in the range between 0.2 and 0.4 eV. The original data given in the above reference have been corrected by Phelps[9] on the basis of additional experiments of Mohler.[10] The value of 1.6×10^{-6} cm^3 sec^{-1} per atom is the result of Phelps' correction. It is of interest to note that this value is almost in exact agreement with the value which is obtained in this article if the total scattering cross section, rather than the momentum transfer cross section (see Table III) is used in calculation of collision frequency.

Preliminary experiments of G. J. Mullaney at General Electric Company indicate values of the cross section much lower than those reported here. Corresponding experiments of C. L. Chen, M. Rather, and colleagues at the University of Illinois suggest cross sections much higher than those reported here (for the energy range less than 1 eV). More experiments are required before this cesium cross section can be known with reliability. Of course, the tentative theoretical cross section given in Fig. 1 should never be used as a basis for comparison with experiment without mentioning that the analysis is tentative and that the limitations of the analysis are fully realized. *It is usually easier to get agreement between theory and experiment, once the experimental result is known.*

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Investigations of Paramagnetic Neutron Scattering from Chromium

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Neutron diffraction investigations on Cr^{52} indicate that localized atomic magnetic moments do not exist in metallic chromium above the Néel temperature. Therefore, the moments which exist in the antiferromagnetic structure are presumably induced during the ordering process.

INTRODUCTION

THE early neutron diffraction experiments on chromium metal[1] showed it to be antiferromagnetic at low temperatures. Since this time numerous investigations have been performed in an attempt to gain a better understanding of the magnetic behavior of this metal and, hence, also of the *3d* metals in general.

It was shown by Corliss, Hastings, and Weiss[2] in experiments on a single crystal of chromium that there exists a long-range modulation of the antiferromagnetic moment distribution. This was first interpreted in terms of an antiphase domain structure, but it is now recognized that other possible magnetic structures are a spiral arrangement of the moments[3,4] and an ordered moment arrangement similar to that found in erbium,[5] in which the magnitudes of the magnetic moments are sinusoidally modulated. Investigations by Shirane and Takei[6] strongly indicate that the magnetic structure belongs to the latter class.

The antiferromagnetic transition in chromium also has a somewhat unusual behavior. Recent neutron diffraction investigations[2,7–9] have shown that single-crystal chromium samples have a Néel temperature near 310°K, whereas this transition occurs near 450°K in certain powdered samples. Since careful studies have revealed anomalies in the electrical resistivity,[10] Hall effect,[11] magnetic susceptibility,[12] and specific heat[13] near the lower temperature, this value is believed to correspond to the antiferromagnetic transition in pure strain-free chromium. It can only be surmised that the higher magnetic ordering temperature in some chrom-

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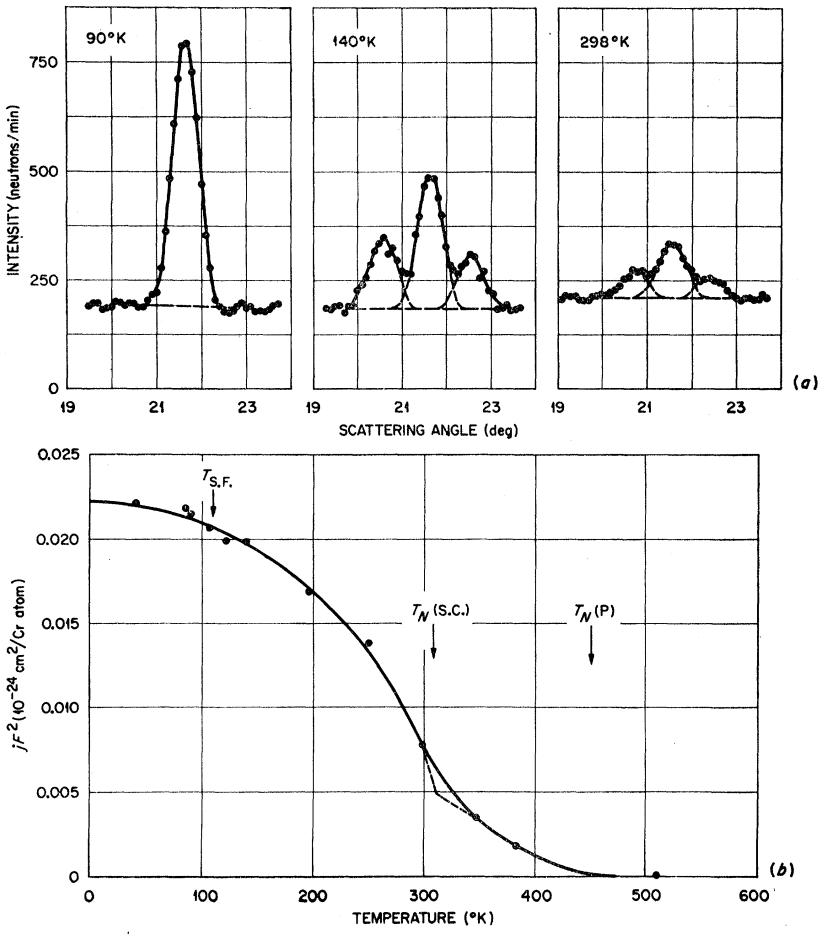

FIG. 1. Temperature variation of antiferromagnetic reflections from Cr^{52} (a) Partial powder diffraction patterns in the angular region near the position of the (001) reflection. (b) Normalized integrated intensities for the total coherent magnetic scattering near the position of the (001) reflection.

ium samples, as observed by neutron diffraction, is in some way related to strains in the sample.

As a part of the over-all investigation of chromium metal it was felt that a determination of its paramagnetic neutron scattering would be of interest since these measurements are related to the value of the localized atomic moments associated with the chromium atoms. Such measurements are of particular importance, because there has been no evidence from other types of experiments for the existence of localized moments in the so-called paramagnetic region.

EXPERIMENTAL PROCEDURES AND RESULTS

Since the atomic magnetic moment that has been determined for antiferromagnetic chromium is small (an average value per atom of about 0.4 μ_B), the intensity of any possible paramagnetic scattering would be very weak, and it was necessary to reduce other types of diffuse scattering to a minimum. The shielding on the diffractometer was modified to eliminate as much background scattering as possible, Soller slits were used to define the scattered beam, most of the second-order contaminant in the scattered beam was removed by an appropriate filter, and radiation shields within the cryostat were reconstructed to minimize the observed scattering from these shields and any condensation on them at low temperatures. Furthermore, an isotopic sample containing 99.6% Cr^{52} was used in the investigation to eliminate isotopic incoherent scattering and nuclear spin incoherent scattering which occur with normal chromium. Twenty-six grams of this isotopic sample were available, and spectrochemical analyses showed that it was very pure chromium with only traces of metallic impurities. The neutron diffraction pattern was found to be a very sensitive indicator of oxide impurities, and, in fact, the first diffraction pattern showed reflections which corresponded to 1.25% Cr_2O_3. The sample was then heated in a hydrogen atmosphere for 72 h,[14] and the absence of detectable oxide reflections in the diffraction pattern limited the maximum possible oxide impurity to 0.1%.

As indicated in Fig. 1, the antiferromagnetic reflections that were observed for this isotopic sample were similar to those observed for other powdered samples of

[14] We are very indebted to D. E. LaValle of the Analytical Chemistry Division, ORNL, for purifying the chromium.

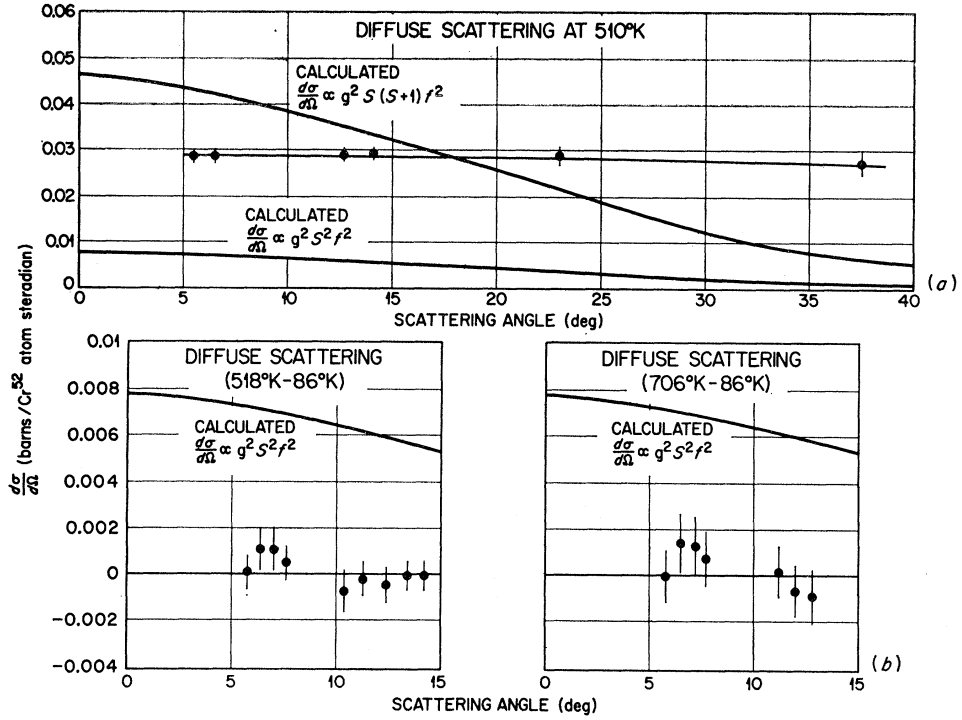

FIG. 2. Diffuse scattering from polycrystalline Cr^{52}. (a) Comparison of experimental values of the total diffuse scattering (corrected for background and thermal diffuse scattering) at 510°K with calculated curves for the expected paramagnetic scattering and the S^2 contribution to the paramagnetic scattering. (b) Results of two experiments showing differences in diffuse scattering at temperatures above and below the Néel transition. (Corrections have been applied for changes in thermal scattering.)

chromium. At temperatures above about 110°K a group of three reflections was found in the angular region near the (001) reflection. This group is characteristic of the complex magnetic structure with one particular moment direction, while the single magnetic reflection below 110°K is the result of a change in the moment direction. The total normalized integrated intensities (for 3 reflections above 110°K and for one reflection below 110°K) are shown at the bottom of the figure, and it is seen that there is no discontinuity at the spin-flip transition. Most of the scattered intensity follows a Brillouin-type dependence toward the Néel temperature obtained from single-crystal measurements, but a small amount exists at high temperatures. The values of jF^2 extrapolated to 0°K are in good agreement with those obtained in previous neutron diffraction investigations. For a spiral moment arrangement and for a model with the moment value modulated sinusoidally, calculations based on the Mn^{+2} form factor give an average moment per atom of $(0.40\pm0.02)\mu_B$. The sinusoidal nature of the moments in the latter model, of course, results in a maximum moment value of 0.57 μ_B per atom.

The analyses of the diffuse neutron scattering from Cr^{52} are shown in Fig. 2. Absolute intensity measurements of the diffuse scattering for a sample temperature of 510°K are plotted in Fig. 2(a) for specific angles where accurate values could be obtained. These points have been corrected for background scattering and for thermal diffuse scattering. For the thermal corrections, the Debye temperature was established from measurements of the nuclear reflections between 706 and 86°K, and the determined value of 485°K is in good agreement with the value of 480°K previously established by other methods.[15] The absolute values of the diffuse scattering are very small, and rough calculations show that they could be due entirely to multiple scattering. The observed values certainly do not agree, either in magnitude or in angular variation, with the calculated curve giving the expected differential paramagnetic scattering cross section for an average atomic moment corresponding to 0.4 μ_B/atom in the antiferromagnetic lattice. The calculated curve, which is proportional to $g^2S(S+1)f^2$, was obtained for an average S value of 0.2. Similar calculations, made on the assumption that 40% of the chromium atoms have $S=1/2$ and the other atoms have $S=0$, show an even greater discrepancy with the observed values.

Since there is some doubt concerning the applicability of the paramagnetic scattering expression for metallic systems, it was also of interest to determine if the S^2 contribution was present in the diffuse scattering. It is this part of the paramagnetic scattering which goes into the magnetic reflections below the ordering transition. As suggested in Fig. 2(a), it is not feasible to make this determination by a close examination of the diffuse scattering above the transition temperature. However, if the S^2 contribution is present in the paramagnetic region, there should be a decrease in the diffuse scattering by the amount as the magnetic lattice becomes

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ordered. In the low-angle region this contribution would be about 18% of the total observed diffuse scattering, and such a decrease should be readily observable. Figure 2(b) shows results from two experiments which measured the difference in diffuse scattering at temperatures above and below the Néel point. The experimental points, which have been corrected for changes in the thermal diffuse scattering, represent average values of several determinations in which the sample was cycled between the indicated temperatures, and the (110) nuclear reflection was carefully monitored at each temperature to insure that there were no changes in the scattering conditions. In the experiments between 518 and 86°K, a direct comparison of the data was possible, because both temperatures could be obtained with the sample mounted in the cryostat. For temperatures above 518°K, the sample was placed in a small furnace, and the data at 706°K were compared to those at 86°K by normalizing both sets of results to room-temperature measurements under similar conditions. Both experiments show that when corrections are applied for changes in thermal scattering, there is essentially no difference in the diffuse scattering from chromium at temperatures above and below the antiferromagnetic transition. The change which would have been observed if the S^2 contribution to the paramagnetic scattering had been present at the high temperatures is shown by the calculated curves.

DISCUSSION

The observation that there is no discontinuity in the coherent magnetic intensities near 110°K, the temperature at which the spin direction changes relative to the direction of the spin modulation, is in accord with the single-crystal results of Shirane and Takei.[6] This result appears to eliminate the spiral arrangement as a possible magnetic structure above the "spin-flip" temperature unless the average magnitude of the Cr moments changes by a factor of $\sqrt{2}$ at this transition. Since their experiments also did not show third-order satellite reflections, Shirane and Takei have concluded that the antiphase domain structure is not possible and that at temperatures both above and below the "spin-flip" transition, the structure is one in which the magnitudes of the atomic moments are sinusoidally modulated.

The results of the paramagnetic scattering investigations indicate that localized atomic magnetic moments do not exist in metallic chromium above the Néel temperature. Consequently, the sinusoidally modulated moments in the antiferromagnetic structure must be induced during the ordering process. This is certainly an unusual situation which has not been observed previously in magnetic systems. Since the mechanism for producing magnetic order is apparently different from that associated with the usual magnetic transition, it is possible that the high Néel temperatures observed in certain chromium samples may be associated with this mechanism. Mention should, therefore, be made of the fact that there has been no evidence for the existence of a long-range periodicity in the magnetic order above 310°K. In those neutron diffraction experiments on powders, which showed a Néel temperature near 450°K, there appeared to be only one reflection near the (001) position at temperatures above 310°K. However, the three reflections, resulting from the long-range periodicity, become so closely spaced above this temperature that this observation could have been caused by insufficient resolution of the weak intensities.

Since there is really no satisfactory theory for explaining some of the more normal aspects of magnetism in the 3*d* metals, it is not surprising that the complicated properties of chromium can not be accounted for at the present time. Overhauser and Arrott[16] have commented on the relation of spin-density waves to the chromium problem, but they assumed the existence of localized atomic moments which could become ordered by the spin-density waves. The absence of localized moments above the transition temperature eliminates these considerations, but very recent calculations by Overhauser[17] suggest that the type of antiferromagnetic transition observed in chromium can possibly be accounted for on the basis of the spin-density-wave mechanism.

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