Magnetic-Structure of UNiGa

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On the magnetic structure of UNiGa

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UNiGa has been studied by powder neutron diffraction together with $UNiAl_{0.6}Ga_{0.4}$ and UNiAl. The collinear magnetic structure of UNiGa consists of U magnetic moments in the [001] direction coupled ferromagnetically within (001) sheets and antiferromagnetic stacking (+ − − + + −) of the sheets along the *c*-axis. In the case of $UNiAl_{0.6}Ga_{0.4}$, a simple (+ − + −) stacking is observed. The very complicated powder pattern of UNiAl prevents determination of the magnetic structure in this case and single-crystal studies are desired.

There has been a long lasting dispute about the type of magnetic ground state of the compound UNiGa, which is magnetically ordered below $T_c = 38$ K. Originally, the magnetization measurements performed on the UNiGa single crystal showed a full remanence [1], which can be interpreted as a sign of ferromagnetic (F) ordering. Complicated magnetic history phenomena and slow relaxations led even to conclusions in terms of spin-glass like or reentrant behaviour [2,3]. Some features of the magnetization of polycrystalline samples, such as the wasp-tailed hysteresis loop [1,4], could be attributed to a metamagnetic transition of an antiferromagnet. For further investigations we have prepared a well-defined single crystal of exact 1 : 1 : 1 stoichiometry checked by a microprobe. The magnetization behaviour of this crystal points to antiferromagnetic (AF) ordering, which undergoes a spin-flip transition in a relatively low field of $\simeq 0.8$ T [5]. It is interesting to determine the magnetic structure of UNiGa in the context of compounds in the pseudoternary UNi(Ga,Al) system, all exhibiting Ising-like behaviour and similar metamagnetic transitions [6]. Here we report on results of neutron diffraction on powder samples of UNiGa, $UNiAl_{0.6}Ga_{0.4}$ and UNiAl performed on the D1B diffractometer at ILL, Grenoble and the HIPD diffractometer at LANSCE, Los Alamos.

Refinement of the 60 K spectrum of UNiGa confirmed that we deal with an ordered ternary compound with ZrNiAl-type structure. The lattice parameters are $a = (670.120 \pm 0.005)$ pm and $c = (400.878 \pm 0.004)$ pm. The atoms are situated as follows: Ga at x, 0, 0; $x = 0.24041 \pm 0.00008$; U at (y, 0, $\frac{1}{2}$), $y = 0.57754 \pm 0.00007$; Ni at (0, 0, $\frac{1}{2}$) and ($\frac{1}{3}$, $\frac{2}{3}$, 0).

The spectrum measured at 3 K and its magnetic part obtained by subtracting the spectrum collected at 41 K ($> T_c$), both shown in fig. 1, demonstrate immediately that UNiGa possesses a long-range magnetic ordering, but it is definitely not ferromagnetic. Magnetic diffraction lines, which disappear at temperatures higher than 37 K, are found between the nuclear Bragg

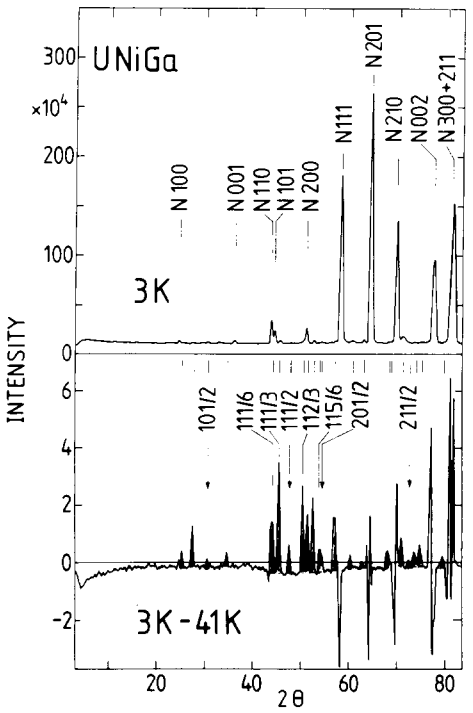

Fig. 1. The neutron powder diffraction pattern of UNiGa measured at 3 K (upper part). The magnetic reflections obtained by subtracting the spectrum collected at 41 K from that measured at 3 K (lower part).

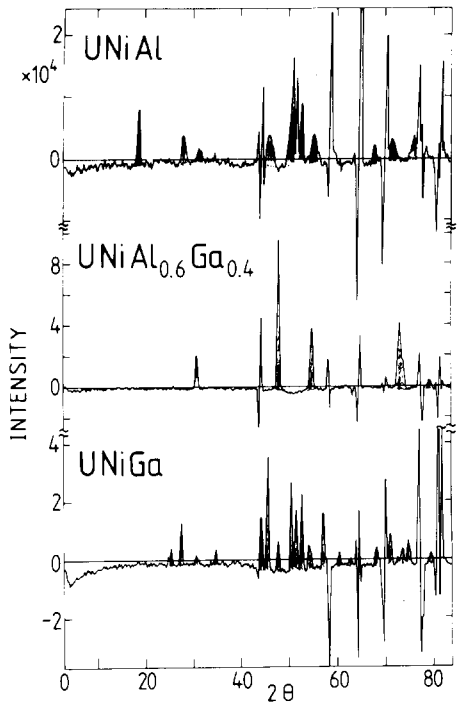

Fig. 2. From the top to the bottom are shown the difference patterns obtained on UNiAl ("3 K"-"28 K"), $UNiAl_{0.6}Ga_{0.4}$ ("3 K" – "27 K") and UNiGa ("3 K" – "41 K").

reflections (marked by N). They correspond to Bragg peaks at (1 0 $\frac{1}{2}$), (1 1 $\frac{1}{2}$), (2 0 $\frac{1}{2}$), (2 1 $\frac{1}{2}$), etc. and to additional positions around them (e.g. those around (1 1 $\frac{1}{2}$) have Miller indices (1 1 $\frac{1}{3}$), (1 1 $\frac{1}{6}$), etc.). This complicated magnetic diffraction pattern is indicative of a magnetic unit cell of the dimensions $a \times 6c$. A collinear model with magnetic moments parallel to the c-axis and the stacking (+ + − + − −) in this direction leads to the best fit of the data. The relatively weak magnetic reflections with respect to the nuclear ones prevent us to extract a precise value of the magnetic moments. The preliminary data obtained recently on a single crystal of UNiGa provide the first estimate of the ordered moment $\mu_0 = (1.4 \pm 0.2)\mu_B$/U-atom is very close to the value of $1.35\mu_B$/U-atom obtained from the saturated magnetization.

Comparison of the magnetic diffraction data obtained on UNiAl, $UNiAl_{0.6}Ga_{0.4}$ and UNiGa, given in fig. 2, emphasizes variations of the magnetic structure in the UNi(Ga, Al) system. By partially substituting Ga by Al, the diffraction pattern becomes simpler and the magnetic Bragg peaks occur only at (1 0 $\frac{1}{2}$), (1 1 $\frac{1}{2}$), (2 0 $\frac{1}{2}$), (2 1 $\frac{1}{2}$), etc., fully consistent with the (+ − + −) AF structure. UNiAl, on the other hand, exhibits an even more complicated magnetic diffraction pattern than UNiGa. Note also the magnetic (0 0 $\frac{1}{2}$) Bragg peak near $2\Theta = 18°$ in UNiAl which is not observed in the other compounds. A reliable interpretation of the UNiAl magnetic structure requires single-crystal studies which are currently under way [7].

The origin of the ferromagnetic behaviour of some "UNiGa" samples remains at the moment unclear. Current studies of non-stoichiometric U–Ni–Ga compounds show that it could be a small deviation from the ideal stoichiometry which may lead to ferromagnetic behaviour [1]. A delicate balance of competing interactions between the U moments along the c-direction is documented, besides a low field necessary for the ferromagnetic alignment, by the existence of a ferrimagnetic phase(s) stable in zero field at temperatures higher than 35 K, which is also indicated by a preliminary neutron diffraction study of the crystal. If the ideal stoichiometry is violated, defects like stacking faults may appear. In this case, the impact on the type of coupling along the c-axis can be very drastic and, consequently, the magnetic behaviour can be easily modified.

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