetization
DRM	Depositional remanent magnetization
IRM	Isothermal remanent magnetization
J_s	Saturation magnetization
NRM	Natural remanent magnetization
PCRM	Partial chemical remanent magnetization
PTRM	Partial thermal remanent magnetization
TRM	Thermal remanent magnetization
VRM	Viscous remanent magnetization

ALTERNATING FIELD DEMAGNETIZATION OF NATURAL REMANENCE

Since the total NRM is generally a combination of components (isothermal and viscous as well as the more stable components), and since some of these components (IRM and VRM) are frequently only a complication in an attempt to unravel the magnetic conditions existing in the early solar system, it is desirable to remove those nuisance components prior to more extensive thermal experiments. This can be accomplished quite simply by subjecting the sample to an alternating magnetic field.

Figure 1 shows the relative stability to alternating field demagnetization of isothermal remanent magnetization (IRM) acquired in a 30 Oe field with that of thermal remanent magnetization (TRM) acquired in a 0.5 Oe field. Viscous remanent magnetization (VRM), which is not shown in Fig. 1, is generally less stable to a.f. demagnetization than isothermal remanent magnetization. It can be seen that as the intensity of the alternating field is increased, the less stable components (IRM and VRM) are erased to a much greater extent than the more stable components (TRM). Furthermore, by noting relative change in intensity of the magnetization vector during a.f. demagnetization, it is possible to gain some insight into the stability and nature of the NRM. Because the NRM of meteorite samples is generally a combination of soft (IRM and VRM) and hard (TRM) components, additional information can be gained by noting changes in the direction of the magnetization vector. The NRM is

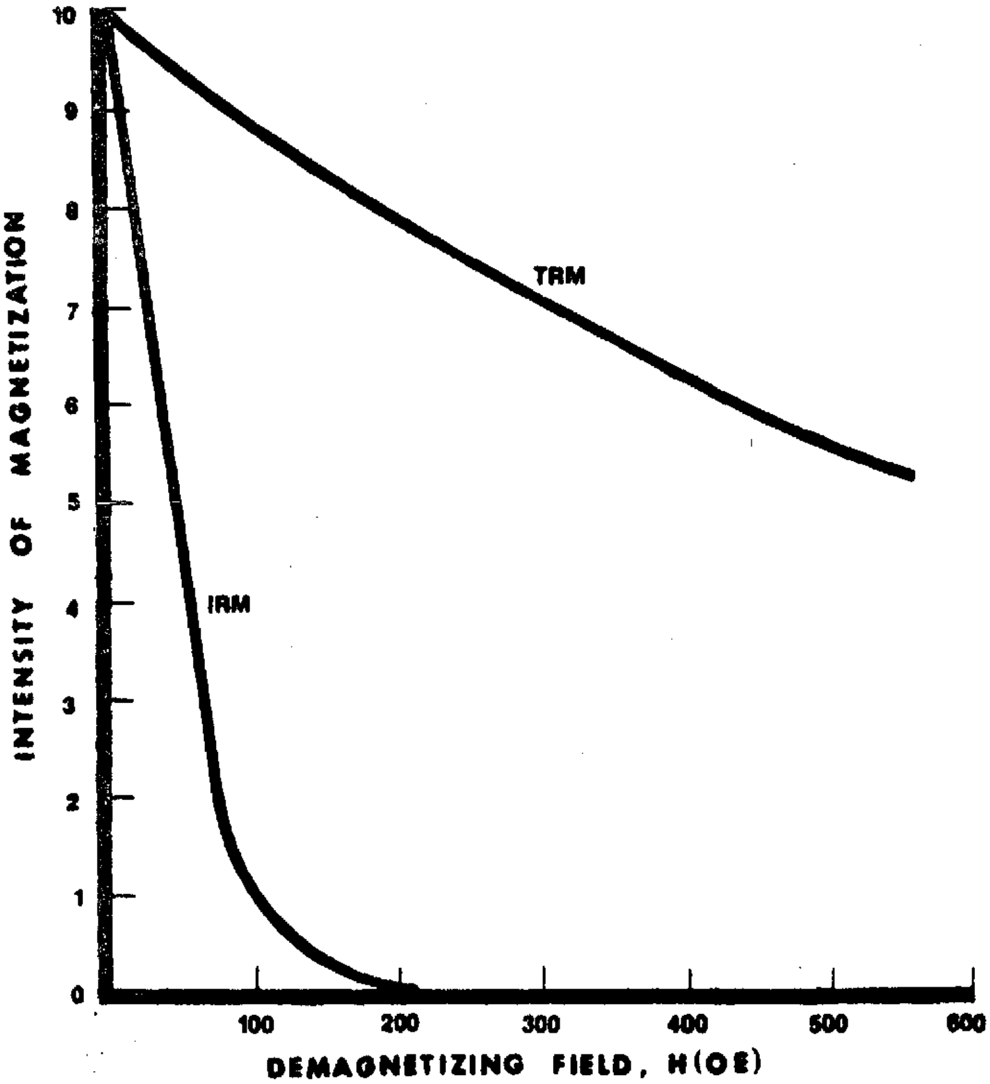

Fig. 1. Relative intensity plot of alternating field demagnetization of thermal remanent magnetization compared with isothermal remanent magnetization.

the vector sum of all the magnetization components. The directions of these components are not necessarily the same. As the soft (less stable) components are erased, their contribution to the direction of the resultant magnetization vector will be removed and the NRM vector may change in direction, ultimately yielding the direction of the resultant magnetization vector of the hard (more stable) components. This technique is used routinely in virtually all magnetic studies and is called either alternating field (a.f.) or alternating current (a.c.) demagnetization.

The ability of a meteorite sample to acquire a soft magnetization (IRM or VRM) depends in part on the relative coercivities of its magnetic minerals. Erratic behavior during a.f. demagnetization may be indicative of minerals with low coercivities which may readily acquire a magnetic moment quickly at ambient temperature from the earth's magnetic field. Such behavior can be demonstrated by storage tests. If a meteorite sample is demagnetized (say to 300 Oe), allowed to remain in the earth's field for several days and remeasured, then a change in the intensity and/or direction of the magnetization vector would indicate that the meteorite sample contains magnetic minerals of low coercivities which can readily acquire a magnetic moment from the earth's magnetic field. Again demagnetizing the sample to 300 Oe would erase the newly acquired soft component and cause the recovery of the direction and intensity that the sample had after the first 300 Oe demagnetization.

The alternating field demagnetization technique, being non-destructive (except as its alters the magnetization), is ideally suited to rare meteorites such as carbonaceous chondrites and ureilites. This technique is useful to gain some insight into the character of the NRM - its nature, direction, stability, and number of components. This information is important in considering which samples should be sacrificed to further (destructive) studies such as thermal demagnetization.

Experimental Procedure: All meteorite samples were removed at least a few mm from the fusion crust to avoid effects of albation heating. The samples, weighing between 0.1 and 1.9 grams, were secured in a plastic sample holder and the intensity of remanent magnetization (NRM) was measured on a 10 Hz spinner magnetometer with a sensitivity of about 5×10^{-6} cgs/cc.

For a.f. demagnetization the samples (still secured in the sample holder) were placed in a 3-axis tumbler (designed by D. E. Watson) within a relatively long solenoid (~25 cm) and tumbled to randomize the sample orientation with respect to the applied field. After a few minutes exposure to a 60 Hz alternating field of predetermined magnitude, the solenoid current was allowed to slowly decay to zero. The tumbler was then stopped, the sample (and sample holder) removed and the NRM remeasured. This procedure was repeated at successively higher alternating fields. Meteorites which displayed an intense NRM were subjected to demagnetizing fields up to

400 Oe. Those with weaker intensities were either demagnetized to a lesser extent or not demagnetized at all. It is possible to completely demagnetize a sample by increasing the field to large enough values. Our use of this technique, however, was simply to remove any unstable nuisance magnetization components which might have been present to simply future thermal demagnetization studies and to investigate the relative stability of the NRM to a. f. demagnetization.

Method of Data Presentation: The a.f. demagnetization data is presented here both in terms of intensity and direction. The intensity is plotted against the value of the applied alternating field. Magnetization, being a vector quantity, necessarily involves direction. But since absolute directional orientation is impossible, an arbitrary orientation was chosen and relative changes in the direction of magnetization were observed. This data is presented here on an equal-area projection.

To understand the equal-area projection, one envisions the meteorite specimen at the center of a sphere. Superimposed on the meteorite is a standard cartesian reference coordinate (X, Y, Z). The magnetization vector in the YZ plane is measured in the X' direction as an angle, θ, relative to the reference coordinate, X. A YZ plane passing through the center of the sphere oriented at an angle θ from X(i.e. in the X' direction) will describe a great circle on the surface of the sphere. Great circles for the Y' and Z' directions are similarly drawn. These three great circles will intersect at a point on the surface of the sphere. A line drawn from the center of the sphere to the point of intersection of the three great circles represents the direction of magnetization of the eteorite specimen relative to the reference coordinate system. The equal-area projection is the projection of this sphere into two dimensions. The convention followed here is to use open circles to represent a vector component out of the page and closed circles to represent a vector component directed into the page.

Results and Discussion: As seen in Figs. 2 - 4, the carbonaceous meteorites exhibit initial NRM intensities which vary over almost four orders of magnitude. The Renazzo and Essebi carbonaceous chondrites and the Goalpara carbonaceous achondrite exceed 5×10^{-2} emu/g in intensity whereas our sample of the Warrenton carbonaceous chondrite was below the limit of detection. While this wide variation in initial NRM intensities may be in part the result of the magnetization having been acquired in fields of significantly different strengths, or of the presence of multiple components of magnetization, it is more likely a reflection of the relative amounts of the various magnetic phases in these meteorites.

Almost all of the carbonaceous meteorites studied here possess an NRM which is stable with respect to both intensity and direction during alternating field demagnetization up to 400 Oe. The small movement in direction during the early stages of demagnetization

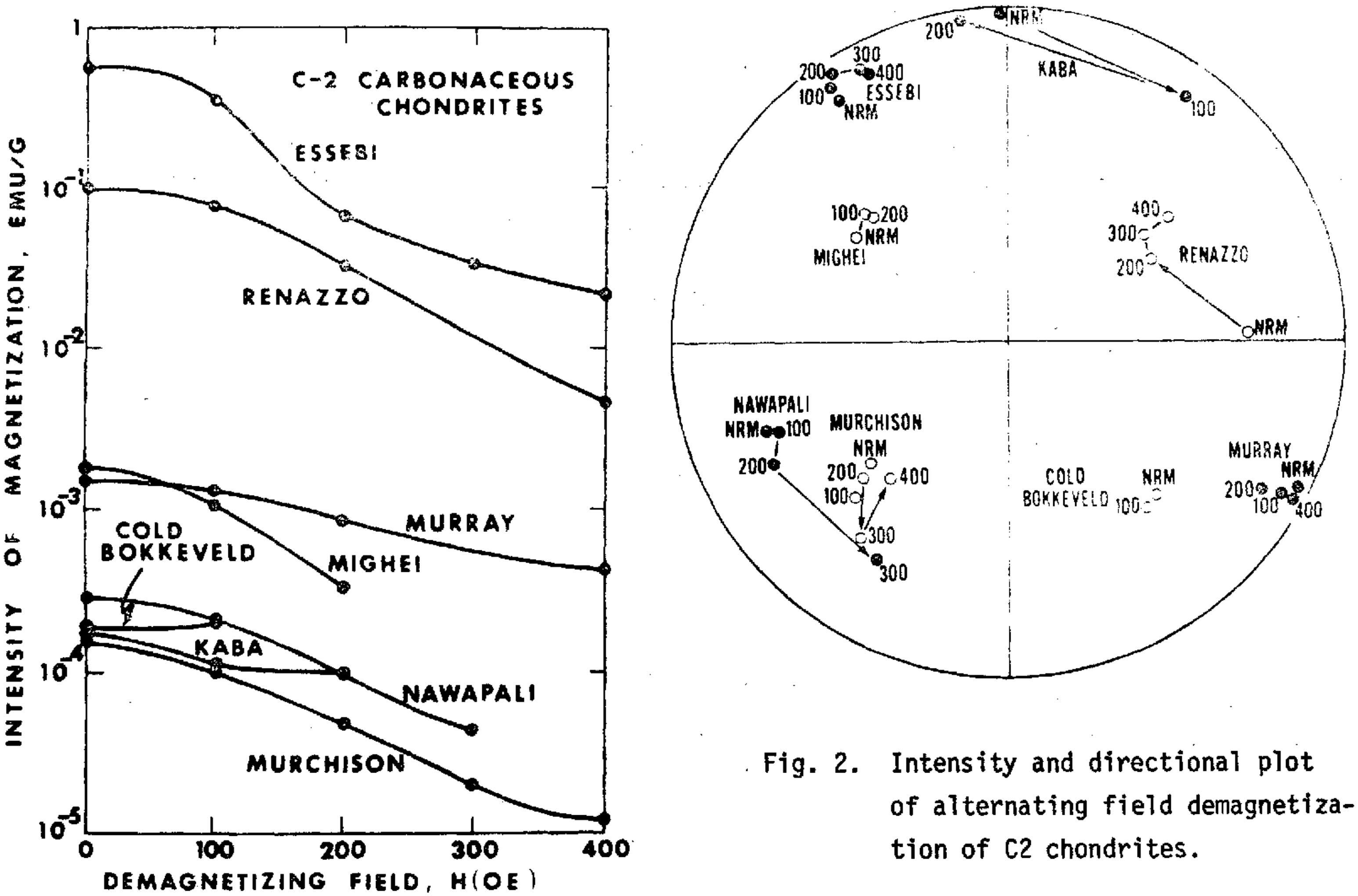

Fig. 2. Intensity and directional plot of alternating field demagnetization of C2 chondrites.

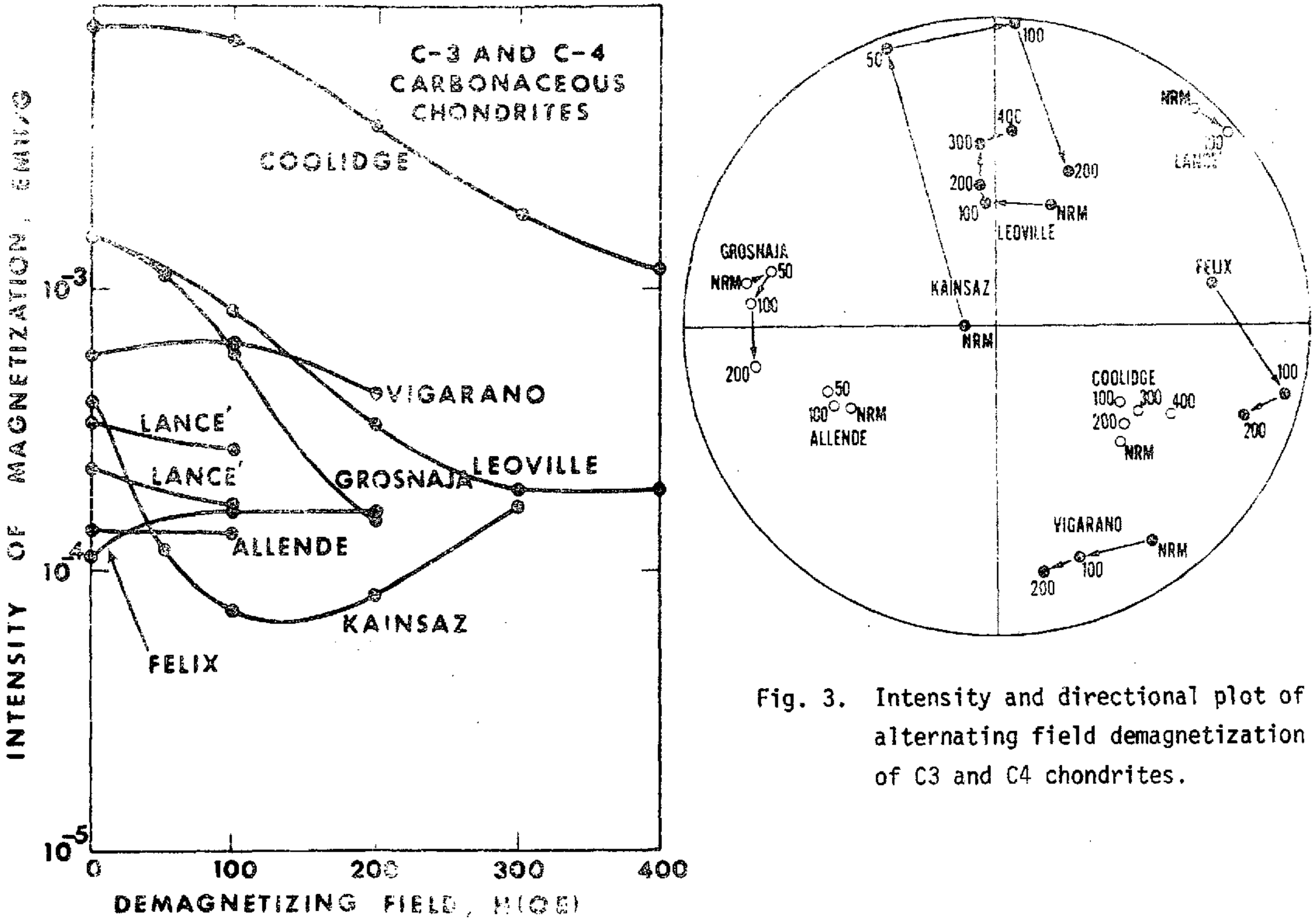

Fig. 3. Intensity and directional plot of alternating field demagnetization of C3 and C4 chondrites.

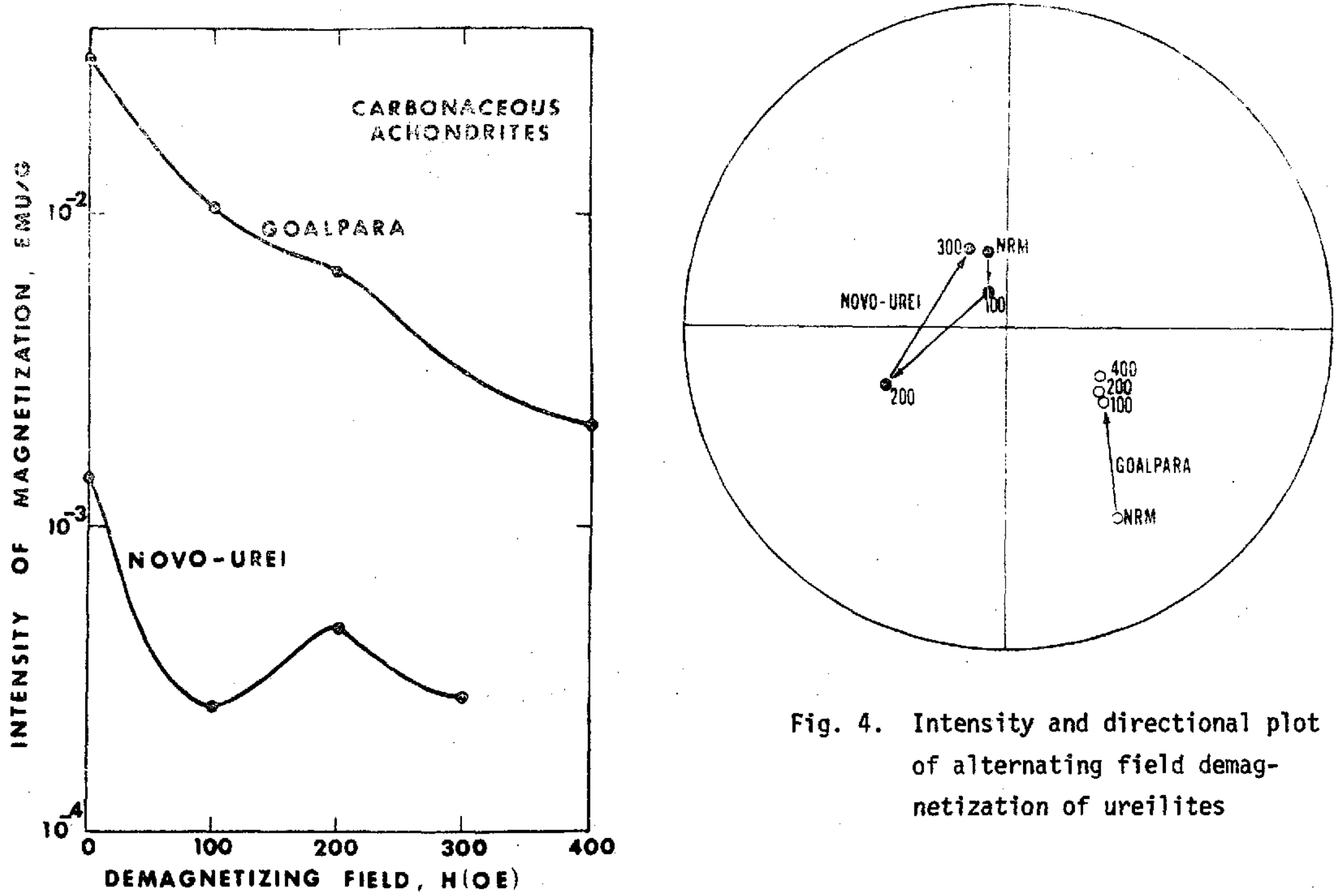

Fig. 4. Intensity and directional plot of alternating field demagnetization of ureilites

observed in some samples is most likely indicative of a small component of viscous remanent magnetization (VRM) which may have been acquired subsequent to the meteorite fall, perhaps in the museum or laboratory. Only the Kainsaz and Felix C3 chondrites (Fig. 3) show directional and intensity evidence strongly indicative of a secondary component. The Novo Urei carbonaceous achondrite (Fig. 4) displays a dip in intensity and an excursion in direction. Subsequent recovery to the initial direction indicates that this is probably the result of a spurious component picked up during demagnetization. Storage tests indicate that Kainsaz and Novo Urei contain components of low coercivity which acquire a moment readily in the earth's field. In general, however, the carbonaceous meteorites shown in Figs. 2 - 4 exhibit principally only one direction of magnetization carried by both lower and higher coercivity magnetic grains. This suggests a thermal remanent magnetization (TRM), thermochemical remanent magnetization (TCRM), or chemical remanent magnetization (CRM), acquired prior to falling to earth.

Previous studies on meteorites (summarized by Anders, 1971) have presented several strong lines of evidence supporting the contention that some carbonaceous chondrites, including at least some of the C2's shown in Fig. 1 were never heated above 250°C. Further evidence suggests that the mineralogical components of carbonaceous chondrites, including at least some of the C2's shown in Fig. 1, were never heated above 250°C. There is also evidence that suggests that the mineralogical components of carbonaceous chondrites respresent relatively unaltered "planetary material" remaining from the early solar system formation (see e.g., Wood, 1967). It is possible that the magnetization of carbonaceous meteorites may be a depositional remanent magnetization (DRM) of materials with a previously acquired TRM. In that case, the TRM would probably of necessity have been acquired during the condensation of the solar nebular dust - occurring therefore before the formation to the planetary bodies of our solar system. A DRM may, however, imply the existence of a magnetic field during the formation of these meteorites. Additional knowledge of the nature of the NRM and its temperature history of acquisition can be obtained through thermal demagnetization studies.

THE THERMAL STABILITY OF THE MAGNETIC CONSTITUENTS OF CARBONACEOUS METEORITES

Thermomagnetic analysis is a technique employed in virtually all magnetic studies for the identification of magnetic components in the samples (see, for example, Banerjee and Hargraves, 1972; Butler, 1972). While frequently this technique is applied simply for qualitative identification, the use of a calibrated system permits quantitative determinations as well. In principle the technique is deceptively simple. A sample of material is suspended from a sensitive balance. Application of a large (~3 kOe), inhomogeneous magnetic field to the sample results in a force of attraction (Faraday effect) between the magnetic components in the sample and the magnetic field. This force of attraction causes the balance to record an apparent weight which is many times that of the actual sample weight. The weight recorded depends on the specific magnetic components present and the respective amount of each present in the sample. For example, 1 mg of metallic iron would appear to weigh about 230 mg with application of the magnetic field. Similarly, 1 mg of magnetite would appear to weight about 95 mg. This apparent weight is effectively a measure of the saturation magnetization. For any given magnetic mineral, the saturation magnetization is temperature dependent - diminishing with increasing temperature until it essentially vanishes at the Curie point. Thus, heating the sample while applying the magnetic field causes the apparent weight to decrease with increasing temperature. At temperatures above the highest Curie temperature of the magnetic components in the sample, the apparent weight is virtually identical to the real sample weight.

Aside from its utility as a method for identifying magnetic components thermomagnetic analysis permits observation of the behavior of magnetic components in the sample upon heating. The significance of this aspect of the technique cannot be overstated. At the present time, all laboratory techniques designed to arrive at an estimate of the intensity of magnetic fields in the early solar system from the remanent magnetism in meteorites are based on the Thellier technique.

In their classic work, Thellier and Thellier (1959) demonstrated that a PTRM obtained in a weak magnetic field develops a memory characteristic with regard to heating, i.e. a rise in temperature from t_2 to t_1 destroys precisely that portion of the magnetization which developed during the original cooling period within the temperature interval from t_1 to t_2. They further demonstrated that for experiments conducted in weak fields comparable in magnitude to the earth's magnetic field (~0.5 Oe), there is a linear relationship between the loss of magnetization in a given temperature interval during laboratory reheating conducted in field free space and the acquisition of magnetization obtained cooling through the same temperature interval in a magnetic field of known intensity. This linearity permits estimation of the intensity of the magnetic field responsible for the initial implantation of the NRM observed in meteorites. At the present time, all methods of paleointensity estimation are based on the Thellier technique (or some variant thereof) and necessitate laboratory reheating of the meteorite samples. The validity of the Thellier technique is dependent on the magnetic constituents of the sample undergoing no significant thermal alteration.

Thermomagnetic analysis is an ideal technique to assess the degree of thermal alteration during laboratory heating of the magnetic phases present in meteorites. The relationship between saturation magnetization and temperature is well known for magnetite and Ni-Fe alloys, the major magnetic constituents of meteorites (Bozorth, 1951). Deviation from the known behavior observed during thermomagnetic anslysis of real meteorite samples is indicative of thermal alteration. Furthermore, thermomagnetic analysis typically necessitates destruction of minute quantities of material (≤ 1 mg). This is contrasted to sample size of ~1 gram typically destroyed during Thellier-type experiments. Thus thermomagnetic analysis is ideally suited as a prelude to other destructive studies.

As one might expect, heating meteorite samples (particularly carbonaceous chondrite material) to temperature as high as 800°C poses major problems - notably oxidation of the magnetic components. Workers in other laboratories have attempted to minimize thermal alteration by conducting thermal experiments under vacuum (Banerjee and Hargraves, 1972; Butler, 1972) or under inert gas (Brecher, 1972). We will demonstrate in the discussion which follows that such techniques are adequate for materials containing only magnetite, but are not satisfactory for materials (such as some carbonaceous meteorites) containing metallic iron in a finely divided state. While some of our studies were conducted under vacuum, the majority were conducted in a mixed gas atmosphere with continuous monitoring of the oxygen fugacity in order to maintain the magnetic components in their stability field. We will demonstrate the advantages of this system over conventional vacuum techniques.

Experimental Procedure: Thermomagnetic analyses, or J_s-T studies, were conducted using a continuously recording Cahn electrobalance equipped with a 3 kOe permanent magnet. Provision was made for vacuum operation for occasional studies. This system is shown in Fig. 5, and is described by Larson et al. (1974). Generally, unless specifically noted in the following discussion, a gas mixing system was employed utilizing a continuously variable gas flow of H_2 and CO_2 in N_2 carriers to yield the proper oxygen fugacity at any instantaneous temperature in order to maintain the magnetic components of the sample in their stability field. The oxygen fugacity was monitored on a real-time basis by employing a yttria stabilized zirconia electrolyte probe according to the technique of Sato (1970). The probe calibration was checked against the known f_{O_2} stability fields for the mineral pairs: Fe-Fe_3O_4; Ni-NiO; and Fe_3O_4-Fe_2O_3 (Fig. 6). It was possible to monitor the equilibrium condition for these pairs with the Curie balance since at any given temperature, the saturation magnetization of Fe > Fe_3O_4 > Fe_2O_3 > FeO and

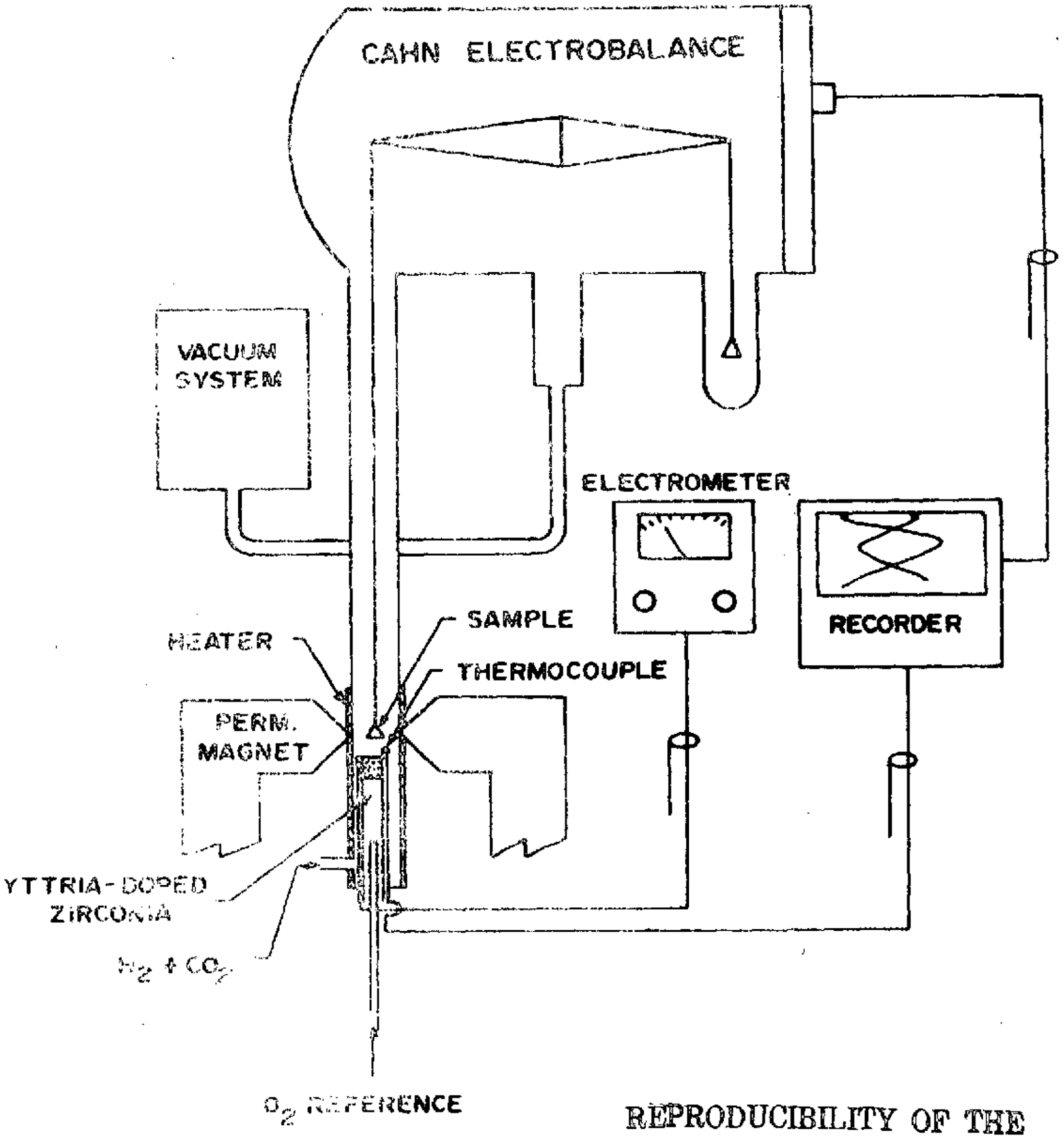

Fig. 5. Schematic drawing of the thermomagnetic system with gas-mixing capabilities.

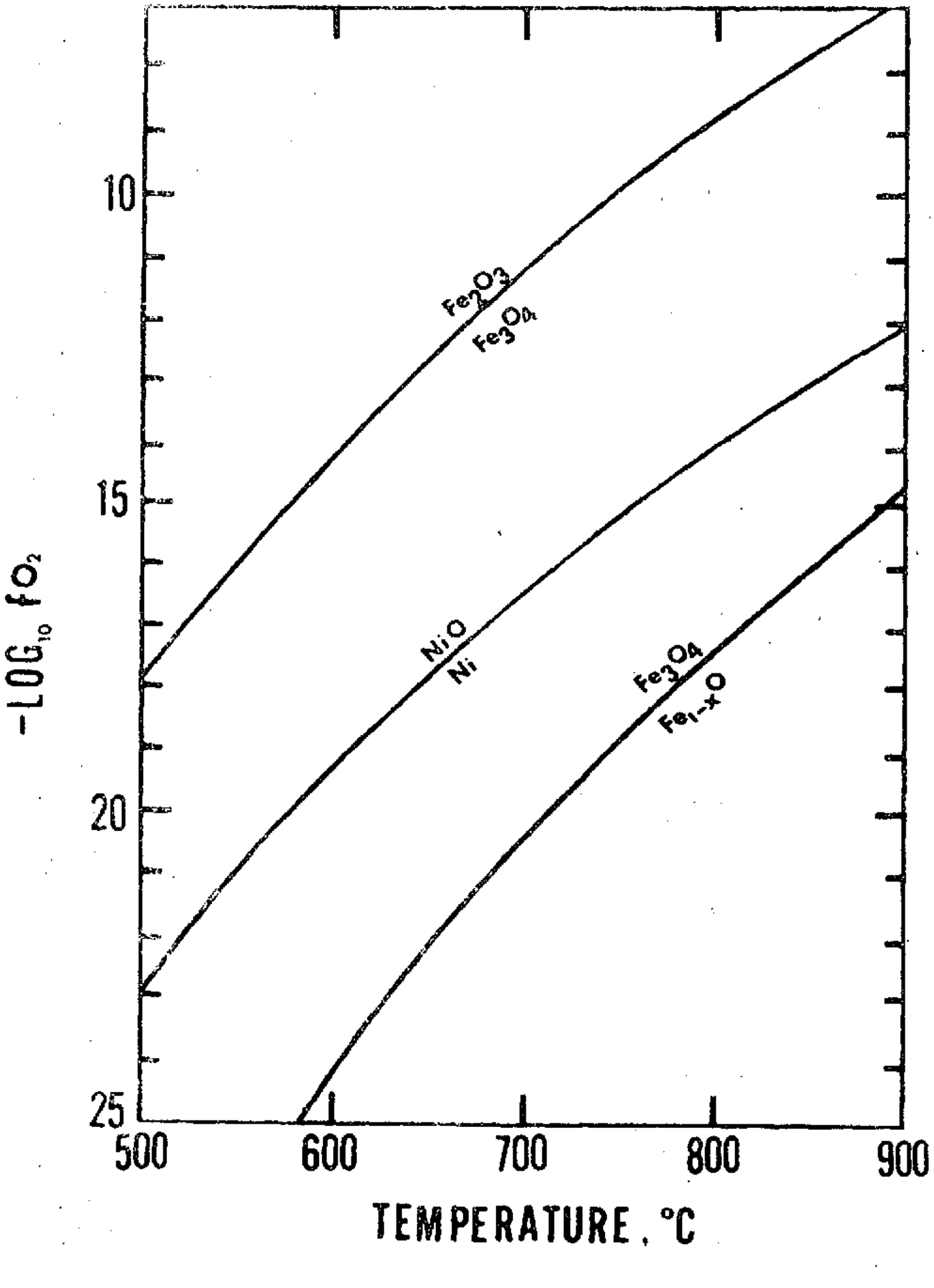

Fig. 6. Oxygen fugacity - temperature stability relationships for mineral buffer pairs. After Buddington and Lindsley (1964).

Ni > NiO. Consequently, any change in the relative concentration of the individual components of any given mineral pair is reflected by a change in the saturation magnetization. Thus, by holding the temperature constant and intentionally altering the gas mixture to create an atmosphere which is either oxidizing or reducing, it is possible to observe mineral synthesis on a real-time basis in the Curie balance as one magnetic component is changed into another with different net saturation magnetization. The advantage of this aspect of the experimental procedure will become obvious in the discussion which follows.

Since the thermomagnetic studies reported in this work were obtained with a new technique, that of employing a gas mixing system for oxygen fugacity control, it is proper before discussing the meteorite results to provide some data which justifies our choice of the system. Figure 7 is the J_s-T curve for iron powder (< 46 μ) which was run under a vacuum (4×10^{-4} torr). The sample was heated to 680°C on the first cycle and then cooled. The lower saturation magnetization observed upon the first cooling is indicative of the oxidation of Fe to Fe_3O_4. This is further verified by the second cooling cycle, the Curie point being indicative of magnetite. Complete oxidation is confirmed by comparing the relative saturation moments at room temperature between the initial iron and final magnetite. This behavior is obviously unsatisfactory if one is dealing with a sample containing iron, especially of small grain size such as found in ureilites and some C2 and C3 chondrites.

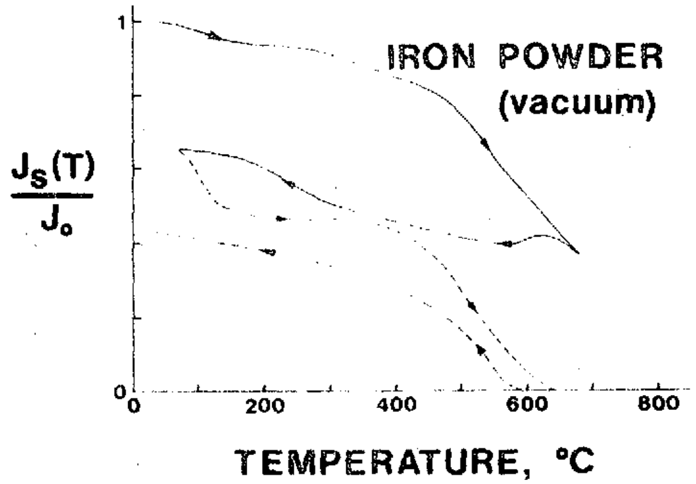

Fig. 7. J_s-T curve for iron powder run in a vacuum (4×10^{-4} torr). The broken curve indicates the second thermal cycle.

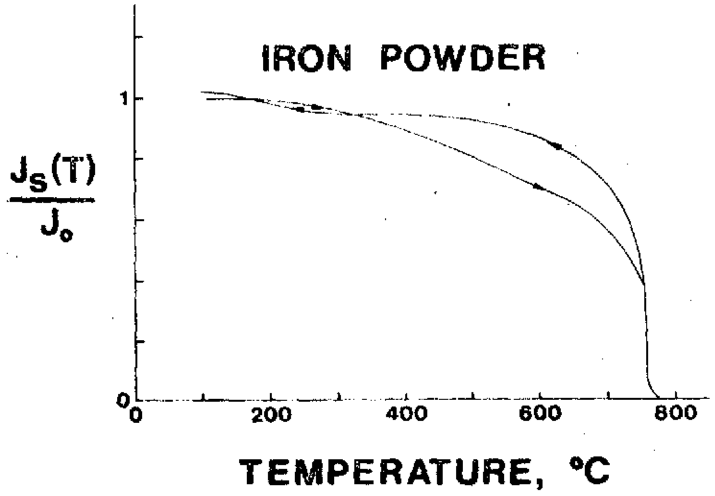

Fig. 8. J_s-T curve for iron powder run at one atmosphere in 8 1/2% in N_2 carrier.

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Figure 8 is the J_s-T curve for iron powder run under stability conditions in our controlled atmosphere (in this case using only 8 - 1/2% H_2 in N_2). The slight decrease in the saturation moment on the heating cycle between 400° and 750°C is probably due to a minor amount of oxidation occurring even in this atmosphere. Undoubtedly a more reducing atmosphere would have yielded more coincident heating and cooling paths. The small amount of magnetite formed during heating appears to have been reduced back to iron upon cooling. The behavior in our atmosphere is far superior to vacuum conditions and is certainly adequate for most purposes. This represents an extreme case, the finely divided powder presenting a large surface for possible oxidation. Better behavior might be expected for meteorite materials run as whole pieces as is demonstrated by the data shown in Fig. 9, the J_s-T curve for the Dyalpur ureilite. The only magnetic component indicated by this data is metallic iron of low nickel content. Figure 10 is the J_s-T curve for synthetic magnetite ($<5\ \mu$) run under a vacuum (4×10^{-4} torr). The unusual behavior upon initial heating is possibly due to rearrangement in the crystal structure of the synthetic magnetite. Aside from this minor anomaly, the behavior is adequate, indicating that even the relatively poor vacuum used resulted in an oxygen fugacity that is approximately in the stability field for magnetite. This further affirms our observations from the iron run under vacuum (Fig. 7). Thus, for meteorite samples containing only magnetite, vacuum techniques appear to be satisfactory. The gas mixing technique is at least equally

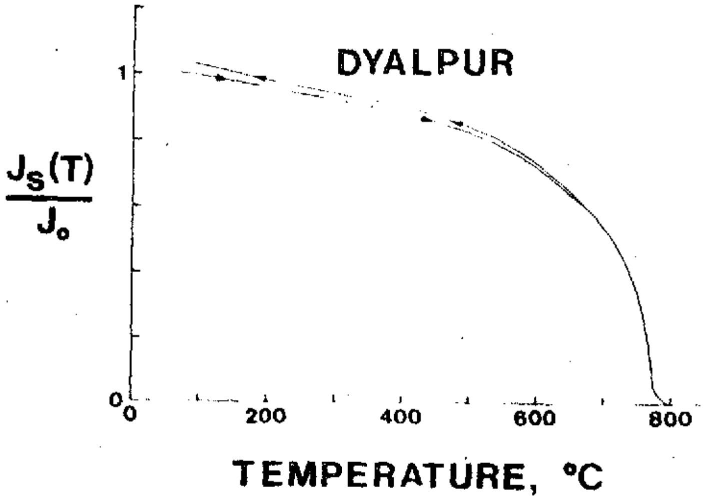

Fig. 9. J_s-T curve for Dyalpur.

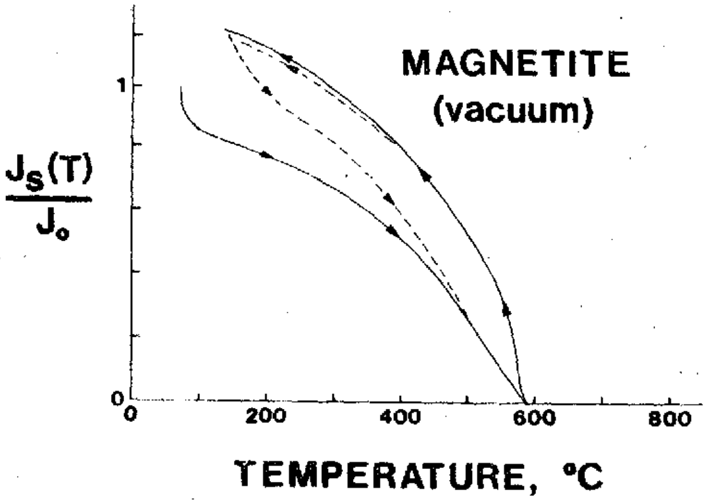

Fig. 10. J_s-T curve for synthetic magnetite run under vacuum (4×10^{-4} torr). The broken curve indicates the second thermal cycle.

satisfactory for meteorite samples containing only magnetite as is demonstrated by the data depicted in Fig. 11. This figure, the J_s-T curve for the Orgueil Cl chondrite, shows that magnetite is the only magnetic component present. Minimal alteration of the magnetite is observed despite having taken this sample through two complete thermal cycles. The major advantage of our controlled atmosphere, i.e., maintaining stability for samples containing both magnetite and iron, is demonstrated by the data depicted in Fig. 12, the J_s-T curve for the Novo Urei ureilite. It can be seen from this figure that two components are present. Upon heating the saturation magnetization decreases rather abruptly at about 580°C as the temperature rises above the Curie point of magnetite. Above that temperature we observe a second component, metallic iron, which is indicated by the Curie temperature of 770°C. This component is almost pure iron ($\leq$ 2% Ni) as indicated by the Curie temperature and lack of $\gamma \rightarrow \alpha$ transition. The most significant aspect of the J_s-T curve in Fig. 12 is demonstrated by the near coincidence of the heating and cooling curves. This near coincidence attests to our ability to heat meteorite samples (typically $\leq$ 1 mg) to temperatures of ~800°C without significantly changing the oxidation state of the magnetite and metallic iron present. As noted previously, the saturation magnetization of iron is greater than that of magnetite. Thus, any change in the relative amount of iron and magnetite present in the Novo Urei sample (due to oxidation or reduction) would have caused the cooling curve to deviate significantly from the heating

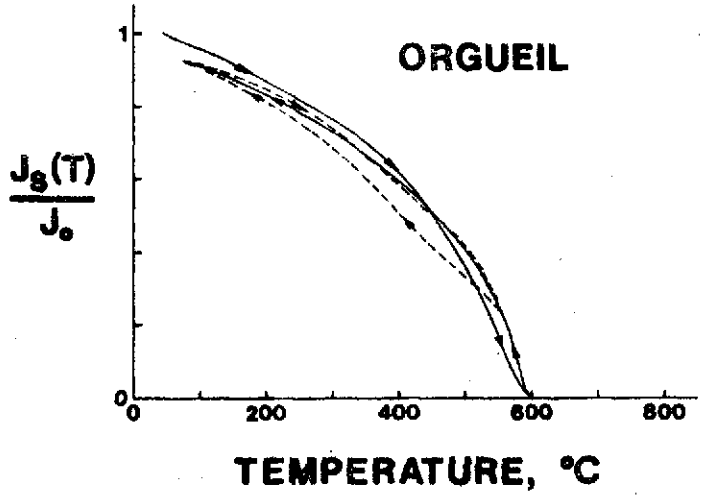

Fig. 11. J_s-T curve for Orgueil. The broken curve indicates the second thermal cycle.

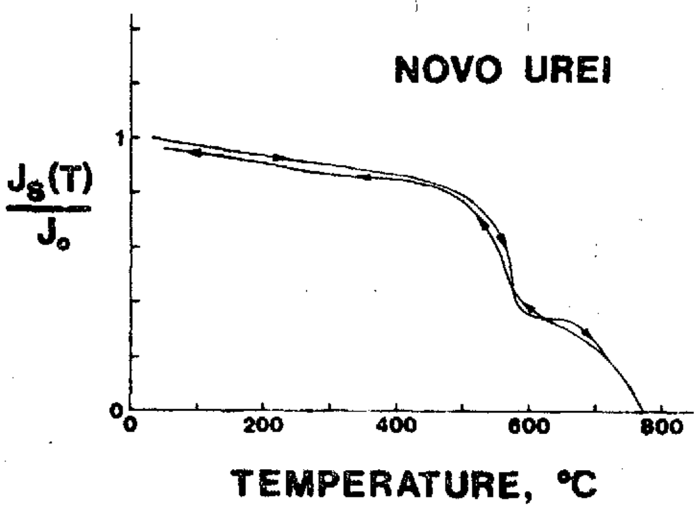

Fig. 12. J_s-T curve for Novo Urei run under mixed atmosphere of CO_2 and H_2 in N_2 carriers.

curve. We have determined experimentally that the optimum oxygen fugacity-temperature relationship that is necessary to maintain stability for a meteorite sample containing both magnetite and iron is, as one might expect, the relationship represented by the lowest curve in Fig. 6. The following experiment demonstrates the sensitivity of this relationship. The heating cycle of a J_s-T curve was run on a sample of the Murray C2 chondrite. The presence of magnetite was indicated but no metallic iron was present in the sample. The heating was then continued until the temperature reached 640°C and held at this temperature. Since no metallic iron was present, the saturation magnetization was zero, 640°C being above the Curie point of magnetite. The atmosphere was then intentionally made reducing. As iron formed from the reduction of magnetite in the sample, the saturation magnetization began to increase. After a small amount of iron had formed the atmosphere was readjusted so that the value of the saturation magnetization remained constant with time. At this point neither net oxidation nor reduction was taking place, as either reaction would have changed the value of the saturation magnetization. It was found from the output of the oxygen fugacity probe that the oxygen fugacity at this temperature (640°C) necessary to maintain equilibrium was $10^{-22.5 \pm 1.0}$. It can be seen from Fig. 6 that the equilibrium point determined falls, within experimental limits, precisely on the lowest curve. Changing the oxygen fugacity at this temperature more than one order of magnitude results in either oxidation of the metallic iron or reduction of the magnetite. This effect is quite pronounced above ~450°C due to the higher reaction rates occuring at these temperatures.

In the previous discussion we have demonstrated that, by maintaining the proper oxygen fugacity, it is possible to heat carbonaceous meteorites containing magnetite, metallic iron, or a combination of the two to temperatures as high as ~800°C without significantly changing the oxidation state of those magnetic components. Were all carbonaceous meteorites to exhibit behavior similar to that observed for our samples of Orgueil, Dyalpur, or Novo Urei, then we should expect that the Thellier technique could be readily applied for paleointensity estimates, providing that the proper oxygen fugacity was maintained during heating. Unfortunately, this is not the panacea it first appears to be as is shown by the thermomagnetic data on the Murchison meteorite.

Figure 13 is the J_s-T curve obtained for the Murchison C2 chondrite. It is obvious that the behavior of the magnetic components depicted in this figure is quite different from that observed for the Orgueil C1 chondrite. The fine structure of the heating curve for the Murchison meteorite is shown by a scale expansion in Fig. 14. The most striking aspect of the J_s-T curve for this meteorite is its irreversibility. The saturation moment after the experiment is about 10 times larger than it was initially. The distinctive Curie temperature of about 550 to 580°C and the generally blocky shape of the cooling curve is indicative of essentially pure Fe_3O_4. It appears that magnetite was produced by the

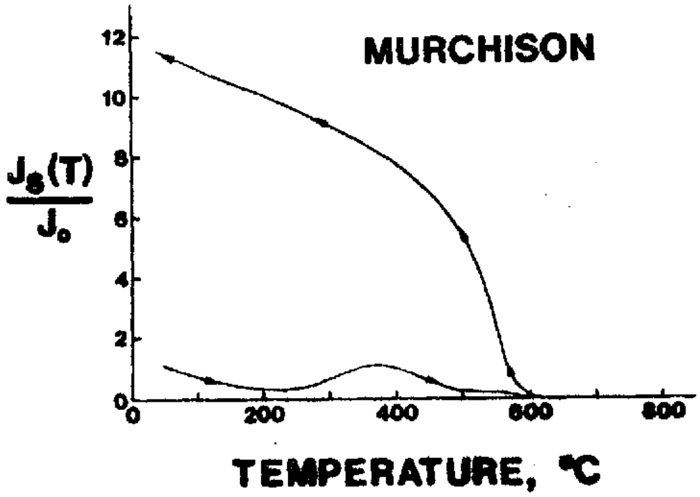

Fig. 13. J_s-T curve for Murchison.

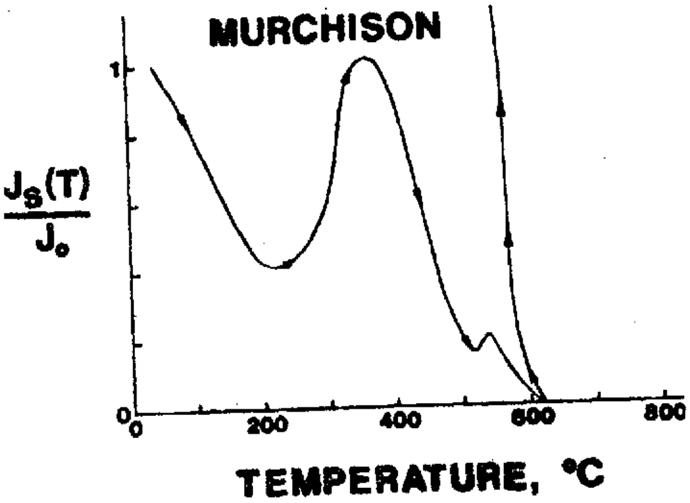

Fig. 14. Detail of heating curve shown in Fig. 13.

breakdown of some non-magnetic phase during the course of the experiment. It further appears that the initial product is not pure magnetite but rather an intermediate phase with a variable Curie temperature which is less than 580°C. If heating is slow, it is possible by the time the temperature has reached 400-500°C for the saturation moment to have increased above the initial value. Continued heating, however, causes J_s to fall as the intermediate Curie temperature are exceeded. If heating is slowed at any temperature above that at which alteration of the phase begins, the J_s will begin to rise. The shape of the heating curve is primarily a function of the heating rate, and the amount and grain size of the unstable phase.

In an extensive study of the chemistry and mineralogy of the Murchison C2 chondrite, Fuchs, et al. (1973) report the presence of three components, any one of which could be the "unstable" phase that alters to magnetite: (1) a layer-lattice silicate, (2) a weakly magnetic "poorly characterized" Fe-S-O phase or, (3) troilite disseminated in a finely divided state throughout the matrix. For investigation of these alternatives, Dr. Edward Olsen kindly provided us with a sample material from a large xenolithic "C3 inclusion" from Murchison. Fuchs, et al. (1973) report that this material is rich in nickel-iron sulfides, but contains neither the layer-lattice silicate nore the "poorly characterized" Fe-S-O phase. The J_s-T curve obtained from the "C3 inclusion" (Fig. 15) is very similar to that obtained from Murchison. This strongly suggests (but does

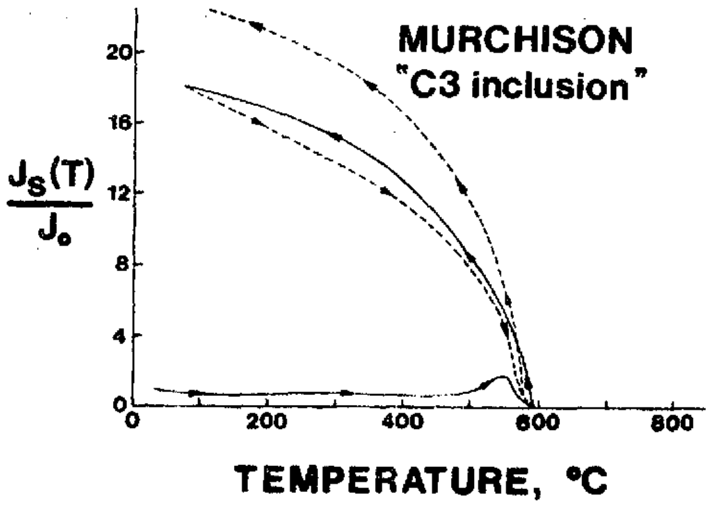

Fig. 15. J_s-T curve for the "C3 inclusion" in Murchison. The broken curve indicates the second thermal cycle.

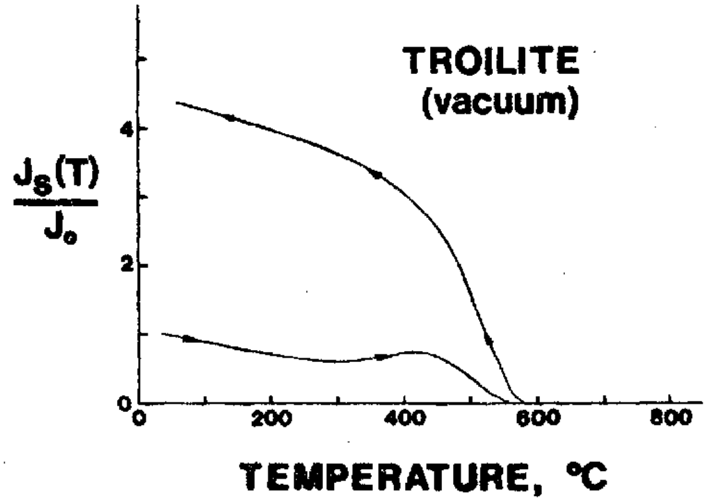

Fig. 16. J_s-T curve for troilite from a troilite nodule in the Staunton iron meteorite. Run under vacuum (4 x 10^{-4} torr).

not prove) that the unstable phase is troilite.

To further verify that the observed magnetite production is characteristic of troilite, we ran J_s-T curves on samples of that mineral removed from the Staunton iron meteorite. One sample was run in a mixed-gas atmosphere and the other was run in a vacuum of about 4×10^{-4} torr. The troilite did break down to produce Fe_3O_4 in both cases. The J_s-T curve of the troilite run under vacuum is shown in Fig. 16 and is essentially identical to that of the same material run in our mixed-gas atmosphere (not shown). The fact that magnetite was produced even when troilite was heated under partial vacuum indicates that the reaction involved is simply oxidation and not a gas-phase reaction peculiar to the gases in our system. In additional experiments we pulverized the troilite to see if the breakdown was grain-size dependent. As the grain size was progressively reduced, the breakdown began at progressively lower temperatures (at least as low as 150°C) and the rate of alteration increased accordingly.

In their discussion of the alternative possibilities which could account for the magnetite production, Fuchs, et al. (1973) point out that their samples of the layer-lattice silicate phase from Murchison were black in color; they suggested that this might be due to the presence of finely divided troilite. They further noted that during heating, magnetite was produced (as determined by x-ray studies) at temperatures below the breakdown temperature of the layer-lattice silicate phase. It seems likely then, that the formation of magnetite, as indicated by their thermal studies, actually resulted from the oxidation of troilite. Perhaps the "poorly characterized" Fe-S-O phase observed by Fuchs, et al. (1973) represents an intermediate stage of the progressive oxidation of troilite, similar in nature to the intermediate alteration phases that we observed forming during the saturation magnetization experiments on the Murchison C2 chondrite.

Throughout the preceding discussion, we have argued that the magnetic phase produced was magnetite. We now present further experimental data to support this assertion. After completing the J_s-T curve on a sample of the "C3 inclusion," the sample was reheated to ~700°C and held ~ 15 minutes in a strongly reducing atmosphere. Figure 17 shows the J_s-T curve obtained, as well as the high temperature portion of the J_s-T curve obtained on the same sample following the reduction experiment. It is evident from the original J_s-T curve that no metallic iron was present prior to subjecting the sample to the strongly reducing atmosphere. The high temperature portion of Fig. 17 (obtained after reduction), however, indicated the presence of metallic iron of low nickel content ($\leq$ 2% Ni) as evidenced by the Curie temperature and lack of a $\gamma \rightarrow \alpha$ transition. It is apparent from this curve (which was run in a reducing atmosphere) that some reduction was still taking place on the heating cycle. The magnitude of the saturation magnetization of the metallic iron was greater than that which would have been obtained from the

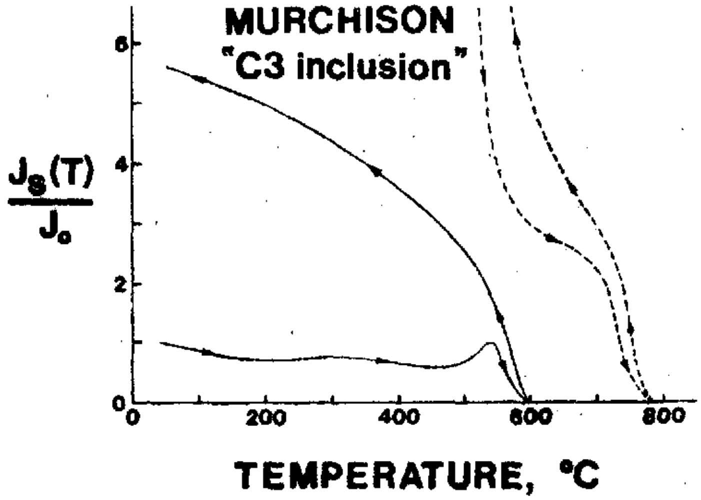

Fig. 17. J_s-T curve for a sample of the "C3 inclusion" in Murchison. The broken curve is the partial J_s-T curve obtained on the sample following reduction.

reduction of the "initial" material if the "initial" material had been entirely magnetite. This demonstrates that indeed magnetite was produced during the original J_s-T run.

Prior to discussing the implications of the thermal instability of troilite as regards paleointensity studies, we present the results of thermomagnetic analyses conducted on samples of all known carbonaceous meteorites.

Thermomagnetic Analysis of Cl Chondrites: Saturation magnetization vs. temperature (J_s-T) curves for each of the five Cl chondrites are given in Figs. 18 - 21 and Fig. 11. Except for that of Revelstoke (Fig. 20), all curves are almost exactly reversible and, from their shape and Curie temperature (575-580°C), indicative of magnetite as the only magnetic phase present. The near coincidence of the heating and cooling curves indicates that essentially no chemical alteration of the magnetic or potentially magnetic phases occurred during the experiment. As a check on our ability to inhibit oxidational alteration, the Orgueil sample (Fig. 11) was subjected to a second thermal cycle after the first had been completed: the J_s-T curves for each run are essentially coincident.

The thermomagnetic curve for the Revelstoke sample (Fig. 20) is generally similar to the other four and indicates Fe_3O_4 as the principal phase. However, the fact that the cooling curve is somewhat elevated above the heating curve (about 20% increase in J_s) is suggestive of the presence in the sample of a thermally unstable subsidiary phase which breaks down on heating to produce at least some additional magnetite - thereby accounting for the single observed Curie temperature of 580°C and the increased saturation moment during cooling. The thermomagnetic behavior of this unstable phase is similar to that observed in Murchison (Fig. 13) and in troilite separates (Fig. 16). Although the Revelstoke sample is the only C1 chondrite sample in which we observed magnetite formation, Banerjee and Hargraves (1972) observed similar behavior for their sample of Orgueil. Whereas every sample of the Murchison C2 chondrite which we examined showed this unstable phase, it was rarely

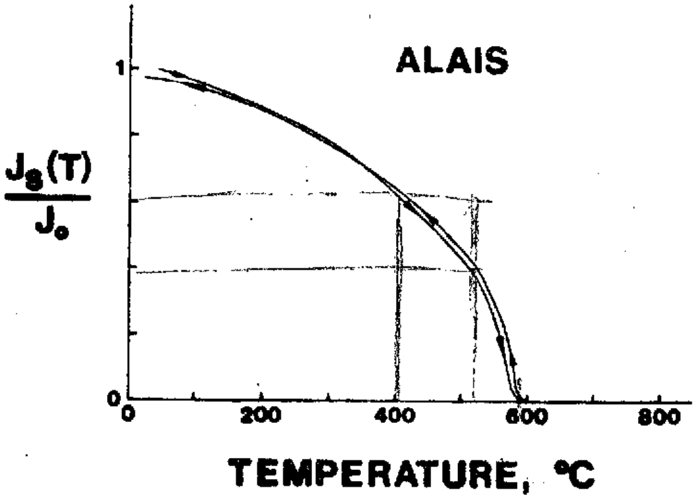

Fig. 18. J_s-T curve for Alais.

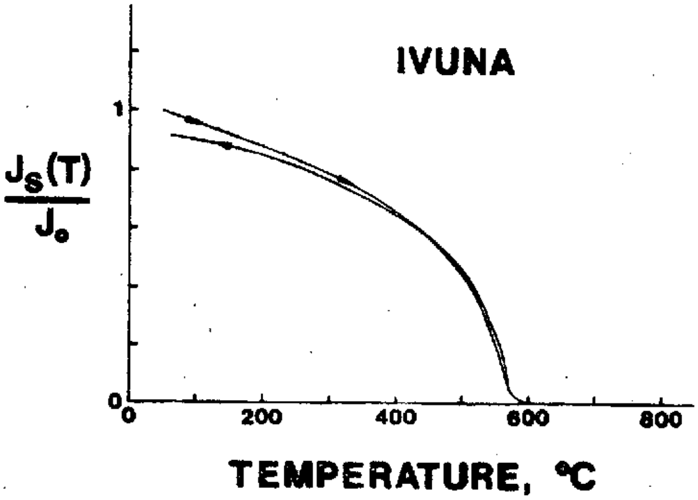

Fig. 19. J_s-T curve for Ivuna.

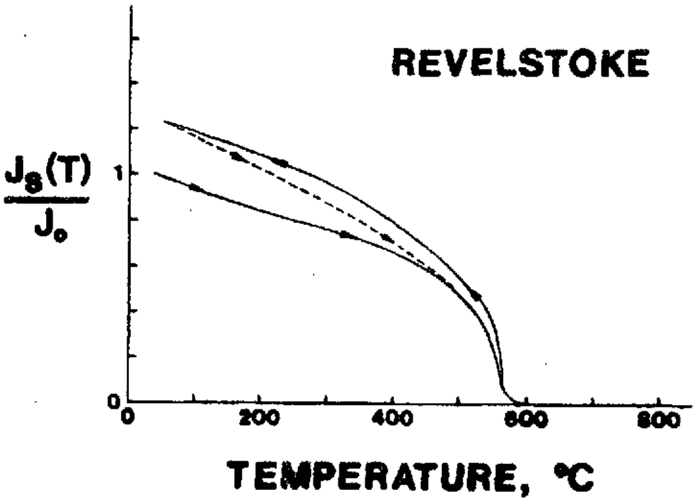

Fig. 20. J_s-T curve for Revelstoke. The broken curve shows reheating prior to conducting the reduction experiment.

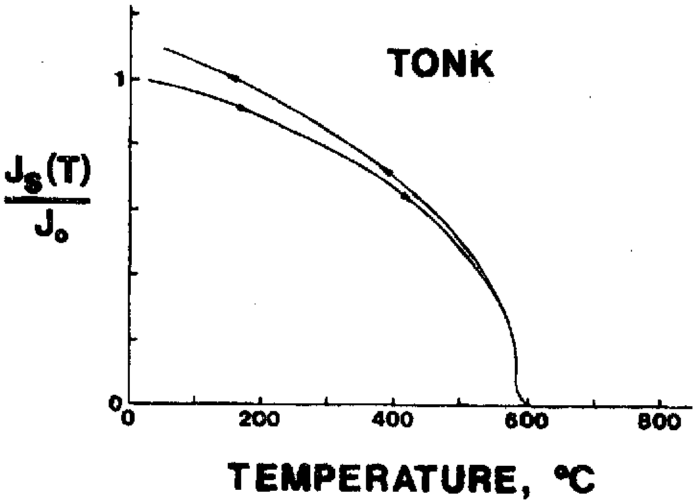

Fig. 21. J_s-T curve for Tonk.

observed in Cl chondrites. That the sulfide content of Cl chondrites is considerably lower than C2 chondrites (Mason, 1962) lends support to our identification of the unstable phase as an iron sulfide mineral.

Although our J_s-T data for Cl chondrites are at first indicative of nearly pure Fe_3O_4 as the only or principal magnetic component, there has been a suggestion by Mason (1962) and by Banerjee and Hargraves (1972), that the iron ferrite phase in Cl chondrites contains appreciable nickel in solid solution. The presence of the nickel-ferrite phase ($NiFe_2O_4$, trevorite) would tend to increase the Curie temperature - to a maximum of 595°C for pure trevorite (Bozorth, 1951). Because of the close similarity in Curie temperatures of the iron and nickel iron ferrites (580°C vs. 595°C) in practice it is difficult to unambiguously determine the presence or absence of nickel in solid solution by standard J_s-T analysis.

By means of the gas-mixing capabilities of our thermomagnetic system, we were able to perform an experiment which could produce unambiguous results. After completion of the J_s-T analysis on each Cl sample, we flowed H_2 (in its carrier gas) only and heated the sample to 700°C, holding this temperature for about 15 minutes. In a reducing atmosphere at this temperature, the spinel phase will reduce to its unoxidized state - Fe or Ni-Fe. If the sample is then heated to about 800°C and subsequently cooled, the Ni content can be estimated by the temperature at which the magnetization is lost and at which it is regained (Bozorth, 1951). In Fig. 22 are shown

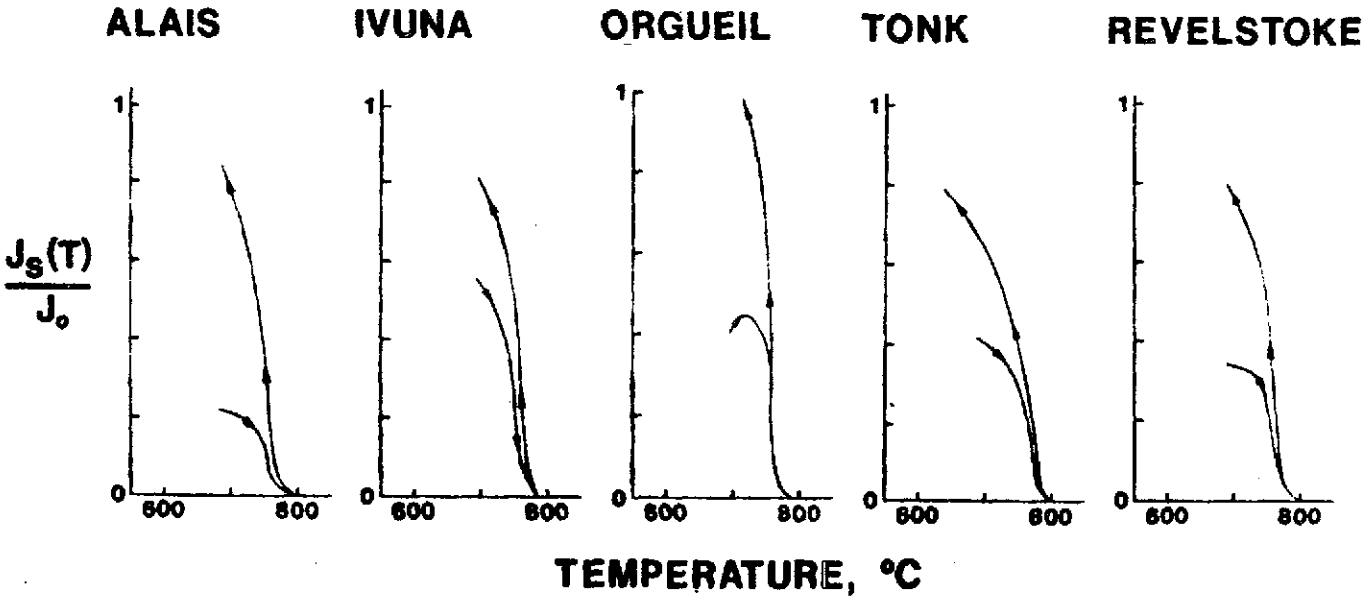

Fig. 22. Partial J_s-T curves for iron formed from the reduction of Cl magnitite.

the results from reduction for 15 minutes of the spinel phase in all five Cl chondrites, and subsequent heating and cooling. The material in each case becomes non-ferromagnetic at about 770°C, and there is no lag of reacquisition of magnetism upon cooling - that is, the heating and cooling curves are coincident. This is indicative of an essentially nickel free metallic iron. Data we have obtained from measurement of synthetic alloys of known Ni-Fe compositions would indicate that this type of behavior is restricted to iron containing 2% or less nickel. A composition of 2% Ni/98% Fe would correspond to less than 6% $NiFe_2O_4$ in the original ferrite solid solution, i.e., essentially pure Fe_3O_4. Recent studies of the magnetic phase in the Orgueil meteorite (Bostrom and Fredriksson, 1966; Kerridge, 1970) indicate that the nickel content of this phase is considerably less than our upper limit determination ($\leq 0.1\%$ Ni).

After determination of the nickel content in the magnetic phase of Cl chondrites, it is possible to estimate the weight percentage of that phase by measurement of its saturation moment. Because of the extremely fine grain size of Cl chondrites, estimates of the Fe_3O_4 content by alternative methods have been rare, and all (as far as we know) have been restricted to the Orgueil meteorite (DuFresne and Anders, 1962; Bostrom and Fredrickson, 1966; Kerridge and Chatterji, 1968; Jeffery and Anders, 1970).

To increase the accuracy of the determination, multiple determinations were made on different samples (each weighing about 1 mg) from the same meteorite, whenever possible. The following is the number of determinations made on different samples from each meteorite: Alais, 11; Ivuna, 5; Orgueil, 7; Revelstoke and Tonk, 1 each. All samples, except that of the Orgueil meteorite, were measured as small whole samples. For the Orgueil, determinations were made on aliquots taken from a 200-mg sample which had been round to < 100 mesh. Error estimates of the results are based on deviations we observed when measuring a standard sample. Our reported error does not reflect the possibility that some of the magnetization is contributed by some magnetic component other than Fe_3O_4, perhaps pyrrhotite. In light of the near-reversibility of the J_s-T curves, the blocky shape of the curves and the presence of only one Curie temperature, it would seem that this is probably a minor source of error. It has been confirmed in the Orgueil meteorite, at least, that the percentage of pyrrhotite is about 2% (Mason, 1962). Since in its most magnetic stoichiometry the saturation moment of pyrrhotite is about 1/5 that of magnetite, small weight percentages of this phase should contribute very little (7% or less) to the total saturation moment.

The magnetite content as determined from saturation magnetizations of the five C1 chondrites is given in Table 3. Of the two C1 chondrites on which multiple determinations were made on different whole rock samples (Ivuna-5 determination; Alais-11 determinations), the Ivuna meteorite shows little variation in Fe_3O_4 content whereas the Alais shows wide variation. Since the weighted average content (5.3 ± 0.4%) is close to the unweighted average (4.9 ± 0.4%), despite

Table 3. Magnetite content of Cl chondrites

Wt. % Fe_3O_4		Sample Weight (mg)
ALAIS		
1.31 ± 0.09		1.65
1.01 ± 0.07		2.32
4.02 ± 0.28		2.52
5.31 ± 0.37		2.38
8.95 ± 0.63		3.67
4.26 ± 0.30		2.85
3.53 ± 0.25		3.30
12.00 ± 0.84		1.25
10.32 ± 0.72		2.18
1.62 ± 0.11		0.55
1.50 ± 0.11		0.80
average	4.9 ± 0.4	
weighted average	5.3 ± 0.4	
IVUNA		
11.29 ± 0.79		3.16
12.67 ± 0.89		1.06
12.01 ± 0.84		1.45
13.22 ± 0.93		1.54
12.94 ± 0.91		1.54
average	12.4 ± 0.9	
weighted average	12.2 ± 0.9	
ORGUEIL (aliquots from 200 mg of powdered sample)		
11.39 ± 0.80		1.86
12.66 ± 0.89		0.31
12.15 ± 0.85		0.57

Table 3. Magnetite content of Cl chondrites (cont'd)

Wt. % Fe_3O_4		Sample Weight (mg)
ORGUEIL (Continued)		
11.34 ± 0.79		0.19
13.17 ± 0.92		0.22
11.78 ± 0.82		0.94
12.73 ± 0.89		0.85
average	12.2 ± 0.9	
weighted average	11.9 ± 0.8	
REVELSTOKE		
7.20 ± 0.50		0.16
TONK		
9.40 ± 0.56		0.67

the large differences in sample weights, it is felt that the former is fairly representative of the average magnetite content of the Alais meteorite. As expected, the 7 measurements on aliquots from the 200-mg powdered sample of the Orgueil meteorite results in relatively small variation between estimates of magnetite content (weighted average - 11.9 ± 0.8%). Because of the large size of the initial sample, we consider the weighted average value to be fairly representative of the Orgueil meteorite as a whole.

We have no way of knowing presently the degree of homogeneity of distribution of magnetite in the Revelstoke or Tonk Cl chondrites and, therefore, how representative are the determinations from measurement of a single small sample of each. Assuming that these two values are not grossly misrepresentative, the mean magnetite weight percentage of the five Cl chondrites is 9.2. It would appear that the magnetite of the Alais sample is not only inhomogeneously distributed, but its abundance is only about half that of the average Cl chondrite.

As far as we know, Fe_3O_4 determinations have been made by other techniques only on the Orgueil Cl chondrite: the reported values are summarized in Table 4. The three recent estimates are in reasonable agreement.

Thermomagnetic Analysis of C2 Chondrites: On the basis of their thermomagnetic behavior, the C2 chondrites can be divided into three groups: (A) those containing little, if any, magnetite initially;

Table 4. Magnetic content of the Orgueil meteorite

Wt. % Fe_3O_4	Reference
6	Bostrom and Fredriksson, 1966
40	DuFresne and Anders, 1962
15.9 ± 3.3	Kerridge and Chatterji, 1968
9.3	Jeffrey and Anders, 1970
$11.9 \pm {}^{0.8*}_{2.0}$	This work

*Error estimate includes possible systematic error due to pyrrhotite interference.

(B) those containing magnetite as the only major magnetic component (similar to Cl chondrites), and; (C) those whose thermomagnetic behavior cannot be classified within the first two groups. The number of C2 chondrites in each group is 11, 4, and 3, respectively.

Group A - The C2 chondrites classified under (A) includes Murchison (Figs. 13 and 14) as well as Boriskino, Cold Bokkeveld, Crescent, Erakot, Mighei, Murray, Nawapali, Nogoya, Santa Cruz, and Pollen. The J_s-T curves for samples of these meteorites are shown in Figs. 23 - 33. The thermomagnetic behavior of these is similar to Murchison, indicating magnetite production upon heating. The saturation moment after the experiment is generally three to four times as large as it was intially; in the case of Murchison it is about ten times larger at the end of the experiment. The distinctive Curie temperature of about 550 to 580°C and the generally blocky shape of the cooling curves is indicative of essentially pure magnetite. The heating curves of Group A samples are quite variable, such that no two heating curves, even from the same sample, are the same. The particular shape of any one heating curve is primarily a function of the heating rate, and amount and grain size of the unstable phase.

The Cold Bokkeveld sample (Fig. 24) and one Pollen sample (Fig. 32) were subjected to reheatings to help evaluate the extent of alteration. Although some additional magnetite was produced during the second heating, it can be seen from the J_s-T curves for these two samples that most of the alteration occurred during the first heating.

The J_s-T curves of the Crescent sample (Fig. 25) and a second sample of the Pollen meteorite (Fig. 33) show the presence of an additional magnetic component which becomes non-ferromagnetic at about 770°C. There is no lag in magnetization upon cooling from 800°C down to the 580°C Curie temperature of magnetite. The loss of magnetization at 770°C and the lack of a $\gamma \rightarrow \alpha$ transition upon cooling are indicative of metallic iron containing $\leq$ 2% Ni.

Because the shapes of the initial heating curves for those C2 chondrites shown in Figs. 23 - 33 are not like those expected for samples containing abundant magnetite, we qualitatively estimate that the magnetite content of these meteorites is initially quite low. If we assume that the initial saturation magnetization is due entirely to magnetite, however, we can determine an upper limit of magnetic content. The upper-limit weight-percent Fe_3O_4 values thusly determined are shown in Table 5. With the exception of Boriskino and Mighei which were measured as whole pieces, the meteorite samples were gently crushed to <100 mesh and measured as aliquots from 50 - 200 mg samples. Care was taken in this homogenization procedure not to subject the material to undue mechanical stress so as to minimize the possibility of maghemite production or physical alteration of the sulfide phases. Error estimates represent analytical uncertainty based on deviations observed in measuring a high purity standard of known content. The actual magnetite content of the C2 chondrites shown in Table 5 is probably much lower than our upper

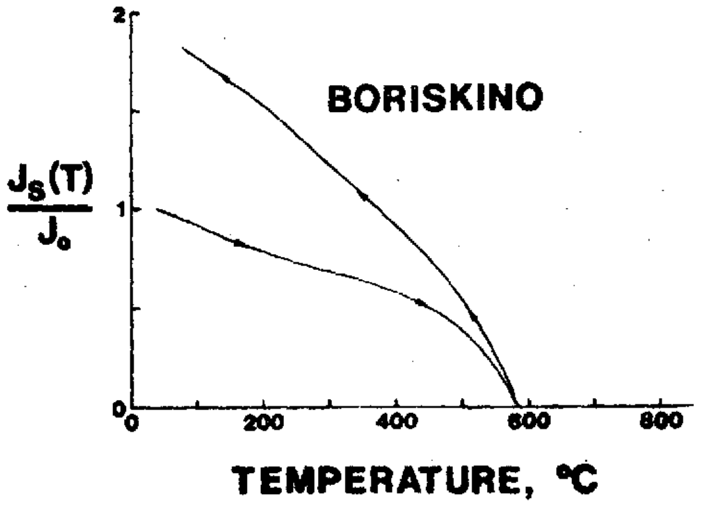

Fig. 23. J_s-T curve for Boriskino.

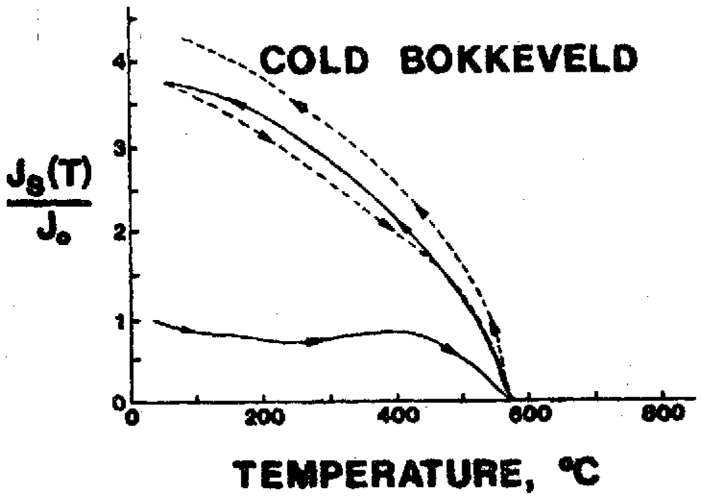

Fig. 24. J_s-T curve for Cold Bokkeveld. The broken curve indicates the second thermal cycle.

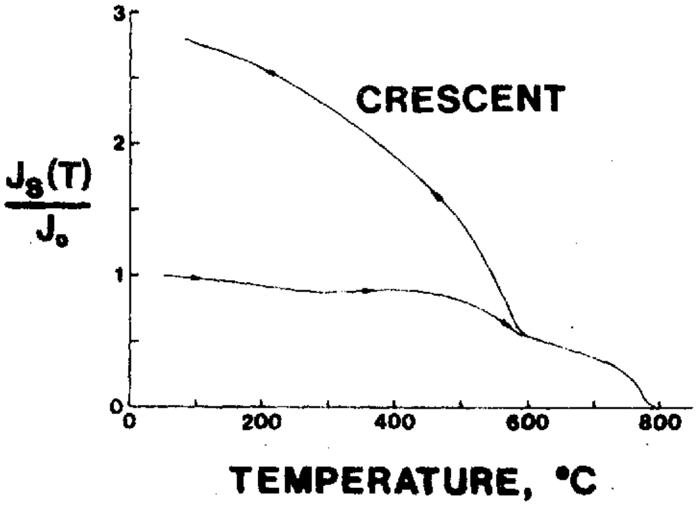

Fig. 25. J_S-T curve for Crescent.

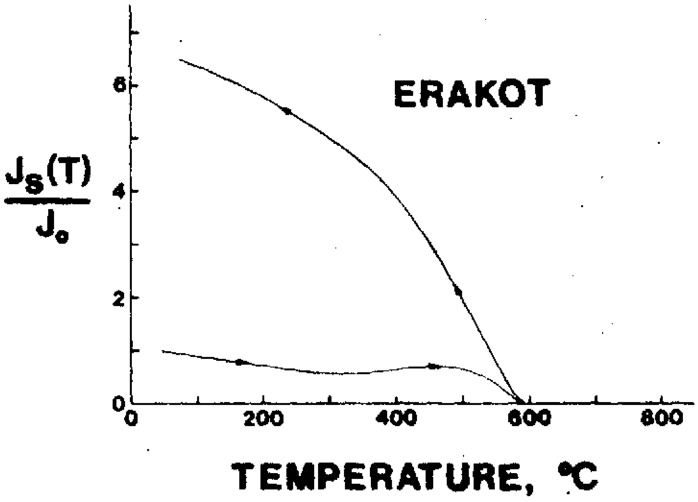

Fig. 26. J_S-T curve for Erakot.

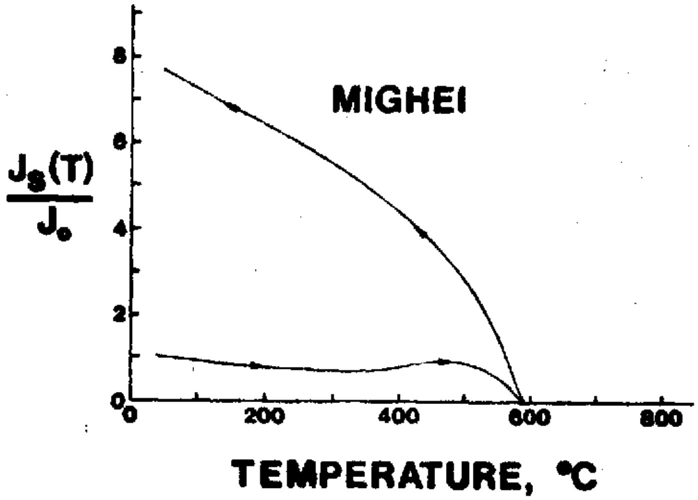

Fig. 27. J_S-T curve for Mighei.

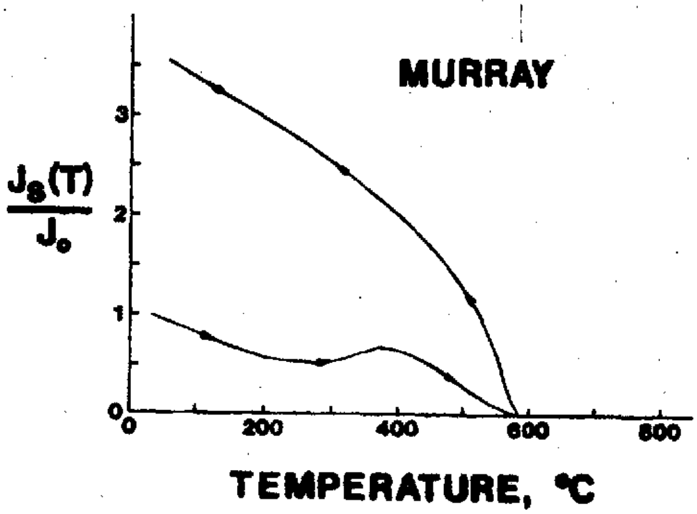

Fig. 28. J_S-T curve for Murray.

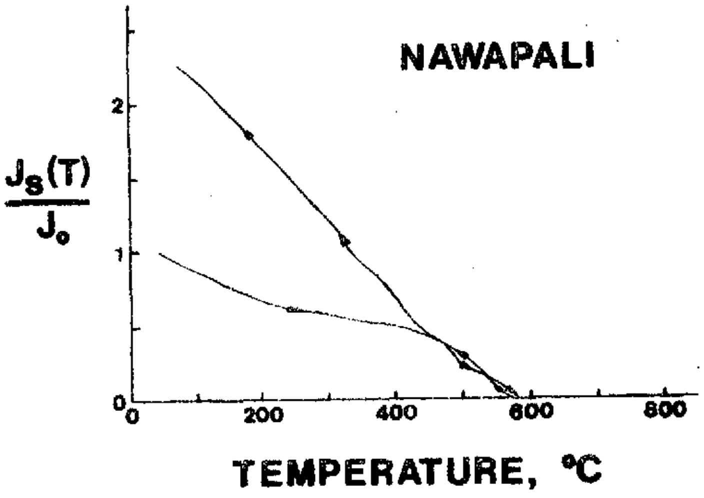

Fig. 29. J_s-T curve for Nawapali.

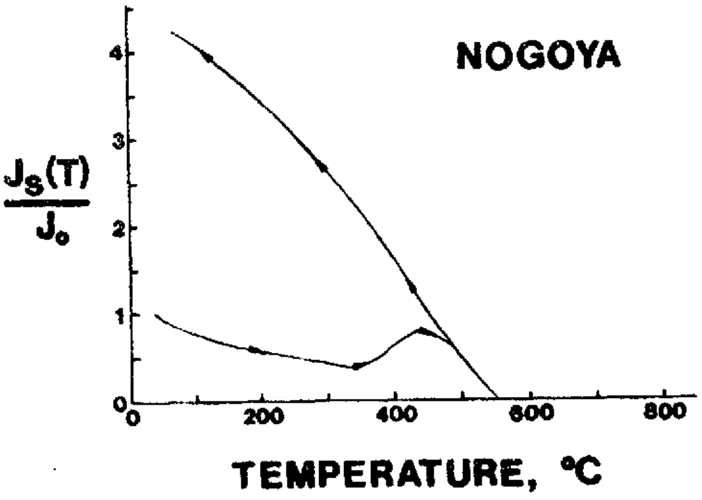

Fig. 30. J_s-T curve for Nogoya.

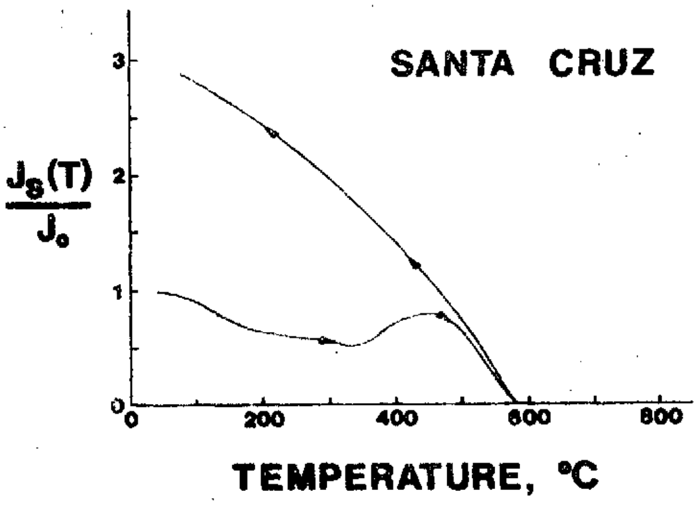

Fig. 31. J_s-T curve for Santa Cruz.

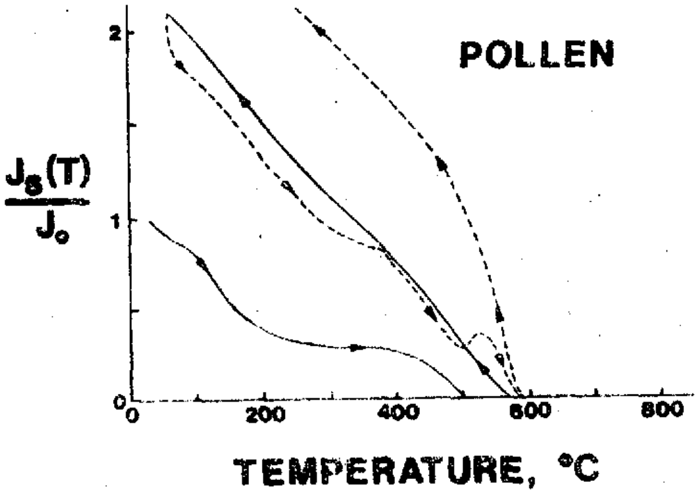

Fig. 32. J_s-T curve for the first sample of Pollen. The broken curve indicates the second thermal cycle.

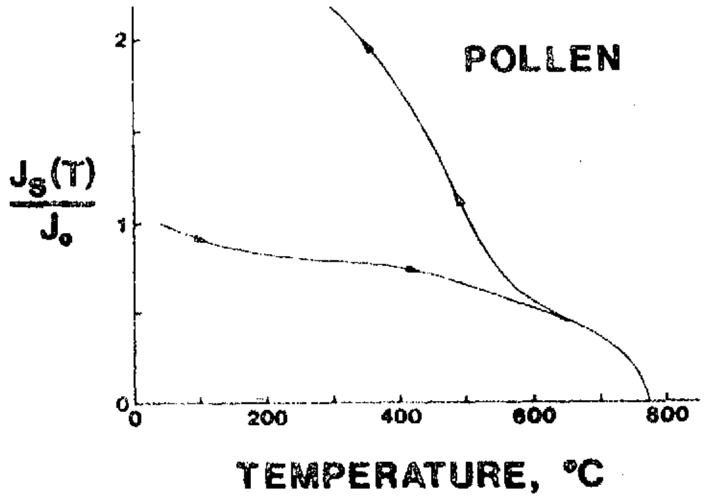

Fig. 33. J_s-T curve for the second sample of Pollen.

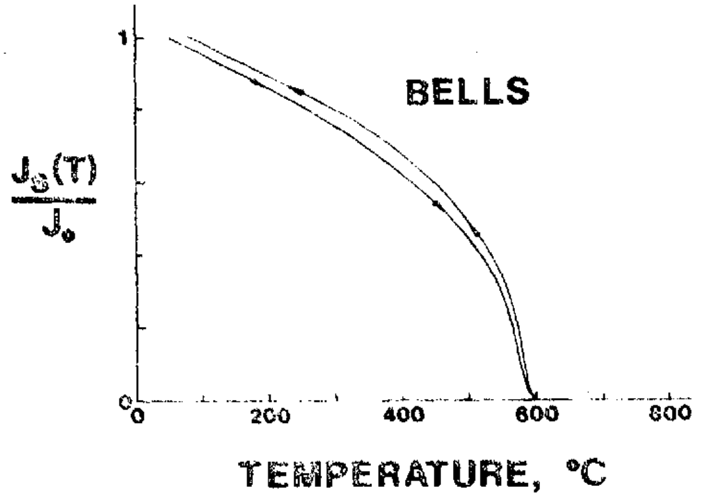

Fig. 34. J_s-T curve for Bells.

Table 5. Upper limit determination of the magnetite content of some C2 chondrites

Upper Limit for Wt. % Fe_3O_4		Sample Weight (mg)
BORISKINO*		
≤0.86 ± 0.05**		0.20
≤0.82 ± 0.05		2.27
weighted average	≤ 0.82 ± 0.04	
COLD BOKKEVELD		
≤ 0.60 ± 0.04		0.46
≤ 0.59 ± 0.04		0.47
≤ 0.45 ± 0.03		4.05
weighted average	≤ 0.48 ± 0.03	
ERAKOT		
≤ 0.36 ± 0.02		2.92
≤ 0.34 ± 0.02		2.59
weighted average	≤ 0.35 ± 0.02	
MIGHEI*		
≤ 0.33 ± 0.02		1.53
MURCHISON		
≤ 1.43 ± 0.04		0.39
≤ 1.16 ± 0.07		0.74
≤ 0.23 ± 0.01		1.10
weighted average	≤ 0.75 ± 0.04	
MURRAY		
≤ 0.64 ± 0.04		1.96
≤ 0.57 ± 0.03		3.36
weighted average	≤ 0.60 ± 0.04	

Table 5: Upper limit determination of the magnetic content of some C2 chondrites (Cont'd)

Upper Limit of Wt. % Fe_3O_4		Sample Weight (mg)
NAWAPALI		
$\leq 0.40 \pm 0.02$		5.84
$\leq 0.59 \pm 0.04$		2.53
weighted average	$\leq 0.46 \pm 0.03$	
NOGOYA		
$\leq 0.68 \pm 0.04$		1.78
$\leq 0.62 \pm 0.04$		1.08
weighted average	$\leq 0.66 \pm 0.04$	
SANTA CRUZ		
$\leq 0.82 \pm 0.05$		2.80
$\leq 0.73 \pm 0.04$		0.83
weighted average	$\leq 0.80 \pm 0.05$	

* Run on whole samples

** Error is analytical only and does not include any attempt to evaluate sample inhomogeneity.

limit determination. Fuchs, et al. (1973) report that magnetite is present in the Murchison meteorite in trace amounts only. Our data indicate that certainly it can be no greater than 0.75 percent, on the average.

Group B: The J_s-T curves for the four Group B C2 chondrites, Bells, Essebi, Kaba, and Mokoia are shown in Figs. 34 - 37, respectively. They are similar to those obtained from C1 chondrites in that for each sample, magnetite is the major magnetic component - as indicated by the Curie temperature (~ 580°C) and the blocky character of the J_s-T curves. The slight increase observed in saturation magnetization during cooling for the Essebi and Mokoia samples is similar to that which we observed for the Revelstoke C1 chondrite and is suggestive of the behavior noted for the eleven Group A C2 chondrites. It appears to be related to the breakdown of a small amount of troilite to magnetite during the experiment.

We have made estimates of the magnetite content of these meteorites based on room temperature saturation magnetization measurements. As with the other C2 chondrites, determinations were made on aliquots taken from gently crushed 50 - 200 mg samples. The results obtained from these measurements are shown in Table 6. The reported error is analytical only and does not reflect sample inhomogeneity or the possible contribution from magnetic pyrrhotite, if present. In its most magnetic stoichiometry, pyrrhotite is only about one-fifth as magnetic as magnetite. If pyrrhotite was present in large quantities, we would expect to see it expressed (Curie temperature

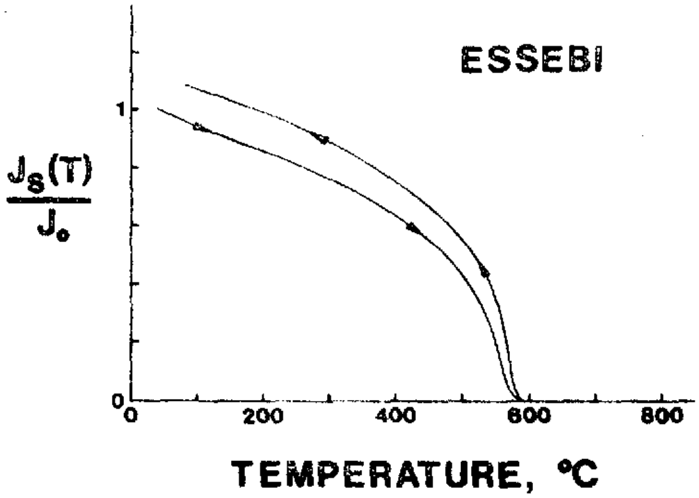

Fig. 35. J_s-T curve for Essebi.

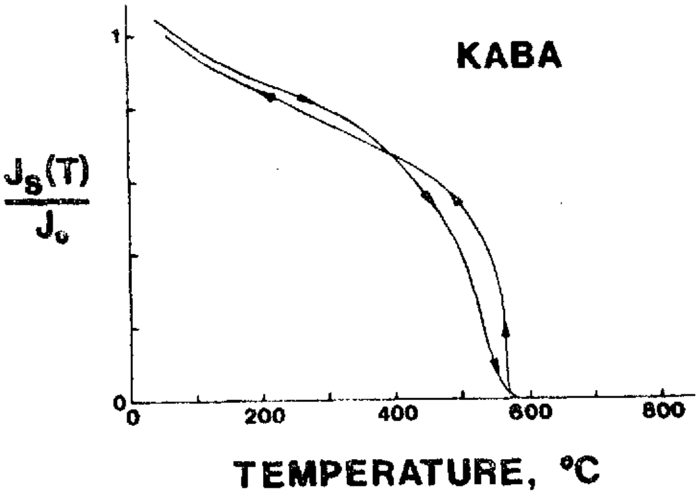

Fig. 36. J_s-T curve for Kaba.

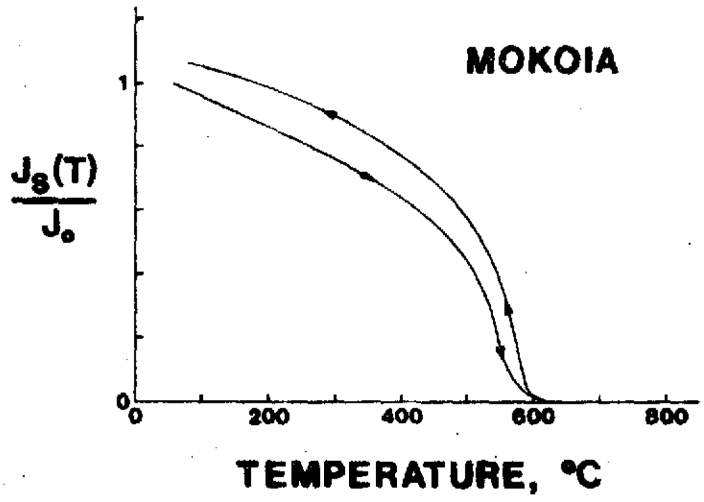

Fig. 37. J_s-T curve for Mokoia.

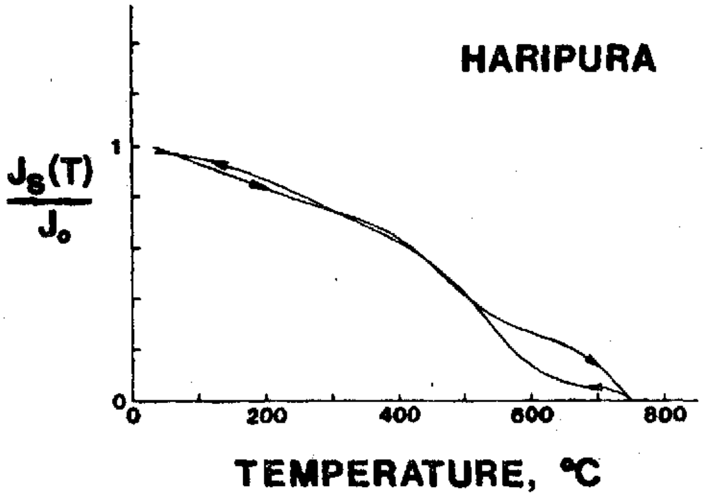

Fig. 38. J_s-T curve for Haripura.

Table 6. Magnetite content of the Bells, Essebi, Kaba and Mokota C2 chondrites

Wt. % Fe_3O_4		Sample Weight (mg)
BELLS		
13.29 ± 0.80		3.93
13.30 ± 0.80		2.60
13.76 ± 0.83		0.88
average	13.5 ± 0.8	
weighted average	13.4 ± 0.8	
ESSEBI		
7.70 ± 0.46		0.34
9.80 ± 0.60		0.82
9.18 ± 0.55		1.10
8.76 ± 0.53		0.55
10.64 ± 0.64		0.55
average	9.2 ± 0.7	
weighted average	9.4 ± 0.6	
KABA		
11.17 ± 0.67		5.62
11.50 ± 0.69		4.25
average	11.3 ± 0.7	
weighted average	11.3 ± 0.7	
MOKOIA		
4.18 ± 0.25		4.98
3.99 ± 0.24		4.49
4.20 ± 0.25		4.44
average	4.1 ± 0.3	
weighted average	4.1 ± 0.3	

~ 350°C) in the J_s-T curves: it is not in evidence.

The Bells, Essebi, Kaba, and Mokoia meteorites have magnetite contents comparable to the values obtained by us for C1 chondrites. The magnetic content of the Group B C2 chondrites ranges from about 4 to 13 weight percent, and averages about 9.6 weight percent. Although the bulk chemical composition of Group A and Group B C2 chondrites is quite similar (Wiik, 1956; Mason, 1962, 1971), the magnetic content varies appreciably. This suggests similarly diverse mineralogies, and hence, different formative conditions.

Group C: The J_s-T curves for the Group C C2 chondrites are shown in Figs. 38 - 42. We suspect that the Al Rais, Haripura, and Renazzo meteorites are inhomogeneous and therefore that the J_s-T curves given in Figs. 38 - 42 may not necessarily be representative of these meteorites as a whole. Due to the high saturation magnetization of metallic iron and the extreme sensitivity of our system, we were constrained to use very small samples (less than 1 mg) of those meteorites, such as Renazzo, which generally contain abundant metallic iron. This practice, of course, accounts for dissimilar results obtained from two samples of the same meteorite, if that meteorite is inhomogeneous. It is also probable that the inhomogeneity observed in samples of Al Rais and Haripura is the combined result of larger grain sizes in these than in other C2 meteorites and the small size of the samples measured.

The J_s-T curve for one sample of Al Rais (Fig. 39) is indicative of a single magnetic component, metallic iron containing some nickel.

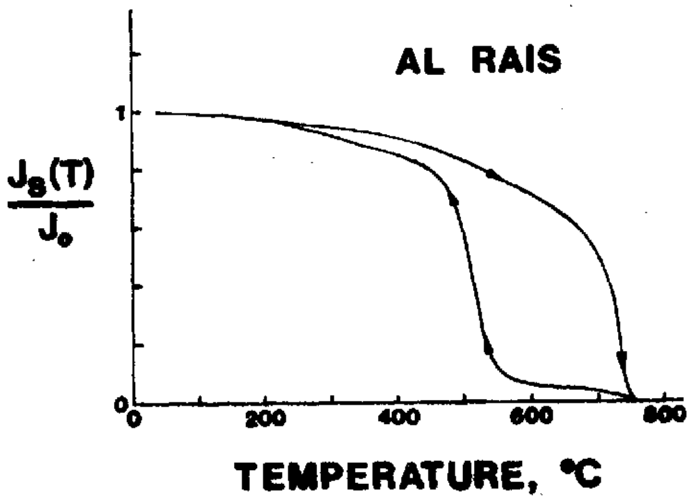

Fig. 39. J_s-T curve for the first sample of Al Rais.

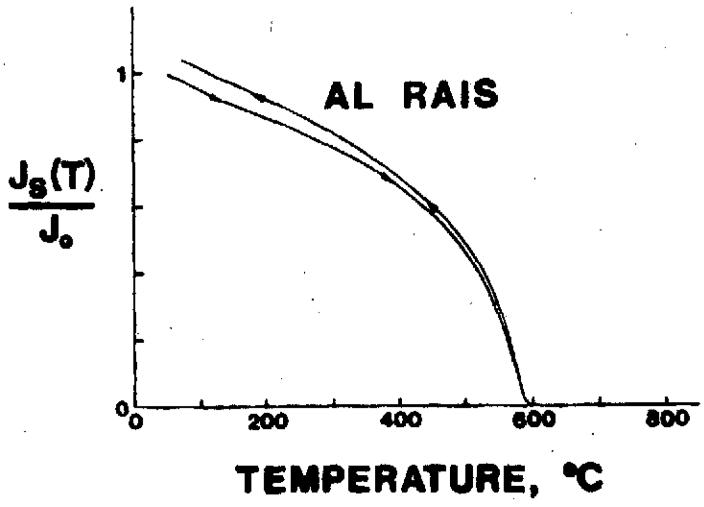

Fig. 40. J_s-T curve for the second sample of Al Rais.

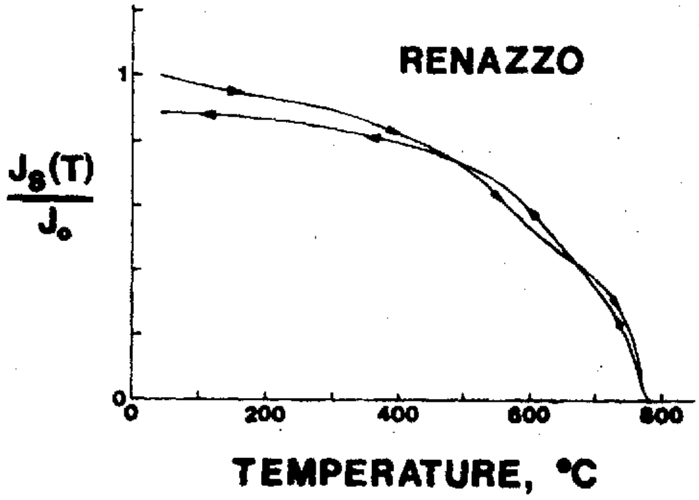

Fig. 41. J_s-T curve similar to several samples of Renazzo.

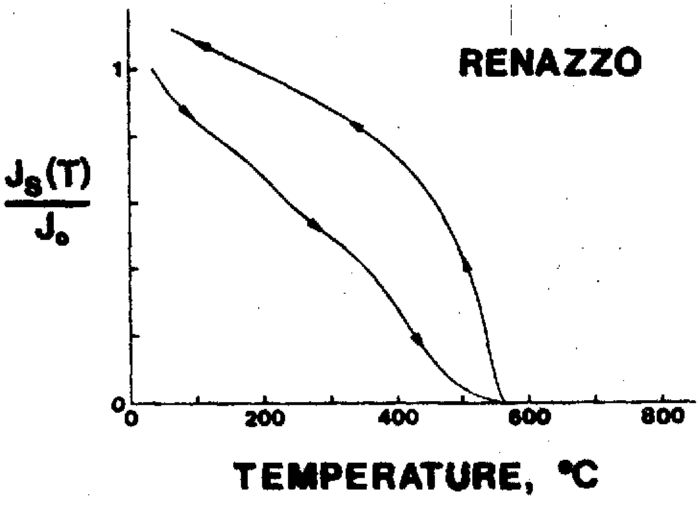

Fig. 42. J_s-T curve for an exceptional sample of Renazzo.

From the particular temperatures at which magnetism is lost on heating and regained during cooling, the nickel content is estimated to be about 6 weight percent. A second sample of Al Rais (Fig. 40) appears to contain pure magnetite. In neither case is there any evidence of troilite breakdown.

With one exception, all samples of Renazzo yielded J_s-T curves similar to that shown in Fig. 41, indicating metallic iron of low nickel content ($\leq 2\%$). The lone exception (shown in Fig. 42) is typical of behavior observed in Group A C2 chondrites and probably represents the alteration of troilite to magnetite.

Thermomagnetic Analysis of C3 and C4 Chondrites: On the basis of their thermomagnetic behavior, we found it convenient for purposes of discussion to divide C2 chondrites into three groups. With the exception of the Renazzo, Al Rais and Haripura meteorites, the C2 chondrites were divided into two groups; those containing < 1 wt.% Fe_3O_4, and those containing > 4 wt. % Fe_3O_4. Taken as a group, the C3 chondrites appear to be considerably more heterogeneous. As only two C4 chondrites are known, the Coolidge and Karoonda meteorites, we include them with the C3's.

Figure 43 shows the J_s-T data on a sample of the Allende meteorite. The behavior indicated is similar to that observed in a majority of the C2 chondrites. The Curie point indicated by the cooling curve in Fig. 43 is slightly higher than that of magnetite. Butler (1972) conducted thermomagnetic analysis on this meteorite and

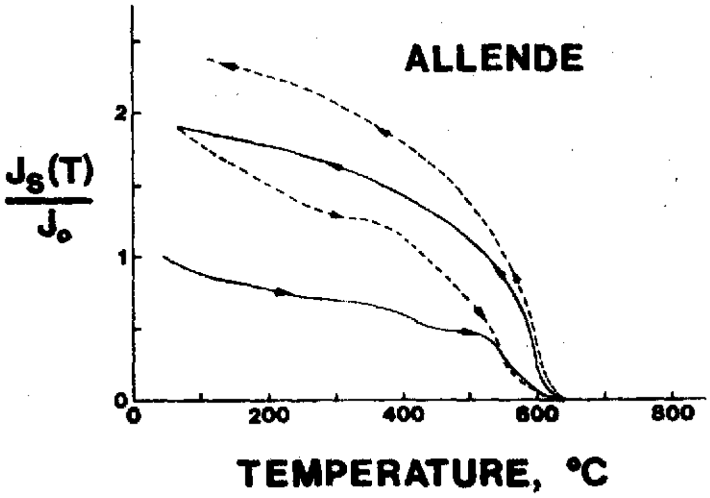

Fig. 43. J_s-T curve for Allende. The broken curve indicates the second thermal cycle.

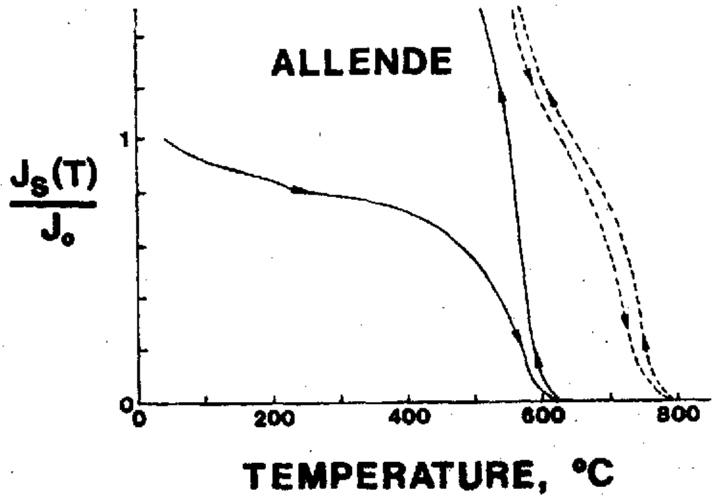

Fig. 44. J_s-T curve for a sample of Allende. The broken curve shown is the partial J_s-T curve for iron formed from the reduction of magnetite in the same sample.

observed a slightly higher Curie temperature on his heating curve. Because of the observed Curie point he attributed the increase in saturation magnetization to a rather complicated "high temperature homogenization" of Ni-Fe alloys. To investigate this possibility as opposed to our thought that the increase in saturation magnetization was due to magnetic production, we ran a J_s-T curve on a powdered sample of this meteorite (Fig. 44). After completing the J_s-T run, we intentionally made the atmosphere in the Curie balance reducing and heated the sample to ~ 700°C, leaving it at this temperature for ~ 15 minutes. The high temperature portion of the curve in Fig. 44 shows the partial J_s-T curve obtained following reduction. It is evident that iron of low nickel content was formed during the reduction experiment. Furthermore, the saturation magnetization of the iron is greater than that which would have been obtained if the initial Allende material had been entirely magnetite. This demonstrates that indeed magnetite was formed, as our reduction procedure would not be expected to affect Ni-Fe alloys. We suspect that the variations noted in the Curie temperatures were due to the thermocouple error.

The J_s-T curves for Efremovka, Leoville, and Vigarano are shown in Figs. 45 - 47. Magnetite formation during the experiment is indicated, although not to the extent observed in several C2 chondrites. The character of the Efremovka J_s-T curve (Fig. 45) is suggestive of some magnetite present prior to heating the sample.

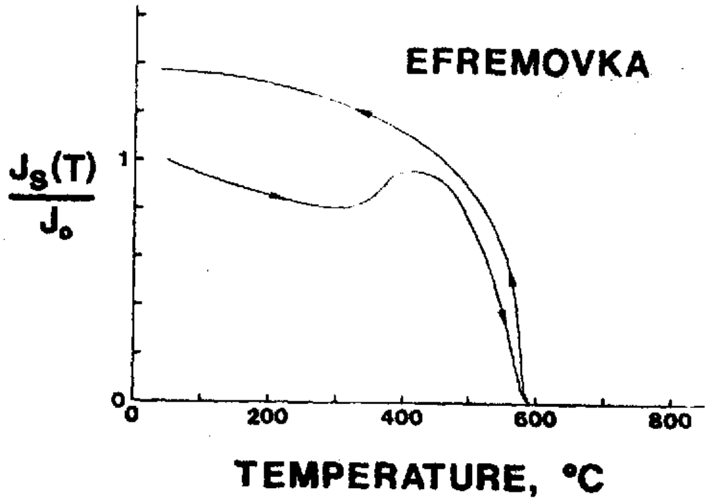

Fig. 45. J_s-T curve for Efremovka.

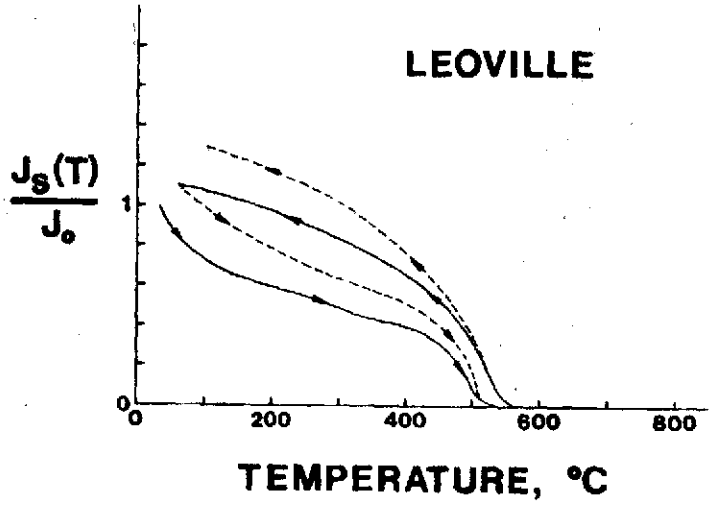

Fig. 46. J_s-T curve for Leoville. The broken curve indicates the second thermal cycle.

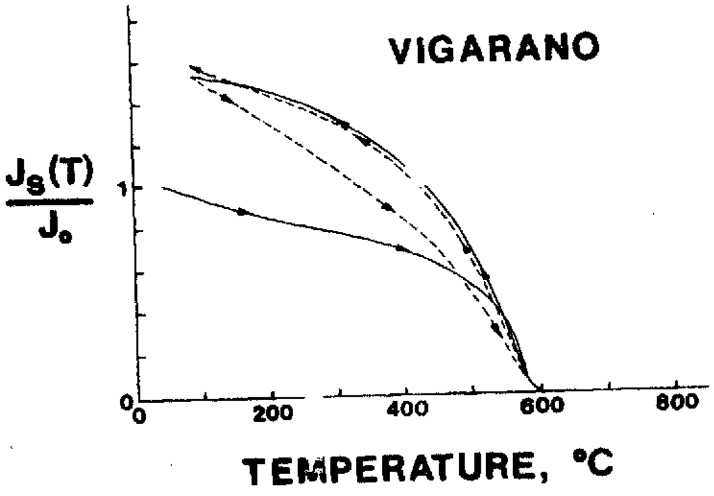

Fig. 47. J_s-T curve for Vigarano. The broken curve indicates the second thermal cycle.

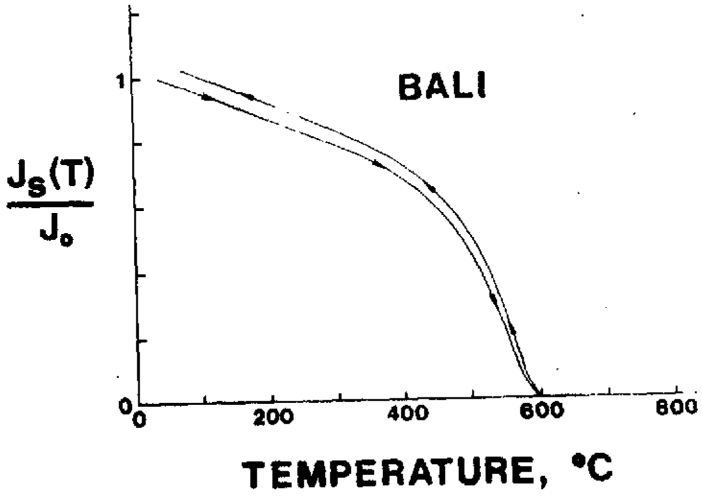

Fig. 48. J_s-T curve for Bali.

As with C2 chondrites, on the basis of saturation magnetization measurements at ambient temperature, we can ascribe upper limits of Fe_3O_4 content for those C3 chondrites discussed above. For Allende and Vigarano determinations were made on aliquots taken from gently crushed 50-200 mg samples. Samples of Efremovka and Leoville were measured as small whole pieces, since these meteorites were found to be too hard to gently crush. The upper limits obtained for the magnetite content of these meteorites is shown in Table 7.

Figures 48 - 51 show the J_s-T curves for Bali, Grosnaja, Ornans, and the C4 chondrite Karoonda, respectively. These curves are quite similar to those obtained for the Cl chondrites and indicate that magnetite is the only magnetic, or potentially magnetic, component. Minimal thermal alteration during the experiment is indicated by these data. Since the J_s-T curves shown in Figs. 48 - 51 display no evidence for the presence of magnetic sulfide phases, magnetite can be determined on the basis of saturation magnetization measurements. These data are shown in Table 8. The Ornans and Karoonda magnetite contents were determined on aliquots taken from gently crushed samples. The Grosnaja samples were run as small whole pieces because of the physical hardness of this meteorite. The Bali sample was also run as a whole piece due to the small amount of available material.

The data shown in Figs. 52 - 56, the J_s-T curves for the Felix, Lance', Kainsaz, and Warrenton C3 chondrites and the Coolidge C4 chondrite were obtained during uncalibrated runs (due to experimental problems). These data, therefore, are useful only in a qualitative

Table 7: Upper limit determination of magnetite content of some C3 chondrites

Upper Limit for Wt. % $\mathrm{Fe_3O_4}$		Sample Weight (mg)
ALLENDE		
$\leq 0.72 \pm 0.04$ **		10.07
$\leq 0.73 \pm 0.04$		7.93
Weighted average	$\leq 0.72 \pm 0.04$	
EFREMOVKA*		
$\leq 10.70 \pm 0.064$		5.37
$\leq 7.01 \pm 0.42$		3.34
Weighted average	$\leq 9.28 \pm 0.58$	
LEOVILLE*		
$\leq 7.52 \pm 0.045$		2.64
$\leq 6.70 \pm 0.40$		2.50
Weighted average	$\leq 7.12 \pm 0.43$	
VIGARANO		
$\leq 8.47 \pm 0.51$		3.20
$\leq 7.11 \pm 0.43$		1.33
$\leq 9.06 \pm 0.54$		2.95
Weighted average	$\leq 8.46 \pm 0.51$	

* Run on whole samples

** Error is analytical only and does not include any attempt to evaluate sample inhomogeneity.

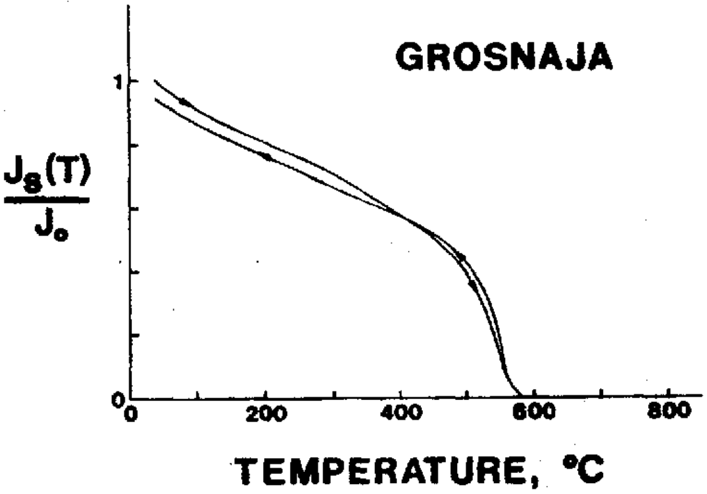

Fig. 49. J_s-T curve for Grosnaja.

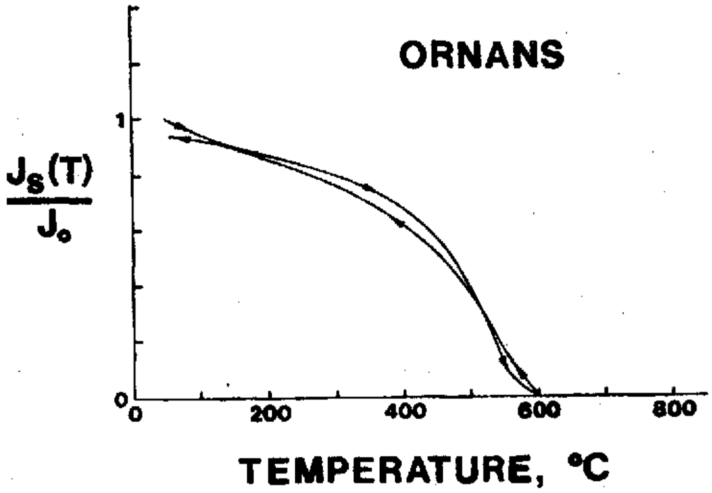

Fig. 50. J_s-T curve for Ornans.

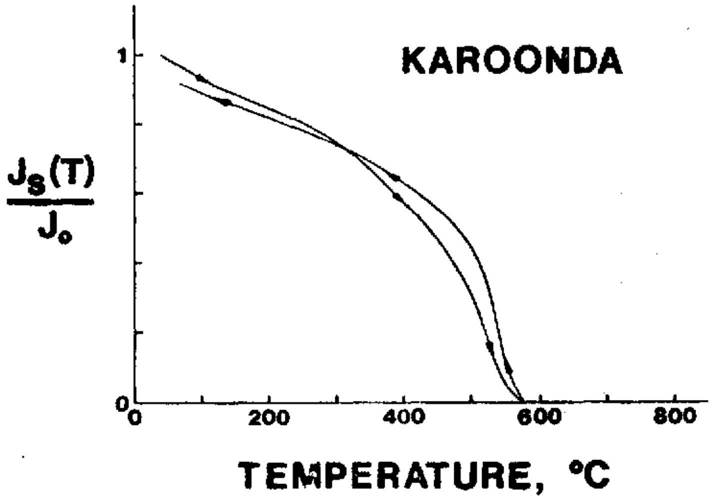

Fig. 51. J_s-T curve for Karoonda.

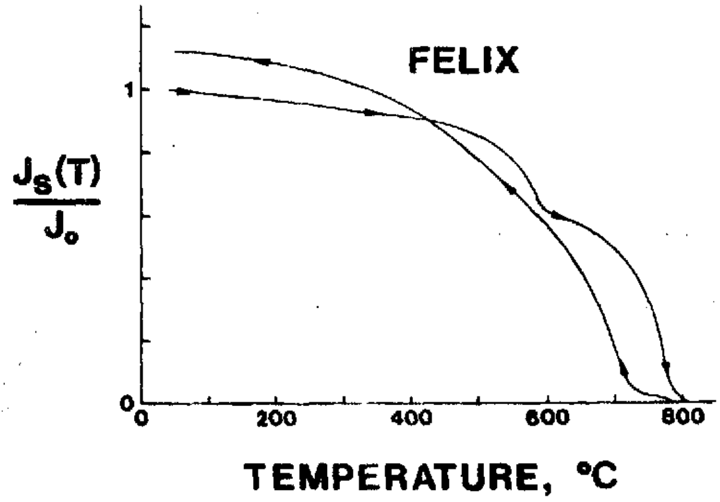

Fig. 52. J_s-T curve for Felix.

Table 8: Magnetite content of the Bali, Grosnaja, Ornans, and Karoonda meteorites

Wt. % Fe_3O_4		Sample Weight (mg)
BALI*		
3.00 ± 0.18 **		0.36
GROSNAJA*		
2.11 ± 0.13		3.42
1.90 ± 0.11		9.62
Weighted average	2.0 ± 0.12	
ORNANS		
4.49 ± 0.27		0.85
4.33 ± 0.26		1.57
4.20 ± 0.25		2.59
Weighted average	4.29 ± 0.26	
KAROONDA		
8.23 ± 0.49		0.97
7.62 ± 0.46		5.52
Weighted average	7.71 ± 0.46	

* Run on whole samples.

** Error is analytical only and does not include any attempt to evaluate sample inhomogeneity.

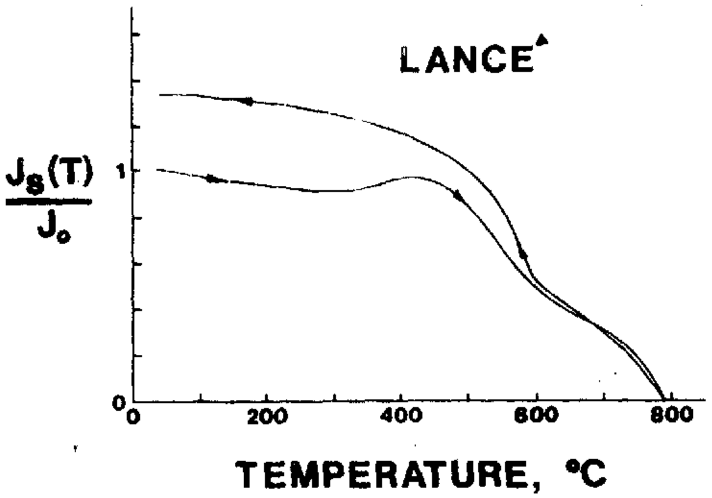

Fig. 53. J_s-T curve for Lance'.

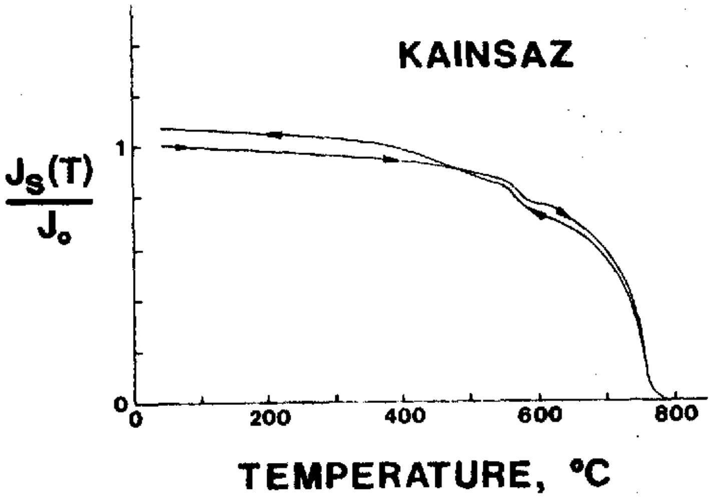

Fig. 54. J_s-T curve for Kainsaz.

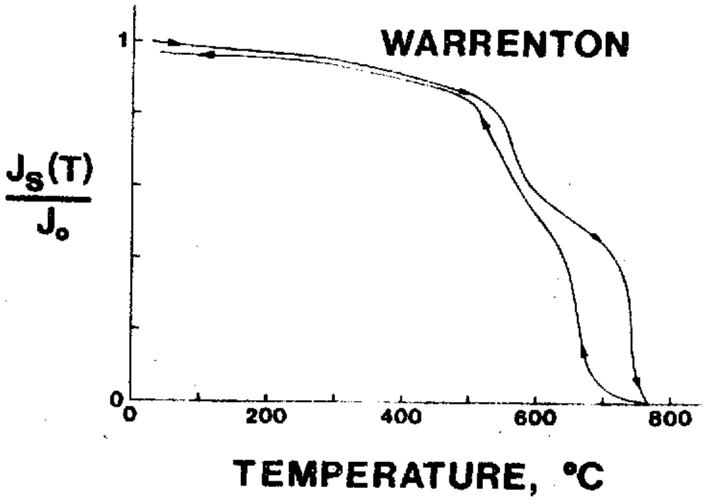

Fig. 55. J_s-T curve for Warrenton.

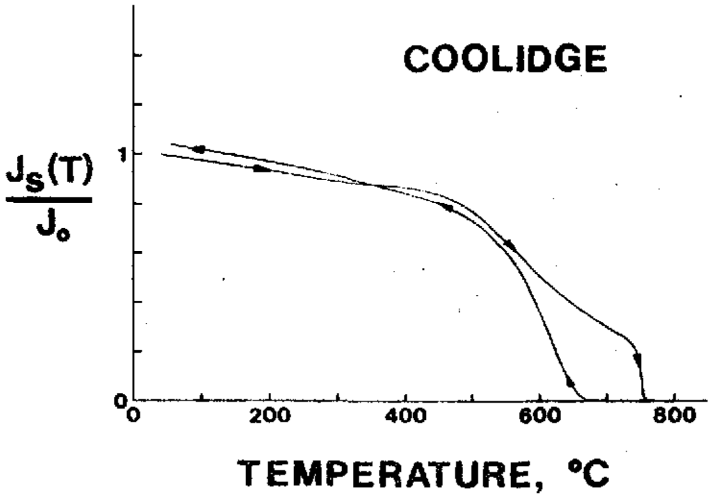

Fig. 56. J_s-T curve for Coolidge.

sense. Of these meteorites the Lance' sample (Fig. 53) is the only one showing pronounced magnetite formation during the experiment. A metallic iron component is present in the sample. The observed Curie temperature of 770°C and the lack of a $\gamma \rightarrow \alpha$ transition upon cooling indicate that the metallic iron contains $\leq$ 2% Ni. The data suggest the presence of some magnetite prior to conducting the exper experiment. The Kainsaz sample (Fig. 54) is indicative of a similarly low nickel, metallic iron component. A small amount of magnetite is indicated by the inflection in both the heating and cooling curves at about 580°C. The samples of Felix (Fig. 52), Warrenton (Fig. 55), and Coolidge (Fig. 56) show evidence for some magnetite in addition to metallic iron containing 4-9% Ni as estimated from the $\gamma \rightarrow \alpha$ transition temperatures.

Thermomagnetic Analysis of Ureilites: We observe the presence of metallic iron of low nickel content ($\leq$ 2% Ni) in all samples which we studied of the ureilites. In fact, this was the only component observed in Dyalpur (Fig. 9), Goalpara (Fig. 57), the Havero (Fig. 58). The J_s-T curve for the Novo Urei sample, shown in Fig. 12, indicates the presence of magnetite in addition to metallic iron. The ureilites Dingo Pup Donga (Fig. 59) and North Haig (Fig. 60) show some degree of magnetite formation during laboratory heating. The J_s-T curves for these two meteorites are suggestive of the presence of some "initial" magnetite in addition to the metallic iron component and the magnetite formed during the experi-

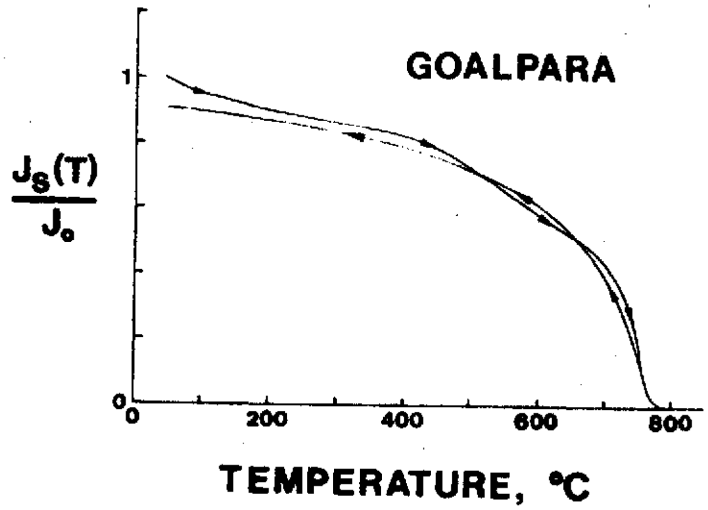

Fig. 57. J_s-T curve for Goalpara.

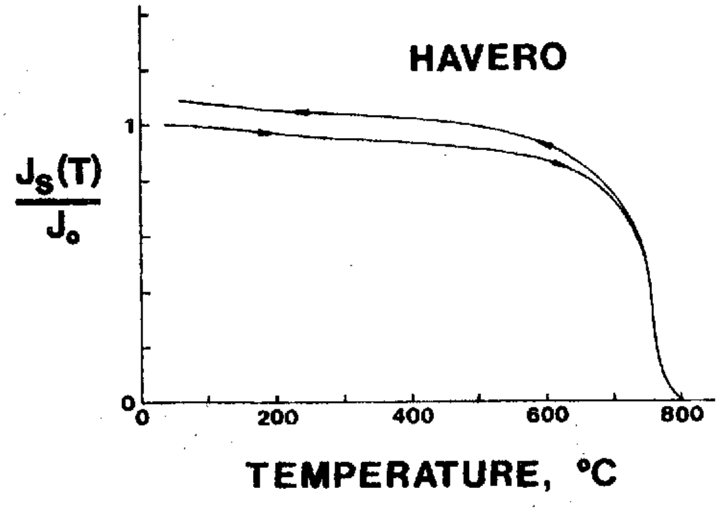

Fig. 58. J_s-T curve for Havero.

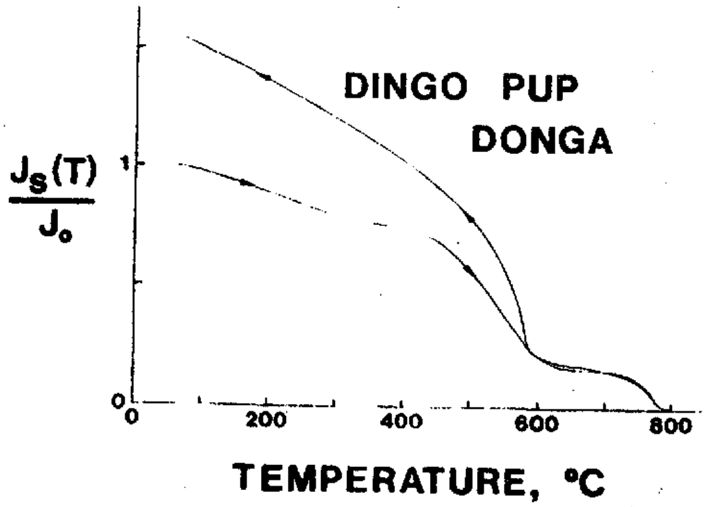

Fig. 59. J_s-T curve for Dingo Pup Donga.

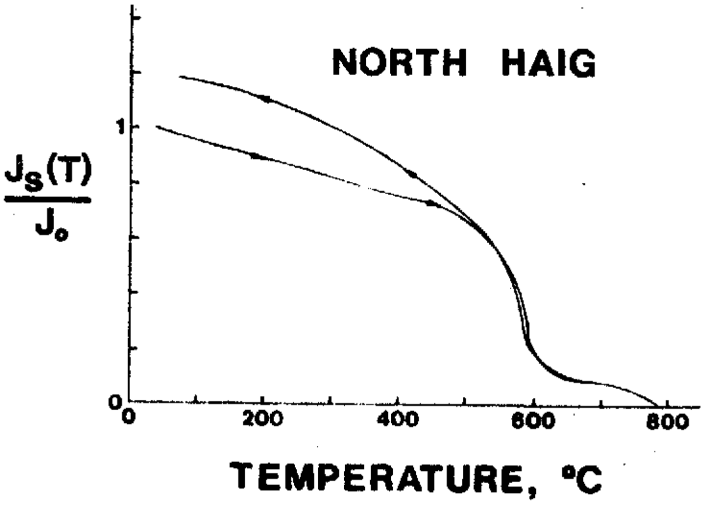

Fig. 60. J_s-T curve for North Haig.

ment. Due to experimental difficulties, quantitative determinations of the magnetite abundance in the ureilites were not made.

Assessment of Thermal Stability: From the data presented above it is apparent that about half of the carbonaceous meteorite samples studied in this work show pronounced thermal alteration during laboratory heating; the alteration being expressed as an increase in saturation magnetization resulting from magnetic formation. We have shown that this alteration is identical to that observed during similar experiments on troilite separates. Some aspects of the oxidation of troilite are discussed in the following sections of this dissertation. As regards the thermal stability of carbonaceous meteorites in Thellier-type paleointensity studies, it is of interest to note that the above thermomagnetic studies were made on milligram size meteorite samples. Since paleointensity studies typically require gram size samples, we cannot reasonably exclude the possibility that even those meteorites (e.g. Orgueil) of relatively low iron sulfide content may in some cases show pronounced magnetite production due to sample inhomogeneity. This conclusion is substantiated by the work of Banerjee and Hargraves (1972).

In designing future paleointensity studies it appears necessary to devise a method to prevent the oxidation of troilite during laboratory heating. We have determined experimentally that control of the prevailing oxygen fugacity is in itself not sufficient to prevent the alteration of troilite. If the oxygen fugacity is maintained within the magnetite stability field, then magnetite will form when troilite is heated. Similarly, if the oxygen fugacity is maintained in the metallic iron stability field, troilite will produce metallic iron upon heating. In order to prevent the oxidation of troilite, it is necessary to maintain a sufficient sulfur fugacity during heating. It may be possible to achieve this by using COS and CO_2 in the gas mixing system or by using an appropriate solid state buffer system. Clearly this is a fruitful area for further study and an area which should be investigated prior to destroying gram size quantities of rare carbonaceous meteorites in paleointensity studies.

THE ORIGIN OF MAGNETITE AND PYRRHOTITE IN CARBONACEOUS CHONDRITES

Although being the subject of numerous inquires since the first observed fall (and recognition) of a carbonaceous chondrite in 1806, the extremely fine-grained structure of these meteorites has presented considerable difficulty in attempts to identify the mineral assemblages present. The mineral phases observed in the carbonaceous chondrites are sufficient unique with respect to other meteorite classes as to pose serious problems in attempting to understand the nature of the physical processes associated with their formation (DuFresne and Anders, 1962). We consider here two minerals previously reported in C1 chondrites - magnetite and pyrrhotite - and propose that magnetite originated from the mild oxidation of troilite. troilite. Pyrrhotite is expected as a direct consequence of magnitite formation via this reaction. Previously meteoritic magnetite was considered to have been derived from the oxidation of metallic iron (Urey, 1952) or from the alteration of olivine (Jeffery and Anders, 1970). No adequate genesis has previously been proposed for pyrrhotite (Kerridge, 1972).

While conducting thermomagnetic analysis under vacuum on meteoritic troilite, Lovering and Parry (1962) observed the formation of magnetite from troilite upon heating. As previously discussed, we have thermomagnetically observed this reaction in some carbonaceous chondrites and in troilite separates (from the Staunton iron meteorite) both under vacuum and in our controlled oxygen fugacity atmosphere. That magnetite was observed to form under vacuum suggests that the reaction involved is simply oxidation and not some gas phase reaction peculiar to the gases in our system. The rapidity of magnetite formation observed in these experiments prompted this author to further investigate the reaction as a potential method by which meteoritic magnetite may have formed in the early solar system.

Previously meteoritic troilite has been considered to be a stable mineral phase below temperatures of about 680°K in the early solar system. Urey (1952) demonstrated on thermodynamic arguments that in an atmosphere of solar composition of H_2 and H_2S that the reaction as written is expected to proceed to the right below

$$Fe(s) + H_2S(g) \rightleftarrows FeS(s) + H_2(g) \qquad (1)$$

the equilibrium temperature of ~680° K. The inferred "stability", however, is relevant only with respect to the gaseous reactants in equation (1) and therefore yields no information on the reactivity of troilite with other gaseous reactants such as oxygen.

Despite the ubiquitous presence of troilite in chondritic meteorites, few inquiries have been made regarding the oxidation of troilite. At temperatures above 413°K troilite converts to hexagonal low-temperature pyrrhotite (Taylor, 1958). Taylor (1970) has determined the approximate path pyrrhotite (and presumably stoichiometric troilite) will take upon oxidation. In Taylor's studies samples of pyrrhotite were oxidized in air. Initially pyrrhotite became increasingly iron deficient. Continued oxidation resulted in the formation of the end member of the oxidation series, pyrite (FeS_2). During the oxidation of pyrrhotite, the effective sulfur fugacity increased without addition of sulfur, whereas the iron activity decreased; the iron being partitioned into the metallic oxide phase.

From thermomagnetic experiments conducted on troilite at constant temperatures, we have observed significant magnetite formation at temperatures as low as 373°K, provided the oxygen fugacity was held in the magnetite stability field and that the troilite was sufficiently fine-grained. As would be expected for a grain surface reaction, the rate and extent of reaction is a strong function of grain size.

To suggest that meteoritic magnetite resulted from the oxidation of troilite, it is first necessary to demonstrate that such magnetite formation is possible under conditions expected in the early solar system and then to examine the petrologic evidence that would be expected to result as a consequence. It is therefore instructive to consider the oxygen fugacity equilibria expected in the early solar system.

Latimer (1950) and Urey (1952) first investigated the oxidation states of iron in the dust phase of the solar nebula, demonstrating for the reaction

$$\frac{3}{4}\,\mathrm{Fe(s)} + \mathrm{H_2O(g)} \rightleftarrows \frac{1}{4}\,\mathrm{Fe_3O_4(s)} + \mathrm{H_2(g)} \qquad (2)$$

that the equilibrium constant $K = H_2/H_2O$ is determined by the relative abundance of hydrogen and oxygen in the solar nebula. The equilibrium constant is further related to the standard free energy change, ΔG^o, by the well known thermodynamic relation

$$\Delta G^o = -RT \ln K \tag{3}$$

Thus, with ΔG^o obtained from thermodynamic data and K determined by the solar H_2/H_2O ratio, the equilibrium temperature of ~ 400°K is calculated. The equilibrium temperature determined from equations (2) and (3), provides a useful "go/no-go" criterion for the formation of magnetite from the oxidation of metallic iron. The formation and subsequent stability of magnetite formed from the oxidation of troilite is subject in part to the thermodynamic considerations implicit in equation (2). With no loss of generality, we examine the separate equilibria involved in equation (2) which are related by the oxygen fugacity, f_{O_2}.

If we assume in an atmosphere of solar composition that the oxygen fugacity is governed by the gas phase reaction

$$H_2(g) + \frac{1}{2} O_2(g) \rightleftarrows H_2O(g) \tag{4}$$

we can calculate the oxygen fugacity as a function of temperature with the expression

$$-\log_{10}(f_{O_2}) = -\frac{2 \Delta G_f^o}{2.3 RT} - 2 \log_{10}(H_2O/H_2) \tag{5}$$

where the ratio H_2O/H_2 is determined from cosmic abundance tables

(Cameron, 1967) and ΔG_f^o is taken from thermodynamic data (JANAF tables, 1962, et seq.). We can further calculate the oxygen fugacity equilibria for reactions

$$3\ Fe(s) + 2\ O_2(g) \rightleftarrows Fe_3O_4(s) \tag{6}$$

and

$$Ni(s) + \tfrac{1}{2}O_2(g) \rightleftarrows NiO(s) \tag{7}$$

with the expression

$$-\log_{10}(f_{O_2}) = -\frac{\Delta G_f^o}{n(2.3)\ RT} \tag{8}$$

using the appropriate thermodynamic data and noting that for equations (6) and (7) n = 2 and n = 1/2, respectively. For the temperature under consideration here FeO equilibria can safely be ignored.

The results of these calculations are shown in Fig. 61. At temperatures below ~ 400°K the oxygen fugacity expected in an atmosphere of cosmic composition of H_2 and H_2O lies within the magnitite stability field. Since we have experimentally determined that oxidation of troilite can proceed at temperatures as low as 373°K, the oxygen fugacity criterion is permissible. Note that the oxygen fugacity expected in the early solar system lies well within the metallic nickel stability field at all temperatures. Under equilibrium conditions this excludes the formation of nickel oxide. Therefore nickel must exist in a metallic state or in some other

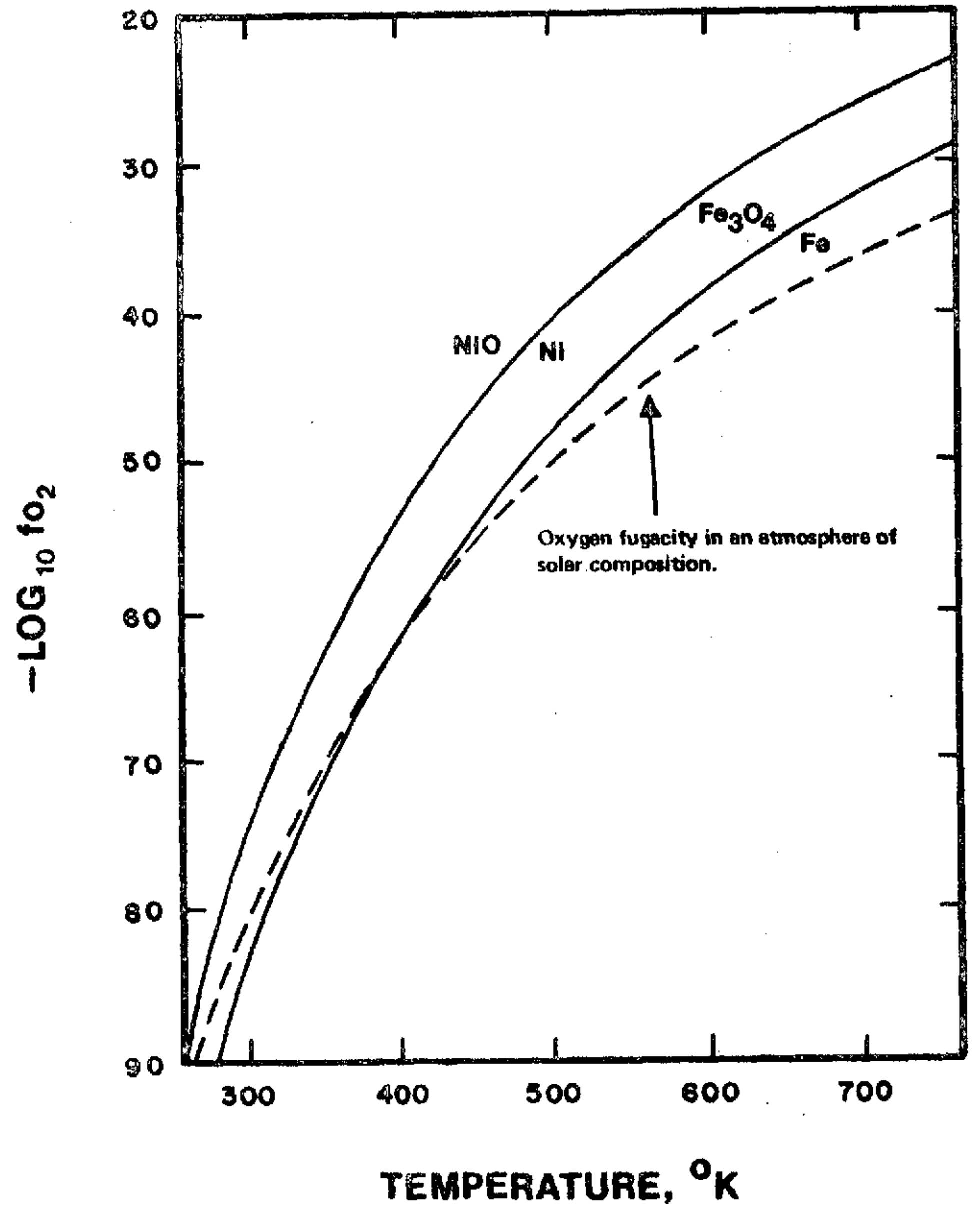

Fig. 61. Ni-NiO, Fe-Fe_3O_4 oxygen fugacity stability fields and the equilibrium oxygen fugacity expected in an atmosphere of cosmic abundance of H_2 and H_2O (as indicated by the broken curve).

chemically stable form such as a sulfide.

Consider now the oxidation of meteoritic troilite under conditions expected in the early solar system. During the oxidation of troilite, the sulfide mineral becomes increasingly iron deficient. The nickel contained in troilite (~ 0.2 wt. %) cannot form an oxide phase at the oxygen fugacities expected and therefore must remain in the sulfide phase. Thus the mineral becomes increasingly rich in nickel, while becoming deficient in iron. The degree to which sulfur is partitioned between the sulfide mineral and the gaseous sulfur phase will be a function of the prevailing sulfur fugacity in the system. One factor difficult to evaluate is the partial pressure of sulfur. In a fully condensed system subjected to mild reheating, the sulfur pressure will depend on whether the system is closed, as would be expected for interior regions of a planetoid; or open (even partially open) as would be expected for small porous bodies, for near surface regions of the planetoid, or for material dispersed in space. In an incompletely condensed system additional complications arise which necessitate knowledge of the amount of sulfur present in the vapor phase. It is obvious that the extent and rate of reaction will be governed in part by the existing partial pressure of sulfur. In any event, it is not difficult to conceive of the possibility of a fully condensed system subjected to mild heating and open to the loss of the gaseous sulfur phase. Such a system closely approximates conditions existing during our experimental studies.

Several consequence can be predicted and should be observed if magnetite formed from the partial oxidation of troilite. On the basis of the chemical arguments presented above we expect that magnetite formed from troilite would contain little nickel. Magnetite in the Orgueil meteorite has been found to contain little nickel (Bostrom and Fredricksson, 1966; Kerridge, 1970). Secondly, we expect the sulfide minerals resulting from the oxidation of troilite to attest to the degree of oxidation; a complex consequence of temperature, duration, grain size, and sulfur fugacity. Large grains subjected to mild oxidation would be expected to form pyrrhotite. Meteorite pyrrhotite has been observed to occur in the Revelstoke and Orgueil Cl chondrites (Folinsbee, et al., 1967; Kerridge, 1972). More extensive oxidation may be expected to form high-nickel sulfide phases such as pendlandite, [$(Fe, Ni)_9S_8$]. The limited data available is suggestive of this (Wood, 1967a). Extreme oxidation should lead to production of pyrite. The only meteorite in which pyrite has been observed is the "recrystallized" carbonaceous chondrite, Karoonda, which contains abundant (8%) magnetite (Ramdohr, 1963). Additionally, mild oxidation of troilite is expected to provide a potential source of SO_2 for the formation of the soluable sulfate species observed in carbonaceous chondrites (DuFresne and Anders, 1962). If so, we would predict that sulfur in the sulfate phase should be isotopically lighter than the sulfur in the sulfide phase. For the Orgueil meteorite, at least, this is the case. Monster et al. (1965) measured average $\delta^{34}S$ values of -1.3% and +2.6% for the sulfate and sulfide phases, respectively. Finally, we expect to observe occasionally meteoritic magnetite associated with sulfur minerals as have been observed in carbonaceous chondrites: magnetite-pendlandite (Sztrokay, 1960); magnetite-pyrite (Ramdohr, 1963); magnetite-sulfate (Nagy, et al., 1962).

With the limited experimental data available on sulfide reactions, we can not exclude other reaction mechanisms for the formation of species such as pendlandite. Since the only two observations of pyrrhotite in carbonaceous chondrites (Orgueil and Revelstoke) have been in two meteorites which contain significant magnetite (11.9% and 7.2%, respectively) it is likely that this mineral resulted singularly as a consequence of the mild oxidation of troilite.

LIMITATIONS ON PALEOINTENSITY STUDIES

The results of our alternating field demagnetization studies as well as results from other laboratories (Banerjee and Hargraves, 1971, 1972; Butler, 1972; Brecher, 1972) point to the same general conclusion - that many, if not all, carbonaceous meteorites possess a stable remanent magnetization of extraterrestrial origin. In 1972, three attempts were made at estimating paleointensities from experimental data on several of these meteorites. Banerjee and Hargraves (1972) and Butler (1972) employed a step-wise variant of the Thellier technique of thermal demagnetization followed by remagnetization in a field of known intensity. This procedure was repeated at successively higher temperatures. Brecher (1972) used a single step variant of the Thellier technique which involved heating the sample in argon to a temperature of 150-250°C and allowing the sample to cool in a known field (thus acquiring a PTRM). The a.f. demagnetization of this PTRM was compared with the previously determined a.f. demagnetization of the NRM. The different choices of experimental technique employed by the above mentioned workers led to different interpretations of their data. Banerjee and Hargraves (1972) and Butler (1972) recognized from their data that at temperatures above 90-130°C "physicochemical changes" occurred, although not associating these changes with the oxidation of troilite. Therefore, they utilized only their low temperature data for paleointensity estimates. Brecher (1972) found that her data on Allende, Murchison, and

Murry did not extrapolate to zero (as would be expected from a single component TRM) and suggested the "presence of one or several (preaccretionary?) CRM components." It is significant that her data on Renazzo did extrapolate to zero. From our thermomagnetic studies, it appears likely that the "physico-chemical changes" and the "non-zero extrapolation" discussed above resulted from the oxidation of troilite during laboratory heating. The fact that Renazzo was the only carbonaceous chondrite studied that was "well behaved" is consistent with this hypothesis. The metallic iron content of this meteorite (~ 12%) is quite high (Mason, 1971). Contributions to the magnetization resulting from troilite oxidation would be expected to be relatively small. Banerjee, Brecher, and Butler (personal communications) have stated that their paleointensity data show the reverse of the effect expected from the simple addition of magnetite during the experiment. However, additional studies (D. E. Watson, personal communication) suggest that at early stages in the formation of magnetite from the oxidation of troilite (in a known magnetic field) the magnetite develops a magnetization antiparallel to the direction of the applied field. If confirmed this would explain the ambiguities mentioned above. The paleointensities determined for the five carbonaceous chondrites studied ranged from 0.2-2.0 Oe, values not significantly different from those found for ordinary chondrites (Stacey, et al., 1961; Weaving, 1962a; Gus'kova, 1963), iron and stony-iron meteorites (Gus'kova, 1965a).

In addition to the experimental problems encountered during laboratory heating, the magnetization of carbonaceous meteorites may be considerably more difficult to interpret than that of other meteorite classes. The possibility that chemical reactions (CRM) or depositional effects (DRM) either altered or contributed to their magnetization is enhanced by the extremely fine-grained (sub-micron) mineralogy of these meteorites. The funamental problem involved reduces to one of establishing the validity of applying the Thellier technique to these primitive meteorites. It must be established that the magnetization of these meteorites was acquired as a PTRM during cooling following their accretion as meteorites (or meteorite parent bodies). Furthermore, it must be demonstrated that the magnetic minerals responsible for the observed NRM are primary phases. Exclusion of possible chemical remanent magnetization effects necessitate additional studies of the magnetic memory involved in reactions with troilite, magnetite, and metallic iron. The observed remanent magnetism of carbonaceous meteorites strongly suggests that magnetic fields of significant intensity were present in the early solar system at some time during their development. Much more work is required before we can hope to elucidate with reasonable certainty the origin, intensity, and duration of these fields.

SUMMARY

On the basis of our alternating field demagnetization experiments we have shown that many (if not all) carbonaceous meteorites possess a stable natural remanent magnetization (NRM). We found that the stability of the NRM is characteristic of a thermal remanent magnetization (TRM), chemical remanent magnetization (CRM), or depositional remanent magnetization (DRM) and is unlike that of a viscous remanent magnetization (VRM) or an isothermal remanent magnetization (IRM). These results are contrary to the result found by Stacey, et al. (1961) on a single sample of the Mokoia meteorite. This implies that the magnetization was acquired extraterrestrially rather than having been induced from either the earth's magnetic field or from exposure to laboratory magnets.

We have conducted thermomagnetic analysis using a system for oxygen fugacity control for the first time on carbonaceous meteorites. As the constituent minerals in carbonaceous meteorites are considerably more fine-grained than those typically found in other meteorite classes, our experiments represent a severe test for the system. By maintaining the appropriate oxygen fugacity during the experiment, we have found that it is possible to heat meteorite samples containing magnetite and metallic iron to temperatures as high as 800°C without significantly reducing the magnetite or oxidizing the metallic iron.

On the basis of our thermomagnetic analyses we have shown that troilite, a component of virtually all chondritic meteorites, is unstable; reacting with gaseous oxygen to form magnetic rapidly at temperatures at least as low as 100°C. We have found that oxygen fugacity control is not in itself a sufficient means to prevent the oxidation of troilite. We have suggested that the results of magnetic studies conducted by Banerjee and Hargraves (1972), Brecher (1972), and Butler (1972) were severely affected by troilite oxidation during the experiment. We have suggested that further destructive thermal demagnetization experiments on rare meteoritic material be deferred until development of a system capable of simultaneously controlling oxygen and sulfur fugacities. We have proposed the use of CO_2 and COS for this purpose.

We have utilized room temperature saturation magnetization measurements and thermomagnetic analyses for determining the magnetite content of many of the carbonaceous chondrites. Estimates of Fe_3O_4 content by alternative methods have been rare and, as far as we know, confined to the Orgueil meteorites.

We have proposed that meteoritic magnetite formed in the early solar system from the oxidation of troilite rather than from the oxidation of metallic iron as proposed by Urey (1952) or from the alteration of olivine as suggested by Jeffrey and Anders (1970). We have shown that the formation of pyrrhotite is an expected consequence of magnetite formation from the oxidation of troilite. Unless an alternative mechanism is found to explain the observed occurrence of pyrrhotite in Cl chondrites, this evidence stands as verification of our thesis. We have further shown that the occurrence of pyrite in the Karoonda meteorite may also result from the oxidation of troilite.

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VITA

James Marvin Herndon was born in [redacted] [redacted] [redacted] [redacted] [redacted] to James M. and Frances G. Herndon. He graduated from Winyah High School, Georgetown, South Carolina. He served on active duty for two years in the United States Naval Reserve. In May, 1970 he received a Bachelor of Arts degree with a major in physics and a minor in music from the University of California, San Diego.

He is married to the former Mary Anne [redacted] and has one son, Oliver Wynn.

His permanent mailing address is 10634 Sanfred Court, Santee, California.

The typist for this dissertation was Mrs. Catherine Mieth.