ELECTRONIC AND NUCLEAR POLARIZATION IN VANADIUM BY SLOW NEUTRON SCATTERING*

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We have studied by means of polarized neutron scattering the magnetization induced in a single crystal of pure vanadium when a magnetic field is applied, with the hope of identifying the particular electron group which is polarized. Vanadium exhibits a weak paramagnetic susceptibility[1] which is essentially temperature independent, and, in assessing the temperature independence of the electronic polarization, additional scattering effects to be associated with nuclear polarization have been encountered at temperatures below 15°K.

With specimen magnetization in a magnetic field H, the magnetic scattering amplitude p for neutrons becomes

$$p = 0.484\,\chi_M H f \times 10^{-16}\ \text{cm}, \qquad (1)$$

with χ_M the molar susceptibility and f the magnetic amplitude form factor. In the polarized neutron technique, the ratio of the Bragg reflection intensity for the two neutron polarization states (parallel and antiparallel to the field on the crystal) is determined, and this polarization ratio R is given by

$$R = \frac{I(+)}{I(-)} = \left(\frac{b+p}{b-p}\right)^2 \cong 1 + 4p/b, \qquad (2)$$

with b the coherent nuclear scattering amplitude which has been determined by separate measurements[2] to be -0.048×10^{-12} cm. The expression (2) is valid when $p \ll b$ characteristic of the present measurements.

Figure 1 summarizes the polarization ratio measurements at 20°C as a function of applied field strength for two reflections, (110) and (220), of vanadium. Values of R less than unity are to be expected because of the negative sign of the coherent nuclear scattering amplitude, and R is seen to vary linearly with H in agreement with Eq. (2) above. Not predicted by this equation, however, is the intercept value as H approaches zero, and this effect is interpreted as the result of a neutron spin-neutron orbit interaction as discussed in a companion Letter.[3] It should be mentioned that the magnetization of our crystals has been established as going strictly to zero with decreasing values of H. From the slopes of the Fig. 1 curves and also for other reflections,

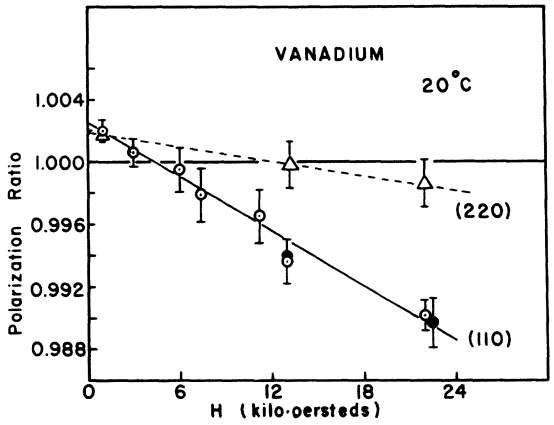

FIG. 1. Polarization ratio versus magnetic field strength for two vanadium reflections. The solid points represent data taken with a second crystal.

values of p/b have been determined, and these are shown as the data points in Fig. 2, plotted as a function of the scattering angle variable $\sin\theta/\lambda$.

The form factor associated with the data of Fig. 2 is characteristic of $3d$ electrons rather than $4s$ electrons, since the form factor for the latter would have fallen to zero value at $\sin\theta/\lambda = 0.17$ with only minor values at higher angles. Theoretical form factors for both $3d$ electrons and $4s$ electrons have been given by Watson and

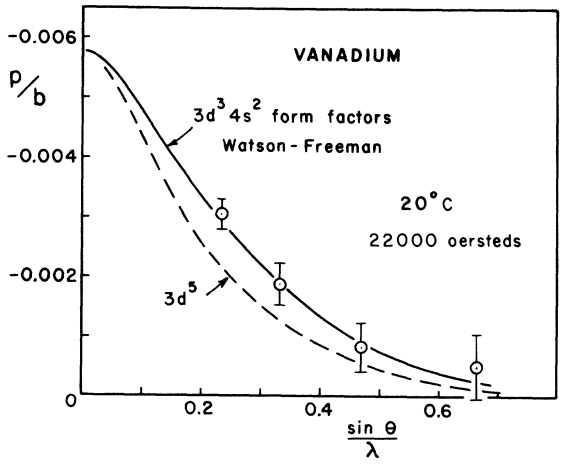

FIG. 2. Ratio of magnetic to nuclear scattering versus scattering angle variable for various vanadium reflections.

Freeman[4] for atoms in $3d^34s^2$ and $3d^5$ configurations, and these may be compared with the data. However, the selection of a particular configuration or a distribution of the $3d$ spin or $3d$ orbital contributions to the magnetization is rather difficult without using additional information. Recently, Clogston et al.[5] have analyzed the measured susceptibility into components arising from $4s$-spin, $3d$-spin, and $3d$-orbital contributions by using low-temperature specific-heat data. If their procedure is followed using the measured susceptibility for our crystal and allowing for a small atomic diamagnetism, the total susceptibility of 2.60×10^{-4} emu/mole is distributed into

$$\chi_{\text{diam}} = -0.15\times10^{-4} \text{ emu/mole},$$

$$\chi_{4s \text{ spin}} = +0.30\times10^{-4} \text{ emu/mole},$$

$$\chi_{3d \text{ spin}} = +0.68\times10^{-4} \text{ emu/mole},$$

$$\chi_{3d \text{ orbit}} = +1.76\times10^{-4} \text{ emu/mole}.$$

Using this magnetization distribution and the component form factors of Watson and Freeman, the calculated curves shown in Fig. 2 are obtained for the two atomic configurations $3d^34s^2$ and $3d^5$. It is seen that good agreement with this susceptibility distribution is obtained if the free atom configuration set of form factors is used.

Since the measured susceptibility is very closely temperature independent, the neutron scattering due to electronic polarization should also be insensitive to temperature changes. In confirmation of this, we have studied the scattering with the crystal held at low temperatures in a field of 13 200 oersteds and find no change until temperatures of 10 or 15°K are reached, below which a pronounced increase in the scattering effect is found. This is ascribed to the development of nuclear polarization because of "brute-force" action of the applied field on the vanadium nuclear moment. Figure 3 illustrates the dependence of the polarization ratio for the (110) reflection upon temperature with an applied field of 13 200 oersteds and for comparison the calculated value of this polarization. It can be shown[6] that nuclear polarization modifies the vanadium coherent scattering amplitude to become

$$b_{\text{coh}}' = b_{\text{coh}} \pm \tfrac{7}{16}P_N(b_+ - b_-),$$

$$= b_{\text{coh}} \pm \tfrac{3}{16}(\mu H/kT)(b_+ - b_-),$$

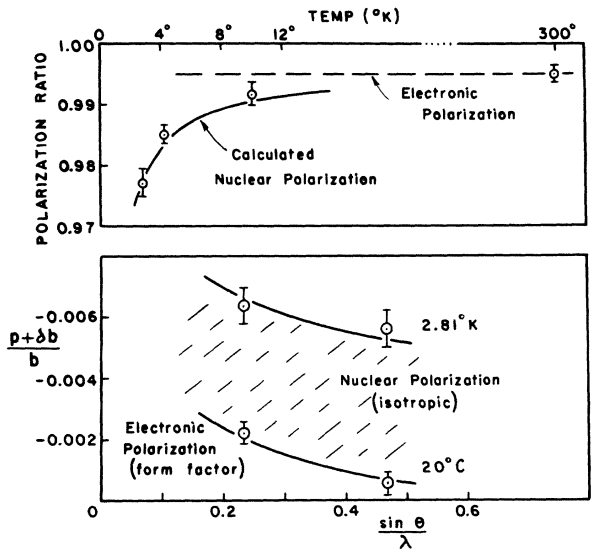

FIG. 3. Polarization ratio versus temperature for the (110) vanadium reflection and form factor curves showing combined electronic and nuclear polarization at low temperature.

with b_{coh} the coherent nuclear scattering amplitude characteristic of unpolarized nuclei, P_N the nuclear polarization, μ the vanadium nuclear magnetic moment, and b_+ and b_- the nuclear spin scattering amplitudes. This expression is valid when the nuclear polarization is very small and the neutron polarization is very high, characteristic of the present measurements. From the incoherent scattering cross section for vanadium (5.1×10^{-24} cm^2/atom), the term $(b_+ - b_-)$ is known in magnitude, and the present experiment determines the sign of this term and hence permits unambiguous assignment of the spin scattering amplitudes.

The additional scattering at low temperatures can be positively identified with nuclear polarization because it is without angular dependence as shown in Fig. 3. It should be mentioned that the nuclear polarization is very small for these conditions of relatively high temperature and low field (0.00040 at 2.81°K with 13 200 oersteds), and yet it can be detected and measured rather easily by this technique.

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[1]A number of susceptibility values for vanadium have been reported, for instance, by C. J. Kriessman, Rev. Modern Phys. 25, 122 (1953), and by B. G. Childs, W. E. Gardner, and J. Penfold, Phil. Mag. 4, 1126 (1959). S. T. Lin has obtained a value of 2.60×10^{-4} per mole for our crystals which were grown by Materials Research Corporation by electron beam zone melting. Our crystals are of 99.97% purity.

[2]In collaboration with D. Murray.

[3]C. G. Shull, following Letter [Phys. Rev. Letters 10, 297 (1963)].

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NEUTRON SPIN-NEUTRON ORBIT INTERACTION WITH SLOW NEUTRONS*

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In experiments with the scattering of polarized slow neutrons from vanadium crystals, it has been found[1] that the intensity of coherent Bragg reflection is neutron-polarization-sensitive, even though no magnetic field is present on the crystal. This suggests the presence of a neutron spin-neutron orbit interaction of the type considered by Schwinger[2] for the fast neutron case. In this spin-orbit interaction the moving neutron magnetic moment senses the nuclear charge, and this produces an asymmetrical scattering potential across the scattering atom along with the symmetrical nuclear scattering potential. This is illustrated schematically in Fig. 1, and it implies that the scattering of neutrons in a given polarization state will differ for scattering to the right or to the left. It also implies that the spin-orbit scattering amplitude will be imaginary (or with 90° phase difference relative to the nuclear scattering amplitude) because of its noncentrosymmetric origin.

The Schwinger treatment shows that this interaction yields an imaginary scattering amplitude which upon combination with the imaginary component of the nuclear scattering amplitude will produce asymmetry. For the slow neutron case, the Schwinger expression for this amplitude must be modified to include effect of electron charge screening of the nuclear charge, so that the total coherent scattering amplitude becomes

$$f(\theta) = b + ib' + i[\vec{P}_n \cdot \vec{n}\gamma \cot\theta],$$

with b the real component of the coherent nuclear scattering amplitude, b' the imaginary component of the total cross section (scattering plus absorption) equal to $(k/4\pi)\sigma(\text{total})$, $\vec{P}_n$ the neutron polarization vector, $\vec{n}$ a unit vector described by $\vec{k} \times \vec{k}_0 = \vec{n}k^2 \sin\theta$, with $\vec{k}_0$ and $\vec{k}$ the incident and scattered neutron wave vectors, and θ the Bragg angle of scattering. γ determines the strength of the spin-orbit interaction, and for slow neutrons

$$\gamma = \tfrac{1}{2}\mu_n (e^2/Mc^2) Z(1-f),$$

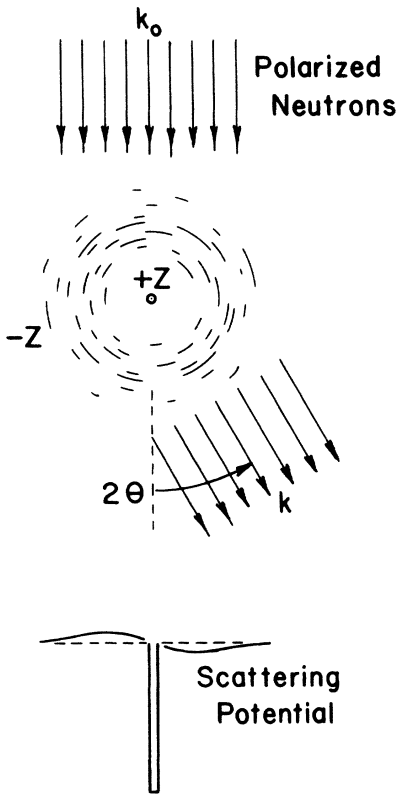

FIG. 1. Schematic diagram of polarized neutron scattering by an atom illustrating neutron spin-orbit scattering potential.