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THE MAGNETIC STRUCTURES OF PRASEODYMIUM AND NEODYMIUM

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Résumé. — Les structures magnétiques de Pr et Nd ont été étudiées par diffraction neutronique dans des champs magnétiques pouvant atteindre 46 kOe. Aucun ordre magnétique spontané n'a été détecté dans Pr, mais un champ magnétique perpendiculaire à l'axe c induit un fort moment ferromagnétique. Le moment induit dans un champ parallèle à l'axe c est notablement plus faible. La susceptibilité est donc très anisotrope. Dans Nd, un champ magnétique modifie considérablement la structure magnétique périodique.

Abstract. — The magnetic structures of Pr and Nd single crystals have been studied by neutron diffraction with applied magnetic fields up to 46 kOe. No spontaneous magnetic order has been detected in Pr, but a magnetic field applied perpendicular to the c-axis induces a large ferromagnetic moment. The induced moment with the field parallel to the c-axis is much smaller, indicating a highly anisotropic susceptibility. In Nd the magnetic field produces substantial modifications of the periodic magnetic structure.

Prasodymium and neodymium have the double hexagonal close packed structure in which the stacking sequence along the c-axis is ABAC. Atoms in the A layers have a local environment of approximately cubic symmetry, while in the B and C layers the atoms have an hexagonal close packed arrangement of nearest neighbours.

The crystal field interactions in the light rare earth metals are comparable in magnitude to the exchange forces. Magnetic anisotropy measurements are therefore essential to the understanding of their magnetic properties. In the present experiments the magnetic ordering properties of Pr and Nd have been studied by neutron diffraction on single crystals in magnetic fields up to 46 kOe. By measuring the effect of the field on Bragg intensities at different reciprocal lattice positions we have been able to separate the contributions to the magnetic anisotropy from the cubic and hexagonal sites.

Bleaney [1] has estimated the crystal field splittings in Pr by fitting the magnetic specific heat anomaly. His calculations show that the ground states of the ions at both the cubic and hexagonal sites are nonmagnetic singlets. No magnetic ordering will occur in this case unless the ratio of the exchange to crystal field interactions exceeds a critical value [2]. Neutron diffraction experiments [3, 4] have shown that polycrystalline Pr is antiferromagnetically ordered at 4.2 °K. In contrast we have detected no trace of spontaneous magnetic order in a single crystal at 4.2 °K. However, a magnetic field applied in the \(b_3 \langle 110 \rangle \) direction is produced large increases in the Bragg intensities at most reciprocal lattice points, indicating a large induced ferromagnetic moment. No change in the intensities of the \(100 \) and \(104 \) reflections was detected up to 46 kOe. This indicates that the moment, \( \mu_h \), induced on the hexagonal sites is just twice \( \mu_c \), the induced moment on the cubic sites, since the structure factor for both of these reflections contains the moment combination \( 2 \mu_c - \mu_h \). This result, combined with the magnetization data of Johansson and al. [5, 6], allows a separate determination of \( \mu_c, \mu_h \) and the form factor for each reflection. Figure 1 shows \( \mu_c \) and \( \mu_h \) as a function of field (points labelled \( \text{H} \perp c \)) determined in this way from the field dependence of the intensities of the \(101 \), \(102 \), \(103 \) and \(004 \) reflections. The moment versus field plots show considerable curvature above 15 kOe, indicating a tendency towards saturation at high fields. Also shown in figure 1 (inset) is the form factor deduced from Bragg intensities at 46 kOe, compared with calculation of Wedgwood [7] for the free \( \text{Pr}^{3+} \) ion.

With the magnetic field applied in the \(b_3 \langle 001 \rangle \) direction the induced moments are considerably smaller, indicating a highly anisotropic susceptibility. The values of \( \mu_c \) and \( \mu_h \) shown in figure 1 (labelled \( \text{H} \parallel c \)) are derived from the field dependence of the \(100 \) reflection, combined with Johansson's [6] magnetization data. The anisotropy at the hexagonal sites is seen to be much larger than at the cubic sites. The cubic anisotropy is probably due to exchange

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**Figure 1.** — Moments induced on hexagonal and cubic sites of Pr by an applied field in \( b_3 \) and \( b_3 \) directions at 4.2 °K. Inset: form factor deduced from Bragg intensities at 46 kOe, compared with calculation of Wedgwood [7] for the free \( \text{Pr}^{3+} \) ion.

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interactions with near neighbours on hexagonal sites. There may also be contributions from anisotropic exchange [8] and the departure from cubic symmetry due to non ideal \(c/a\) ratio. Bleaney's molecular field calculations [1] predicted an anisotropy of the susceptibility for the hexagonal ions in the opposite sense to that observed. To account for the experimental results, it is necessary to invoke highly anisotropic molecular field parameters. It is interesting to note that the low lying excitations in Pr, which have been observed recently by inelastic neutron scattering [9], have a dispersion that is consistent with a ferromagnetic exchange along \(b_2\), and a strong antiferromagnetic exchange along \(b_4\).

The magnetic structure of Nd in zero field has been investigated by Moon and al. [10], using neutron diffraction. The moments on the hexagonal sites order at 19.2 °K forming a periodic antiferromagnetic structure in which the moments in the B and C layers are sinusoidally modulated, with both the moment, \(\mu_h\), and the modulation vector \(Q_b\) parallel to the \(b_1\) direction. The cubic moments order in a similar fashion at 7.5 °K. The modulation vector \(Q_b\) is again along \(b_4\), but the moments are parallel to a \(b_2\) direction. Because of crystal field effects, the ordered moments on both sites are smaller than the value \(gJ\mu_B = 3.3\mu_B\). At 4.2 °K we find \(\mu_h = 2.1 \pm 0.3\mu_B\) and \(\mu_b = 2.0 \pm 0.3\mu_B\).

The satellite reflections that correspond to the modulated moments on the hexagonal sites are observed to split at the lower transition temperature into two components having roughly equal intensity and modulation vectors that differ by about 8%. When a magnetic field is applied in the \(b_3\) direction at 4.2 °K, one of the two components of the hexagonal satellites gradually disappears. This transition, which is finally complete at 26 kOe, is accompanied by a marked hysteresis, which can be seen in the behaviour of the modulation vectors of the two components, shown in figure 2a. The field has very little effect on the integrated intensity of the hexagonal satellites. The cubic satellite intensity decreases uniformly with increasing field and simultaneously a ferromagnetic moment develops on the cubic sites (Fig. 2b). This is consistent with the tilting of the cubic moment out of the basal plane. It appears, then, that the anisotropy at the cubic sites is relatively weak, whereas at the hexagonal sites, the anisotropy is large enough to constrain the moments to lie in the basal plane. This result is also contrary to the predictions of a simple molecular field theory [11], suggesting that anisotropic exchange interaction may also be important in Nd.

Preliminary measurements [12, 5] with the field in the \(b_3\) direction at 4.2 °K show a more pronounced modification of the moment distribution. The cubic satellite intensities decreases with field and disappear at about 35 kOe. At the same time the cubic sites develop a ferromagnetic moment that is consistent with the complete alignment of the cubic moments above 35 kOe. One component of the hexagonal satellites disappears in a field of 12 kOe, and at 25 kOe there is an abrupt increase in both the periodic and ferromagnetic moments on the hexagonal sites. This may result from a level-crossing as the splitting of the crystal field energy levels by the magnetic field increases.

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