Noncollinear Magnetization Density in Cobalt Metal*

M. BLUME
Physics Department, Brookhaven National Laboratory, Upton, New York 11973
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It is shown that a recent experiment by Moon and Koehler designed to detect a noncollinear magnetization density in cobalt metal should give a null result on symmetry grounds.

RECENTLY, Moon and Koehler[1] have reported a null result in a neutron scattering experiment designed to detect a noncollinear magnetization density in hexagonal cobalt metal. It is the purpose of this note to point out that a null result is to be expected on symmetry grounds with the experimental arrangement described in their paper.

We briefly restate the notation of Ref. 1: The magnetization density $\boldsymbol{\varrho}(\mathbf{r})$ may be divided into two parts

$$\boldsymbol{\varrho}(\mathbf{r})=\hat{\eta}\rho_{11}(\mathbf{r})+\boldsymbol{\varrho}_{\perp}(\mathbf{r}), \tag{1}$$

where $\hat{\eta}$ is the direction of magnetization. The parallel and noncollinear components of magnetization density have the properties[2]

$$\int_{V_0}\rho_{11}(\mathbf{r})\,d\mathbf{r}=M_0$$

and

$$\int_{V_0}\boldsymbol{\varrho}_{\perp}(\mathbf{r})\,d\mathbf{r}=0,$$

respectively, and $\hat{\eta}\cdot\boldsymbol{\varrho}_{\perp}(\mathbf{r})=0$. Here V_0 is the volume of the unit cell and M_0 is the magnetic moment per unit cell.

The amplitude for neutron scattering in a Bragg peak is then proportional to

$$A\sim\mathbf{K}\times\left[\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}}\boldsymbol{\varrho}(\mathbf{r})\,d\mathbf{r}\times\mathbf{K}\right], \tag{2}$$

where $\mathbf{K}$ is the scattering vector. In the experimental arrangement of Ref. 1 the scattering vector was taken parallel to the direction $\hat{\eta}$ of magnetization, so that $\mathbf{K}\cdot\boldsymbol{\varrho}_{\perp}(\mathbf{r})=0$, and (2) becomes

$$A\sim\mathbf{K}\times\left[\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}}\boldsymbol{\varrho}_{\perp}(\mathbf{r})\,d\mathbf{r}\times\mathbf{K}\right]. \tag{3}$$

i.e., only the noncollinear component of the density contributes to the scattering amplitude. This arrangement is quite reasonable in that it isolates the quantity to be studied by eliminating contributions from the (much larger) collinear component $\rho_{11}(\mathbf{r})$ of the density. Unfortunately, the integral in (3) vanishes with this direction of $\mathbf{K}$ except when the noncollinearity is due to magnetic anisotropy *and* if the Bragg vector $\mathbf{K}$ does not point along a symmetry axis of the crystal.

To see this, we first consider the case when the noncollinearity is due to spin-orbit coupling in a free ion. The noncollinear part of the density would then have cylindrical symmetry about the direction of magnetization, which is, in Moon and Koehler's experiment, also the direction of $\mathbf{K}$. The $\mathbf{K}$ vector in the integral in (3) does not single out any special direction along which the magnetization can lie, so the integral must vanish. Mathematically there is a position vector $\mathbf{r}'$ for each vector $\mathbf{r}$ such that $\boldsymbol{\varrho}_{\perp}(r')=-\boldsymbol{\varrho}_{\perp}(r)$, and also such that $\mathbf{K}\cdot\mathbf{r}'=\mathbf{K}\cdot\mathbf{r}$. Since

$$\int_{V_0}d\mathbf{r}'=\int_{V_0}d\mathbf{r},$$

we then have

$$\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}}\boldsymbol{\varrho}_{\perp}(\mathbf{r})\,d\mathbf{r}=-\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}}\boldsymbol{\varrho}_{\perp}(\mathbf{r}')\,d\mathbf{r}$$

$$=-\int_{V_0}e^{i\mathbf{K}\cdot\mathbf{r}'}\boldsymbol{\varrho}_{\perp}(\mathbf{r}')\,d\mathbf{r}'=0.$$

In the event that the noncollinearity is produced by magnetic anisotropy $\boldsymbol{\varrho}_{\perp}(\mathbf{r})$ need no longer have cylinridical symmetry about the direction of magnetization. If $\mathbf{K}$ and $\hat{\eta}$ coincide, however, *and* if $\mathbf{K}$ lies along a symmetry axis of the crystal, the transformation discussed above can still be made and the conclusion holds. Since the two reflections measured by Moon and Koehler had scattering vectors along the hexagonal axis and along a twofold axis in the basal plane, we conclude that a noncollinear density could not be detected in their experiment.

It should be noted that noncollinearity due to anisotropy could be detected with their arrangement by a measurement on a Bragg peak whose scattering vector does not lie along a symmetry axis. The effect would be maximized by looking at peaks with $\sin\theta/\lambda\sim0.8$, since it is expected that the form factor for noncollinear (and anisotropic) densities would increase with scattering angle. Indeed, calculations on terbium[3] show that the

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noncollinear form factor increases in this way. Finally, we note that the noncollinearity due to spin-orbit coupling in free atoms can never be observed with **K** parallel to $\hat{\eta}$. This type of density contributes to the scattering amplitude only in conjunction with the collinear part.

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Low-Temperature Magnetic Susceptibilities of $Al_2O_3:V^{3+}$ †

DONALD J. ARNOLD, ALFRED R. SMITH, AND RAYMOND W. MIRES
Department of Physics, Texas Technological College, Lubbock, Texas 79409
(Received 31 July 1969)

Previous measurements of the magnetic anisotropy of single-crystal $Al_2O_3:V^{3+}$ in the temperature range 1–5°K have already been used to obtain more accurate values for the zero-field splitting δ and for the perpendicular splitting factor $g_\perp$. The results of measurements by the Faraday method of $\chi_{\parallel}$ and $\chi_\perp$ are reported here along with some new anisotropy measurements. The results for all the parameters agree very well between the two experiments, and the values found are $\delta = 8.06 \pm 0.15$ cm^{-1}, $g_\perp = 1.720 \pm 0.003$, and the spin-orbit parameter $\lambda = 91 \pm 1$ cm^{-1}.

SEVERAL years ago, Brumage, Quade, and Lin[1,2] reported results of magnetic-susceptibility measurements on vanadium-doped Al_2O_3 in the temperature range 4–300°K. From these results, they were able to determine the zero-field splitting δ of the trigonal ground state, the perpendicular g factor $g_\perp$, the trigonal field splitting Δ_T, and the spin-orbit coupling parameter λ. Later, Brumage, Seagraves, and Lin[3] extended some of these measurements to 1000°K to make a more direct determination of Δ_T. More recently, Smith[4] has measured the magnetic anisotropy of this crystal system using one of Brumage's samples in the temperature range 1–5°K, a region in which the susceptibility expressions are most sensitive to δ and $g_\perp$. Reported here are results of measurements by the Faraday method of the parallel and perpendicular susceptibilities, $\chi_{\parallel}$ and $\chi_\perp$, respectively, in the same low-temperature range. For consistency, however, the measurements have been extended up to 300°K. Also, new anisotropy results are included and, not only are they compatible with the Faraday results, but they are believed to be more accurate than those reported in Ref. 4 for two reasons: (i) The sensitivity of the torsion balance has been greatly improved, and (ii) a new method of analysis of the data has been used which is independent of the reference value.

To obtain a more direct determination of $g_\perp$ and the zero-field splitting of the trigonal ground state of the V^{3+} ion in the octahedral field of the Al_2O_3 host crystal, measurements by the Faraday method have been made down to about 1°K for the magnetic field parallel and perpendicular to the c axis of the crystal. The apparatus was similar to that used in magnetic studies[5] of V_2O_3 but equipped with a low-temperature Dewar and manifold for pumping on the liquid-He vapor. Helium gas at a pressure of about 2–3 cm of Hg was inserted in the sample chamber when the system was at 77°K for the purpose of exchanging heat between the liquid He and the sample. However, in selected temperature ranges less exchange gas was used, but a longer time between temperature changes was allowed so that the sample could reach thermal equilibrium with its environment. The same procedure

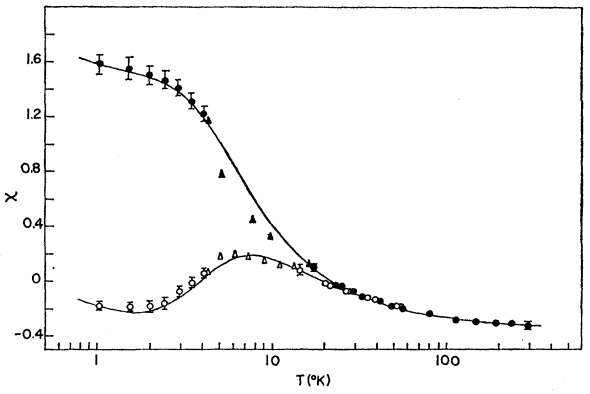

FIG. 1. Measured magnetic susceptibilities of $Al_2O_3:V^{3+}$ in units of 10^{-6} cgs emu. Only the circles, both filled ($\chi_\perp$) and unfilled ($\chi_{\parallel}$), were used in the data analysis. The solid curves represent the theoretical values (including $C/T+\chi_{\text{dia}}$) obtained using the parameters in Table I. Above 80°K, filled circles represent both $\chi_{\parallel}$ and $\chi_\perp$.

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