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 temperature phase, the spiral structure is not possible because of this spin direction.

In order to test the validity of the antiphase structure, a neutron diffraction study was carried out on several chromium single crystals grown either from chromium iodide vapour or by grain growth during annealing. The annealed crystal has a cylindrical shape, 4.8 mm in diameter and 3.6 mm in height. This crystal was grown by Dr. M. J. Marcinkowski of U.S. Steel Research Center

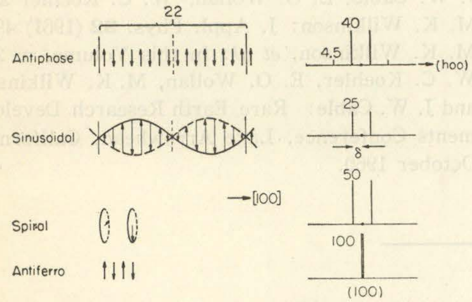

Fig. 1. Models for Cr spin arrangement with their magnetic intensities, calculated with the periodicity of 22 unit cells. This spin direction corresponds to the high temperature phase.

by annealing a chromium ingot containing less than 0.02% of oxygen[7]. An extensive study of elastic anomalies of this crystal has been carried out by Bolef, *et al.*[8], through the Néel temperature at 310°K as well as the low temperature transition at 121°K.

The neutron intensities from this annealed crystal for several ($hk0$) reflections, for wave length λ=1.14Å and $\lambda/2$=0.57Å, indicate that the extinction effect is negligible. The main body of diffraction data was collected on this crystal and the magnetic intensities were normalized by using $b_{\mathrm{Cr}}=0.352\times10^{-12}$cm. The crystals grown from vapour suffered severely from extinction although the results are in general agreement.

2. Low temperature phase

The magnetic scattering near the (100) and (210) reciprocal lattice points was investigated at 78°K, together with several nuclear peaks. The intensity distribution among "satellites" is consistent with the assumption that the spin direction is perpendicular to the antiphase boundary, and the magnetic form factor is close to that of Mn^{2+}. The possibility of the existence of additional satellites, required by the antiphase structure, was carefully examined. In Fig. 2, the crystal was rotated with a fixed counter angle, 2θ. The $(1, 3\delta, 0)$ reciprocal lattice point was also scanned by conventional 2 : 1 coupling. It can be concluded that 3rd satellites (the second possible pair) at (100) are, if they exist, less than 1% of the first ones, while the antiphase structure with a periodicity of 22 unit cells should give 11%. Moreover, the line widths of the magnetic peak are identical, within the experimental uncertainty of 10%, with the nuclear peaks—namely, no additional line broadening was observed due to a magnetic origin. This seems to exclude the possibility that 3rd satellites are not observed because of a fluctuation in periodicity of antiphase domains.

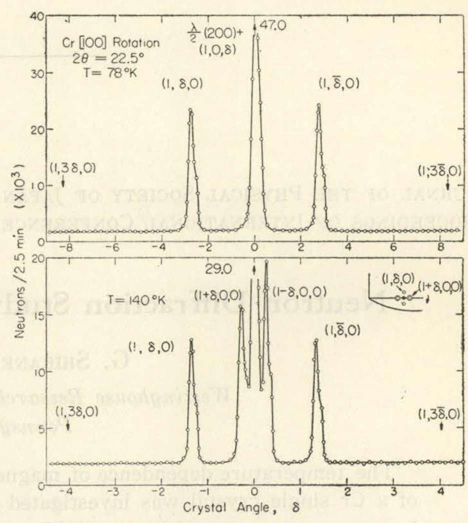

Fig. 2. Neutron diffraction patterns from Cr single crystal at 78° and 140°K, with λ=1.14Å. The crystal is rotated around the [001] axis while the counter is set at 2θ=22.5°. The slight asymmetry of the peak shape is due to a lineage structure of the monochromator. Beam is not filtered, thus contains ~0.5% of $\lambda/2$ component.

Thus, we are forced to a model of a sinusoidal modulation of the magnetic scattering amplitude with the spin direction parallel to the propagation vector. In this model, 50% of the magnetic intensities are "lost" because of the sinusoidal nature. With this model, we obtained the true Bohr magneton number as 0.59 at 78°K. If we had assumed the antiphase model, in which 19% of the intensities are lost from the first satellites, we would have obtained 0.46 μ_B, in good agreement with the previous reports[1),2),3),9)].

3. High temperature phase

The temperature dependence of the intensities of $(1, \delta, 0)$ satellites is shown in Fig. 3. If we take into account the factor 2 resulting from the change of spin direction, there is relatively a small change ($\sim$10%) in intensities through the transition at 121°K. Moreover, it appears that the intensities of both phases are saturating to approximately the same value at 0°K. The drop of the intensities near the Néel temperature at 310°K is somewhat steeper than the curve calculated from Brillouin function with spin 1/2. The temperature dependence of the magnetic periodicity is shown in Fig. 4. This was determined by the crystal angle φ of $(1, \pm\delta, 0)$, instead of conventional 2θ measurements on $(1\pm\delta, 0, 0)$, and higher accuracy is attained because of the diffraction geometry.

The possible existence of 3rd satellites in

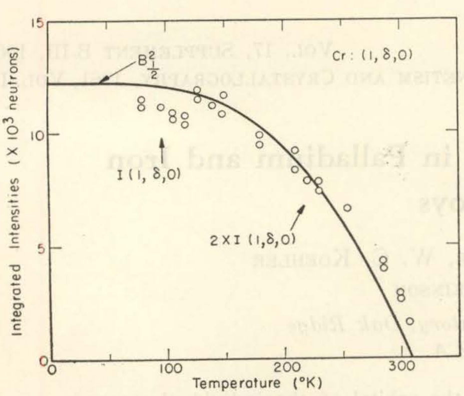

Fig. 3. Temperature dependence of the intensities of $(1, \delta, 0)$ reflections. Solid line is calculated from Brillouin function for spin 1/2.

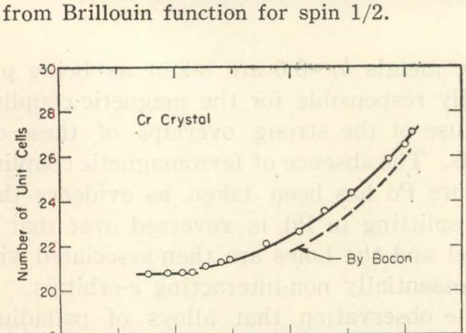

Fig. 4. Temperature dependence of magnetic periodicity of Cr crystal.

the high temperature phase was examined at 140°K (see Fig. 2). It can be concluded that these satellites cannot possess intensities more than 2% of the first pair, thus favoring the spiral structure. The moment calculated from the spiral structure, in which no intensities are "lost," is 0.41 μ_B at 125°K.

This spiral model results in one serious difficulty: namely, the atomic moment of Cr must change by a factor of $\sqrt{2}$ at the low temperature transition. If we make the reasonable assumption that the moment should not change appreciably through the transiton, the available data imply that the high temperature structure is identical to the low temperature one, namely, sinusoidal, except for the spin direction perpendicular to the propagation vector.

Among the theoretical models proposed for chromium, the spin wave model by Overhauser and Arrott[10)] is in line with the results reported above. However, the field cooling effect which they predicted did not give conclusive results. Two Cr crystals were cooled through the Néel temperature, while being subjected to a magnetic field of 10 koe along the [100] direction. The asymmetries of the intensities created among (100) satellites were less than 5% and no definite trend could be established.

Acknowledgment

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DISCUSSION

T. NAGAMIYA: You don't have the possibility that the low temperature form is cycloidal?

G. SHIRANE: The cycloidal spiral model does not explain the observed magnetic scattering in the low temperature phase, because it should give (00*l*) satellites by the ordering of a perpendicular component.

R. J. ELLIOTT: The sine wave structure is stable in Er at high temperature, and this has been interpreted as arising because this arrangement has high entropy, and hence lower free energy at high temperature. The fact that this phase is stable at low temperature in Cr is surprising and would seem to be support for the Overhauser mechanism.

G. SHIRANE: In this connection, it may be interesting to see, as suggested by Dr. T. Kaplan, whether an additional ordering develops at lower temperature. The available powder data do not seem to indicate this.

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Magnetic Moment Distribution in Palladium and Iron Group Alloys

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The distribution of the magnetic electrons in the orbital of the individual atoms in alloys of palladium and nickel with iron and cobalt are discussed. These data suggest the change in orbital splitting which gives rise to ferromagnetism in dilute iron group-palladium alloys.

The magnetic moment distributions in face centered alloys of Fe and Co with Pd and Ni have been studied in an attempt to throw light on the magnetic coupling properties in such systems. The ferromagnetic coupling in face centered 3*d* metals has been previous discussed[1] in terms of the splitting of the *d*-shell into *e*-orbitals and *t*-orbitals and the respective occupation and overlap properties of these orbitals. The proposed occupation of these orbitals for Fe, Co, and Ni is shown in Table I. The holes in the *t*-orbitals of these metals $h_t \sim 0.6$ are taken as being primarily responsible for the magnetic coupling because of the strong overlaps of these orbitals. The absence of ferromagnetic coupling in pure Pd has been taken as evidence that the splitting in Pd is reversed over that in nickel and the holes are then associated with the essentially non-interacting *e*-orbitals.

The observation that alloys of palladium with very small quantities of the 3*d* metals Fe, Co, and Ni are ferromagnetic and that the Curie temperature increases with increased