Magnetic-Moment Distributions for 3*d*-Transition-Metal Impurities in Cobalt†

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Neutron diffuse scattering measurements were made on Co alloys containing 5 at.% V, Cr, Mn, and Ni impurities in order to determine the distribution of magnetic moment around the impurity sites. The measurements were made at room temperature on polycrystalline samples with 1.09-Å neutrons. The observed cross sections are of two types. The cross section of the Ni alloy is independent of the scattering vector and indicates that Ni impurities produce no moment disturbances on surrounding Co atoms. With V, Cr, or Mn impurities, the cross section decreases with increasing scattering vector in a manner which indicates that these impurities produce moment disturbances on their Co neighbors out to distances of approximately 5 Å. All three of these metals produce a net moment reduction when added to Co. For V and Cr, this is mostly due to the negative disturbances produced on the neighboring Co atoms. For Mn impurities, an oppositely aligned Mn moment makes an equally significant contribution to the observed moment reduction.

INTRODUCTION

The elastic, diffuse scattering of thermal neutrons from ferromagnetic alloys provides valuable information about the spatial distribution of the magnetic moment on an atomic scale. In this paper, we describe scattering measurements and moment distributions for Co-based alloys containing 5 at.% of the 3*d*-transition metals V, Cr, Mn, and Ni. Bulk magnetization measurements show that the average moment of Co-Ni alloys follows the Slater-Pauling curve with a decrease of about 1 μ_B per Ni atom. For Co-Cr and Co-Mn alloys, however, there are sharp departures from the Slater-Pauling curve with moment decreases of 6.4 μ_B per Cr atom and 4.5 μ_B per Mn atom.[1] Neutron scattering measurements[2] for the Co-Cr system show that the decrease in moment is predominantly due to a decreasing Co moment rather than to an oppositely directed Cr moment. These measurements were not, however, extended to sufficient dilution or range of scattering vector for a determination of the detailed spatial aspects of the Co moment decrease. Such details for this and other systems emerge from the present study.

EXPERIMENTAL

The arc-melted and drop-cast alloys were machined into 2×3-cm flat plates of 3-mm thickness. Neutron and x-ray examination at this stage in the preparation showed equal amounts of the hcp and fcc phases for all samples. The samples were hot rolled to 1.5-mm thickness and annealed. This resulted in the conversion to the pure hcp phase for all samples except the Co-Mn alloy, for which only 80% conversion to the hcp form was obtained after repeated attempts. We did not attempt to prepare an hcp Co-Fe sample because of the rapidly decreasing fcc to hcp transformation temperature with increasing Fe content in this system. Electron probe analysis showed homogeneity for all samples within the 1-μ range of the instrument.

The cross-section measurements were made at room temperature with 1.09-Å neutrons. These were of the field-off–field-on type with a 12-kOe field used to extinguish the magnetic scattering. Bulk magnetization measurements on the same samples showed about 90% saturation at this field and indicate that the diffuse scattering separation procedure is effective to that extent. Absolute cross sections were obtained by calibration with a standard V scatterer, and are shown in Fig. 1. Here, the apparent magnetic cross section for pure Co provides an important check on the multiple Bragg scattering effects for these samples. The absence of any K dependence in this cross section ensures that multiple Bragg scattering introduces no K dependence into the alloy cross sections. The observed cross section is 7±2 mb while that calculated by the method of Brockhouse, Corliss, and Hastings[3] is 4 mb. We have used the calculated value to correct the alloy cross sections. Transmission measurements with the two field conditions showed negligible single transmission effects for all samples. The Co-Ni cross section is quite small and provides no quantitative information. However, the absence of any K dependence indicates a relatively uniform moment distribution without fluctuations due to environmental effects. This conclusion agrees with the neutron scattering results[4,5] for more concentrated Ni-Co alloys which indicate that both the Co and Ni moments remain constant at about 1.7 μ_B/Co and 0.6 μ_B/Ni over the entire composition region.

Cross-section measurements were obtained for samples of different phase composition for the Co-V and Co-Mn alloys. The results are included in Fig. 1 and show no significant dependence on

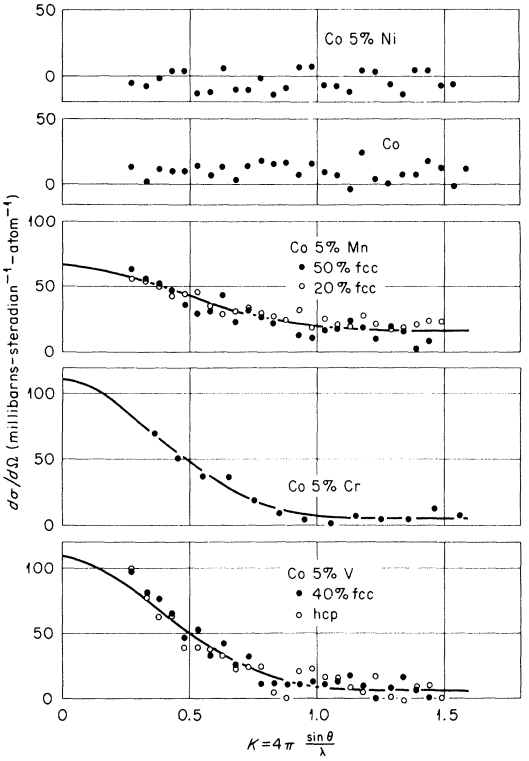

FIG. 1. Neutron diffuse scattering cross sections for cobalt-based alloys.

phase composition. This is not surprising since both are close-packed phases with similar atomic separations. The cross sections for Co-V, Co-Cr, and Co-Mn all show a pronounced enhancement at small K values. These are characteristic of magnetic-moment disturbances localized near the impurity atoms in dilute ferromagnetic systems.

The spatial distribution of the magnetic-moment disturbance produced by an impurity atom in a ferromagnet has been treated by Marshall.[6] The fractional impurity content c is considered sufficiently small that all impurity atoms have the same immediate environment and therefore the same moment μ_i. The impurity atoms produce a moment disturbance $g(R)$ on those host atoms located at distance R. The differential cross section for the magnetic diffuse scattering of unpolarized neutrons from such a system is

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2\gamma}{2mc^2}\right)^2 q^2 f(K)^2 c(1-c)\left[\mu_i - \mu_h + (1-c)\,G(\vec{K})\right]^2 \quad (1)$$

in which $e^2\gamma/2mc^2 = 0.27\times10^{-12}$ cm, q is the magnetic interaction vector, and $f(K)$ is the unpaired d-electron form factor for the host atom. The average moment of the host atom is μ_h, and the transform of the moment disturbance is

$$G(\vec{K}) = \sum_{\vec{R}} g(\vec{R})\, e^{i\vec{K}\cdot\vec{R}} \quad . \quad (2)$$

The data were least-squares fitted to Eq. (1) with the parameters $\Delta\mu = \mu_i - \overline{\mu}_{Co}$ and $g(R)$. In this analysis, we group the neighboring atoms into nearly symmetric shells and assume a spherical average for the exponential function in Eq. (2). Thus, the first shell contains the 6 atoms at 100 plus the 6 atoms at $\frac{1}{3}\frac{2}{3}\frac{1}{2}$. The second shell is taken to include the 2 atoms at 001 plus the 6 atoms at 210, etc. With such grouping of atoms, and since the measurements are at small K values, this spherical average should be a good approximation. We also assume a spherical average of q^2 for the unmagnetized state. This is strictly valid for such uniaxial and magnetically anisotropic systems only if the crystallites are randomly oriented. The observed degree of preferred orientation is not considered sufficient to significantly alter the spherical average of $q^2 = \frac{2}{3}$. Convergence problems were encountered in the least-squares analyses. The inclusion of too many parameters led to oscillatory behavior and unrealistic values for the $g(R)$ parameters. Nevertheless, a minimum number of parameters are required to fit the observed cross sections. We have therefore taken the solution which fits the observations with the smallest number of parameters. This is the three-parameter fit represented by the solid curves in Fig. 1 and for which the parameters are given in Table I. There is a sign ambiguity inherent in this analysis, but the negative solutions are tabulated in accordance with the bulk-magnetization measurements for the Co-Cr and Co-Mn alloys. All parameters are in Bohr magnetons per atom. The $\Delta\mu$ parameter can also be obtained directly from the cross section at large K values, where the $G(K)$ modulation becomes negligible . The same $\Delta\mu$ values as those given by the least-squares analyses are obtained. There remains some concern about the $g(R)$ parameters, however, because of the apparent lack of convergence with increasing R. We therefore examine the physical validity of the parameters by comparison of the calculated and observed magnetic behavior. We assume that all impurity atoms have the same moment but that the Co atoms have moments dependent on their immediate environment. The average Co moment is given by

TABLE I. Cross-section parameters for Co-3d alloys.

Impurity	$\Delta\mu = \mu_i - \overline{\mu}_{Co}$	$g(R_1)$	$g(R_2)$
V	-1.4 ± 0.7	-0.25 ± 0.06	-0.32 ± 0.10
Cr	-1.0 ± 0.7	-0.27 ± 0.06	-0.35 ± 0.10
Mn	-2.6 ± 0.7	-0.12 ± 0.03	-0.16 ± 0.05

$$\bar{\mu}_{Co} = \mu^0_{Co} + c\,G(0) \qquad (3)$$

in which μ^0_{Co} is the moment in pure Co and $G(0)$ is the net change in the host moment induced by the impurity atom. The average alloy moment is then

$$\bar{\mu} = (1-c)\,\bar{\mu}_{Co} + c\,\mu_i \quad , \qquad (4)$$

and the change in average alloy moment per impurity atom is

$$\frac{d\bar{\mu}}{dc} = \mu_i - \bar{\mu}_{Co} + (1-c)\,G(0) \quad . \qquad (5)$$

This is just the bracketed term in the neutron cross section evaluated at $K = 0$. A comparison of the $d\bar{\mu}/dc$ values from the cross-section parameters with those from bulk-magnetization data[1] is given in Table II. The agreement is quite satisfactory for the two alloy systems for which magnetization measurements have been reported.

The impurity and the average Co moment for the Co-Cr and Co-Mn alloys are given in Table III. The average Co moment can be calculated either by use of the $g(R)$ parameters in Eq. (3) or by use of the $\Delta\mu$ parameter in Eq. (4). Both values are given in Table III.

The Co-moment behavior given by the $g(R)$ parameters also agrees with that observed by Shull and Wilkinson.[2] They report 1.27 and 0.89 μ_B/Co at 9 and 13.6 at. % Cr, respectively. The corresponding values given by the parameters obtained in this study are 1.16 and 0.88 μ_B/Co.

Each of these comparisons supports the physical validity of the neutron-cross-section parameters. We conclude that V, Cr, and Mn impurities in hcp Co induce negative magnetic-moment disturbances extending at least to the second-neighbor shell of the impurity atom.

TABLE II. Comparison of neutron-cross-section and magnetization values of $d\bar{\mu}/dc$ for Co-3d alloys.

Impurity	$\Delta\mu$	$(1-c)\,G(0)$	$d\bar{\mu}/dc$ (neutron)	$d\bar{\mu}/dc$ (magn.)
V	−1.4	−5.4	−6.8±1.3	
Cr	−1.0	−5.8	−6.8±1.3	−6.4
Mn	−2.6	−2.6	−5.2±0.9	−4.5

TABLE III. Moment values of Co-Cr and Co-Mn alloys.

Impurity	$\Delta\mu$	$\bar{\mu}$	μ_i	$\bar{\mu}_{Co} = \bar{\mu} - c\Delta\mu$	$\bar{\mu}_{Co} = \mu^0_{Co} + cG(0)$
Cr	−1.0	1.40	0.45±0.7	1.45±0.04	1.41±0.06
Mn	−2.6	1.50	−0.97±0.7	1.63±0.04	1.58±0.03

DISCUSSION

These diffuse scattering measurements provide a rather detailed descripton of the magnetic-moment distribution around 3d-transition metal impurities in hcp Co. The distribution has two components: the moment change at the impurity site, and the disturbance produced at the neighboring Co atom positions. The net moment change per impurity atom produced by V and Cr impurities is largely associated with the Co moment disturbance. With Mn impurities, however, there are nearly equal contributions from the moment difference and the Co moment disturbance. Finally, Ni impurities produce no Co moment disturbance, and the entire moment change is associated with the moment difference term.

There is a striking similarity between these results and those for impurities in Ni.[7,8] In both alloy systems, the disturbance of the host moment is large for large change separation between host and impurity. Furthermore, the disturbances are similar in range to those for impurities in nickel, and also do not change in extent with different impurities. The Friedel bound-state model,[9] as modified to explain the magnetic behavior of Ni-based alloys,[8] is therefore probably also applicable to Co-based alloys.

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