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Magnetic alignment of nonmagnetic silicates caused by paramagnetic anisotropy: origin of polarization observed in planetary formation region

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Possible dust alignment in dense regions due to paramagnetic anisotropy $\Delta\chi_{PARA}$ is discussed based on the results of a laboratory experiment on ortho-pyroxene grains containing a small amount of Fe^{2+} ions (1.8 mol%). Ortho-pyroxene has been detected in circum-stellar regions by infrared emission spectroscopy. Our experiment was performed at room temperature using He gas as the dispersing medium. Although the grains do not contain strong magnetic moments, alignment was achieved at low field strength (<2000 G). The alignment efficiency of ortho-pyroxene was compared with those of various rock-forming minerals. The magnitude of diamagnetic anisotropy $\Delta\chi_{DIA}$, free of paramagnetic ions, is generally $<10^{-8}$ emu/g for various silicates. In contrast, $\Delta\chi_{PARA}$ increases considerably with increasing Fe^{2+} concentration, reaching 10^{-5} emu/g for many of the silicates when the concentration of Fe^{2+} is >1 mol%; this increasing $\Delta\chi_{PARA}$ is the cause of the above-mentioned alignment obtained at the low field strength. Based on our observations, we infer the field intensity needed to obtain partial dust alignment of sufficient size to explain the observed polarization in astrophysical environments. Due to temperature dependences caused by a Curie Law and a rotational Brownian motion, the field intensity required to cause the alignment is expected to decrease considerably at low-temperature conditions assumed for a proto-planetary disk. The results of our experiment performed at room temperature provide a technical basis to reproduce grain alignment under such temperature conditions.

Key words: Magnetic field direction, dust alignment, ortho-pyroxene grains, proto-planetary disks, paramagnetic anisotropy.

1. Introduction

The direction of magnetic field B in a diffuse region is frequently estimated from infrared or visible polarization P caused by partial dust alignment; a weak galactic field of approx. 3 μG is considered to be the cause of alignment. Because the geometrical relationship between P and B strongly depends on the mechanisms of alignment, it is important to firmly establish this mechanism to obtain a reliable field direction from polarimetry data. The conventional mechanism of alignment (the so-called a "Davis-Greenstein model") is based on a paramagnetic relaxation process of magnetic moments included in the dust (e.g. Spizer, 1978). This model requires an extremely high rotational energy relative to the thermal energy—a condition that is only met in a diffuse interstellar region. The model is not realistic for planetary formation regions wherein thermal equilibrium is achieved between dust and gas (Whittet, 1992). Consequently, alternative mechanisms for the origin of magnetic alignment should be considered for solid materials that do not possess a strong magnetic moment. According to data compiled on solar elemental abundance and infrared emission observational data, high concentrations of magnetic ions cannot be expected in the dust particles. The current theory of magnetic alignment of solid particles is that it occurs only for materials possessing a strong magnetic moment.

Alignment of micron-size crystals caused by the anisotropy of magnetic susceptibility $\Delta\chi$ was recently observed for common rock-forming silicates, namely kaolinite, talc, muscovite and biotite (Uyeda *et al.*, 1991, 1993). The experiment was performed at room temperature (T = 300 K) using liquid ethanol as the dispersing medium. Partial alignment occurred by a simple balance between Brownian energy $1/2\,k_BT$ and field-induced anisotropy energy $1/2\,M\Delta\chi B^2$, as was first proposed by Langevin and Curie (1910); where M denotes the mass of the particle and $\Delta\chi$ describes anisotropy per unit mass. This process of grain alignment has been quantitatively studied on inorganic material, such as clay and mica minerals (Uyeda *et al.*, 1993, 2003, 2005a) where it was explicitly shown that alignment was controlled by $\Delta\chi$, T and M. This result means that the minimum field intensity required to achieve alignment is predictable, provided that the numerical values of these three parameters can be determined. This mechanism of aligning nonmagnetic particles was considered to be a promising technique to develop new types of industrial material (e.g. Maret and Dransfield, 1985). When an axis of

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a grain material possesses high material functionality, such as in terms of hardness, conductivity or optical efficiency, among others, this functionality can also be preserved in a grain aggregate provided that the grain axes are orientated in one direction. Based on this principle, attempts were made to produce large single crystals of protein, synthetic bone materials or aggregations of carbon nano-tubes. However, to date, only a limited number of materials have been investigated. Alignment is believed to occur only at a field intensity >10,000 G, which requires a superconducting magnet.

Recent observations of infrared emissions have enabled the dust in the planetary formation region to be identified as rock-forming minerals, such as forsterite, ortho-pyroxene, diopside or hibonite; these are frequently observed in chondritic meteorites (e.g., Hanner *et al.*, 1995; Malfait *et al.*, 1998; Sitko *et al.*, 1999; Honda *et al.*, 2003; Hofmeister *et al.*, 2003). The observations indicated that these silicates had Mg-rich components. Based on the observed 69-μm feature, the Fe concentration of the forsterite grains was estimated to be <5% (e.g. Molster *et al.*, 2002). On the other hand, a small shift observed for the 11.44-μm feature of Vega-like star HD 14563 indicated the existence of Fe-bearing forsterite (Honda *et al.*, 2004). Fe-bearing silicates are commonly seen in chondritic meteorites as well as in interplanetary dust particles (IDP) that are derived from the early solar system. It is likely that sufficient amount of silicate dusts in the circum-stellar region contain a small amount of Fe^{2+} ions (below several percent).

Here we report on an experimental study on the process of magnetic alignment. The experiment was carried out at room temperature on micron-sized ortho-pyroxene crystals dispersed in He gas, and the result was analyzed based on the $\Delta\chi$ values measured for bulk single crystals. Because temperatures of proto-planetary disks are estimated to range between $T = 400$ K and 50 K at a radius between 2 AU and 50 AU (Willacy and Langer, 2000), we discuss the effect of reducing the temperature on the magnetic alignment process. The practicability of dust alignment due to the above-mentioned process proposed by Langevin and Curie is quantitatively evaluated for the proto-planetary disks since reliable values of the three parameters are obtained for this region; values of T and M (dust size) are estimated from astronomical observation, whereas the $\Delta\chi$ value is determined by the magnetic-anisotropy measurement of a bulk single crystal composed of the same material as that identified in proto-planetary disks.

2. Experimental

A method to measure the alignment process in He gas recently developed by Uyeda *et al.* (2001) is shown schematically in Fig. 1. A large Dewar (inner diameter: 150 mm; inner height: 400 mm) serves as a sample chamber. This cell size was necessary to maintain the dispersed particles in the gas phase during the time of the experiment. The cell was placed at the center of a Helmholtz coil system having an inner diameter 1000 mm; the coil produced a homogeneous field up to 700 G within a spherical area 500 mm in diameter at the center of the system. The inhomogeneity of the field was less than 1000 ppm inside the sphere. The interior of the Dewar was placed where the field is homogeneous. Single-crystal grains of ortho-enstatite ($Mg_{0.983}$, $Fe_{0.017}$, from Morogoro, Tanzania) were used. The sample was free of ferromagnetic impurities and contained 1.8 mol% paramagnetic Fe^{2+} ions. For the bulk single crystal, χ at $T = 300$ K was measured using standard procedures (Uyeda *et al.*, 2005b). Measured anisotropy between the *c-a*, *c-b* and *b-a* axes were $\Delta\chi_{c\text{-}a} = 1.2 \times 10^{-5}$ emu/g, $\Delta\chi_{c\text{-}b} = 1.0 \times 10^{-5}$ emu/g and $\Delta\chi_{b\text{-}a} = 0.2 \times 10^{-5}$ emu/g, respectively. Magnetic susceptibility of the sample measured by a vibrating sample magnetometer (VSM) was $\chi_{\text{PARA}} = 4.4 \times 10^{-5}$ emu/g.

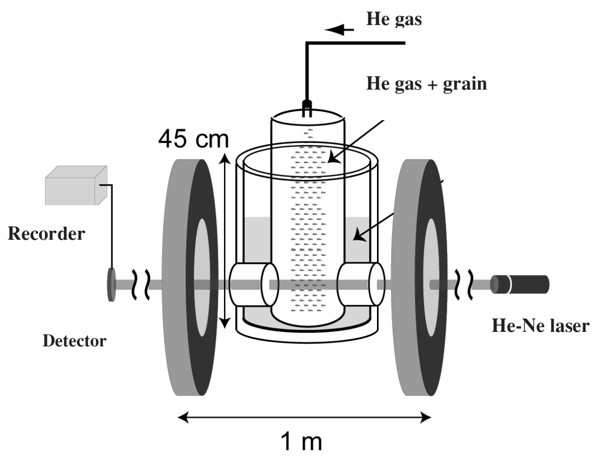

Fig. 1. Schematic view of apparatus developed to measure magnetic grain alignment under low-temperature conditions between $T = 100$ K and 10 K (Uyeda *et al.*, 2001, 2004c). He gas is used as the dispersing medium. A set of Helmholz coils produces a homogeneous magnetic field in a spherical area of 100 mm in diameter; the sample suspension is included in this area. Alignment of graphite grain at $T = 110$ K was recently performed using this setup (Uyeda *et al.*, 2004c).

The bulk crystal sample was ground to a micron size and placed in the ampoule. The ampoule was connected to the sample chamber by a copper pipe. The interior of the chamber, the ampoule and the pipes were filled with He gas at 1.0×10^5 Pa. Thereafter, additional He gas was supplied to the ampoule at an enhanced pressure of about 1.1×10^5 Pa for about 1 s, and the sample grains were dispersed into the He gas. The gas with dispersed grains was injected into the chamber. The degree of grain alignment at arbitrary field intensity is obtained from the intensity of the He-Ne laser beam transmitted through the sample suspension in a direction parallel to the magnetic field. This can be done because the direction of an optical principle axis of a crystal grain coincides with the direction of the magnetic principle axis. The efficiency of the above method has been established in earlier experiments (Uyeda *et al.*, 2001, 2003). A small portion of the dispersed grains was collected on a copper plate immediately after each experimental run was completed. M values at the time of the experiment were estimated as $M = (1.9 \pm 1.0) \times 10^{-14}$ g, which was obtained from the size distribution (1.1 ± 0.5 μm in diameter) observed on scanning electron microscope (SEM) images and volume density $\rho = 3.2$ g/cm^3.

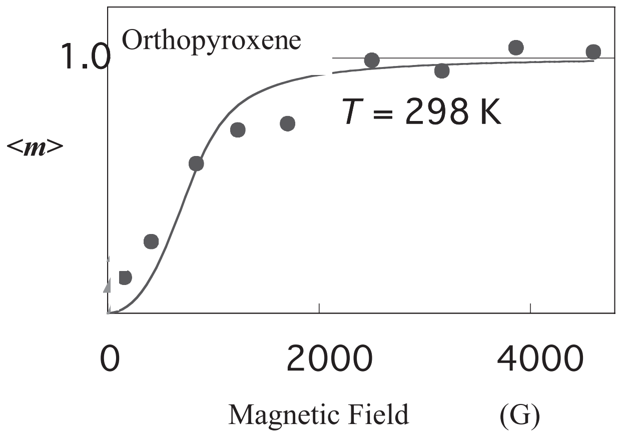

Fig. 2. Magnetic alignment of ortho-pyroxene grain observed at room temperature ($T = 300$ K). Solid symbols indicate measured relationships; solid curve represents theoretical fit to the measured data. Degree of alignment is described by an order parameter $\langle m \rangle$; its definition is given in the text.

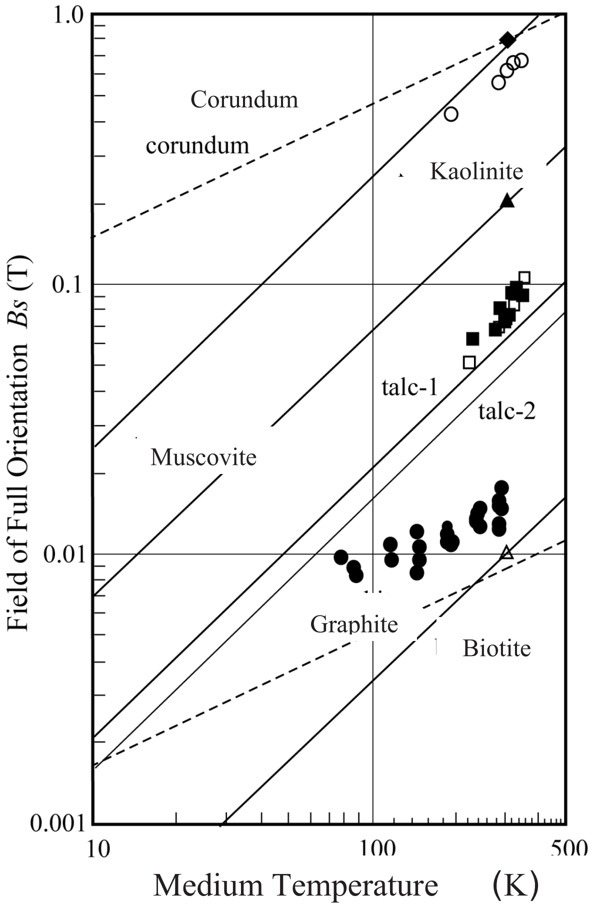

Fig. 3. Relationship between field of orientation B_s and temperature compiled for various silicate (Uyeda *et al.*, 2005b). Definition of B_s is given in the text. Experimental data of graphite (solid circles) are measured by the system described in Fig. 1.

3. Measured Results of Magnetic Grain Alignment

Results of the grain alignment are shown in Fig. 2 for ortho-pyroxene grains. The degree of the alignment is defined by an order parameter $\langle m \rangle$, which is an average of $1/2\left(3\cos^2\theta - 1\right)$ over an ensemble of measured grains. Here θ denotes an angle between a magnetically stable axis of a grain and B. The average θ value at a completely random state is 54.7°, whereas θ is equal to 0° at a completely ordered state. The completely ordered and random states are described as $\langle m \rangle = 1$ and $\langle m \rangle = 0$, respectively. The parameter $\langle m \rangle$ has been conventionally used to describe an aligned state of liquid crystal. An almost full alignment is achieved at a field below 2000 G. It has not been considered possible for magnetic grain alignment to occur at such a low field intensity for oxide particles without spontaneous magnetic moment; however, we have achieved alignment in the field of a simple hand magnet. A similar effect caused by paramagnetic ions was also observed for common silicates, such as biotite, muscovite, orthoclase, talc and kaolinite (Uyeda *et al.*, 2003, 2005a).

A theoretical fit to measured data is calculated from a Boltzmann average of $1/2\left(3\cos^2\theta - 1\right)$ in terms of a field-induced free energy of particle $U = -\left(MB^2/2\right)\left\{\chi_\perp + \Delta\chi\cos^2\theta\right\}$; magnetic anisotropy $\Delta\chi$ is described as $\Delta\chi = \chi_\parallel - \chi_\perp$, where $\chi_\parallel$ and $\chi_\perp$ are the susceptibilities parallel and perpendicular to the stable axis, respectively. Here, a $M\Delta\chi$ value which gives the best fit to the measured data is used to describe the curves. The adopted $M\Delta\chi$ value in the above calculation was $M\Delta\chi = (4.3 \pm 2.1) \times 10^{-19}$ emu, which is consistent with the value obtained from the experimental M and $\Delta\chi$ values; $M\Delta\chi = (2.3 \pm 1.1) \times 10^{-19}$ emu. The consistency indicates that alignment proceeded by a Langevin process.

4. Effect of M, $\Delta\chi$ and T on the Alignment Process

A field intensity where the anisotropy energy $(1/2)M\Delta\chi B^2$ of the particle was equal to $(15/2)k_BT$ was defined as field of alignment B_s (Uyeda *et al.*, 1993), which was directly calculated from the Langevin process as

$$B_s = (15k_BT/N\Delta\chi)^{1/2}. \quad (1)$$

The value of $\langle m \rangle$ is equal to 0.78 when the external field is equal to B_s. The equation explicitly shows that the field required to align a particle will decrease as T decreases and also decrease as either M or $\Delta\chi$ increase. Complete alignment is achieved only at a infinite field intensity. Therefore, it is necessary to introduce an indicator, such as B_s, in order to carry out quantitative analysis on the alignment process (Uyeda *et al.*, 2005b). In a typical study of magnetic grain alignment, a theoretical B_s value is calculated by inserting observed M, $\Delta\chi$ and T values in Eq. (1), and the obtained value is compared with an experimental B_s obtained from the measured $\langle m \rangle$-B relationship. When the alignment proceeds by the Langevin process, the two independently obtained B_s values show fairly good agreement. The effect of parameters M, $\Delta\chi$ and T on B_s, which is expected from Eq. (1), has been systematically examined by measuring common sheet-silicates, such as muscovite, talc and kaolinite (Uyeda *et al.*, 1991, 1993). The results of these experiments confirmed that the field of alignment could be reduced considerably by increasing M or $\Delta\chi$, and also by decreasing T, as expected from Eq. (1) (Uyeda *et al.*, 2005a).

The effect of M on the B_s value was pointed out by Yamagishi *et al.* (1989) in the course of their analysis of the polymerization process of fiblin fibers in a magnetic field. The effect of M was also discussed by Sazaki *et al.* (1989) when they analyzed the field dependence of the growth of lythozyme crystals. However, these latter researchers found that the reduction rate of B_s did not exceed 10% due to increased M, since the maximum size of the particle that could not be maintained in the dispersed fluid did not exceed several microns in terrestrial gravity. Chihara *et al.* (1998) measured the temperature dependence of B_s for graphite grains free of paramagnetic ions in a temperature range of $T = 300$ K and 180 K, as shown in Fig. 3. The grains were dispersed in liquid ethanol. A $T^{1/2}$ dependence, expected from Eq. (1), was confirmed in this experiment.

For the effect of $\Delta\chi$, paramagnetic anisotropy $\Delta\chi_{\mathrm{PARA}}$ is a dominant factor compared to diamagnetic anisotropy $\Delta\chi_{\mathrm{DIA}}$ for most of the natural oxides. The origin of $\Delta\chi_{\mathrm{DIA}}$ was attributed to preferential orientation of bond direction with respect to a magnetic principle axis of crystal (Uyeda, 2004a). Accordingly, the $\Delta\chi_{\mathrm{DIA}}$ of various silicates was expected to distribute in a range between 10^{-8} and 10^{-10} emu/g. In contrast, $\Delta\chi_{\mathrm{PARA}}$ is derived from the anisotropy of the local crystalline field of a site that is occupied by an isolated paramagnetic ion (Ballet and Coey, 1982). Hence, $\Delta\chi_{\mathrm{PARA}}$ is proportional to the concentration of the paramagnetic ion; it reaches a level of 10^{-5} emu/g when the concentration of Fe^{2+} is about 1 mol%. Such concentrations of Fe^{2+} are often observed in major rock-forming minerals. The effect of paramagnetic Fe^{2+} ions in reducing in B_s at room temperature was specifically observed for biotite grains, which is a typical sheet-silicate (Uyeda *et al.*, 2005a). The value of B_s was found to be as low as 220 Oe for biotite, which had a susceptibility of $\chi_{\mathrm{PARA}} = 4.4 \times 10^{-5}$ emu/g. This B_s was less than 1% of the B_s value observed for synthetic phlogopite that was free of paramagnetic ions, whereas the B_s was 6.2 kOe for phlogopite with $\chi_{\mathrm{PARA}} = 1.2 \times 10^{-6}$ emu/g. The concentration of Fe^{+2} is proportional to χ_{PARA} in the above three minerals. As almost equivalent crystal structures are reported for these minerals, they should have similar amounts of $(\Delta\chi)_{\mathrm{DIA}}$. Hence, the large variation of B_s observed between the three materials can be attributed to the difference in the paramagnetic $(\Delta\chi)_{\mathrm{PARA}}$.

Variations in the temperature are expected to change the values of $(\Delta\chi)_{\mathrm{PARA}}$ and B_s considerably since the paramagnetic moment follows a Cuire-Weiss law. The $\Delta\chi$-T relationships were previously measured for bulk single crystals at between $T = 300$ K and 100 K for biotite and muscovite that contained Fe^{2+} ion at a level of 1 mol%. As temperature decreased from $T = 300$ K to 80 K, the $\Delta\chi$ of biotite increased from 1.2×10^{-5} emu/g to 8.0×10^{-5} emu/g, while the $\Delta\chi$ of muscovite increased from 0.9×10^{-6} emu/g to 5.1×10^{-6} emu/g (Uyeda *et al.*, 2004b). The temperature dependence of B_s was measured between $T = 300$ K and 200 K for crystal grains of talc and kaolinite that contained paramagnetic ions (Uyeda *et al.*, 2003). As shown in Fig. 3, observed B_s of kaolinite and talc was nearly proportional to T since $(\Delta\chi)_{\mathrm{PARA}}$ is proportional to T^{-1} following Curie's law. Here, the magnitude of $(\Delta\chi)_{\mathrm{DIA}}$ was negligible compared to $(\Delta\chi)_{\mathrm{PARA}}$. Hence, when a material contains a finite amount of paramagnetic ion, the reduction rate of B_s with decreasing T becomes much larger than the above-mentioned $T^{1/2}$ dependence of a diamagnetic material. The measurement below $T = 273$ K involves using liquid ethanol as the dispersing medium; at the present time, pure water is used as the dispersing medium in most measurements.

In conclusion, most of the diamagnetic oxides have a potential to cause magnetic alignment, with the exception of cubic crystals and amorphous materials. The underlying reason for this is that the resultant $\Delta\chi$ may be enhanced considerably by increases in the concentration of paramagnetic ions. $\Delta\chi$ increases even more with decreasing temperature.

5. Conditions Required to Reproduce Dust Alignment in Planetary Formation Region

The values of χ_{PARA}, $(\Delta\chi)_{\mathrm{PARA}}$ and B_s observed for ortho-pyroxene are comparable to those of the biotite mentioned above. Consequently, B_s is expected to be considerably reduced in the low-temperature region below $T = 100$ K, which is now in progress for ortho-pyroxene. Precise measurements of both $\Delta\chi$ and B_s are required in the low-temperature region for ortho-pyroxene as well as for other dust minerals. These results may provide quantitative information that can be used to assess whether dust alignment in planetary formation region is caused by a simple Langevin process or not.

The magnetic field is known to be a major factor that controls the evolution of stars and planets. For example, reconnection of magnetic field associated with the protoplanetary disk was recently proposed to be a heat source that produces high-temperature condensates of primitive meteorites (Shu *et al.*, 1997). In order to assess this hypothesis on planetary evolution, it is necessary to carry out high-spatial-resolution polarimetery mapping (about 1 AU). Near infrared (Lucas *et al.*, 2004) and sub-millimeter (Tamura *et al.*, 1999) imaging polarimetry was recently reported for YSO regions as well as for massive young objects (Momose *et al.*, 2001). Spatial resolutions in these observations were about 100 AU although the data indicate that partial magnetic alignment of dust particles does take place in these systems. It is expected that polarimetry surveys with high-spatial resolution will be realized by forthcoming projects such as that of the Akatama Long Mand Association (ALMA).

It is necessary to reproduce the temperature conditions of planetary formation regions, which range between 400 K and 50 K; the setup shown in Fig. 1 is a step towards realizing alignment in a temperature range between $T = 200$ K and 50 K. In addition, our apparatus is capable of performing magnetic alignment in a reduced temperature condition down to $T = 10$ K, which is comparable to that of the molecular cloud. The experiment was possible because He gas is a unique medium that can disperse micron-sized particles and remain in the liquid state above $T = 4.2$ K. At the present time, measurements are only possible at temperatures above 160 K for silicate grains since liquid ethanol is used as a dispersing medium. As mentioned before, it is essential to know the geometrical relationships between B and P for determining the field direction from polarimetry data. This relationship differs considerably depending on the alignment model used. The apparatus shown in Fig. 1 is capable of examining the relationship between P and the direction of the crystalline axes of mineral grains, which is not clearly understood in the dense region.

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