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Magnetic hardening of high-energy ball-milled nanocrystalline $LaMn_2Si_2$

Ayhan Elmali[a,*], Simsek Tekerek[a], Ilker Dincer[a], Yalcin Elerman[a], Ralf Theissmann[b,c], Helmut Ehrenberg[b,d], Hartmut Fuess[b]

[a]*Department of Physics Engineering, Faculty of Engineering, Ankara University, 06100 Besevler-Ankara, Turkey*
[b]*Institute for Materials Science, Darmstadt Technology University, Petersenstr. 23, 64287 Darmstadt, Germany*
[c]*Institute for NanoTechnology, Forschungszentrum Karlsruhe, Germany*
[d]*Institute for Solid State and Materials Research, Institute for Complex Materials, Helmholtzstr. 20, D-01069 Dresden, Germany*

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Abstract

Nanocrystalline $LaMn_2Si_2$ powders have been obtained by high-energy ball milling for 30 min from bulk alloys. After milling a high coercivity about 6 kOe is observed at 10 K in contrast to neglectable coercivity for the bulk $LaMn_2Si_2$ at 5 K. The average grain size of the optimum particles which is obtained from X-ray diffraction pattern and HRTEM picture is about 20 nm. The magnetic hardening is observed for the nanocrystalline $LaMn_2Si_2$, reflected in the coercivity field strength of 6 kOe at 10 K.

Keywords: Intermetallic phase; Nanocrystalline material; Magnetic property; Coercivity

1. Introduction

Interest in the rare-earth intermetallics RT_2X_2 (R: rare-earth, T: transition metal, X: Si or Ge) and related compounds has continued unabated in recent years [1,2]. These compounds have a body-centered tetragonal $ThCr_2Si_2$-type crystal structure with space group I4/mmm. In this crystal structure, R, T and X atoms are stacked in layers along the *c*-axis in the R–X–T–X–R sequence [3,4]. The RMn_2X_2 compounds form a distinct group since ferromagnetic and antiferromagnetic Mn sublattice ordering is observed in these compounds unlike in other transition metals [5–7]. The magnetic structure of these compounds contains two interacting magnetic sub-systems. One of them is composed of rare-earth atoms and manganese atoms form the other. The R sublattice orders magnetically at low temperatures (below ~100 K). According to the earlier results of the investigations, the interlayer Mn–Mn exchange is very sensitive to the lattice parameter *a*, leading to ferromagnetic and antiferromagnetic ordering of the Mn sublattice, depending on the specific R element and temperature [7–13].

In $LaMn_2Si_2$, the Mn moments are coupled antiferromagnetically with neighbored Mn moments in the (0 0 1) plane [12]. The Néel temperature of this antiferromagnetic arrangement has been found to be $T_N^{intra} \approx 470\,K$. It has been shown that below $T_C^{intra} \approx 310\,K$, a canted ferromagnetic spin structure of Mn moments with an antiferromagnetic component within *ab*-planes takes place. Cooling to temperature below 45 K leads to a helical magnetic arrangement in which conical and canted magnetic structures coexist down to 2 K [12,14–17].

Recently, nanoscaled permanent magnets have attracted considerable attention because of their super hard magnetic properties [18]. High coercivity is expected to develop in magnetic materials with a nanoscale grain size, because it goes through a maximum at the single-domain size considering the size dependence of coercivity. High-energy ball milling is powerful technique to develop fine-grained particles of intermetallic compounds and has been successfully used to prepare various permanent magnets including

*Corresponding author. Fax: +90 312 2127343.
E-mail address: elmali@eng.ankara.edu.tr (A. Elmali).

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Sm–Co, Nd–Fe–B and Sm–Fe–N(C) magnets [18–20]. In this article, we report on magnetic hardening of $LaMn_2Si_2$ by nanoscaling.

2. Experimental

Bulk $LaMn_2Si_2$ was prepared by arc melting of the constituent materials La, Mn and Si (with a purity of at least 99.9%) under highly purified argon atmosphere. A 3% excess of Mn was added to compensate for the empirically estimated loss during the arc-melting process. The specimens were melted sequentially three times for better homogeneity and in every step were weight controlled. The as-cast bulk $LaMn_2Si_2$ was crushed and then high-energy ball milled for 30 min in the hardened steel vial under purified argon atmosphere in a high-energy ball-milling system D-SPEX 8000 M. The diameter of the steel ball is about 12 mm and the weight ratio of ball to powder is about 15:1. The crystal structure was identified by X-ray diffraction with CuKα radiation (Rigaku D-Max 2200). The microstructure was studied by high-resolution transmission electron microscopy (HRTEM). The as-milled powders for TEM characterization were immersed into alcohol and dispersed using an ultrasonic vibrator for about 45 min. Holey carbon grids were then used to hold the dispersed powders and the powders were examined using a JEOL 3010 UHR transmission electron microscope operated at 300 kV. Magnetization data were measured using a SQUID magnetometer (Quantum Design) in the temperature range 5–350 K in an external magnetic field of 50 Oe. The measurements were performed in a zero-field-cooled (ZFC) and field-cooled (FC) mode. The magnetic hysteresis loops were measured at different temperatures from 10 to 300 K.

3. Results and discussion

The X-ray diffraction pattern obtained at room temperature revealed the characteristic reflections of a tetragonal body-centered $ThCr_2Si_2$-type structure with space group I4/mmm. The refined unit cell parameters for bulk and nanocrystalline specimens are given in Table 1. Fig. 1 shows the Rietveld refinements of the X-ray diffraction patterns of as-cast and as-milled $LaMn_2Si_2$ powders. The X-ray diffraction pattern of the milled powders indicates that these powders consist of 1:2:2-type phase. This structure is preserved for nanocrystalline grains as seen from a high-resolution transmission electron micrograph (HRTEM) in Fig. 2. The reflections from milled $LaMn_2Si_2$ appear broadened, indicating a significant decrease in the grain size due to the high-energy mechanical collision

Table 1
Refined unit cell parameters for bulk and nanocrystalline specimens

	a (Å)	c (Å)	c/a	V (Å^3)
Bulk $LaMn_2Si_2$	4.108	10.596	2.580	178.806
Nanocrystalline $LaMn_2Si_2$	4.103	10.601	2.583	178.497

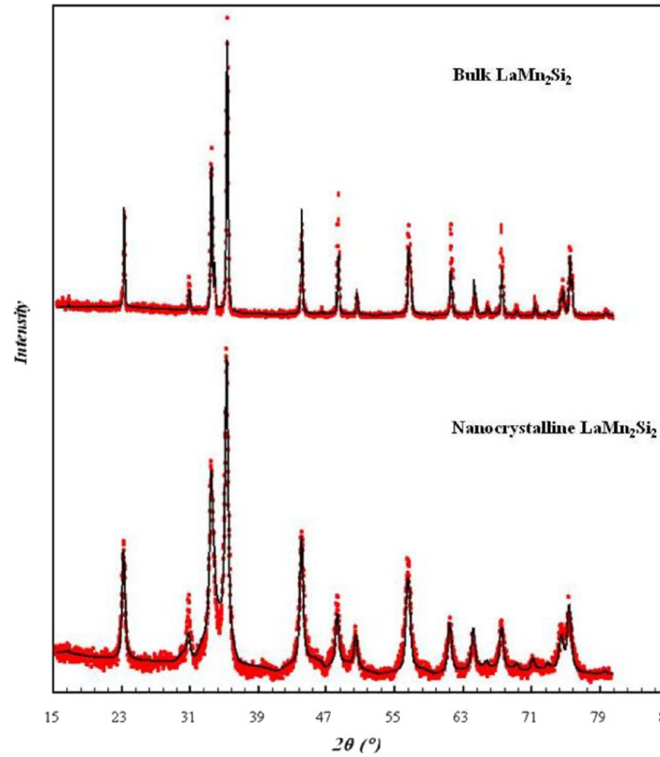

Fig. 1. The Rietveld refinement patterns of the bulk and nanocrystalline $LaMn_2Si_2$.

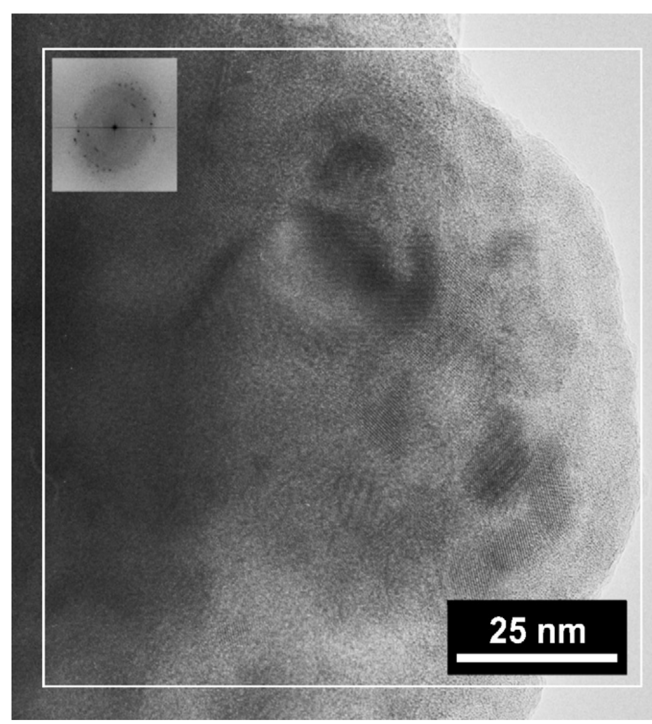

Fig. 2. HRTEM image of the nanocrystalline $LaMn_2Si_2$.