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Search for Noncollinear Spin Density in Hexagonal Cobalt*

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The interpretation of the magnetic form factors for the $3d$ metals has been based on the assumption that the magnetic moment is everwhere collinear, so that a scalar density function may be used. It has been suggested that if the spin density is noncollinear, the form-factor measurements should be reinterpreted. We have conducted experiments, by means of the new technique of neutron-polarization analysis, designed to detect a noncollinear spin component in hexagonal cobalt, but have seen no evidence for scattering from such a component. Upper limits on the magnitude of such scattering have been established for the (002) and (110) reflections. Further, it is shown that the polarized-beam technique used in the form-factor measurements is not very sensitive to a small noncollinear spin component. Ignoring the presence of noncollinear spin scattering, of magnitude given by the upper limit of the polarization analysis experiment, produces an error in the Co form factor smaller than the experimental error due to other effects. For Fe and Ni, it is shown that the form-factor experiment is much less sensitive to a noncollinear spin density than is the case for Co. We conclude that the form-factor measurements need not be reinterpreted, provided it is understood that they apply to the component of the total spin density which is parallel to the average spin density.

INTRODUCTION

THE interpretation of the magnetic form-factor experiments on the ferromagnetic $3d$ metals[1–3] has been based on the assumption that the spin density is everywhere collinear so that the distribution of spin can be described by a scalar function of position multiplied by a unit vector. Blume[4] has suggested that this may not be true, particularly in the case of hexagonal Co, and has shown that the form-factor measurements should be reinterpreted if there is an appreciable noncollinear contribution to the spin density. Our motivation in searching for a noncollinear spin density in Co was partly the intrinsic interest in such an effect, and partly to discover whether the previous form-factor measurements need to be reinterpreted.

We have used the technique of neutron-polarization analysis[5] in this experiment. The sample was magnetized to saturation along a Bragg scattering vector and the incident neutrons were polarized along this same direction. Under these conditions the spin density parallel to the magnetization has a zero cross section and any spin density oriented perpendicular to the magnetization will scatter neutrons with reversal of the neutron spin. The non-spin-flip scattering will be proportional to the square of the coherent nuclear scattering amplitude. The basic experiment is a measurement of the ratio of spin-flip to non-spin-flip scattering at a Bragg peak. The interpretation of this measurement is complicated by the presence of nearly elastic magnon scattering, which makes a contribution to the spin-flip cross section.

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THEORY

We wish to describe the experiment more formally in order to see exactly what is measured and to be able to relate this measurement to the results of the form-factor experiments. We will use Blume's[4] notation, but with a slight revision in the formulation which helps to relate the calculation to the experiments. We split the spin density into two parts,

$$\boldsymbol{\rho}(\mathbf{r})=\hat{\eta}(0)\rho_{\parallel}(\mathbf{r})+\boldsymbol{\rho}_{\perp}(\mathbf{r})\,, \tag{1}$$

where $\hat{\eta}(0)$ is a unit vector in the direction of the average spin, $\rho_{\parallel}(\mathbf{r})$ is a scalar function describing the spin distribution in the direction of $\hat{\eta}(0)$, and $\boldsymbol{\rho}_{\perp}(\mathbf{r})$ describes the distribution of spin which is oriented perpendicular to $\hat{\eta}(0)$. On integrating over the unit cell we have

$$\int_{V_0}\rho_{\parallel}(\mathbf{r})dV=nS\,, \tag{2}$$

$$\int_{V_0}\boldsymbol{\rho}_{\perp}(\mathbf{r})dV=0\,, \tag{3}$$

where n is the number of atoms per unit cell and S is the total ordered spin per atom. The magnetic scattering amplitude is given by

$$\mathbf{p}(\mathbf{K})=\frac{\gamma e^2}{nmc^2}\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}}[\hat{\eta}(0)\rho_{\parallel}(\mathbf{r})+\boldsymbol{\rho}_{\perp}(\mathbf{r})]dV\,, \tag{4}$$

where γ is the magnitude of the neutron moment in nuclear magnetons. Making the obvious identifications, we write the magnetic amplitude in the form

$$\mathbf{p}(\mathbf{K})=\hat{\eta}(0)p_{\parallel}(\mathbf{K})+\hat{\eta}_{\perp}(\mathbf{K})p_{\perp}(\mathbf{K})\,, \tag{5}$$

where $\hat{\eta}_{\perp}(\mathbf{K})$ is a unit vector in the direction of the second term of Eq. (4). The partial cross section for Bragg scattering from a ferromagnet in which the

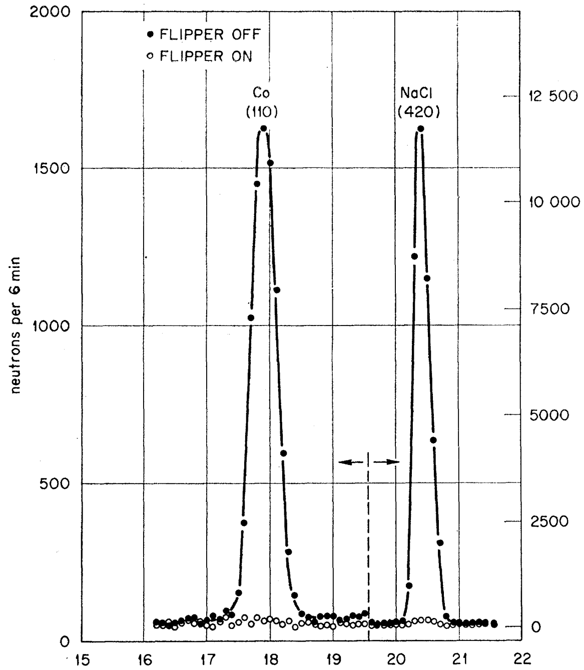

FIG. 1. Polarization analysis of the Co (110) and NaCl (420) reflections. $|\hat{K}\cdot\hat{\eta}(0)|=1$, $|\hat{K}\cdot\hat{z}|=1$.

neutron spin goes from state s to s' is

$$\frac{d\sigma^{ss'}}{d\Omega}\propto |\langle s'|b-\hat{K}\times[\mathbf{p}(\mathbf{K})\times\hat{K}]\cdot\boldsymbol{\sigma}|s\rangle|^2\delta(\mathbf{K}-\boldsymbol{\tau}), \quad (6)$$

where $\boldsymbol{\tau}$ is a reciprocal-lattice vector and $\boldsymbol{\sigma}$ is the Pauli-spin operator (the magnitude of the neutron spin has been absorbed in the scattering amplitude). Taking $\hat{z}$ to be the direction of the neutron polarization and using the properties of the Pauli-spin matrices, we find that

$$\frac{d\sigma^{\pm\pm}}{d\Omega}\propto |\{b\mp[p_{\parallel}(\mathbf{K})\mathbf{q}_{\parallel}(\mathbf{K})+p_{\perp}(\mathbf{K})\mathbf{q}_{\perp}(\mathbf{K})]\cdot\hat{z}\}|^2 \times\delta(\mathbf{K}-\boldsymbol{\tau}), \quad (7)$$

$$\frac{d\sigma^{\pm\mp}}{d\Omega}\propto |[p_{\parallel}(\mathbf{K})\mathbf{q}_{\parallel}+p_{\perp}(\mathbf{K})\mathbf{q}_{\perp}(\mathbf{K})]\cdot(\hat{x}\pm i\hat{y})|^2 \times\delta(\mathbf{K}-\boldsymbol{\tau}), \quad (8)$$

where $\mathbf{q}_{\parallel}=\hat{K}\times[\hat{\eta}(0)\times\hat{K}]$, with a similar definition for $\mathbf{q}_{\perp}$. In the experiment we have performed $\hat{\eta}(0)\cdot\hat{K}=|\hat{z}\cdot\hat{K}|=1$, so that $\mathbf{q}_{\parallel}=0$ and $\mathbf{q}_{\perp}=\boldsymbol{\eta}_{\perp}$. Since $\boldsymbol{\eta}_{\perp}$ must be in the x,y plane, it follows that

$$\frac{d\sigma^{\pm\pm}}{d\Omega}\propto b^2\delta(\mathbf{K}-\boldsymbol{\tau}), \quad (9)$$

$$\frac{d\sigma^{\pm\mp}}{d\Omega}\propto p_{\perp}^2(\mathbf{K})\delta(\mathbf{K}-\boldsymbol{\tau}). \quad (10)$$

In the absence of other scattering processes the ratio of the spin-flip to non-spin-flip scattering will be $(p_{\perp}/b)^2$.

In the form-factor experiment, the sample is magnetized to saturation perpendicular to the scattering vector. For this case we have $\mathbf{q}_{\parallel}=\hat{\eta}(0)=-\hat{z}$ (the negative sign enters because the neutrons are polarized antiparallel to the sample spin) and $\mathbf{q}_{\perp}\cdot\hat{z}=0$. The partial cross sections become

$$\frac{d\sigma^{\pm\pm}}{d\Omega}\propto [b\pm p_{\parallel}(\mathbf{K})]^2\delta(\mathbf{K}-\boldsymbol{\tau}), \quad (11)$$

$$\frac{d\sigma^{\pm\mp}}{d\Omega}\propto q_{\perp}^2(\mathbf{K})p_{\perp}^2(\mathbf{K})\delta(\mathbf{K}-\boldsymbol{\tau}). \quad (12)$$

In the form-factor experiment, the total cross section, summed over final spin states, is measured for both initial neutron-spin states. The ratio of these two observations is given by

$$R=\frac{(1+p_{\parallel}/b)^2+q_{\perp}^2(p_{\perp}/b)^2}{(1-p_{\parallel}/b)^2+q_{\perp}^2(p_{\perp}/b)^2}. \quad (13)$$

The interpretation of the form-factor measurements has been made on the assumption that $p_{\perp}$ is everywhere zero. We can check the validity of this assumption by measuring $(p_{\perp}/b)^2$ in the polarization analysis experiment. We should then be able to put limits on the influence of a noncollinear spin density on the form-factor measurement. Note that $q_{\perp}^2<1$ by definition.

EXPERIMENTAL

The experimental arrangement has been described elsewhere.[5,6] Basically the apparatus consists of a triple-axis spectrometer with polarization-sensitive crystals on both the first and third axes. The flipper was located before the sample. Two reflections were measured: the (002) and (110). A disk-shaped crystal was used for the (002) reflection, magnetized in the plane of the disk. A smaller pillar-shaped crystal was used for the (110) reflection with the long axis parallel to the (110). In this case, the shape anisotropy helped in magnetizing perpendicular to the easy axis. The neutron wavelength was 1.07 Å for the (002) measurement and 0.77 Å for the (110) measurement.

Inspection of Eq. (8) shows that it is important that the magnetization be accurately aligned along the scattering vector to avoid a contribution to the spin-flip scattering from the parallel component of the spin density. The crystals were mounted in a goniometer which allowed orientation of the crystal relative to the magnet. Final adjustments were made by observing the flipping ratio without the analyzer. If the magnetization makes a small angle δ with the scattering vector, this ratio should be $R=1+4(p_{\parallel}/b)\sin^2\delta$. At the same time, the spin-flip scattering (assuming for the moment that $p_{\perp}=0$) becomes $p_{\parallel}^2\sin^2\delta$. We believe that a

[6] T. Riste, R. M. Moon, and W. C. Koehler, Phys. Rev. Letters 20, 997 (1968).

reasonable maximum value for δ, including effects of field nonuniformity, is $\frac{1}{2}°$, so that this contribution to the spin-flip scattering is negligible. Much more serious is the problem of simultaneous reflections. If the crystal is oriented so that other Bragg reflections are occurring, neutrons will be scattered into the direction corresponding to the reflection under study by a double-reflection process. The scattering vectors corresponding to these simultaneous reflections will not be aligned along the magnetization direction, so the total cross section will show a polarization dependence, and there will be some spin-flip scattering. The flipping ratio without the analyzer was used as a test for the presence of simultaneous reflections. A large deviation, either positive or negative, from the expected value of unity indicated the presence of simultaneous reflections. In our final alignment, this ratio measured 0.9999±0.0018 for the (002) reflection and 1.001±0.001 for the (110) reflection.

It was apparent from the beginning that the spin-flip cross section was quite small. This meant that instrumental corrections would be very important. These could be determined by moving the analyzer into the beam transmitted through the test crystal and measuring the flipping ratio. A decrease in this ratio when the analyzer was in the reflected beam indicated either the presence of spin-flip scattering or that there was some depolarization of the neutrons along the reflected-beam path. To remove this uncertainty, the (420) reflection of NaCl was used to evaluate the instrumental corrections used for the Co (110) measurement. Both crystals were mounted in the magnet gap in close proximity. Because the d spacing for the (420) reflection is nearly identical to the Co (110), the neutrons follow the same trajectory in both cases. The small fraction of $\frac{1}{2}\lambda$ contaminant in the beam was measured so that appropriate corrections could be applied. Both methods of evaluating the instrumental corrections gave equivalent results for the (110) reflection. For the (002) reflection, the instrumental correction was based on the transmitted beam flipping ratio. All the measured ratios were about 85.

RESULTS AND DISCUSSION

Some typical data are shown in Fig. 1, which illustrates the polarization analysis of the Co (110) and the NaCl (420) reflections. These data were obtained by rocking the crystals through the Bragg reflections with the analyzer set for elastic scattering. The small peak in the "flipper-on" data for the NaCl peak is a measure of the instrumental imperfection. Basically, the experiment consisted of measuring the ratio of "flipper on" to "flipper off" for both peaks. To a good approximation, the difference in these two ratios is equal to the ratio R_A of spin-flip to non-spin-flip scattering from Co. In practice, these ratios were determined by measuring the peak counting rates above background rather than the integrated intensity.

TABLE I. Ratio of spin-flip to non-spin-flip scattering for hexagonal Co.

	(002)	(110)
$(I^{-+}/I^{++})_{\text{tot}}$	0.0006±0.0002	0.0009±0.0003
$(I^{-+}/I^{++})_{\text{mag}}$	0.0009±0.0004	0.0003±0.0002
$(p_\perp/b)^2$	−0.0003±0.0004	0.0006±0.0004

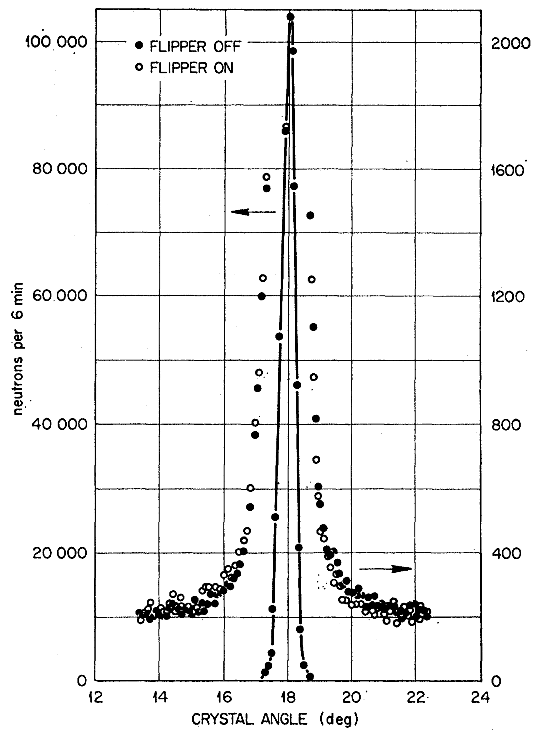

FIG. 2. Total scattering near the Co (110) reflection. $|\hat{K}\cdot\hat{\eta}(0)|=1$, $|\hat{K}\cdot\hat{z}|=1$.

We obtained the values of R_A listed in Table I. The possible sources contributing to the spin-flip scattering are a noncollinear spin density, magnon scattering, simultaneous reflections, and alignment errors. We neglected the last two possible sources and attempted to measure the magnon contribution to get an upper limit on the scattering due to noncollinearity. The spin-flip neutron incoherent scattering from Co was subtracted out in making the background correction.

The polarization dependence of the total magnon cross section[7] was used to get an estimate of the magnon scattering at the Bragg peak position. Figure 2 shows the total scattering (no analyzer) as the Co crystal is rocked through the (110) peak. Note that on the low-angle side of the peak the "flipper-on" data are consistently higher than the "flipper-off," while the reverse is true on the opposite side of the peak. As shown in Eqs. (9) and (10), the total cross section for scattering from a noncollinear spin density is independent of the initial neutron-spin state, so that the observed polarization dependence is attributed to magnon scattering. For the geometry we were using, scattering with

[7] A. W. Sáenz, Phys. Rev. **119**, 1542 (1960).

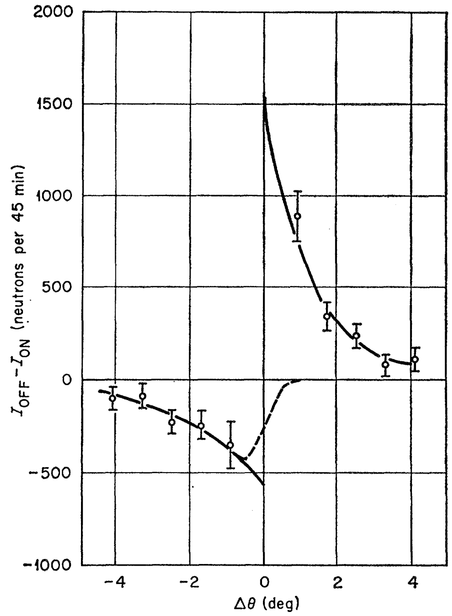

FIG. 3. Magnon scattering near the Co (110) reflection. The peak intensity is at $\Delta\theta=0$. $|\hat{K}\cdot\hat{\eta}(0)|=1$, $|\hat{K}\cdot\hat{z}|=1$.

creation of magnons involves a neutron-spin transition from (+) to (−), while magnon annihilation involves a (−) to (+) transition. It is only the annihilation scattering, for which $\theta-\theta_B<0$, that can contribute to the spin-flip scattering observed with the analyzer. In this case the scattered neutrons have their spins in the proper direction to be reflected by the analyzer. We wish to extrapolate the observed annihilation scattering towards the center of the Bragg peak to get an estimate of the magnon contribution to the spin-flip scattering observed at peak position. This extrapolation is shown in Fig. 3. The data of Fig. 2 were averaged over several points and the "off-on" difference is plotted as a function of $\Delta\theta=\theta-\theta_B$. The intercept of the magnon annihilation curve (solid line) at $\Delta\theta=0$ is an overestimate of the desired correction. When consideration is given to the instrumental resolution and the fact that the magnon annihilation scattering falls to zero very rapidly for $\Delta\theta>0$, the dashed line is a more reasonable extrapolation. This line has been drawn such that the $\Delta\theta=0$ intercept is $\frac{1}{2}$ the solid-line intercept. The estimates of the magnon contribution to the observed spin-flip scattering obtained in this manner are also shown in Table I.

The difference between the observed values of R_A and the magnon correction gives the measurement of $(p_\perp/b)^2$. As shown in Table I, we find that $p_\perp$ is very small for both reflections. No significant difference from zero was detected.

To examine the effect of a small noncollinear spin density on the form-factor measurement, we ask the following question: Given a measured value of R in Eq. (13), what error in $p_{\parallel}/b$ is introduced by setting $p_\perp/b=0$? This error is given by

$$\Delta(p_{\parallel}/b)=\left(\frac{\partial R}{\partial(p_{\parallel}/b)}\right)^{-1}\frac{\partial R}{\partial\epsilon}\bigg|_{\epsilon=0}\epsilon=-\frac{p_{\parallel}/b}{1-(p_{\parallel}/b)^2}\epsilon, \quad (14)$$

where $\epsilon=q_\perp^2(p_\perp/b)^2$. We have calculated this error for all the reflections observed in the form-factor measurements on hexagonal Co, assuming a value for ϵ consistent with the polarization analysis experiment (we took $\epsilon=0.0007$). Of course, we do not expect ϵ to be constant for all reflections, but this calculation serves to give an indication of the magnitude of the possible effect. It is clear that the correction will be appreciable only when $p_{\parallel}/b\cong 1$. For the (110) reflection, with $p_{\parallel}/b=0.704$, the calculated error due to noncollinearity is five times smaller than the quoted experimental error[2] in $p_{\parallel}/b$. For all other reflections, the correction is even less significant. For example, it is 140 times smaller than the experimental error for the (140) reflection.

Although we can say nothing definite about noncollinearity in Fe and Ni, it is clear from Eq. (14) that the form-factor measurements for these metals are less sensitive to a nonzero $p_\perp$ than is the case in Co. This follows from the much lower $p_{\parallel}/b$ values for the Fe and Ni cases. If we scale the value of ϵ used in the Co calculation to the Fe and Ni cases, assuming $p_\perp$ is proportional to the total magnetic moment, we can obtain an estimate of the noncollinearity correction to $p_{\parallel}/b$. For Fe, this correction is 83 times smaller than the experimental error for the (110) reflection.[1] For Ni, the noncollinearity correction is 1200 times smaller than the experimental error for the (111) reflection.[3] It seems highly improbable that there could be a noncollinear spin density in Fe or Ni which is large enough to have any effect on the form-factor measurements.

SUMMARY

We have detected no departure from collinearity in hexagonal Co. The experimental error in our measurement enables us to place a reasonable limit on the effect of a possible small noncollinear spin density on the form-factor measurement. This effect is negligibly small in comparison to experimental errors in the form-factor determination. For Fe and Ni, it is shown that the form-factor experiment is much less sensitive to a noncollinear spin density than is the case for Co. We believe that the form-factor measurements for Fe, Co, and Ni need not be reinterpreted, even if there is a very small noncollinear spin density, provided that it is understood that they apply to the component of $\boldsymbol{\rho}(\mathbf{r})$ which is parallel to the magnetization.

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