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Research articles

Anomalous softening of magnon modes in the reentrant state in $Cr_{70}Fe_{30}$ thin films

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ABSTRACT

In the reentrant (RE) state at low temperatures, the functional dependence of magnetization on temperature at fixed magnetic fields, $M_H(T)$, in the $Cr_{70}Fe_{30}$ thin films deviates considerably from the well-known Bloch power law, predicted by the spin-wave (SW) theory for a conventional three-dimensional (3D) ferromagnet. Strong departures from the SW predictions are shown to be a manifestation of an anomalous softening of magnon modes in the RE regime. The model that invokes the Bose–Einstein condensation (BEC) of magnons provides a satisfactory explanation for this unusual behavior. The magnon BEC transition temperature, $T_c(H)$, the volume, $V(H)$, over which the condensate wavefunction retains its phase coherence, the chemical potential, $\mu(T, H)$, and the average number of magnon condensates in the ground state, $\langle n_0(T, H)\rangle$, are accurately determined from $M_H(T)$ using a self-consistent approach. Reduction in the film thickness from 978 nm to 21 nm enhances the BEC transition temperature at zero field, $T_c(H = 0)$, from 0.1 K to 0.33 K but drastically reduces the phase coherence length of the BE condensate wavefunction and the spin-wave stiffness at $T = 0$ and $H = 0$. The variation of T_c with magnetic field has the form that is characteristic of the magnon BEC in a 3D spin system. Regardless of the film thickness, the BE condensate fraction at $H = 0$, $\langle n_0(H = 0)\rangle$, turns out to be zero at $T \simeq 2\ K \gg T_c(H = 0)$, as expected. The 'zero-field' quantities $M_0(H = 0)$, $D_0(H = 0)$, $V(H = 0)$ and $T_c(H = 0)$ are found to vary with the film thickness as $M_0(H = 0) \sim t^{3/4}$, $D_0(H = 0) \sim t^{-3/4}$, $V(H = 0) \sim t^{2/5}$ and $T_c(H = 0) \sim t^{-1/3}$. These power laws assert that the film thickness is the fundamental length scale so far as the magnon BEC phenomenon in the $Cr_{70}Fe_{30}$ thin films is concerned.

1. Introduction

According to the spin-wave (SW) theory, thermally-excited spin-wave (collective) excitations cause the $T^{3/2}$ power law decay of magnetization in a three-dimensional (3D) conventional ferromagnet. Spontaneous as well as 'in-field' magnetizations routinely follow this SW power law behavior at low temperatures in ferromagnets with homogeneous long-range magnetic order. Nevertheless, there exist numerous ferromagnetic systems in which strong departures from the $T^{3/2}$ power law have been observed particularly at low temperatures. These systems include: amorphous $(T_xNi_{1-x})_{80}B_{16}Si_4$ ($T = Fe, Co$) alloys [1], polycrystalline Ni_xAl_{100-x} (74 at.% $\leqslant x \leqslant$ 76 at.%) alloys [2], elongated Fe nanoparticles [3], ferrite nanoparticles [4], Co-Pt nanopillars [5], nanograin Fe Powder [5], and bulk nano-crystalline Gd [6–8]. The deviations from the $T^{3/2}$ law have found different interpretations in different systems: (i) magnon-fracton crossover in $[(Fe/Co)_xNi_{1-x}]_{80}B_{16}Si_4$ [1] and Ni_xAl_{100-x} [2], (ii) particle shape-dependent modifications in the spin-wave spectrum [3] in elongated Fe nanoparticles, (iii) discrete, quantized spin-wave energy spectrum brought about by the reduced particle size in confined systems [4,9] such as ferrite nanoparticles [4], and (iv) Bose–Einstein condensation of magnons in Co-Pt nanopillars [5], nanograin Fe powder [5] and nanocrystalline Gd [6–8].

In this paper, we demonstrate that, out of the models (i)-(iv) mentioned above, only the magnon Bose–Einstein condensation (BEC) model provides an adequate description of the observed thermal decline in magnetization in the reentrant state at low temperatures in $Cr_{70}Fe_{30}$ thin films.

2. Results and discussion

Before proceeding on with the presentation, and subsequent discussion, of the results, we briefly summarize the structural/microstructural findings, based on the previously reported [10–12] X-ray reflectivity (XRR), grazing incident-angle X-ray diffraction (GIXRD) and atomic force microscopy (AFM) data on the $Cr_{70}Fe_{30}$ thin films with

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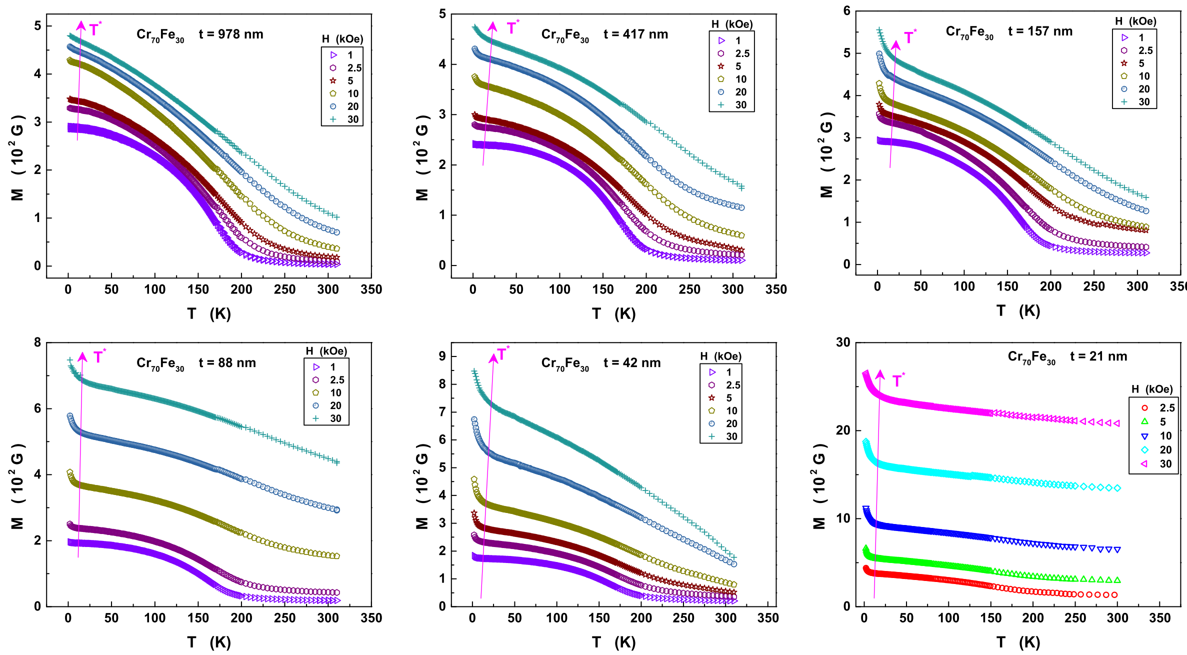

Fig. 1. Thermal decline of magnetization, $M(T)$, at fixed magnetic fields for the $Cr_{70}Fe_{30}$ thin films of thickness t = 978 nm, 417 nm, 157 nm, 88 nm, 42 nm and 21 nm. The upward arrows serve to highlight the field-induced shift in the temperature T^*, below which an upturn in $M(T)$ is observed, to higher temperatures.

thickness t ranging from 11 nm to 978 nm. Excellent structural quality of the films has been inferred from well-defined Kiessig fringes over the incident angle, 2θ, range 1° to 7° in the XRR spectra of even the thinnest film with $t = 11$ nm as well as from the sharp (110) (the most intense peak), (200), (211), (220) and (310) Bragg peaks in the XRD patterns of the films with $t = 978$ nm and 417 nm. For both these films, the Scherrer formula yields the average crystallite size (d) as 20(1) nm. AFM pictures reveal a columnar grain growth along the film normal in all the films. For details, the reader is referred to the reference [10].

Thermal decline of magnetization, $M(T)$, was measured in the 'field-cooled' mode at 1 K intervals over the temperature range $2\,K \leqslant T \leqslant 300\,K$ at fixed magnetic fields, H, ranging between 1 kOe and 70 kOe on well-characterized [10–12] crystalline $Cr_{70}Fe_{30}$ thin films. To facilitate an enlarged view of the variation of magnetization with temperature, only the $M(T)$ curves taken in the field range 1–30 kOe are shown in Fig. 1. Instead of a concave downward curvature of $M(T)$, normally observed in a ferromagnet, an upturn in $M(T)$ is apparent at low temperatures ($T \lesssim 25$ K), where these thin-film samples are in the reentrant state [10,13]. This upturn becomes more prominent as the film thickness decreases but with increasing field strength, it gets progressively smeared out and displaced to higher temperatures. In a conventional three-dimensional (3D) ferromagnet, thermally-excited spin-wave (SW) excitations give rise to the $T^{3/2}$ power law decay in magnetization at low temperatures. The M versus $T^{3/2}$ plots, shown in Fig. 2, serve to highlight the *strong departures* from the SW $T^{3/2}$ variation of magnetization at $T \lesssim 30$ K. As a first step towards understanding this unusual behavior, the temperature dependence of the spin wave (SW) stiffness, D, is obtained by adopting the procedure [14] (described below) that makes use of the SW relation [15–20]

$$M(T, H) = M(0, H) - g\mu_B\, Z\left(\frac{3}{2}, t_H\right)\left[\frac{k_B T}{4\pi D(T)}\right]^{3/2} \quad (1)$$

where the Bose-Einstein integral function $Z\left(\frac{3}{2}, t_H\right) = \sum_{n=1}^{\infty} n^{-\frac{3}{2}} exp(-n t_H)$ with $t_H = T_g/T = \Delta/k_B T$, $\Delta = \Delta_0 + g\mu_B H$ is the gap introduced in the spin-wave spectrum by the dipole–dipole interactions and/or magnetocrystalline anisotropy (Δ_0) and by the external magnetic field ($g\mu_B H$); $g = 2.1$ is the Landé splitting factor while the thermal 'renormalization' of D, $D(T)$, is of the form [21]

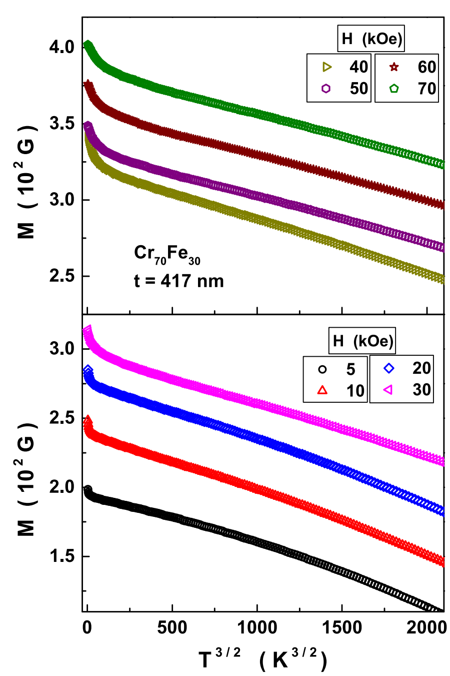

Fig. 2. Magnetization versus plots at different but fixed fields for $Cr_{70}Fe_{30}$ thin film with thickness t = 417 nm. Magnetization as a function of temperature is found to exhibit strong departures from the Bloch power law (predicted by the spin-wave theory) at low temperatures.

$$D(T) = D(0)\,[1 - D_2\, T^2 - D_{5/2}\, T^{5/2}] \quad (2)$$

where $D(0)$ is the spin-wave stiffness at $T = 0$ K and the $T^{5/2}$ (T^2) term arises from the *direct* (*indirect*) magnon-magnon interactions (mediated by the conduction-electron spins).

To begin with, an optimum fit to the $M(T)$ data, taken at a given field over a temperature range as narrow as 4 K (e.g., $2\,K \leqslant T \leqslant 6\,K$), is attempted based on the modified form of Eq. (1) in which the spin-wave stiffness, D, is treated as a constant over this sufficiently narrow temperature range. Such a fit yields the value of D corresponding to the *mid*