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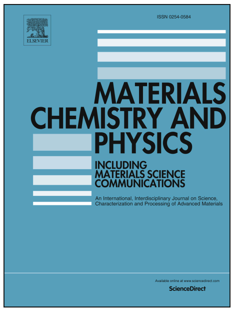

Phase composition (Reviewer 1) and growth mechanisms of half-metal Heusler alloy produced by Pulsed Laser Deposition: From core-shell nanoparticles to amorphous randomic (Reviewer 2) clusters

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Phase composition (Reviewer 1) and growth mechanisms of half-metal Heusler alloy produced by Pulsed Laser Deposition: from core-shell nanoparticles to amorphous randomic (Reviewer 2) clusters

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(Dated: April 21, 2017)

We report on a systematic study of the experimental parameters to control size and morphology of half-metals Heusler nanoparticles synthesized by pulsed laser deposition (PLD) technique. Our findings suggest the existence of a threshold across which the mechanism of particle growth changes: core-shell NPs for higher values of laser energy and randomic (Reviewer 2) amorphous structure for lower values of laser energy. The growth mechanisms are discussed in deep based on the high resolution transmission electron microscopy (HR-TEM) results; and a phase diagram was developed to summarize our findings.

Keywords: Pulsed Laser Deposition-PLD, Heusler alloys, nanoparticles, half-metal, spintronics

I. INTRODUCTION

Half-metal (HM) materials have attracted much interest as promising materials to enhance several kinds of spin-dependent phenomena, since they generate a highly spin-polarized current due to an energy band gap in one spin-channel at the Fermi level (E_F) [1, 2]. This property makes these materials very promising for applications on spin-injection and spin-manipulation in spintronic devices [3]. Therefore, the exploration of half-metallic materials with as high spin-polarization as possible at room temperature is critical for developing practical devices with 'spin-mediated functionalities' [4]. In particular, Si-containing full-Heusler alloys are promising materials for spin injector/detector of Si-based spin devices, such as metal-oxide-semiconductor field-effect transistors [5].

Full Heusler alloys are of the form X_2YZ, where X and Y are *d*-block elements and Z is a *p*-block element [6]. In the ordered $L2_1$-phase, it has a cubic structure with X occupying a simple-cubic sub-lattice (or, equivalently, two fcc sub-lattices), and Y and Z occupying two distinct interlocking fcc cubic sub-lattices [7]. Generally, the Heusler structure can be looked on as four interpenetrating face-centred-cubic (fcc) lattices, in which there are four crystal sites represented as A(0,0,0), B(1/4, 1/4, 1/4), C(1/2, 1/2, 1/2) and D(3/4, 3/4, 3/4) in Wyckoff coordinates [8]. In addition, A and C sites are similar in chemical surroundings and usually created as equilibrium. Conventionally, (A, C) and B sites are occupied by X and Y atoms, respectively, while D site is occupied by Z atoms. A fully ordered atomic arrangement in full-Heulser alloys is the $L2_1$ structure, however, the partially disordered B_2 and fully disordered A_2 structures also exist[9, 10]; and this atomic disorder induces significant effects on the physical properties of the material, in particular on the magnetization and half-metallicity [11].

Pulsed Laser Deposition (PLD) is a method to produce metal nanoparticles (NPs) free from products of chemical reactions [12]. NPs are formed at relatively low gas pressure (0.2-10 Torr) of inert or reactive gas. During expansion and cooling, condensation starts within the ablated vapour and the condensed particles undergo multiple collisions with ambient gas molecules, leading to the stabilization of the nanoclusters before they arrive to the substrate surface [13]. On the other hand, the magnetic properties of a system of NPs are strongly dependent on the particle size distribution, crystalline structure, shape, and the spatial arrangement of the particles [14]. Many basic phenomena observed in such systems are related to the interaction among NPs, such as the displacement of the blocking temperature and spin glasslike properties, for example [15].

The present paper deals with the optimization of PLD parameters to obtain nanoparticles of Heulser alloys to spintronic devices. To this purpose, the material must have the highest magnetic saturation moment and Curie temperature as possible; as well as follow the Slater-Pauling rule and, in addition, be ferromagnetic. Fe_2MnSi has a low Curie temperature (220 K [16]), fails to follow the Slater-Pauling rule [16] and, in addition, has an antiferromagnetic arrangement of Mn ions [7, 9]. Due to these reasons (Reviewer 1), this material is a good choice, since it is possible to verify if the nano-topology will improve those non-optimized physical properties.

In brief, we found two different NPs morphology, depending on the laser energy: a core-shell pattern, with an atomically disordered/ordered shell/core, for higher values of laser energy; and a fractal agglomerate of nanoparticles atomically disordered, for lower values of laser energy. We also found a limiting threshold containing both morphologies and all these findings are summarized on the proposed phase diagram. The growth mechanism and compositional analysis of the nanoparticles are also discussed in deep. These results are able to guide further

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