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Exploring the effects of dimensionality on the magnetic properties of intermetallic nanowires

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ABSTRACT

Correlated electron intermetallic bulk systems exhibit exciting phenomena, such as unconventional superconductivity, heavy fermion behavior, magnetic ordering, and quantum criticality. However, such exciting properties in related systems with reduced dimensionality are rather unexplored and unpredictable. In this work, we explore the routes for synthesizing nanowires of the intermetallic antiferromagnet compound $GdIn_3$ by an innovative method: the metallic-flux nanonucleation (MFNN). This technique allows the simultaneous synthesis of bulk $GdIn_3$ single crystals ($T_N^{3D} = 45$ K) and their low-dimensional (LD) analogs, which nucleate with diameter $d \approx 200$ nm and length $l \approx 30$ μm inside pores of an Al_2O_3 template. Both systems were studied by means of Energy Dispersive Spectroscopy (EDS), magnetic susceptibility, heat capacity and electron spin resonance (ESR) measurements. Interestingly, the metallic nanowires show a drastic suppression of the antiferromagnetic ordering to $T_N^{LD} = 4$ K. These observations suggest the presence of LD magnetic frustration in this compound and possibly open a new route to explore the role of low-dimensionality in strongly correlated materials.

1. Introduction

In general, interacting many-body systems obey the symmetry properties of periodic lattices when the particles are confined within a crystalline solid [1]. The microscopic description of these systems relies on the existence of translational symmetry in three dimensions, which allows Bloch's theorem to apply, leading to the formation of valence/conduction bands. Furthermore, it is desirable to understand the properties of interacting condensed-matter systems by unveiling spontaneous symmetry breaking, such as magnetic and crystalline orderings, superfluidity and superconductivity; elementary excitations, such as quasiparticles in heavy fermion systems; collective modes such as plasmons; and phase transitions [2]. In all cases, the system dimensionality and the disruption of translation symmetry, when one of the solid dimensions becomes comparable to the important length scale of the problem, plays a fundamental and unpredictable role in determining the system ground state. Moreover, in reduced spatial dimensions, many-body correlation effects due to the Coulomb interaction between electrons tend to become more relevant.

In particular, the series $R_mM_nIn_{3m+2n}$ ($R=$ rare-earth, $M=$ Co, Rh, Ir; $n=0$, 1; $m=1$, 2) of intermetallic compounds would be a fantastic system to study in low dimensions since they have several remarkable physical properties such as complex magnetic ordering, Ruderman–Kittel–Kasuya–Yoshida (RKKY) magnetic interaction, crystalline electrical field (CEF), Fermi surface (FS) effects and, for R=Ce, non-Fermi-liquid behavior, quantum criticality (QC) and the interplay between antiferromagnetism and unconventional superconductivity (USC) [3–5]. This variety of interesting physical properties in structurally related series represents a great opportunity to explore systematically the role of the each interaction in determining the system properties, specially in favoring USC in many Ce-based members of these series. As the properties of the heavy-fermion superconductors in their family are presumably magnetically mediated, the study of non-Kondo isostructural $R_mM_nIn_{3m+2n}$ ($R=$ Nd, Gd, Tb) magnetic materials has been used to elucidate the role of the RKKY interactions and CEF effects in the evolution of the magnetic properties [6–10].

The cubic intermetallic compound $GdIn_3$ [11] is a promising candidate to start a new route to study LD systems since it allows one to individually investigate the dimensionality effects on the

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RKKY magnetic interaction, i.e. the intersite exchange interaction mediated by the conduction electrons (*ce*). In fact, it has been already shown by resonant X-ray magnetic diffraction that the $GdIn_3$ antiferromagnetic (AFM) ordering temperature (T_N) has a dependence with the X-ray penetration depth, displaying a larger T_N in the surface as compared to the bulk [12]. By making use of macroscopic measurements, such as magnetic susceptibility and heat capacity, one can study the evolution of the bulk AFM order at $T_N^{3D}=45$ K with the dimensionality. Additionally, electron spin resonance (ESR) is a highly sensitive microscopic technique that has been used to investigate spin fluctuations and magnetic interactions in such compounds. In particular, CEF effect is a higher order effect in the Gd^{3+} S-state ($S=7/2$, $L=0$) ground state. As such, Gd ions are excellent ESR probes to study magnetic properties which purely reflect the details of RKKY magnetic interaction and FS effects in intermetallic magnetic materials. Thus, ESR experiments can reveal details about the microscopic interaction J_{fs} between the 4*f* electrons and the *ce*.

However, the growth of intermetallic nanowires containing a rare-earth element has been challenging [13–18]. In the present work we have successfully synthesized Gd-In nanowires close to 1:3 ratio by an innovative method called metallic-flux nanonucleation (MFNN). Our results show a drastic suppression of the antiferromagnetic transition from the bulk ($T_N^{3D}=45$ K) to the nanowire system ($T_N^{LD}=3.8$ K) which we speculated to be due to a change in the magnetic RKKY exchange interaction. These observations indicate the presence of magnetic frustration driven by low-dimensionality in this compound and may open a new field for the research of the role of low-dimensionality in strongly correlated materials.

2. Experiment

Intermetallic nanowires with 1Gd:3In stoichiometry were successfully grown by the metallic flux nanonucleation (MFNM) method. This innovative method is based on the conventional flux-growth technique [19] performed in a nanometric template that mediates the preferential nucleation of the single crystals in the desired geometry [20]. Particularly, in this work we have used Al_2O_3 membranes fabricated via hard anodization process, described in detail in Ref. [21]. The difference between MFNN method and the classical flux-growth technique is the presence of this anodized Al_2O_3 membrane fixed in the base of an alumina crucible enclosing the involved metals. The metals were weighted in the ratio 1 Gd to 10 In. The crucible containing the elements and the membrane was covered with quartz wool and sealed inside an evacuated quartz tube. The tube was placed in a furnace and

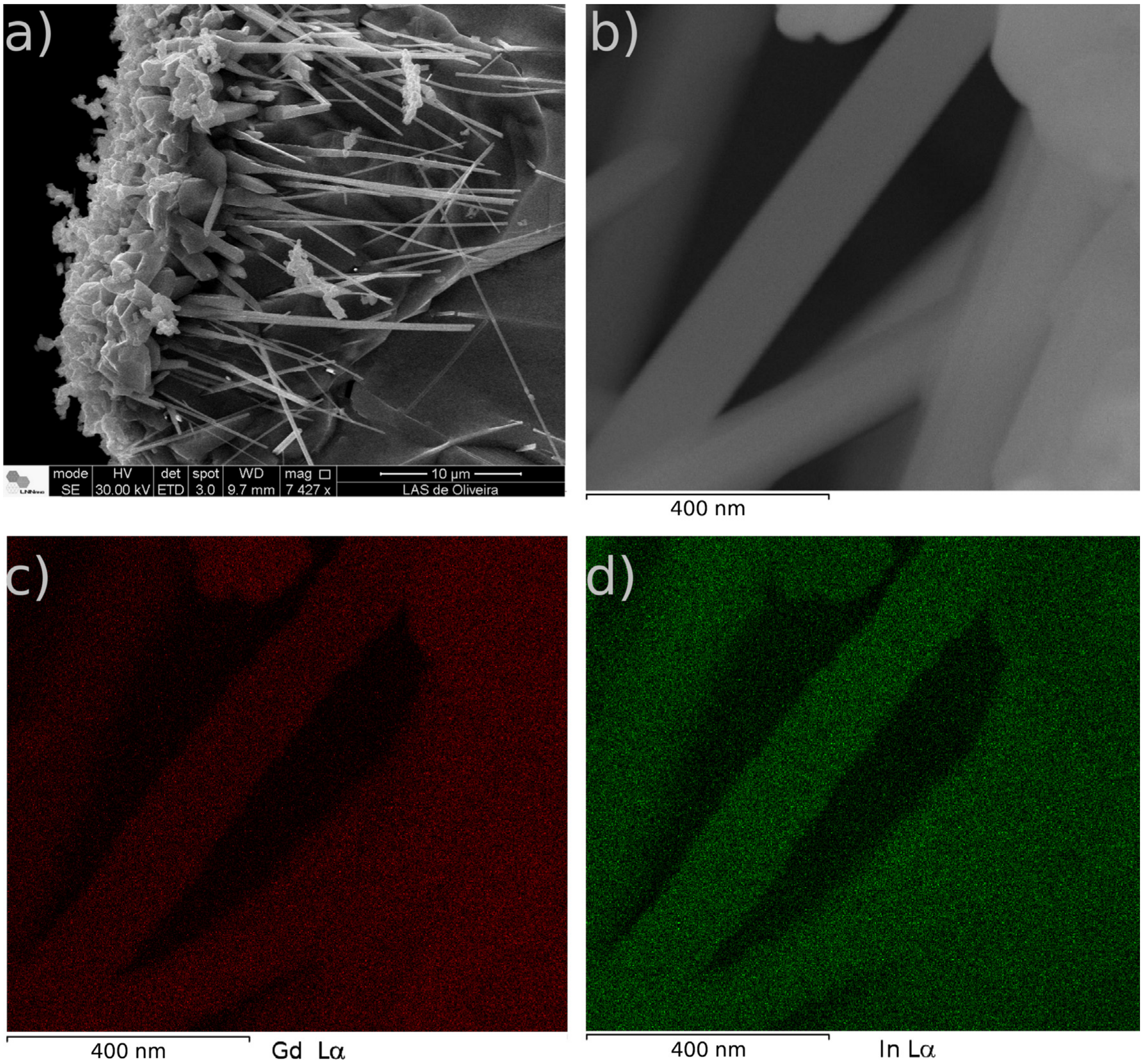

Fig. 1. (Color online) Scanning electron microscope (FE-SEM) image and Energy Dispersive X-Ray Spectroscopy (EDS) mapping of $GdIn_3$ nanowires. (a) FE-SEM image of $GdIn_3$ nanowires grown by the innovative MFNN method, (b) SEM image of a $GdIn_3$ nanowire, and EDS composition mapping for (c) Gd Lα and (d) In Lα.