Metastable magnetic behaviour in UFe_4Al_8

2003 J. Phys.: Condens. Matter 15 S2029

(http://iopscience.iop.org/0953-8984/15/28/320)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) S2029–S2033 PII: S0953-8984(03)62667-6

Metastable magnetic behaviour in UFe_4Al_8

D X Li and Y Shiokawa

Institute for Materials Research, Tohoku University, Oarai, Ibaraki 311-1313, Japan

E-mail: dxli@imr.tohoku.ac.jp

Received 12 November 2002
Published 4 July 2003
Online at stacks.iop.org/JPhysCM/15/S2029

Abstract
Experimental results of ac susceptibility, dc magnetization, magnetic relaxation, and specific heat measurements on a well annealed polycrystalline UFe_4Al_8 sample establish that this compound is a long-range magnetic exchange interaction-dominated ferromagnet (Curie temperature $T_C = 138$ K) with strong magnetic anisotropy. The typical metastable magnetic characteristics—the irreversibility of the temperature dependence of the magnetization and the long-time magnetic relaxation behaviour at low temperatures—mainly result from domain wall pinning effects. The downshift of the ac susceptibility peak position with increasing frequency suggests that the effect of the spin-glass state, if it exists at all, is rather weak in this system.

1. Introduction

In the last decade, the intermetallic compound UFe_4Al_8 with the tetragonal body-centred crystal structure of $ThMn_{12}$ type has been extensively investigated by means of magnetization [1, 2], magnetoresistance [3], Mössbauer spectroscopy [4, 5], x-ray [6], and neutron diffraction measurements [4–9]. These studies led to contradictory explanations of the spin configuration, ranging from simple one-sublattice ferromagnetism to an unusual spin-glass state. Recently, a polarized neutron diffraction experiment was carried out on a stoichiometric single crystal of UFe_4Al_8, and the results revealed an ordered magnetic structure with two magnetic sublattices involving the Fe and U magnetic moments, respectively, and no spin-glass properties [9]. Although some conflicting results were obtained, which may be explained by deviations from ideal stoichiometry, the presence of a certain disordering on the Fe and Al sublattices, as well as the difference in synthesis conditions, magnetic history-dependent feature is a behaviour common to polycrystalline and single-crystalline samples of UFe_4Al_8 prepared in different laboratories.

In order to obtain more complete knowledge of the metastable magnetic properties, we have systemically measured the ac susceptibility $\chi_{ac}(T)$ at various frequencies, the dc magnetization $M(T)$ in various magnetic fields, the magnetic relaxation $M(t)$ at different temperature, the high-field magnetization $M(H)$ at 4.2 K up to 230 kOe, and the specific

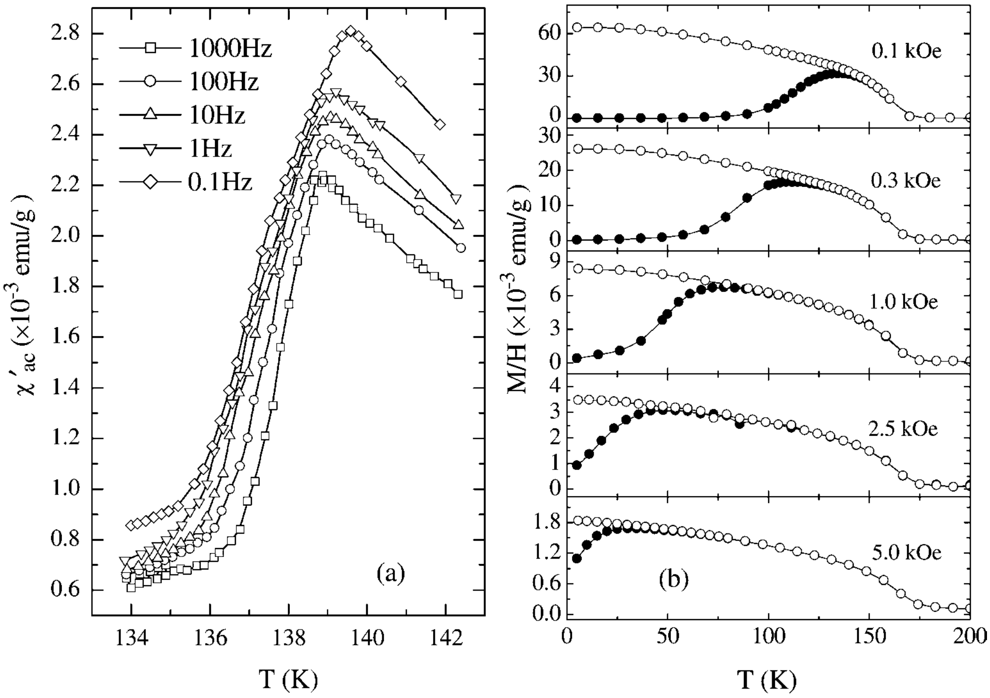

Figure 1. (a) The real component χ'_{ac} of the ac susceptibility of UFe_4Al_8 versus temperature between 134 and 143 K at frequencies $0.1 \leqslant \omega/2\pi \leqslant 1000$ Hz in an applied ac field of 1 Oe. (b) Magnetization data M/H versus temperature for UFe_4Al_8 measured in the FC (○) mode and in the ZFC (●) mode in various magnetic fields.

heat $C(T)$ up to 220 K for a well annealed polycrystalline UFe_4Al_8 sample. The sample was prepared by arc melting the starting elements (U: 3N; Pt: 4N; Al: 5N) in an argon atmosphere, and was annealed at 900 °C for 23 days in high vacuum. Low-field magnetization, ac susceptibility, and magnetic relaxation measurements were carried out using a SQUID magnetometer. The high-field magnetization up to 230 kOe was measured in a steady magnetic field. The adiabatic heat pulse method was employed for the specific heat measurement.

2. Results and discussion

Many rare-earth or uranium compounds show irreversible magnetic behaviours and long-time relaxation effects. These phenomena are usually interpreted as reflecting either domain wall pinning effects in highly anisotropic materials or the formation of spin-glass states. To clarify the existence of above two effects in UFe_4Al_8, the complex susceptibility was studied in an ac field of 1 G at several frequencies ranging from 0.1 to 1000 Hz. The results for the real component χ'_{ac} are displayed in figure 1(a). It is clear from this figure that χ'_{ac} shows a pronounced maximum around T_C, and T_C is evidently dependent on the frequency, ω, in the range between 0.1 and 1000 Hz. With increasing ω, we observed a downshift of T_C contrary to the behaviour observed for a usual spin glass, which shows an upshift of the spin-freezing temperature T_f with increasing ω [10]. This suggests that the pronounced maximum in $\chi'_{ac}(T)$ around T_C originates from a ferromagnetic phase transition. For convenience, in this paper the position of the peak in χ'_{ac} at $\omega/2\pi = 0.1$ Hz (the lowest frequency of the measurement) is defined as the Curie temperature T_C (=138 K).

The temperature variations of the field-cooled (FC) and zero-field-cooled (ZFC) dc magnetizations $M(T)$ of UFe_4Al_8, measured in magnetic fields between 0.1 and 5 kOe, are compared in figure 1(b). Thermomagnetic irreversibility is observed below a characteristic temperature $T_{ir}(H)$ which is strongly field dependent. The FC curve is stable, reversible, and has a tendency to approach a constant value at low temperatures. In contrast, metastable behaviour is observed for the ZFC curve, which shows a broad maximum at a temperature T_{max} near T_{ir} due to the long-range ferromagnetic ordering. As the field is increased, both T_{max} and T_{ir} shift to lower values, and a much broader magnetic phase transition and a larger difference between T_C and T_{max} can be observed in higher applied magnetic fields. It is known that systems with high magnetic anisotropy have narrow domain walls [11, 12]. The domain wall energy is different when its centre coincides with an atomic plane from when the centre is located between the two planes [13, 14]. The difference ΔE between the different domain wall energies hinders the wall motion, leading to domain wall pinning. On the basis of this model, the observed magnetic properties on UFe_4Al_8 can be understood as follows: in the ferromagnetically ordered state, two competing effects on the temperature dependence of the magnetization come into play. Firstly, the thermal fluctuation is reduced with decreasing T and thereby increases M_{ZFC}. Secondly, the movement of the domain walls also slows down, which results in decrease in M_{ZFC}. These two opposite processes could give rise to the observed peak in the M_{ZFC}-curve for UFe_4Al_8. The FC curves shown in figure 1(b) exhibit a tendency to saturate at low temperatures rather than a maximum, indicating the absence of domain wall effects. This means that at T_C, even a small field could cause domain wall movements.

Figure 2(a) shows the magnetic field variations of the magnetization $M(H)$ recorded at different temperatures for UFe_4Al_8. The existence of high magnetic anisotropy is further confirmed by the distinct crossover phenomenon, with an $M(H)$ value that does not saturate up to $H = 230$ kOe at $T = 4.2$ K (see the inset of figure 2(b)). The low-temperature $M(H)$ curves show a small step in the low-magnetic-field range, which is much more pronounced at 2 K and shifts to the low-field side with increasing temperature. Such a behaviour appearing for ferromagnetic UFe_4Al_8 may be considered as a spin reorientation of the U or the Fe atoms, or a domain effect [2].

The temperature dependence of the specific heat, $C(T)$, of UFe_4Al_8 is illustrated in figure 2(b). $C(T)$ shows an anomaly around $T_C = 138$ K due to the ferromagnetic ordering in the compound. At low temperatures, the C/T versus T^2 plot (inset (i) of figure 2(b)) yields for $T \rightarrow 0$ K a γ-value of 97 mJ mol^{-1} K^{-2}. This value is evidently larger than that of a normal metal, and may be related to a certain random distribution on Fe and Al sublattices.

It is interesting to note that from the measured hysteresis loop (not shown here) the coercive field H_C of UFe_4Al_8 is determined as 3.7 kOe at 10 K. At the same temperature, we have also observed a long-time magnetic relaxation effect even when measuring with a magnetic field of 8 kOe as shown in inset (ii) of figure 2(b). In general, if the magnitude of the applied magnetic field for magnetic relaxation measurements is much larger than the coercivity H_C, the observed magnetic relaxation is usually attributed to spin-glass phenomena, although the relaxation behaviour is also typical of ferromagnetic materials [15]. Therefore, the present results seem to be surprising. Recent studies [16], however, indicate that a peak or maximum in the $M_{ZFC}(T)$ curve can be observed in a given temperature range for some ferromagnets when measured with magnetic fields below a critical value H_{cr}. Unlike the coercivity H_C, H_{cr} corresponds to the field below which the contribution to the initial magnetization is mainly from domain wall displacements, and can be obtained by extrapolating the linear region below and above the knee of the virgin $M(H)$ curve [16]. This means that the irreversible magnetic behaviour and long-time magnetic relaxation effect could also be observed in these compounds when measured with a field less than H_{cr}. H_{cr} for UFe_4Al_8 is determined as 9 kOe

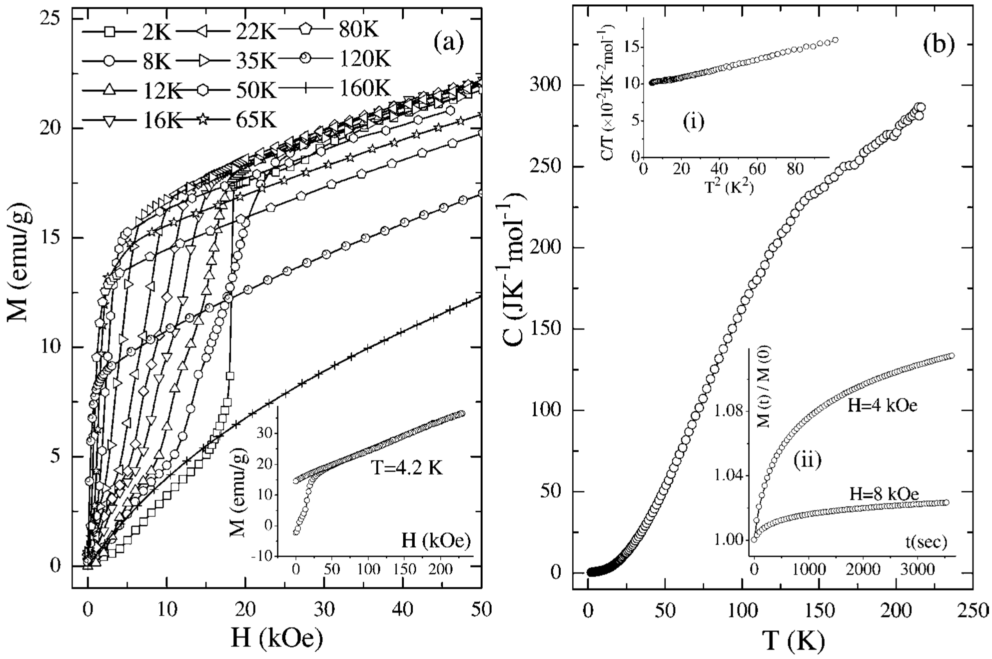

Figure 2. (a) Magnetizations of UFe_4Al_8 at different temperatures. The inset shows the high-field magnetization data up to 230 kOe at 4.2 K. (b) The temperature dependence of the specific heat of UFe_4Al_8 between 1.7 and 220 K. Inset (i) shows the plot of C/T versus T^2. Inset (ii) displays the magnetic relaxation behaviour measured at 10 K after ZFC the sample from 250 K.

at 10 K, and thus a magnetic relaxation measurement under much larger fields seems to be necessary.

3. Summary

We have investigated the magnetic properties of UFe_4Al_8 by means of ac susceptibility, dc magnetization, magnetic relaxation, and specific heat measurements. The large peak in the $\chi'_{ac}(T)$ curve and the downshift of the peak position T_C (=138 K at $\omega/2\pi = 0.1$ Hz) with increasing frequency, the sharp rise of the low-temperature magnetization $M(H)$ at low fields, the broad peak in the $M_{ZFC}(T)$ curve and evident difference between $M_{FC}(T)$ and $M_{ZFC}(T)$ below T_{ir}, the broad anomaly in the temperature-dependent specific heat $C(T)$ around T_C, and the long-time magnetic relaxation effects at low temperatures all indicate that UFe_4Al_8 is a typical ferromagnet with a large magnetic anisotropy and large coercive field. The metastable magnetic properties (time-dependent magnetic phenomena) could be interpreted as domain wall pinning effects. No spin-glass behaviour can be observed for our UFe_4Al_8 sample, suggesting that the contribution from the spin-glass state, if it exists at all, is extremely weak.

References

[1] Andreev A V *et al* 1992 *J. Alloys Compounds* **182** 55
[2] Godinho M *et al* 1995 *J. Magn. Magn. Mater.* **140–144** 1417
[3] Bonfait G *et al* 1996 *Phys. Rev.* B **53** R480
[4] Gal J *et al* 1990 *Phys. Rev.* B **42** 8507

[5] Waerenborgh J C *et al* 1999 *Phys. Rev.* B **60** 4074
[6] Dobrzynki L *et al* 1996 *J. Alloys Compounds* **236** 121
[7] Ptasiewicz-Bak *et al* 1988 *J. Magn. Magn. Mater.* **76/77** 439
[8] Schäffer W *et al* 1989 *J. Less-Common Met.* **149** 237
[9] Paixão J A *et al* 1997 *Phys. Rev.* B **55** 14370
[10] Mydosh J A 1993 *Spin Glass: an Experimental Introduction* (London: Taylor and Francis)
[11] Sampaio L C and da Cunha S F 1991 *J. Magn. Magn. Mater.* **99** 145
[12] Alves K M B *et al* 1994 *J. Alloys Compounds* **210** 325
[13] Van den Broeck J J and Zijlstra H 1971 *IEEE Trans.* **7** 226
[14] Rao T V C *et al* 1996 *Phil. Mag.* B **74** 275
[15] Majumdar S *et al* 1999 *J. Phys.: Condens. Matter* **11** L329 and references therein
[16] Joy P A and Date S K 2000 *J. Magn. Magn. Mater.* **218** 229 and references therein