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Neutron diffraction study of the magnetic ordering in NpBi

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The magnetic ordering of NpBi has been determined by neutron diffraction on a single crystal. NpBi orders antiferromagnetically in the triple-$\boldsymbol{k}$ type-I structure ($\boldsymbol{k} = |0\,0\,1|$, $\boldsymbol{m}_k // \boldsymbol{k}$) below $T_N = 192.5$ K. The ordered magnetic moment develops at T_N with a critical exponent $\beta = 0.31 \pm 0.02$ an increases continuously on cooling to reach a value of $2.48 \mu_B$/Np at low temperature.

NpBi belongs to the homogeneous series of actinide monopnictides MX (M = U, Np, Pu; X = N, P, As, Sb, Bi) which crystallize in the NaCl type structure. In these compounds, due to the partially localized character of the 5f electrons, the hybridization with the p or d electrons leads to peculiar magnetic interactions [1]. A characteristic of these interactions is their anisotropy which consists in strong ferromagnetic coupling in a (0 0 1) plane and much weaker interactions in the perpendicular direction. This results, without any exception, in a magnetic ordering characterized by wave vectors and Fourier components $\boldsymbol{k} = |0\,0\,k|$ and $\boldsymbol{m}_k // \boldsymbol{k}$. The particular value of k depends on the details of the interplane interactions. In many cases a local moment direction, different from the cubic axis, is satisfied by stabilizing multi-$\boldsymbol{k}$ structures; double-$\boldsymbol{k}$ if $\boldsymbol{m} // \langle 1\,1\,0 \rangle$ and triple-$\boldsymbol{k}$ if $\boldsymbol{m} // \langle 1\,1\,1 \rangle$ [2].

Since the pioneering work of the Argonne group [3], who studied by powder neutron diffraction the magnetic ordering of some Np monopnictides, the availability of large and good quality single crystals has allowed much progress in the description of the magnetic behaviour of these compounds, such as a detailed description of the nature of the magnetic ordering and the determination of the field versus temperature phase diagrams [4].

The NpBi single crystal ($1 \times 2 \times 3\,\text{mm}^3$) was grown by the mineralization technique [5]. It was encapsulated in a sealed double wall aluminium sample holder. The neutron experiments were performed at the Siloe reactor of CEN-Grenoble, using a double axis spectrometer equipped with a moving up counter arm. The crystal was mounted in a cryomagnet with a $|1\,1\,0|$ direction vertical and parallel to the magnetic field direction. A neutron wavelength $\lambda = 1.54$ Å, provided by an HOPG monochromator and filtered by pyrolitic graphite, was used.

Results and discussion

At low temperatures, superlattice magnetic Bragg reflexions are observed corresponding to the wave vectors $\boldsymbol{k} = \langle 0\,0\,1 \rangle$ and to a longitudinal polarization ($\boldsymbol{m}_k$ parallel to $\boldsymbol{k}$). The magnetic intensities from the three equivalent $\boldsymbol{k}$-vectors $|0\,0\,1|$, $|0\,1\,0|$ and $|1\,0\,0|$ have the same values and give Fourier components $m_k = 1.4 \pm 0.07 \mu_B$. This situation can correspond either to a collinear structure, with an equipartition of domains, or to multi-$\boldsymbol{k}$ ordering [6]. In any case the Np ordered moment value is $m = \sqrt{3} m_k = (2.48 \pm 0.1) \mu_B$/Np, which is in good agreement with Mössbauer's results [6].

The same magnetic ordering is observed in the whole ordered temperature range below T_N (fig. 1). Accurate intensity measurements in the vicinity of T_N, in the range of reduced temperature $10^{-3} < (T_N - T)/T_N < 10^{-1}$, have been made. A log–log plot of these intensities versus temperature gives a straight line (corresponding to $T_N = 192.5 \pm 0.1$ K and a slope $2\beta = 0.62 \pm 0.01$) which characterizes the critical exponent β (fig. 2). The temperature dependence of

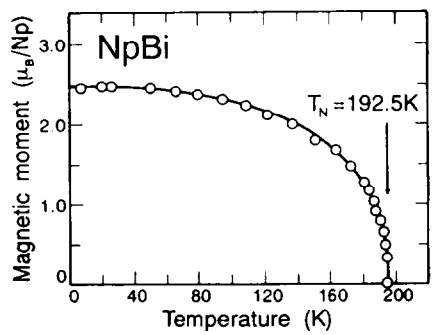

Fig. 1. Temperature dependence of the Np magnetic moment in NpBi in zero applied field.

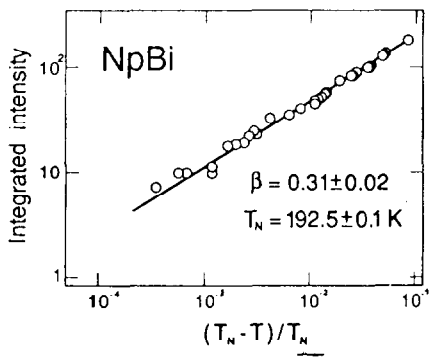

Fig. 2. Log–log plot of the magnetic intensities versus reduced temperature $T_N - T/T_N$ in NpBi near the ordered temperature.

the magnetic intensities has also been studied for a magnetic field of 4.5 T applied along a |1 1 0| axis. As shown in fig. 3 the magnetic intensities, arising from the 3 equivalent k-vectors measured on cooling the sample from above T_N, have the same values and the same temperature dependence. This implies the absence of any domain motion. Collinear and double-k structures of tetragonal symmetry give rise to three domains which are not equivalent with respect to this applied field. This situation gives rise to domain motion in a rather small applied magnetic field which

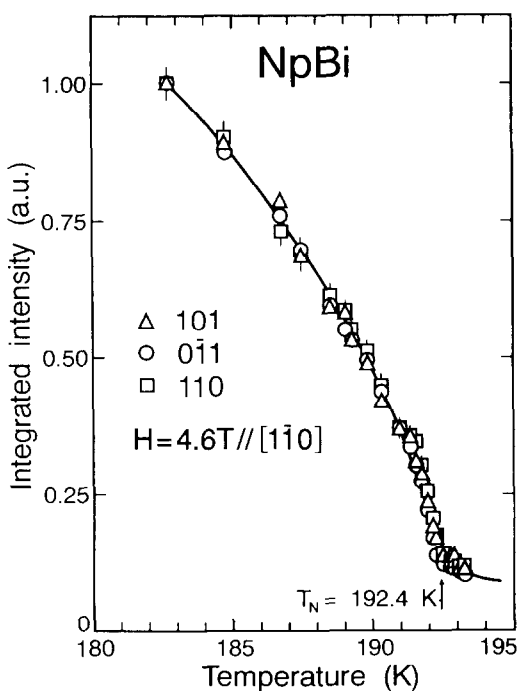

Fig. 3. Temperature dependences of the magnetic intensities in NpBi under a magnetic field $H = 4.5$ T along the |1 1 0| direction.

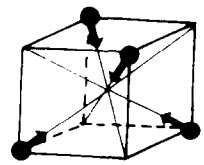

Fig. 4. Multiaxial (triple-k) structure of NpBi.

leads to a change of intensities. The triple-k ordering, which preserves the full symmetry of the cubic group, presents only one domain and the magnetic field cannot have any effect, unless the interaction terms stabilizing the triple-k structure were dominated by the Zeeman energy ($-\chi H^2$) of a collinear structure. The present experimental results then allow us to deduce a triple-k ordering in NpBi. The corresponding multiaxial structure is shown in fig. 4. This triple-k type-I structure is common to USb, NpSb and NpBi. These compounds have similar Néel temperatures (212 K in USb, 199 K in NpSb and 192.5 K in NpBi). The type-I triple-k ordering is very stable because no metamagnetic transition can be induced in the range of experimentally available magnetic field and, moreover, even near to T_N, large fields ($H > 5$ T) are needed to overcome the coupling resulting in the triple-k ordering [6]. The critical exponent $\beta = 0.31$ corresponds to the theoretical one expected for a 3d Ising system and has also been observed in many other NaCl compounds of Ce, U and Np which all are characterized by the same anisotropic exchange interaction [7]. However in NpSb $\beta = 0.257$; a value much lower than that found in NpBi in spite of their similarities [7].

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