Journal of Magnetism and Magnetic Materials 352 (2014) 56–60

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Unusual dilution effect of non-magnetic Al-substitution in intermetallic DyAg

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ARTICLE INFO

Article history:
Received 10 July 2013
Received in revised form
9 September 2013
Available online 12 October 2013

Keywords:
Intermetallic
Supercell
High magnetic field
Dilution effect

ABSTRACT

This paper introduces a supercell model to explain the field-induced multiple transitions and the anomalous dilution effect in the nonmagnetic-substituting intermetallic compounds $Dy_{50-x}Al_xAg_{50}$, in which the shift of the field-induced transitions in the Al-doped samples results from the change of the magnetic structure. At the doping level of $x=1.8$ (the ratio of Al/Dy approximates 1/26), an anomalous magnetic dilution effect was observed, in which the dilution effect becomes weak and the magnetic behavior is fairly similar to that of pure DyAg. We assumed that the anomalous dilution effect mainly results from the formation of uniform distribution of supercell in the doped $Dy_{48.2}Al_{1.8}Ag_{50}$.

1. Introduction

The rare-earth intermetallic compound DyAg was attractive due to its good ductility and magnetoelastic property at room temperature. Usually, the stress-induced phase transformation has great contribution to the ductility of the material. On the other hand, DyAg, which is an antiferromagnet with the CsCl-type cubic structure, shows complicated magnetic properties and has three obvious magnetic transitions at $T_N=56$ K, $T_1=49$ K and $T_2=46.5$ K [1,2]. Magnetization measurements of RAg and RCu (R=Dy, Tb and Gd) under pulsed high magnetic field up to 300 kOe show that DyAg has an AF1–AF2 transition between T_N and T_2 [3]. The susceptibility and magnetization have also been measured in the amorphous alloys $RE_{50}Ag_{50}$ (RE=Gd, Tb, Dy, Ho, and Er) [4]. The anisotropic multistep metamagnetic jumps of magnetization measured in DyAg single crystal samples along [100], [110] and [111] directions have been reported [5,6]. The interesting properties can be well explained using the theoretical model with effective quadrupole interactions. In a quadrupole arrangement, *f* electron charge's distribution which diagonalizes certain quadrupole moments orders spontaneously and spatially as the temperature is lowered [7]. According to the neutron diffraction and the magnetization measurement, some magnetic structure models of DyAg in high magnetic fields were proposed [8,9]. The temperature and magnetic field induced multiple magnetic transitions have also been observed in other rare earth intermetallic compounds $DyPt_2$ [10], $DyAg_2$ [11], and $Dy(Pt_{0.94}Ag_{0.06})_2$ [12]. The magnetic field dependence of the multi-step magnetization of the DyAg polycrystalline samples with different doping levels of Al was measured up to 300 kOe at a higher temperature region [13]. In this paper, we report the influence of Al-substitution on the magnetic structure and the magnetic properties of polycrystalline compounds $Dy_{50-x}Al_xAg_{50}$ ($x=0$, 0.3, 1.2 and 1.8) by X-ray diffraction and detailed susceptibility measurements, meanwhile, we expanded the measurements to the pulsed high magnetic field. Based on the experimental results, a supercell model was proposed to interpret the observed unusual dilution effect.

2. Experimental

Polycrystalline samples of $Dy_{50-x}Al_xAg_{50}$ ($x=0$, 0.3, 1.2 and 1.8) were synthesized by conventional arc-melting stoichiometric amounts of high purity Dy (99.99 wt%), Al (99.99 wt%) and Ag (99.99 wt%) in argon atmosphere, which were annealed at 1173 K. For better homogeneity the ingots were re-melted several times by flipping over each time. The room temperature X-ray diffraction (XRD) studies confirmed the composition of the obtained samples. The zero-field-cooled (ZFC) and field-cooled (FC) magnetization were measured as a function of temperature (2–300 K) by using a commercial superconducting quantum interference device magnetometer (SQUID-VSM Quantum Design, Inc.). The high field magnetization measurement was carried out at 4.2 K using a pulsed magnet of 500 kOe built in the Wuhan National High Magnetic Field Center.

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http://dx.doi.org/10.1016/j.jmmm.2013.10.007

3. Results and discussion

Fig. 1(a), (b), (c) and (d) shows the X-ray diffraction patterns of the intermetallic compounds $Dy_{50-x}Al_xAg_{50}$ with $x=0$, 0.3, 1.2 and 1.8, respectively. The lattice parameters calculated from the X-ray diffraction patterns are listed in Table 1. Fig. 1(e) presents the diffraction pattern of DyAg got from the standard PDF database. When we compared the patterns of the doped samples with pure DyAg shown in Fig. 1(a), a new peak around 37.5° was found in Fig. 1 (b)–(d), which may result from the non-magnetic Al^{3+} ions' substitution; when the doping level increases, the intensity of the new peak increases. On the other hand, the intensities of the peaks near 50° and 65° are directly related to the doping level, the change of the relative intensity of the three main peaks shows that non-magnetic ion substitution has some effect on the preferred orientation of the lattice.

The crystalline and magnetic structure of the ground state of pure DyAg proposed by Yoshii etc. [5,6] are presented in Fig. 2(a), in which, to be clear, only Dy^{3+} ion sites are shown. It is a typical structure with strong antiferroquadrupolar coupling interaction, accompanied with multistep metamagnetism which has non-collinear magnetic field induced phases [5]. Fig. 2(b) presents an assumed supercell consisting of 8 unit cells, in a low doping level, for a given Dy^{3+} ion site labeled with dark ball, there are three possible Al^{3+} ion substitution sites: the nearest-neighbor substitution site, the second-nearest-neighbor substitution one and the third-nearest-neighbor substitution one, which are labeled with arrows.

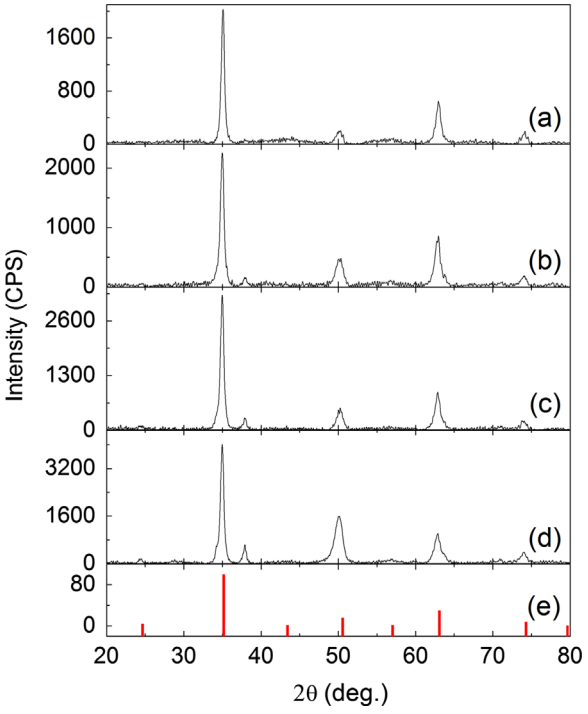

Fig. 1. X-ray diffraction results of the polycrystalline compounds $Dy_{50-x}Al_xAg_{50}$: (a) $x=0$, (b) $x=0.3$, (c) $x=1.2$ and (d) $x=1.8$; (e) the standard PDF pattern of DyAg.

Table 1
Structural parameters of polycrystalline samples $Dy_{50-x}Al_xAg_{50}$ ($x=0$, $x=0.3$, $x=1.2$ and $x=1.8$).

$Dy_{50-x}Al_xAg_{50}$	$x=0$	$x=0.3$	$x=1.2$	$x=1.8$
Space group	*Pm-3m*	*Pm-3m*	*Pm-3m*	*Pm-3m*
a (Å)	3.6133	3.6217	3.6204	3.6221
Volume (Å^3)	47.18	47.5	47.45	47.52

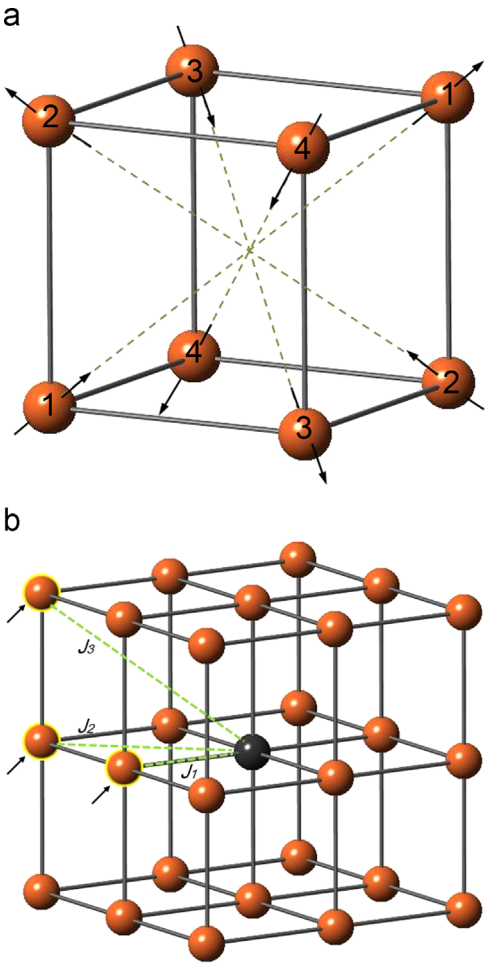

Fig. 2. (a) Ground state of the proposed magnetic structures of DyAg at 4.2 K. The arrows show the directions of magnetic moments. (b) A supercell of DyAg consisting of 8 unit cells. The arrows show three different substitution sites for the central dark ion: the nearest-neighbor substitution site, the second-nearest-neighbor substitution site and the third-nearest-neighbor substitution site.

Fig. 3(a) shows temperature dependence of magnetization (M–T) of $Dy_{50-x}Al_xAg_{50}$ ($x=0$, 0.3, 1.2 and 1.8) measured with both ZFC and FC modes in a 50 Oe magnetic field. Above 60 K, all the samples show a similar paramagnetic (PM) behavior with slightly difference in the magnetic moment. Below 60 K, the M–T curves are dependent on the Al doping level and temperature; the experimental results are shown in the inset of Fig. 3(a). Fig. 3(b) shows the temperature dependence of DC susceptibility (magnetization over field) measured at 50 Oe, an obvious PM behavior is shown above 60 K and a deviation from the PM behavior is observed below 60 K.

In order to analysis the low temperature magnetic behaviors, the temperature dependence of dM/dT was calculated, in which the transition temperature is defined by a peak or a zero-crossing point as presented in Fig.4. The dotted arrows indicate the temperature T_N, where dM/dT equals zero [6]. The gray arrows indicate the temperature T_3 where dM/dT returns to zero from the positive value, indicating the transition from antiferromagnetic (AFM) ordering to ferromagnetic (FM) ordering. Below T_N, the AFM ordering dominates when dM/dT is positive, and the FM ordering dominates when dM/dT is negative. For pure DyAg, as shown in Fig.4, three kinks were observed at T_N, T_1 and T_2, which show a little difference than the value reported previously [1,2]. Two more transitions at T_3 and T_4 were observed in the lower temperature range (about ~17 K and ~11 K respectively), indicating the