JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-III, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. III

Recent Progress in Magnetic Structure Determinations of Rare Earth Metals

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This paper summarizes recent work on single crystal specimens of rare earth metals which has been carried out at the Oak Ridge National Laboratory during the last two years. Results are presented for thulium and for terbium. Experiments designed to investigate the magnetization process in holmium are described.

Introduction

The results of neutron diffraction studies of single crystal specimens of Dy[1], Ho[2], and Er[3], and the magnetic structures of these metals derived therefrom are summarized in another contribution to this conference[4]. In this paper, we report results of recent investigations of single crystal Tb and Tm, which metals had heretofore been studied only in polycrystalline form[5], and results of further studies of single crystal Ho.

Terbium

According to magnetic and thermal data, terbium is ferromagnetic below about 218°K and antiferromagnetic in the range 218°K to 230°K. From measurements which have been made on a large single crystal of Tb, it has been established that the magnetic structure in the antiferromagnetic region is a helical structure similar to that observed for Dy and Ho. The magnetic moments are parallel to the c-planes, and the c-axis is the screw axis.

The diffraction pattern produced by such a structure is characterized by the presence of a single pair of magnetic satellite reflections symmetrically disposed about allowed nuclear reflections on lattice rows parallel to the $\boldsymbol{b}_3$ direction of the reciprocal lattice. The spacings of the satellite reflections are determined by the condition

$$\boldsymbol{s}=\frac{2\pi}{\lambda}(\boldsymbol{u}-\boldsymbol{u}_0)=\frac{2\pi}{\lambda}(\boldsymbol{B}_H\pm\boldsymbol{\tau}_1)\,, \qquad (1)$$

where $\boldsymbol{s}$ is the scattering vector, $\boldsymbol{u}$ and $\boldsymbol{u}_0$ are unit vectors in the direction of the scattered and incident beams respectively, and $\boldsymbol{B}_H=H_1\boldsymbol{b}_1+H_2\boldsymbol{b}_2+H_3\boldsymbol{b}_3$ is a reciprocal lattice vector. In the case of Tb, the vector $\boldsymbol{\tau}_1$ is parallel to $\boldsymbol{b}_3$ and has a magnitude related to the interlayer turn angle ω by the expression $\tau_1=\omega/\pi b_3$.

The magnitude of the vector $\boldsymbol{\tau}_1$ was observed to vary with temperature over the antiferromagnetic temperature range in the manner shown in Fig. 1. For convenience $|\boldsymbol{\tau}_1|$ is expressed in units of b_3, and the temperatures are given as reduced temperatures T/T_N.

The Néel temperature as measured from the temperature variation of intensity in the satellite reflections was found to be 230°±1°K. With decreasing temperature the intensities of these reflections increased sharply to a maximum at about 224°K, and then fell off to unmeasureably small values at about 217°K. At 223°±1°K the normal lattice reflections first exhibited a magnetic contribution, indicating the onset of ferromagnetism. As in the case of Dy, the transformation helix-ferromagnet takes place over a finite temperature range in the absence of an external magnetic field.

Holmium

Recent work on holmium has been directed toward the study of (1) the magnetization process in this substance and (2) the origin of the higher order satellites of the (OOH_3) reflections which had been observed. The bulk of the magnetization process work was carried out on a small crystal cut from a grain found in a cast ingot. This crystal we designate as Ho(A). Most of the work on the higher harmonics was done with a second crystal grown at Ames by a strain-anneal method and which we denote as Ho(B).

After a complete reinvestigation of Ho with the second crystal, Ho(B), it was found that the general conclusions reached from the earlier study were confirmed. However, for reasons not yet known, there were significant differences in the details of the diffraction effects from the two crystals. The second crystal, for example, has a Néel temperature of 133°±1°K, in good agreement with the Ames magnetization data, instead of 122°K which was measured with the first crystal. In addition, the temperature variation of τ_1, the fundamental wave vector, is markedly different in the two cases. As shown in Fig. 1, τ_1 has approximately the same values for the two crystals for $T/T_N > 0.6$. For Ho(A), τ_1 falls to a value of 0.204 (a spacing somewhat less than $5.0a_3$ periods) at a T/T_N value of about 0.3 and remains constant at this value for all temperatures down to 4.2°K. The corresponding curve for Ho(B) shows a change in slope near $T/T_N = 0.3$, but then exhibits an abrupt decrease near the Curie point to a value which corresponds to $6.0a_3$ periods.

Since the magnitude of the fundamental

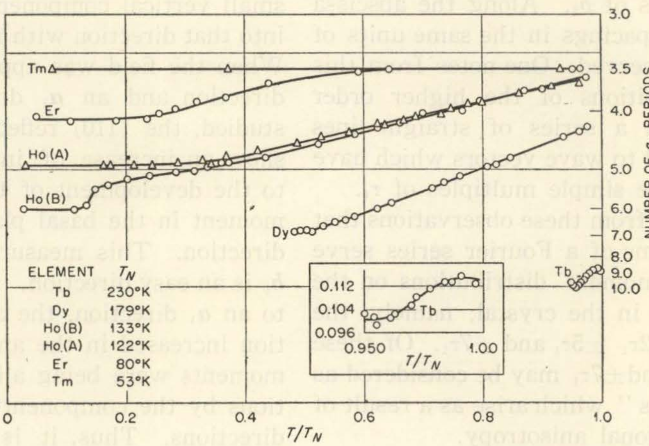

Fig. 1.

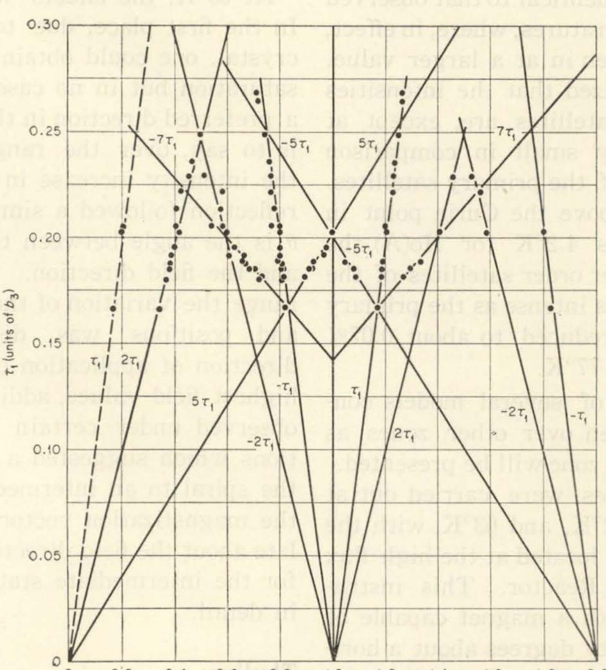

Fig. 2.

wave vector for the two crystals is quite different in the range T/T_N 0.30 to 0.025, the distribution of higher harmonics also might be expected to differ in this range. This was indeed observed to be the case.

With the crystal Ho(B) a series of surveys of scattering density along $\boldsymbol{b}_3$ between the origin and the (002) reciprocal lattice point was carried out at various temperatures, and the positions of the higher order statellites so observed were mapped in the manner shown in Fig. 2. In the figure the ordinate is the magnitude of the spacing of the first harmonic in units of b_3. Along the abscissa are plotted the spacings in the same units of the reflections observed. One notes from this plot that the positions of the higher order satellites fall on a series of straight lines which correspond to wave vectors which have lengths which are simple multiples of τ_1.

One may infer from these observations that at most eight terms of a Fourier series serve to describe the moment distributions on the two sets of sites in the crystal; namely, the terms in $\pm\tau_1$, $\pm 2\tau_1$ $\pm 5\tau_1$ and $\pm 7\tau_1$. Of these the terms $\pm 5\tau_1$ and $\pm 7\tau_1$ may be considered as " beat frequencies " which arise as a result of the planar hexagonal anisotropy.

The distribution of satellite positions at $\tau_1=0.204$ for Ho(B) is identical to that observed for Ho(A) at low temperatures, where, in effect, the turn angle is frozen in at a larger value.

It must be emphasized that the intensities in the higher order satellites are, except at 4.2°K for Ho(B), very small in comparison with the intensities of the primary satellites. For example, just above the Curie point in Ho(B), and as low as 4.2°K for Ho(A) the strongest of the higher order satellites of the origin is one percent as intense as the primary satellite, and this is reduced to about 0.07% at temperatures near 77°K.

Detailed discussion of several models consistent with data taken over other zones as well as over the $(00H_3)$ zone will be presented.

Magnetization studies were carried out at two temperatures, 4.2°K, and 63°K, with the magnet diffractometer located at the high flux Oak Ridge Research Reactor. This instrument is equipped with a magnet capable of rotation through ninety degrees about a horizontal axis, so that the effect of the direction of application of the field on the diffraction pattern could be observed.

The results of these measurements at 4.2°K may be briefly summarized as follows. With the field applied parallel either to a $\boldsymbol{b}_1$ direction or to an $\boldsymbol{a}_1$ direction the intensities of the satellites of all reflections were reduced to zero with no observable shift in position. With the field parallel to a $\boldsymbol{b}_1$ direction, the (100) normal lattice reflection corresponding to that direction was reduced in intensity an amount corresponding to the ferromagnetic c-axis component. This suggests that the $\boldsymbol{b}_1$ direction is an easy direction and that the small vertical component may be pulled over into that direction with relatively small fields. When the field was applied parallel to a $\boldsymbol{b}_1$ direction and an $\boldsymbol{a}_1$ direction normal to it studied, the (110) reflection was observed to show an increase of intensity corresponding to the development of the full ferromagnetic moment in the basal plane parallel to the $\boldsymbol{b}_1$ direction. This measurement confirms that $\boldsymbol{b}_1$ is an easy direction. With the field parallel to an $\boldsymbol{a}_1$ direction, the associated (110) reflection increased in the amount expected if the moments were being aligned along $\boldsymbol{b}_1$ directions by the component of the field in those directions. Thus, it is possible to conclude that the $\boldsymbol{a}_1$ directions are hard directions.

At 63°K, the effects were quite different. In the first place, due to the shape of the crystal, one could obtain only about 70% of saturation but in no case was there observed a preferred direction in the basal plane. That is to say, over the range of fields studied the intensity increase in a given (100) or (110) reflection followed a simple $\sin^2\theta$ law where θ is the angle between the scattering vector and the field direction. In this temperature range the variation of the satellite intensities and positions was dependent upon the direction of application of the field. At the highest field values, additional satellites were observed under certain experimental conditions which suggested a transformation from the spiral to an intermediate state in which the magnetization vectors of the layers oscillate about the field direction. Possible models for the intermediate state will be discussed in detail.

Thulium

Preliminary measurements on thulium

have been made and the following results may now be given. At 53°K the moments form an oscillating z-component structure similar to that found in the high temperature region for Er. The value of τ_1 corresponds very closely to $3.5a_3$ periods over the whole range of temperatures from 53°K to 4.2°K.

At about 33°K, there are observed additional reflections of appreciable intensity, which observation suggests that a process similar to that observed in Er is taking place; namely, the tendency toward a "squaring up" of the sinusoidal wave.

Acknowledgment

It is a great pleasure for the authors to express their gratitude and their indebtedness to Professor Sam Legvold and his students and to Professor F.H. Spedding for making available to them their single crystal specimens, for the privilege of seeing their data prior to publication, and for stimulating correspondence and conversation.

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Neutron Diffraction Study of Chromium Single Crystals

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The temperature dependence of magnetic intensities and spacings of (100) "satellites" of a Cr single crystal was investigated through the Néel temperature at 310°K and the low temperature transition at 121°K. No higher order satellites, characteristic of the antiphase structure, were observed in either phase. If we make the reasonable assumption that the atomic moment does not change appreciably through the low temperature transition, the available data implies a sinusoidal modulation of the magnetic scattering amplitude with the spin direction parallel to the propagation vector below the 121°K transition and perpendicular to it above. This model gives the "true" moment of Cr as $0.59\,\mu_B$ at 78°K.

1. Introduction

The unusual magnetic scattering of neutrons by a chromium single crystal was first reported by Corliss, Hastings and Weiss[1] and subsequently investigated by several authors[2,3,4]. The antiferromagnetic reflections exhibited characteristic splittings below its Néel temperature of 35°C, and these were interpreted in terms of an antiphase antiferromagnetic domain structure with a periodicity of 28 unit cells. An alternate possibility is a spiral spin arrangement with the same periodicity[5,6]. The antiphase structure is characterized by a 3rd "satellite" (even orders have zero intensity) with intensity approximately 11% of the first pair, while the uniform spiral gives only one pair of satellites (see Fig. 1).

The spin direction at room temperature is parallel to the antiphase domain boundary. At 120°K, the spin direction changes to one perpendicular to the boundary. In this low