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MAGNETIC PHASE DIAGRAMS OF UNiGe

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UNiGe undergoes two magnetic transitions in zero field. Here, the magnetic diagrams of UNiGe for B ||*b* and B ||*c* are reported. We performed temperatures scans of the magnetization in static magnetic fields up to 19.5T applied along the *b* and *c* axes. For both orientations 3 magnetic phases have been identified in the B-T diagrams. We confirmed the previously reported phase boundaries for B || *c*, and in addition we determined the location of the phase boundaries for B || *b*. We discuss a possible relationship of the two zero-field antiferromagnetic phases (commensurate: T<42K; incommensurate: 42K<T<50K) and the field-induced phase, which, at low temperatures, occurs between 18 and 25T or 4 and 10T for B || *b* or B || *c*, respectively. Finally, we discuss the field dependence of the electronic contribution γ to the specific heat for B ||*c* up to 17.5T, and we find that its field dependence is similar to the one found in more itinerant uranium compounds.

Keywords: magnetic-phase diagram, antiferromagnetism, uranium compounds

UNiGe exhibits two antiferromagnetic transitions at about 50 and 42K in zero field [1]. Neutron-diffraction studies established that both zero-field phases are well described in terms of single-**q** non-collinear arrangement of the moments which form in two domains [2]. The magnetic phase below 42K is commensurate with **q**=(0,1/2,1/2), while the phase between 42 and 50K is incommensurate with **q**=(0,δ,δ) and $\delta \approx 0.35$. Bulk magnetic investigations of UNiGe revealed a highly anisotropic behaviour both in the magnetically-ordered and in the paramagnetic states [3]. At low temperatures, spin-flip transitions occur for fields applied along *b* and c axis. At 4.2K, two transitions are found in both the *b*-axis magnetization (~17 and ~25T) and the c-axis magnetization (~4 and ~10T). For fields applied along *c*, it was shown that a magnetic structure with **q**=(0,1/3,1/3) forms [1], while the second transition presumably leads to a forced-ferromagnetic alignment of the moments. On the other hand, it was found that the *a*-axis response is much weaker and cannot be aligned even in pulsed fields up to 38T [3].

For B ||*c*, the magnetic-phase boundaries of the two zero-field phases and the field-induced (0,1/3,1/3) phase have been well established by previous magnetization [4], magnetoresistance [5] and specific-heat measurements [6]. For B ||*b*, on the other hand, far less work has been done and the studies have been restricted to field sweeps at fixed temperatures [4]. These studies had led to a relatively complex *b*-axis B-T diagram with 5 different magnetic phases. However, Purwanto et al. [7] pointed out that some of the 'phase lines' may be due to a misaligned crystallite, which was found in neutron-diffraction studies on the same sample. Therefore, it was argued that the intrinsic *b*-axis B-T phase diagram exhibits only 3 different magnetic phases similar to the *c*-axis B-T phase diagram. [7].

Here, we want to clarify the phase boundaries for B || *b* using a better-quality single crystal, which was used also previously in magnetoresistance studies for B || *c* [5]. We performed temperatures scans of the magnetic response in static magnetic fields up to 19.5T applied along the *b* and *c* axes. In addition, we studied the field dependence of the electronic contribution γ to the specific heat in magnetic fields up to 17.5T. The magnetization studies were performed in the 20T superconducting magnet at the Los-Alamos Pulse Field Facility of the National High Magnetic Field Laboratory; specific-heat studies in applied fields were done using the 17.5T superconducting magnet at the University of Amsterdam [8].

Fig. 1 shows the temperature dependence of the magnetization for various magnetic fields applied along the *b* and the *c* axis. For B || *b*, the most prominent feature is the drop in the magnetization at the 42K transition. We find a smooth and continuous shift of this transition toward lower temperatures with increasing field until it disappears for B>18T. There is a second anomaly in the *b*-axis magnetization: It is a barely visible increase in the magnetic response around 50K (at 2T) which becomes slightly more pronounced, but for fields above 10T it changes sign and a weak drop in the magnetic response appears at similar temperatures. Taking the temperatures where the anomalies occur, we can construct the B-T phase diagram for B || *b* and B || *c*. The results are shown in Fig. 2, where previous results have been included as well. It should be noted that we do not find any other obvious anomalies for the *b*-axis response, and this seems to confirm the picture proposed in ref. 7. Comparing the *b*-axis and *c*-axis magnetizations (Fig. 1) with the B-T phase diagrams (Fig. 2), the following picture emerges: For sufficiently low fields (B<10T for B || *b* and B<4T for B || *c*), one enters the incommensurate (0,δ,δ) phase, which causes the magnetic response to increase possibly suggesting changes in δ and/or domain repopulation effects. For higher magnetic fields, but below the second spin-flip transition ($B \approx 25T$ for $B \,||\, b$ and $B \approx 9.5T$ for $B \,||\, c$), one enters the field-induced (0,1/3,1/3) phase, which is accompanied by a slight drop in the magnetic response. A very similar behaviour in such intermediate fields for $B \,||\, b$ and $B \,||\, c$ may thereby indicate that the same (0,1/3,1/3) phase forms for both field orientations. Entering the (0,1/2,1/2) gives rise to a pronounced drop in the magnetic response. Application of fields in close proximity of the 'ferromagnetic' phase boundary (9T for $B \,||\, c$) gives rise to a relatively complex behaviour, which likely can be attributed to domain effects.

Altogether, we believe that our magnetization results are consistent with a relationship between the 3 magnetic phases which has been proposed earlier [2]. The incommensurate phase is believed to consist out of ferromagnetic sheets with a (++-) stacking with a spin-slip every 20th layer or so. The (0,1/3,1/3) has the same (++-) stacking of ferromagnetic sheets, but there is no spin-slip. The commensurate (0,1/2,1/2) phase, on the other hand, has ferromagnetic sheets stacked in a (+-) sequence.

Furthermore, we studied the field dependence of the electronic contribution γ to the specific heat. This provides some qualitative measure of Fermi-surface effects. For that, we measured c/T vs B for $B \,||\, c$ at 850mK using a semi-adiabatic method. Data were taken with increasing field. As can be seen in Fig.3, γ is somewhat reduced at the fields where the spin-flip transitions (~3T and ~11T at 850mK) occur. However, reduced values of γ at the first transition are present only in close proximity to the spin-flip transition. In fact, excluding the data points at 3 and 4T, one can fit all other values up to 11T to a quadratic behaviour (solid line in Fig. 3). Above the second transition, however, one finds almost linearly decreasing γ values up to the highest field applied (17.5T). This behaviour (quadratic increase below the transition, linear decrease above the transition) is surprisingly similar to the findings for UNiAl [9], which one can consider as a moderately enhanced heavy-fermion system, whereas the 5*f* electrons in UNiGe are more localized. At present, we have very little idea how to interpret this observation. Furthermore, we find some evidence that γ is slightly enhanced (~2-3mJ/K^2mol) between 3 and 9T after application of a field, but we did not study such hysteresis effects in details. Nevertheless, our results provide some evidence that the basic features of the Fermi surface in UNiGe survive the first spin-flip transition, while the nature of the Fermi surface seems to be modified at the second spin-flip transition.

In conclusion, we have confirmed the phase boundaries between the long-range ordered and the paramagnetic phases for B || *c*, and we also established the location of the phase boundaries for B || *b*. Finally, we found a field dependence of γ in UNiGe similar to the one found in more itinerant uranium compounds.

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figure captions:

Fig. 1: Temperature dependence of the magnetization for various fields (a) applied along the *b* axis and (b) fields applied along the *c* axis. Note the presence of various anomalies (see text), which determine the magnetic phase boundaries.

Fig. 2: Magnetic phase diagram for fields (a) applied along the *b* axis and (b) applied along the *c* axis. The plots comprise the data from previous magnetization (x)[4], magnetoresistance (Δ)[5] and present magnetization data (•). The shaded area in (b) represents the region where hysteresis effects occur.

Fig. 3: Field dependence of the electronic contribution γ to the specific heat for fields applied along the *c* axis, determined by C/T values at 850mK. The arrows indicate the locations of metamagnetic transitions in the c-axis magnetization (~4 and ~11T at 850mK). The error bars represent an estimate of the absolute error (~1mJ/K^2mol), while the relative error is much smaller (~0.1mJ/K^2mol). The solid line represents the fit of the data for B<10T (and excluding 3 and 4T) to a quadratic field dependence. Data have been taken with increasing field.

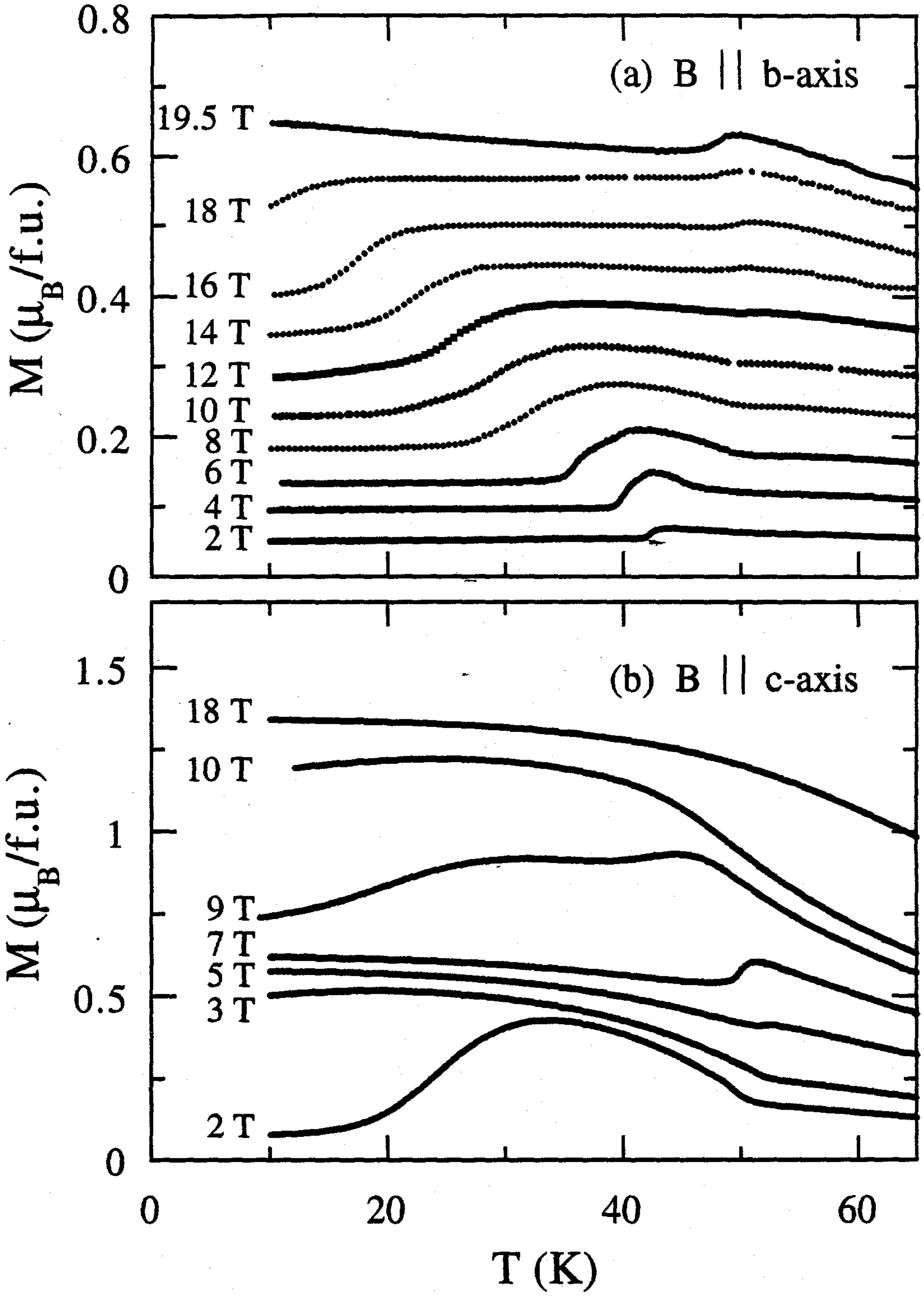

Fig. 1 H. Nakotte et al

Magnetic phase diagrams...

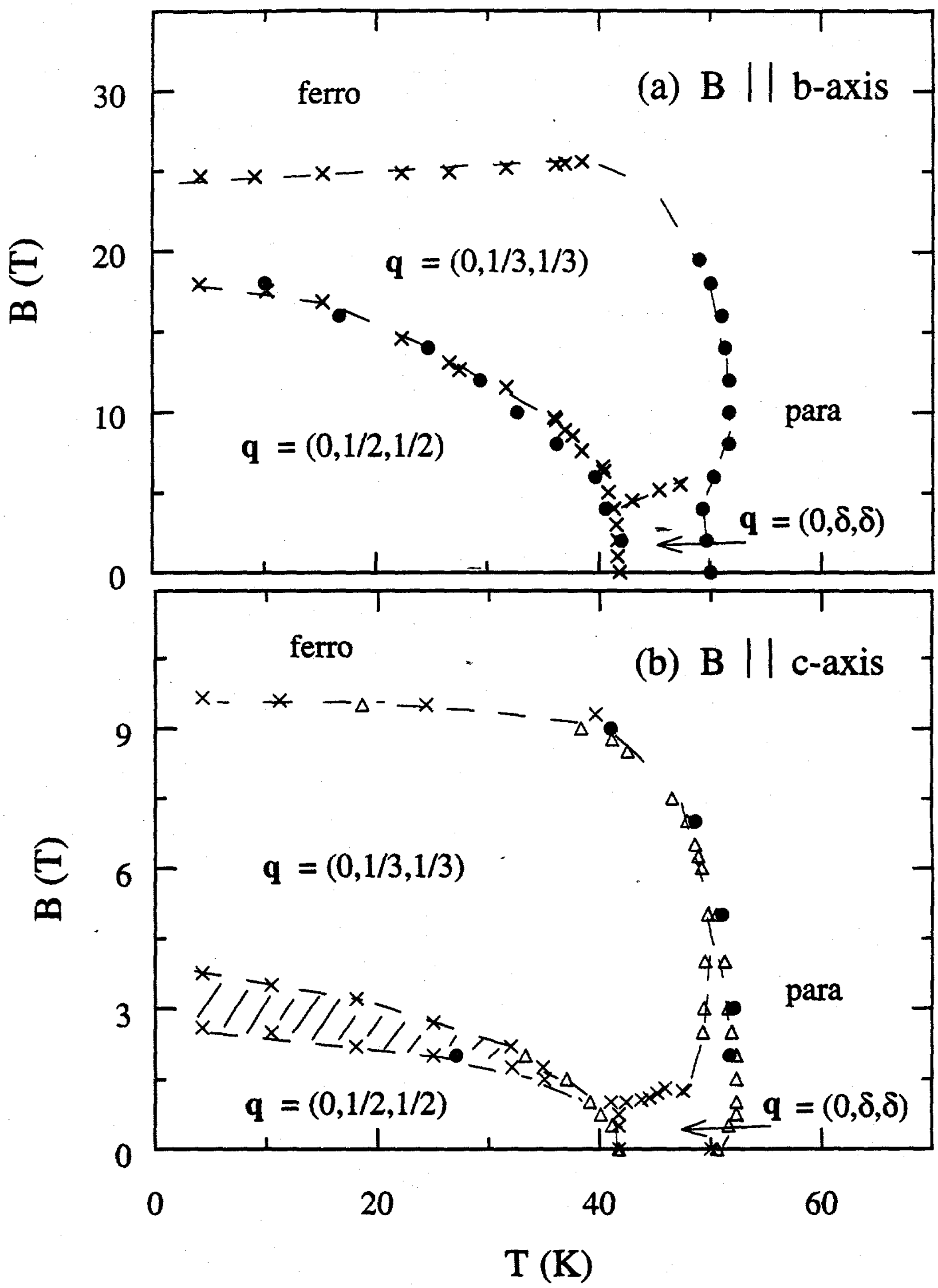

Fig. 2: H. Nakotte et al.
Magnetic phase diagrams ...

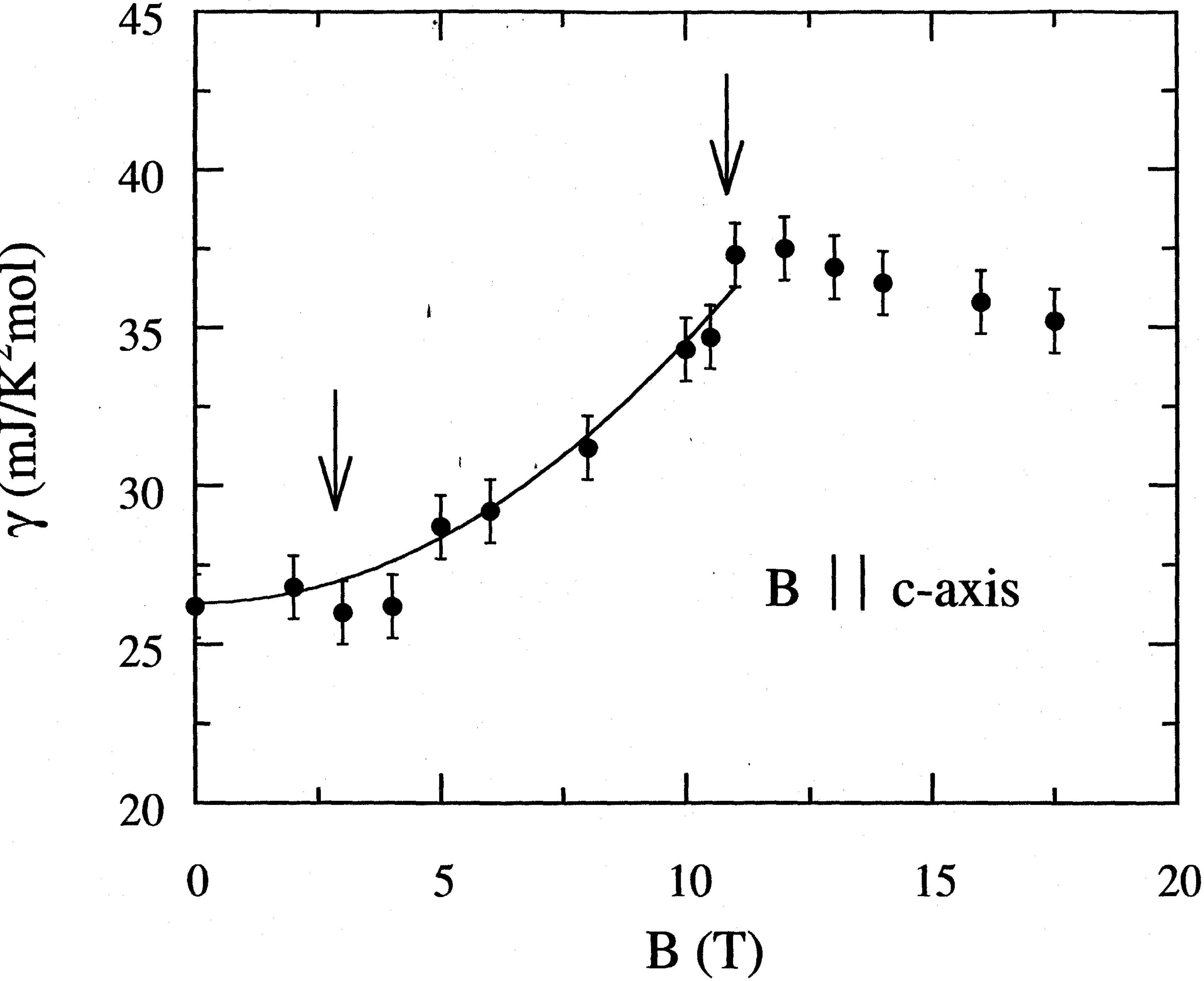

Fig. 3: H. Nakotte et al.
Magnetic phase diagrams ...