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Size effect on magnetic ordering in Ce_3Al_{11}

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Abstract

To study the size dependence of magnetic ordering, magnetic measurements have been made between 1.8 and 300 K on Ce_3Al_{11} particles having an average particle size of 1400 Å. The nanoparticles were single phase as confirmed by X-ray diffraction. At low temperatures a ferromagnetic transition occurs at $T_C = 6.2$ K, which is the same as that for the bulk material. On the other hand, the antiferromagnetic transition at $T_N = 3.2$ K for the bulk material is not visible down to 1.8 K. Meanwhile, the slightly smaller Curie constant of nanoparticles as compared to that of the bulk indicates a certain degree of demagnetization of Ce ions when the particle size is sufficiently reduced.

Keywords: Nanoparticles; Ce_3Al_{11}; Magnetic order

1. Introduction

It has been well demonstrated that magnetic ordering in certain materials can be suppressed by reducing their sample size [1]. Experimental studies on (Ce, Al) compounds with different crystal structures even show a crossover from magnetic ordering to enhanced Kondo behavior when particle size was sufficiently reduced [2]. It would be of interest then to investigate the difference in size effects on ferromagnetic order and antiferromagnetic order, especially if both of them happen to occur in a same material such as Ce_3Al_{11} being described here.

Ce_3Al_{11} in its bulk form undergoes a ferromagnetic transition at $T_C = 6.2$ K, followed by an antiferromagnetic ordering at $T_N = 3.2$ K. The two magnetic ordering processes are associated with the two different Ce sites, Ce_I and Ce_{II}, in the lattice [3]. A preliminary study on fine particles of the compound having an average diameter of about 100 Å revealed no trace of any magnetic order. In this report, a specimen with an intermediate particle size was prepared and evaluated.

2. Experiments and analysis

Bulk Ce_3Al_{11} was first prepared by arc-melting in argon thoroughly mixed Ce (99.9%) and Al (99.9999%). Fine particles were then formed on a liquid-nitrogen cold trap by sputtering a bulk ingot in a 0.5 Torr of high purity Ar atmosphere. According to X-ray diffraction patterns in Fig. 1, no second phase was detected, except the particle size-induced broadening of diffraction peaks. Judging from the peak positions, there is little change in lattice constants. High resolution transmission electron microscopy (HRTEM) was further employed to directly observe the nearly spherical particles and their size distribution. As shown by a representative HRTEM image in the inset of Fig. 1, the average particle size can be estimated to be 1400 $\pm$500 Å, and each nanoparticle actually contains polycrystals with grain size of about 250 Å.

Magnetic measurements were performed in a quantum design SQUID magnetometer. Reduced-magnetization M/H data for bulk Ce_3Al_{11} at magnetic fields of 100–3500 G are shown in Fig. 2. The ferromagnetic and antiferromagnetic ordering temperature T_C and T_N are determined to be 6.2 and 3.2 K, respectively, in agreement with literature values. The antiferromagnetic transition was suppressed to lower temperatures by

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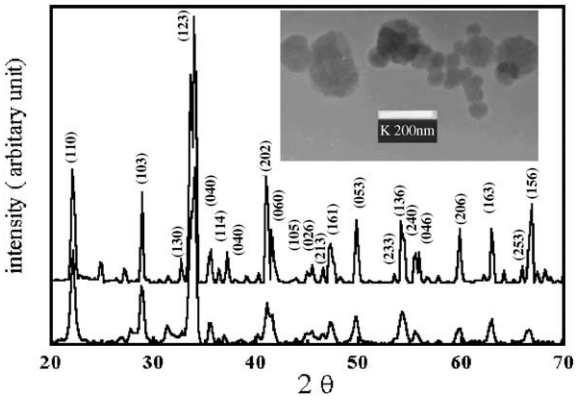

Fig. 1. X-ray Diffraction patterns of nanoparticle (bottom) and bulk (top) Ce_3Al_{11}. Inset: the HRTEM image of Ce_3Al_{11} nanoparticles.

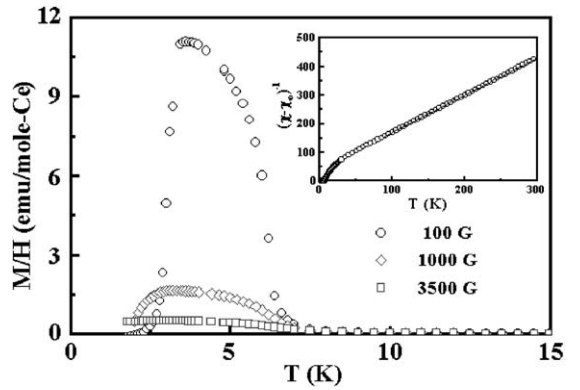

Fig. 2. ZFC reduced-magnetization of bulk Ce_3Al_{11} at various fields. Inset: $1/(\chi-\chi_0)$ at 1000 G fitted to a Curie–Weiss relation for $T > 100$ K.

increasing magnetic field, whereas the ferromagnetic transition remains at the same T_C. A similar result was also reported by Boucherle et al. [3] and Gavilano et al. [4]. Inset in Fig. 2 shows a Curie–Weiss fit above 100 K : $\chi(T) = \chi_0 + C/(T-\theta)$ with $C = 0.77$ emu K, $\theta = -32$ K and a constant $\chi_0 = -0.0047$ emu. The derived effective moment of $2.48\mu_B$ is approximately equal to the value of $2.54\mu_B$ for Ce^{3+}. The reduced-magnetization of nanoparticles exhibits very different behavior from the bulk. From field-cooling (FC) and zero-field-cooling (ZFC) only a ferromagnetic order can be identified (Fig. 3). The strong thermal irreversibility of magnetization is a characteristic of superparamagnetic behavior, with a blocking temperature T_B around 5.7 K. Based on the temperature of magnetization upturn and the higher value of T_C than T_B, the ferromagnetic transition appears to remain about the

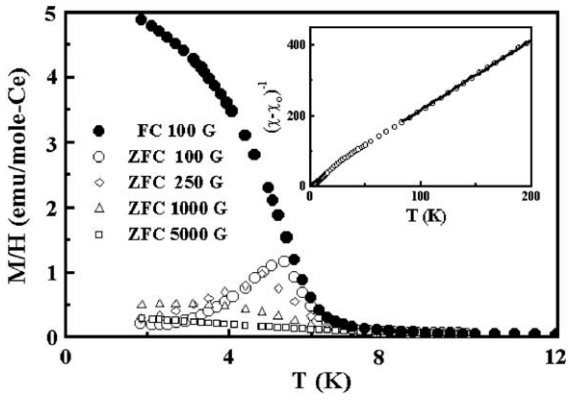

Fig. 3. ZFC and FC reduced-magnetization of nanoparticle Ce_3Al_{11} at various fields. Inset: $1/(\chi-\chi_0)$ at 5000 G fitted to a Curie–Weiss relation fot $T > 100$ K.

same as that for the bulk. The shift of peak position to lower temperature with increasing magnetic field is also a typical behavior of superparamagnetism. Most important, there is no further magnetic transition visible down to 1.8 K. This is quite different from the situation in the bulk material, which undergoes an antiferromagnetic transition at $T_N = 3.2$ K. The inset of Fig. 3 shows the Curie–Weiss fit above 100 K with $C = 0.54$ emu K, $\theta = -11$ K and a constant $\chi_0 = -0.0011$ emu. The slightly smaller Curie constant of nanoparticles is about 70% of the bulk, suggesting a certain degree of demagnization of Ce ions in nanoparticles.

Acknowledgements

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