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Magnetostriction of Dy and Ho studied by X-ray diffraction

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Abstract

A change of the crystal lattice of Dy and Ho which is induced by a magnetic field along the easy direction of magnetization in the *c*-plane has been studied by X-ray diffraction in a temperature range of the helix phase. Continuous and discontinuous changes of the *c*-lattice parameter accompanying magnetic phase transitions have been observed and satellite reflections caused by the lattice modulation have been found in the helix phase of Dy and Ho and the helifan phase of Ho. The results can be explained by using the magnetoelastic effect and the magnetic structure calculated by a self-consistent mean field method.

Keywords: Metals; Magnetostriction; Lattice modulation; Phase transitions; X-ray diffractions

Rare earth metals [1] Dy and Ho show helical magnetic structures below the Néel temperatures and they transform to a ferromagnet or a cone helix structure at the Curie temperature. A giant magnetostriction was observed [2] at low temperatures by applying a magnetic field in the *c*-plane of the hexagonal close packing crystal structure. This is caused by the large magnetoelastic effect which induces the crystal phase transition by changing the magnetic structure. The magnetization process of the helical structure was theoretically studied [3], and a helix transforms to a fan magnetic structure and finally reaches a ferromagnet in a relatively high temperature region of Dy. In Ho, a new complicated magnetic structure, a helifan, is expected to appear between the helix and fan structures [4] and the reflection from the helifan was really detected by neutron diffraction [5]. The purpose of the present work is to study the effect of various magnetic structures on the crystal lattice of Dy and Ho by means of X-ray diffraction techniques. From the observations with X-rays, novel microscopic phenomena of the magnetostriction in rare earth metals is expected to appear, which were impossible to find by macroscopic observations such as by the strain gauge method.

Measurements have been performed with an X-ray triple-axis diffractometer installed at a conventional rotating anode X-ray source (Copper target, 18 kW). The (002) reflection of a vertically bent pyrolytic graphite is used as a monochrometer (before the sample) so as to focus an X-ray beam on the sample position. A split pair type superconducting magnet which produces a maximum magnetic field of 8 T perpendicular to the scattering plane has been installed on a large goniometer head. The (006) reflection of Dy and Ho single crystals has been observed at various temperatures in the helix phase as a function of a magnetic field along the easy direction of magnetization, the *a*-axis for Dy and the *b*-axis for Ho. For the measurement of the (006) Bragg reflection, a flat germanium (111) reflection was used as an analyser (after the sample) so as to eliminate the reflection from the $K\alpha_2$ incident beam, while a flat pyrolytic graphite was used for the measurement of satellites.

For Ho [6,7], examples of the magnetic field dependence of the diffraction pattern along the c^*-direction at 80 K are illustrated in Fig. 1. The line profile for a single phase appears to have a Gaussian shape, and the profiles in Fig. 1 were analyzed by fitting with sums of Gaussian functions. The solid lines are the results of fitting and dotted straight lines correspond to the positions of the Bragg reflections for various magnetic phases having different *c*-lattice parameters. Two successive first order phase transitions accompanied by the discontinuous changes in the *c*-lattice parameter have been observed in a narrow region of the magnetic field around 2 T, and a second order transition occurs around $H=4$ T. As shown later, the discontinuous changes correspond a helix to helifan and a helifan to fan transitions, and the continuous change corresponds to a fan to ferromagnetic transition. The magnetic field dependence

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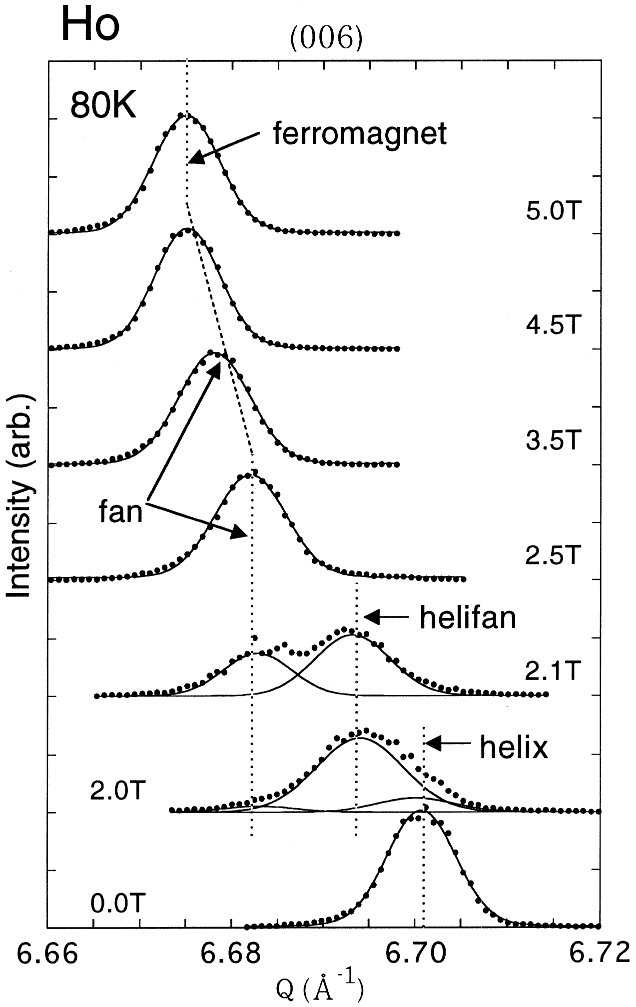

Fig. 1. Diffraction patterns of the (006) reflection of Ho at 80 K in various magnetic fields. Measurements were made with increasing magnetic field.

of the *c*-lattice parameter obtained from the (006) reflection is illustrated in Fig. 2, together with the results for 40 and 60 K. Solid lines in the figure are the result of calculation at 80 K as described below. In the helix and the helifan phase in Ho, satellite reflections have been observed around the (00*l*) reflection along the *c**-direction where *l* is an even integer. The magnetic field dependence of the $(0,0,6-2q_m)$ satellite in the helifan phase at 80 K is illustrated in Fig. 3, where q_m is the fundamental wave number of the magnetic modulation. The pairs of sharp peaks correspond to $K\alpha_1$ and $K\alpha_2$ incident X-ray beams. The solid lines represent the results of fitting with the squared Lorentzian functions. As is shown in Fig. 3(a), two broad satellites having different wave numbers seem to exist at 2.0 T with increasing magnetic field. One of the two wave numbers is twice the value of the fundamental wave number q_m for the helifan(3/2) which was reproduced by the calculation. The satellite disappeared above 2.2 T and no satellite is detected in the higher magnetic field region. With decreasing magnetic field, a

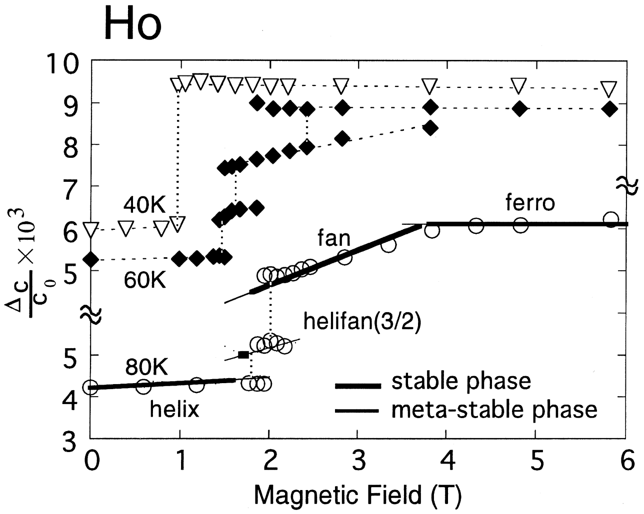

Fig. 2. Magnetic field dependence of the *c*-lattice parameter at 40, 60 and 80 K. The solid thick and thin lines are calculated results.

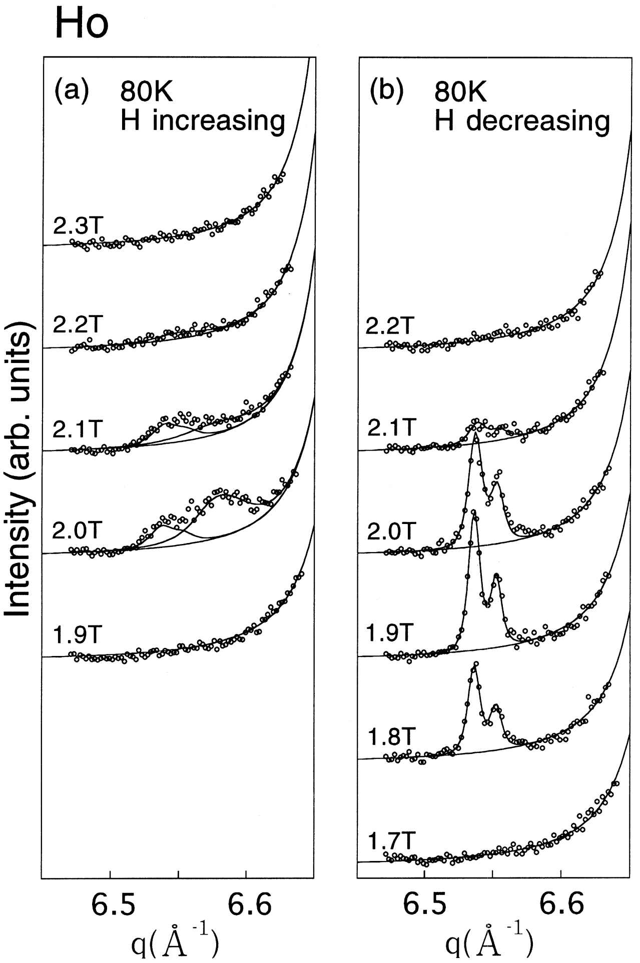

Fig. 3. Satellite reflections along the *c**-direction around the (006) of Ho at 80 K in various magnetic field in the helifan phase. Measurements with (a) increasing and (b) decreasing magnetic field.

sharp satellite appeared in a range between 2.1 and 1.8 T, as shown in Fig. 3(b), which makes a contrast with the increasing process. The intensity is approximately 1/10 000 of the (006) reflection and corresponds to 0.01 Å for the amplitude of the lattice modulation. These satellites caused by the helifan structure have been detected in the temperature range between 35 and 110 K within a narrow range of magnetic field of 0.4 T [7].

In Dy [8], the magnetic field dependence of the c-lattice parameter has been also measured, and the discontinuous or continuous change has been detected accompanying the transition from the helical to fan or ferromagnetic structure. Search for the helifan structure as in Ho has been carefully made but the extra Bragg reflection as shown in Fig. 1 could not be detected. Satellite reflections have been observed at 120, 150 and 173 K in the helix phase. An example of the satellite reflection $(0,0,6-q_{\mathrm{m}})$ at 150 K is shown in Fig. 4. It is noted that the lattice is modulated with the same wave number as the magnetic modulation in the helix structure. The intensity increases with increasing magnetic field and decreases abruptly at 1 T where the phase transition to the fan phase occurs. The magnetic field dependence of the amplitude at 150 K is illustrated in Fig. 5. The amplitude increases linearly with increasing magnetic field, and in the fan phase at 1.2 T, the satellite disappears at the $\pm q_{\mathrm{m}}$ position. The maximum amplitude of the lattice modulation along the c-direction is approximately 0.002 Å. In the fan magnetic structure, no appreciable satellite intensity could be detected both in Ho and Dy. The satellite observed in the helix phase of Ho shows also the same characteristic behaviors as in Dy.

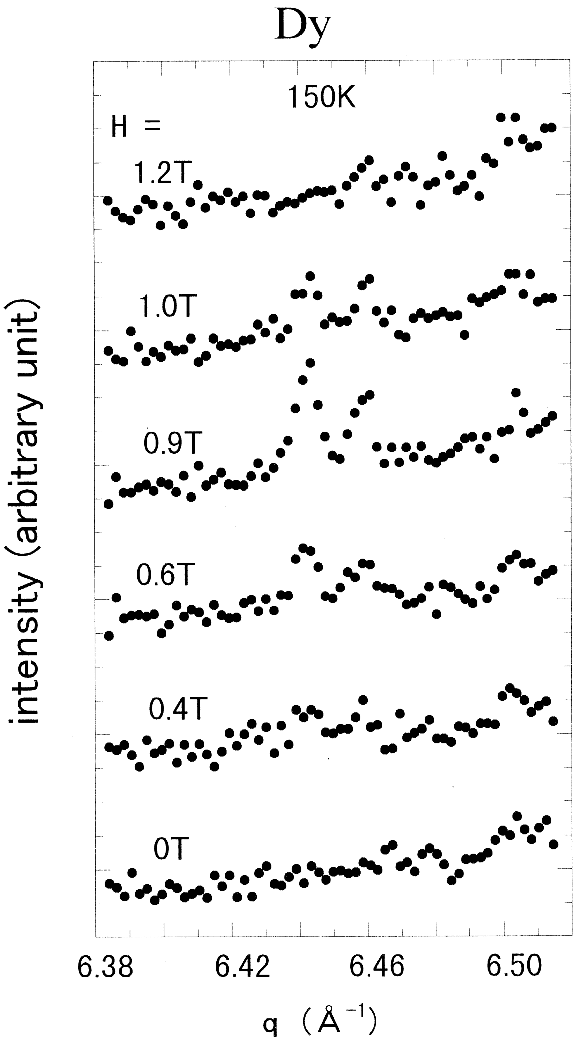

Fig. 4. Satellite reflections along the c^*-direction around the (006) of Dy at 150 K in various magnetic fields in the helix phase.

The field dependence of the lattice parameter and the behavior of the crystal lattice modulation can be described within the formalism of magnetostriction [9]. If exchange interactions up to the second neighbors are included, the change of the lattice spacing ϵ_i at the i-th site caused by the appearance of magnetic ordering can be obtained from minimizing the energy E,

$$E=\frac{1}{2}\phi_i\epsilon_i^2-\epsilon_i(K_1S_i\cdot S_{i+1}+2K_2S_i\cdot S_{i+2}) \tag{1}$$

where the first term is the elastic energy and the second term is the magnetoelastic energy which arises from the distance dependence of the exchange interaction. K_1 and K_2 are parameters which correspond to the derivatives of the exchange parameter with respect to the distance, and ϕ_i is the elastic constant. We have assumed here the period of the lattice modulation is sufficiently long compared with the lattice spacing. We obtain

$$\epsilon_i=\frac{1}{\phi_i}(K_1S_i\cdot S_{i+1}+2K_2S_i\cdot S_{i+2}) \tag{2}$$

The change of the c-lattice parameter Δc is given by Eq. (2) averaged over a magnetic unit cell. The magnetic structure gives the strain at the i-th site through the magnetoelastic interaction. The scalar products of the

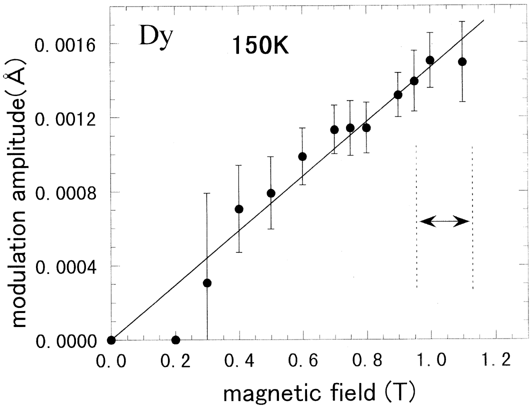

Fig. 5. The magnetic field dependence of the amplitude of the lattice modulation obtained from the satellite intensity. The arrow between dotted lines indicates the coexistence region of the helix and fan phases and the satellite intensity in this region is corrected for the domain fraction of the helix phase.

magnetic moments in Eq. (2) depend on sites in the *distorted* helix, helifan and fan structures. Therefore, the lattice modulation is induced by the magnetic structure, which is really observed in the present measurements except in the fan structure. Magnetic structures in various magnetic fields have been calculated using a self-consistent mean field method [10]. The magnetic moment at the i-th site has been determined self-consistently for the helix, helifan, fan and ferromagnetic arrangements. The stable magnetic structure at a given magnetic field has been determined as that having the lowest free energy among various magnetic structures. In Ho, Δc has been calculated by Eq. (2) averaged over sites using the magnetic structure obtained above. The solid line in Fig. 2 is the result of the best fitting to the experiment at 80 K and a good agreement between the calculation and the observation can be obtained with this model. The helifan structure has been found to be stable in a very narrow range of magnetic field and the effect of this structure on lattice is shown around 2 T in Fig. 2. Lattice modulations in the distorted helix and helifan phases have been also calculated using Eq. (2) with the values for parameters K_1 and K_2 obtained from the fitting for Δc in Fig. 2. Fig. 6 shows the result of the calculations thus obtained for Ho in the helifan at 2.2 T (a) and helix in various magnetic fields (b). The amplitude of the modulation in the helifan is 0.015 Å which is nearly in agreement with the observed value mentioned above. In the helix phase, the amplitude is found to increase linearly with magnetic field, which describes well the experimental results both in Ho and Dy, as shown in Fig. 5 for Dy. However, the calculated values of the amplitude in the helix phase of Dy and Ho are several times larger than those of the observations. This difference may be partly due to the assumption that the lattice modulation is characterized by small wave numbers in obtaining Eq. (2). The elastic energy in modulated lattice may not be taken into account correctly in the present assumption especially for large wave numbers. The lattice modulation in the helifan has 1.5-times smaller wave number than in the helix, which may give relatively good results in calculations with Eq. (2). In the fan phase, the same order of magnitude for the satellite intensity of the lattice modulation as in the helix phase is expected to appear, but no intensity could be detected. The wave number of the lattice modulation in the fan phase is $2q_{\mathrm{m}}$ which is twice that of the helix, and the intensity may be much reduced from that given by the calculation with Eq. (2).

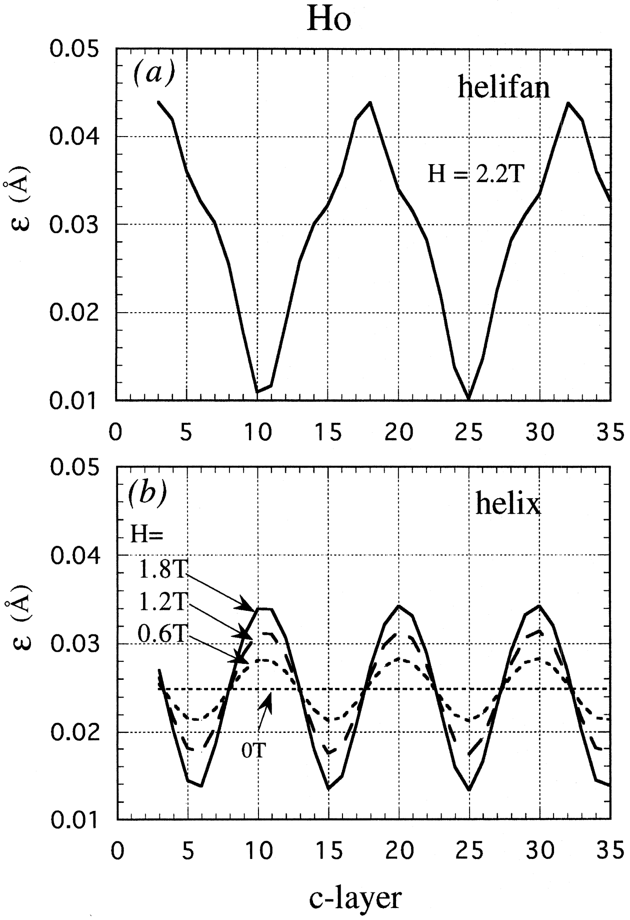

Fig. 6. The calculated lattice modulation ϵ_i in the helifan(3/2) (a) and the helix (b) phases using Eq. (2). The magnetic structure used in the calculation was obtained from a self-consistent mean field method.

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References

[1] R.J. Elliot (Ed.), Magnetic Properties of Rare Earth Metals, Plenum, New York, 1972.
[2] S. Legvold, J. Alstad, J. Rhyne, Phys. Rev. Lett. 10 (1963) 509.
[3] Y. Kitano, T. Nagamiya, Prog. Theor. Phys. 31 (1964) 1.
[4] J. Jensen, A.R. Mackintosh, Phys. Rev. Lett. 64 (1990) 2699.
[5] D.A. Jehan, D.F. McMorrow, R.A. Cowley, G.J. McIntire, J. Magn. Magn. Mater. 104–107 (1992) 1523.
[6] H. Ohsumi, K. Tajima, N. Wakabayashi, K. Kamishima, T. Goto, J. Phys. Soc. Jpn. 66 (1997) 1896.
[7] H. Ohsumi, K. Tajima, J. Phys. Soc. Jpn. 67 (1998) 1883.
[8] Y. Kida, K. Tajima, Y. Shinoda, K. Hayashi, H. Ohsumi, J. Phys. Soc. Jpn. 68 (1999) 650.
[9] E.W. Lee, Proc. Phys. Soc. 84 (1964) 693.
[10] J. Jensen, J. Phys. F 6 (1976) 1145.