Novel Magnetic Domain Structure in Iron Meteorite Induced by the Presence of L1₀-FeNi

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Iron meteorite shows an extraterrestrial pattern termed as the Widmanstätten structure [Fig. 1(a)]. Its metallographic feature has been of great benefit to planetary scientists for studying the history of the solar system.¹⁻⁴ The scientists have believed that the Widmanstätten structure was formed by the long-range thermal diffusion of Fe and Ni in asteroid’s core over a period of 4.6 billion years.⁵ Meanwhile, the iron meteorite is also characterized by remarkable magnetic properties, namely large magnetic anisotropy and strong coercivity, differing from those of synthetic Fe–Ni alloys.⁶ However, there is no explanation how the magnetic properties are associated with the Widmanstätten structure. From the viewpoint of materials science, the Widmanstätten structure is regarded as Fe–Ni alloy segregated α (bcc-FeNi, Kamagate) and γ (fcc-FeNi, Taenite) lamellae on the micrometer scale [Fig. 1(b)].¹ The Ni concentration in the γ lamella rapidly increases toward the interface,⁷ resulting in several laminated Fe–Ni alloys, namely invar alloy (Fe₃Ni₅₅),⁸ tetrataenite (Fe₆₀Ni₄₀)⁹⁻¹¹ and permalloy (Fe₅₀Ni₅₀).¹²,¹³ The [110]bcc axis maintains its parallel orientation to [111]γ̅, and either [001]bcc or [111]bcc orients parallel to [010]bcc, known as the Nishiyama–Wassermann (NW) or Kurdjumov–Sachs (KS) orientation, respectively.⁶ Such a heterogeneous structure near boundary can be considered as same sort of magnetic multilayer system. In this study, we investigate the magnetic properties of iron meteorite resulting from the Widmanstätten structure for the first time.

Among the Fe–Ni alloys, we pay particular attention to the tetrataenite phase, which is described as a chemically ordered FeNi alloy with L1₀-type superstructure.⁹,¹⁰ It is also well known that tetrataenite exhibits significantly different magnetic properties from synthetic Fe–Ni alloys. Néel et al. reported the magnetic anisotropy energy of tetrataenite as 3.2 × 10⁵ J/m³,⁸ which is more than one order of magnitude larger than those of Fe, invar alloy, ordered permalloy and pure Ni (4.8 × 10⁴, 5 × 10⁴, –2.5 × 10⁴, and –6 × 10⁴ J/m³, respectively).⁷,⁸,¹²,¹³ Our pilot magnetic hysteresis measurement for tetrataenite flakes shows coercive force of more than 1.0 × 10⁵ A/m, which is entirely larger than common Fe–Ni alloys.⁸,¹² In other words, tetrataenite is characterized as a hard ferromagnet with a strong anisotropy despite the fact that common Fe–Ni alloys are classified as softmagnetic materials.

To elucidate the relationship between the Widmanstätten structure and the magnetic property, we used Gibeon iron meteorite, which is one of the typical iron meteorites showing a clear Widmanstätten structure.¹⁵ Figure 1(c) shows the image of the Widmanstätten structure; a thin γ lamella of 4 µm width is separated from a thick γ lamella of 40 µm width by α lamellae. Recent progress in photoelectron emission microscopy (PEEM) using synchrotron radiation enables us to obtain the spatial information on composition, electronic state,¹⁴ crystallographic structure,¹⁵,¹⁶ and magnetic domain structure in the combination with X-ray absorption fine structure (XAFS) or magnetic circular dichroism (MCD). The spatial resolution of apparatus reaches well below 100 nm.

Specimen was sliced nearly parallel to the [001]bcc plane of the α lamella. The surface was carefully prepared using automatic mechanical polisher with a 6 µm diamond slurry for rough treatment and finished by buff polishing with a 1 µm diamond slurry. AC demagnetization field is applied to the specimen to cancel residual magnetization or improper magnetic treatment since 19th century. We examine the surface irregularity using AFM that shows the scratches with a typical width of 100 nm and depth of 10 nm. We adopted even deeper scratch in the numerical calculation, but the scratch does not influence observed magnetic domains. The influence of shape anisotropy energy is negligibly smaller than exchange energy to produce the scratch induced magnetic domain structure. PEEM measurement was performed in the region with the lowest density of scratches.

The spatial distribution of Ni is shown in Figs. 2(a) and 2(b) for the boundary regions indicated by circles in (a).
Fig. 2. Characterization of Widmanstätten structure. (a) Ni distribution in boundary region between α and γ lamellae observed by PEEM. (b) Ni distribution in thick γ lamella segregated, (c) XAFS spectrum at Ni K edge probed for each lamella in (a). (d) Line profile of Ni concentration over the boundary in (b). (e) Magnetic domain structure for same region in (a). Transversal striped magnetic domains arranged parallel to thin γ lamella. (f) Magnetic domain structure for same region in (b). Head-on magnetic domains identified over interface. On both sides of the interface, the magnetizations align opposite to each other.

Fig. 1(c). PEEM images were continuously obtained with scanning photon energy over K absorption edge of Fe (7.10 keV) and Ni (8.35 keV) using BL39XU of SPring-8. The typical exposure time per image was 10 s. Ni composition was estimated by edge-jump of the XAFS spectrum. The Ni composition in α lamella in Fig. 2(a) shows spatially homogeneous profile of 6.6 at.%. On the other hand, that in γ lamella is highly condensed of 28 at.%. Figure 2(c) is typical XAFS spectra obtained for α and γ lamella. As denoted by arrows, the spectral change from a single to a double peak on the crest is ascribed to the structural alternation from bcc to fcc over the transition threshold at 25 at. % Ni.21 Figure 2(d) shows the Ni line profile at the boundary region in Fig. 2(b), indicating that local Ni composition is rapidly increasing from 20 to 35 at.% toward the interface. L1₀ structure was not directly recognized here because of the limit of resolution, however such structural alternation associated with chemical composition suggests that tetrataenite is segregated at the boundary from the metallurgical viewpoint.22 To confirm the presence of tetrataenite, chemical etching with 5% HCl for several minutes reveals the tetrataenite at this region. Scanning electron microscopy and electron probe microanalyzer (SEM–EPMA) estimates the chemical composition as to be Fe₃₀Ni₅₀.

Next, the surface magnetic domain structure was probed by MCD-PEEM for the same area. The circularly polarized light from BL25SU was used to illuminate the specimen along [110]bcc of α lamella as shown by a thick arrow in Figs. 2(e) and 2(f). The exposure time of PEEM was set to be about 20 min to accumulate the image at the Fe L₃ edge (708.4 eV). The red-to-blue color scale indicates the MCD intensity. As shown in Fig. 2(e), a transversal domain lying parallel to the interface is observed clearly at 6 and 12 µm from the interface. In common bcc-Fe, such as whiskers, the magnetic domain shows a wide rectangular structure with a sharp domain wall; thus the transversal stripe in meteoritic iron shows a behavior different from that of the bcc Fe–Ni alloy. By considering its direction, the striped domain may be associated with the boundary.

Over the α/γ interface, as shown in Figs. 2(e) and 2(f), a fine structure of about 2 µm width is also observed; this structure is characterized by an elongated shape oriented parallel to the [110]bcc direction, and the direction of magnetization orients parallel or antiparallel to the [110]bcc direction, as indicated by arrows in Fig. 2(f). The magnetizations on both sides of the interface align opposite to each other and orthogonal to the domain wall, and then this magnetic domain eventually forms a ‘head-on’ structure, which requires a large amount of magnetostatic energy for demagnetizing field.23,24 For a typical 180° domain structure, the magnetization orients parallel to the domain wall so as to reduce the static magnetic energy. For the polycrystalline Fe–Ni alloy, the magnetization over the grain boundary aligns in the same continuous direction,23,24 thus the head-on configuration in the iron meteorite is completely different from the case of Fe–Ni alloy. In epitaxially grown Fe ultrathin films on Ni(111) system, Fe moment align perpendicularly or parallel to Ni moments.25,26 However, this is also not the case of iron meteorite. The head-on configuration is not simply explained by interface mismatch or atomic relaxation. It is concluded, therefore, that the striped magnetic domain and head-on magnetic coupling are unique properties of the magnetic domain in iron meteorite.

To verify such a magnetic domain, we performed micromagnetics simulation solving the Landau–Lifshitz–Gilbert (LLG) equation.27,28 Numerical study is achieved fully three dimensional with a functional form of boundary condition. We used two simple theoretical models, namely Fe/Ni [Fig. 3(a)] and Fe/tetrataenite/Ni [Fig. 3(b)] interface. We assumed a spatially uniform composition for the Ni lamella here. Downward (−z) of the specimen uses a continuous boundary, and upward (+z) free. Longitudinal direction (x) uses a continuous boundary, and transversal (γ) periodic. 1.6 × 6.4 × 1.6 mm³ with 100 nm grid is adopted for simulating boundary region. Magnetic moment is referred to 2.2 and 0.6 μB/atom for Fe and Ni, respectively, and 1.33 μB/atom evaluated by superconducting quantum interference device (SQUID) is used for tetrataenite. Exchange stiffness is adopted as 1.3 × 10⁻¹¹, 1.0 × 10⁻¹¹, and 0.8 × 10⁻¹¹ J/m for Fe, tetrataenite and Ni respectively. Averaged value is used for the interface exchange stiffness here. Magnetic anisotropy energy is referred to the values as described above.7,8,12,13 Calculation runs from random magnetization to a cooled equilibrium state under zero magnetic field. Numerical simulation was performed for both NW and KS configurations and for 1.4-, 1.2-, 1-, 0.8-, 0.6-, 0.4-, and 0.2-µm-thick tetrataenite films to examine the dependences of orientation and thickness. Here, we present the NW configuration of the 1-µm-thick tetrataenite film as a representative result. Top layer is responsible to experimental results. To confirm the entire region, we also performed the calculation for large area of 16 × 72 × 8 µm³ with 1 µm grid, and the result was consistent with that for 100 nm grid and experimental result.

As indicated in Fig. 3(a), the Fe/Ni interface shows a simple magnetic domain, and no head-on magnetic domain is formed. Most magnetic moments in both Fe and Ni
lamellae align to the bulk-like easy axis as \( (100)_{bcc} \) and \( (111)_{fcc} \). The Fe moment gradually cants while approaching the interface, because of the requirement for continuity of the magnetization in \( x \)-direction. On the other hand, magnetic domain is disarranged in the Fe/tetraenaite/Ni system [Fig. 3(b)], and head-on structure definitely reveals at nearby interface. Such head-on domains are commonly formed at any tetraenaite film thickness and in both the KS and NW configurations.

According to technical magnetization, magnetic domain structure is determined so as to minimize the total energy. The magnetic anisotropy of tetraenaite is extremely larger than that of surrounding soft magnetic Fe and Ni. Thus, magnetization in tetraenaite remains in the direction of an easy axis. As shown in the inset, the magnetic pole with \( z \)-component is produced at bare surface of tetraenaite, resulting in the increase of magnetostatic energy. In order to cancel the surface pole at tetraenaite (N-pole in Fig. 3), the S-pole is created at the surface of Ni. These upward and downward configurations of magnetization increase the exchange energy at the interface between Ni/tetraenaite. However, the exchange energy between Fe/tetraenaite is larger than that of Ni/tetraenaite. Accordingly, the configuration in Fig. 3 produces the lower energy. On the other hand, in \( x \)-direction, the S-pole is created at the interface between Ni/tetraenaite, because the magnetization of tetraenaite is larger than that of Ni. In order to cancel the influence of that pole, the generation of N-pole is required at the interface between tetraenaite/Fe. Therefore, the magnetization in Fe shows the opposite direction against the magnetization in tetraenaite. This configuration causes the head-on domain, and increases the exchange energy. However, the cost of the exchange energy in head-on domain wall is equivalent to that in \( 180^\circ \) domain wall, assuming the same wall width. Thus, head-on domain is agreeable to reduce the magnetostatic energy in the system. Consequently, we can conclude that the observed magnetic domains in iron meteorite are induced by the large magnetic anisotropy of the tetraenaite phase at the boundary. Tetraenaite will play a key role in the magnetic anisotropy of iron meteorite.

Synthesis of tetraenaite phase is currently attracting new attention because of inexpensive and abundant resource of Fe and Ni, thus tetraenaite phase will offer potential applications in magneto-electronic devices.

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