Observation of spin-wave characteristics in the two-dimensional ferromagnetic ordering of in-plane spins

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The role of dipolar interactions and anisotropy are important to obtain, otherwise forbidden, ferromagnetic ordering at finite temperature for ions arranged in two-dimensional (2D) arrays (monolayers). Here we demonstrate that conventional low temperature magnetometry and polarized neutron scattering measurements can be performed to study ferromagnetic ordering of in-plane spins in 2D systems using a multilayer stack of non-interacting monolayers of gadolinium ions. The spontaneous magnetization is absent in the heterogeneous magnetic phase observed here and the saturation value of the net magnetization was found to depend on the applied magnetic field. The net magnetization rises exponentially with lowering temperature and then reaches saturation following a $T \ln(\beta T)$ dependence. These findings verify predictions of the spin-wave theory of 2D in-plane spin system with ferromagnetic interaction and will initiate further theoretical development.

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Ferromagnetic materials confined in ultra-thin films and multilayered structures are being studied extensively for the development of high-density magnetic data storage devices and to refine our basic knowledge in low-dimensional physics. Recent advances in growth techniques such as molecular beam epitaxy and magnetization ($M$) measurement techniques based on the magneto-optical Kerr effect have enabled us to measure small magnetic signals as a function of applied magnetic field ($H$) and temperature ($T$) even from one atomic monolayer of a ferromagnetic material and a wide range of ordering effects has been observed. These measurements have also demonstrated the existence of a spontaneous magnetization and have revealed hysteresis curves in two- and one-dimensional systems, where magnetic ions are arranged in a grid or in a line within a monolayer. A recently generalized theorem showed, following spin-wave theory, that long-range ferromagnetic order and hence spontaneous magnetization cannot exist at finite temperature in a two-dimensional systems provided spin-spin interactions are isotropic and short range. A theoretical formalism and computer simulations have been developed to include anisotropy and dipolar interactions to explain the apparent contradiction between theory and experiment in low-dimensional systems. A 2D array of magnetic ions with lattice parameter ‘a’ of spins $S$ can be described by a Hamiltonian,

$$\mathcal{H} = \mathcal{H}_{ex} + \mathcal{H}_d + \mathcal{H}_k$$

The strength of the three terms arise from exchange, dipolar and magneto-crystalline anisotropic interactions respectively, and have been approximated by expressing these terms in equivalent magnetic field units as,

$$2\mu_B H_{ex} = JS, \quad 2\mu_B H_d = 4\pi\alpha g S, \quad 2\mu_B H_k = 6KS$$

In the above expression $\alpha$ (≈1) depends on the lattice type and $g$ is equal to $(2\mu_B)^2/a^3$. $K$ is the anisotropy constant. The magnetization reduction due to thermally activated spin waves was calculated with this Hamiltonian and one obtains a non-zero temperature for long-range ferromagnetic ordering as a gap of width $\Delta_s = 2\mu_B H_k^\text{eff}$ opens up at the bottom of the spin wave spectrum for an easy-magnetization axis ($z$) perpendicular to the film plane. The easy-magnetization axis is determined by the sign of the effective anisotropy field ($H_k^\text{eff} = H_k - H_d$) defined by

$$H_k^\text{eff} = \frac{1}{2\mu_B} (6K_{eff} S) \quad \text{with} \quad K_{eff} = K - \frac{2\pi\alpha g}{3}$$

This explains the observation of hysteresis curves in a monolayer with spins oriented normal to the surface. However, the situation is quite different for spins oriented in an in-plane direction with $H_k^\text{eff} < 0$ as the spin-wave spectrum remains gapless. The long-range character of the dipole interactions was found to be responsible for creating a pseudo-gap $\Delta_{xy} = (\pi S g/2)\sqrt{6(6K_{eff}/J)}$ in the spin-wave spectrum that may give rise to long-range ferromagnetic order in 2D in-plane spins provided $|K_{eff}| > K_c = \pi^2 g^2/(6J)$. The temperature dependence of the magnetization $M(T)$ above a transition temperature $T_c = 6S|K_{eff}|/|K_B|$ takes the same form

$$M(T) = M_0[1 - AT \ln(\beta T)]$$

for the ordering of in-plane spins (with $\beta = K_B/\Delta_{xy} = 2\sqrt{6|K_{eff}|/J/(\pi g T_c)}$) and out-of-plane spins (with $\beta = $...
Here \( A = K_B/(4\pi J S^2) \) and \( M_0 \) is the saturation value of the net magnetization that depends on the field applied to carry out measurements \cite{12}. Below \( T_c \) spin wave theory predicts \cite{8} an enhancement of \( M(T) \) as \([1 - A \exp (-1/(\beta T))] \) and \([1 - CT^\nu] \) for out-of-plane and in-plane ordering respectively where \( C \) depends on \( \Delta_{xy} \) and \( \nu \) expected \cite{9, 10, 11} to be \( 3/2 \).

For \( 0 < |K_{\text{eff}}| < \pi^2 g^2/(6J) \) in-plane spins can not stabilize in a homogeneous phase as the magneto-crystalline anisotropy becomes large enough to pull some of the spins in the out-of-plane direction and create a ripple like instability \cite{8, 10}. It is known that both the average magnetization \( M(T) \) as well as the initial susceptibility \( \chi(T) \) is proportional to the physical extent of the ordered phase \( l^* \) that minimizes the zero-field energy \cite{9} and can be written as

\[
M \propto H l^* \quad \text{and} \quad \chi \propto l^* \propto \exp(-\gamma T) \quad (5)
\]

It is expected that at low enough temperature, \( l^* \) reaches saturation either because \( l^* \) becomes comparable to the sample size or due to a freezing of the ripple walls. The net magnetization \( M(T) \) of the ordered domains then should follow the spin-wave prediction (Eq. (4)) to reach saturation.

Here we present the verification of this theoretical prediction of 2D ferromagnetic ordering of in-plane spins using Langmuir-Blodgett (LB) films of gadolinium stearate. The presence of a large multilayer stack of non-interacting monolayers of gadolinium has enabled us to carry out conventional quantitative magnetization measurements at sub-Kelvin temperatures. We could also use polarized neutron scattering measurements \cite{12, 13, 14}, to show that the ordered phase of the in-plane spins is inhomogeneous and that the monolayers remain uncorrelated even when the net magnetization reaches a saturation value.

In the metal-organic structure formed by LB techniques \cite{13, 14, 15}, gadolinium ions are separated by approximately 5Å within a monolayer to form a distorted hexagonal 2D lattice and the monolayers are separated from each other by 49Å by organic chains (Fig. 1). Films having 50 such monolayers of gadolinium ions were deposited on 1 mm thick Si(001) substrates and characterized using x-ray reflectivity technique, as discussed earlier \cite{17}.

Neutron reflection measurements (refer Fig. 2) were carried out on the CRISP reflectometer at the Rutherford Appleton Laboratory (RAL), UK using a cold, polychromatic neutron beam \cite{15, 16} and at the ADAM beamline \cite{20, 21} of the Institut Laue-Langevin (ILL), Grenoble, France, using a monochromatic cold neutron beam. In Fig. 2(b) we have shown spin-polarized neutron reflectivity data taken at 2K by applying a magnetic field of 13 kOe along an in-plane direction (refer Fig. 1). It is known that, if the polarization of the neutrons defined to be parallel (++) or antiparallel (--) to the applied field direction (along +y axis) \cite{13, 14, 22}, the intensity of a multilayer Bragg peak in this geometry increases with effective scattering length \( b_{\text{eff}} = b \pm A \mu_y \) with \( A = 0.2695 \times 10^{-4} \) A/\( \mu_B \). In the left inset of Fig. 2(b) we have shown intensity profiles of parallel (+) and anti parallel (-) incident neutrons at the first peak position. The average of (+) and (-) profiles represent the non-magnetic contribution to the reflectivity. Systematic analysis of all these profiles provide us the value of \( \mu_y \), the component of average moment per gadolinium ion along +y direction. The obtained values of \( \mu_y \) as a function of \( H \) at 4.2K and at 1.75K using the CRISP and ADAM spectrometers respectively are shown in Fig. 2(a) along with data obtained from the magnetization measurements \cite{12} at 2K and 5K. Results of these two independent measurements show that the obtained average saturated moment per gadolinium ion is much less than the expected 7\( \mu_B \) and confirms the existence of a heterogeneous phase. In the right inset of Fig. 2(b) we have shown the transverse diffuse neutron scattering intensity profile at the first Bragg peak. The hyper-geometric line shape profile confirms that the in-plane correlation is logarithmic in nature and that the interfaces are conformal \cite{22, 24}. It should be noted that unlike in x-ray measurements the scattering here originates primarily from the metal heads. The line shape and the associated parameters were found to be independent of \( T, H \) and hence exhibit the absence of conformality in the magnetic correlations between interfaces. This again confirms that the LB films represent a collection of isolated 2D spin-membranes of gadolinium ions.

Now we present the results of conventional magnetization measurements carried out at sub-Kelvin temperature to understand the nature of the ordering. These measurements were performed in a conventional way by measuring forces exerted on a sample situated in a spatially varying magnetic field with a Faraday balance \cite{25}. In Fig. 2(a) we have shown \( M \) vs. \( H \) data taken at 100mK and 500mK temperature in an in-plane (+y) direction. This data reconfirms the absence of hysteresis and remanence (\( M = 0 \) at \( H = 0 \)). The saturation value of the net magnetization at 100mK and 500mK found to be \( 12.7 \times 10^{-6} \) emu/mm\(^2\) \( \cong 5.4 \mu_B / \text{Gd atom} \); much lower than the expected 7\( \mu_B / \text{Gd atom} \) for a homogeneous ferromagnetic phase.

In Fig. 3(a) we have shown the magnetization data taken with different fields as a function of temperature. At higher temperatures these data were fitted with Eq. (5), and the expected exponential dependence is also observed in the magnetization data extracted from our neutron reflectivity measurements (inset of Fig. 3(a)). The values of \( \gamma \) obtained from fitting were found to increase with the reduction of \( H \) and at 0.25 kOe it is found to be \( 2.162 \text{ } K^{-1} \). It is also observed that at a lower field, the magnetization at a fixed temperature is nearly proportional to the applied field (5.03 \( \times 10^{-7} \) at 0.25 kOe...
and $1.29 \times 10^{-6}$ emu/mm² at 0.5 kOe and at a temperature 0.9 K) as predicted for inhomogeneous striped phases (refer Eq. (5)). The amount of majority phase grows exponentially as we lower the temperature for each applied field but below a certain temperature this growth stops as the walls of the majority phase freeze. Below this temperature measured data do not follow Eq. (5) and the net magnetization of the majority phase $M(T)$ increases with lowering temperature following the thermally activated spin waves as given in Eq. (4). We fitted all the data with Eq. (4) and obtained $M_0$ values as $0.9 \times 10^{-6}$, $1.6 \times 10^{-6}$, $3.2 \times 10^{-6}$, $4.5 \times 10^{-6}$, $7.9 \times 10^{-6}$, $9.5 \times 10^{-6}$ emu/mm² with 0.15, 0.25, 0.5, 1.0, 2.5, 5.0 kOe magnetic fields respectively. These saturation values of net magnetization indicate that the percentage of the ferromagnetic phase is increasing from 7.1% to 74.8% as we approach the maximum saturation value of net magnetization obtained of $12.7 \times 10^{-6}$ emu/mm² ($\geq 5.4 \mu_B$/Gd atom) as shown in Fig. 2(a). We extracted the value of exchange $J$ as $8.76 \times 10^{-19}$ erg (or $H_{ex}=0.165$ kOe) from the fitted value of $A (= 1.02 K^{-1})$ for the 0.25 kOe data. We obtained $\beta$ as 3.4 for all the data and hence $|K_{eff}|$ was calculated to be $1.7 \times 10^{-19}$ erg (or $H_{k}^{eff}=0.19$ kOe) for 0.25 kOe data giving $T_c=26$ mK. In this calculation $g$ was $6.88 \times 10^{-18}$ erg, assuming that one gadolinium atom occupies 2.5 Å×20 Å², as obtained from neutron and x-ray analysis (refer Fig.1).

This proves that we are dealing with an inhomogeneous phase as $0 < |K_{eff}| < K_{c} (= 8.89 \times 10^{-17}$ erg). Although Eq. (4) describes the temperature dependence of the magnetization for ferromagnetic ordering of both in-plane and out-of-plane spins, the argument of the logarithmic function can become less than 1 only for in-plane ordering. Unusually low values of $H_{ex}$ and $H_{k}^{eff}$ with a rather large value of $H_d (= 16.3 kOe)$ make $\beta T < 1$ even for $T > T_c$ - this situation has not been reported earlier to the best of our knowledge. It is interesting to note that all the magnetization data shown in Fig. 3(a) attains respected saturation values $M_0$ at temperature $T_0 = 1/\beta (\simeq 0.29 K)$ and a maximum magnetization at temperature $T_m = 1/(e\beta) (\simeq 0.108 K)$. Our experimental uncertainties below 100 mK prohibit us from commenting on any reduction of magnetization below this $T_m$ but Eq. (4) with $\beta T < 1$, all the data is fitted quite well. Further theoretical development is required to understand the thermal activation of spin-waves with in-plane ordering especially as we approach below $T_c (= 26$ mK).

In Fig. 3(b) we have shown zero-field-cooled (ZFC) and field-cooled (FC) magnetization data taken with 0.15 kOe and 0.5 kOe field along with the fitted curve from Eq. (4). We observe a blocking temperature ($T_b$) of 125 mK below which there is branching in both the ZFC and FC data. This result reconfirms that the observed ferromagnetic ordering requires an external field to stabilize and such a low $T_b$ indicates the existence of very small spin clusters in the ZFC phase. Application of the field lowers the activation energy required for the randomly oriented domains to increase the number of spins in the ferromagnetically ordered majority phase and increases the activation energy of the reverse transition $T_a$. As a result we do not observe $T_b$ in the measured temperature range for 0.5 kOe.

In conclusion, we have demonstrated that polarized neutron scattering and conventional magnetization measurements can be used to study 2D ferromagnetic ordering of in-plane spins using a stack of magnetically uncorrelated spin membranes formed with gadolinium stearate LB film. The in-plane ordering observed here shows that a spontaneous magnetization is absent even at 100 mK and saturation value of the net magnetization increases with a lowering in temperature. The magnetization is found to increase exponentially with a lowering in temperature due to the exponential increase of the physical extent of the ferromagnetic domains in the heterogeneous phase. The ferromagnetic domains ultimately saturate following $T \ln(\beta T)$: characteristic of thermally activated spin-waves and are found to be valid for even $\beta T \leq 1$. We believe these experimental results will initiate further theoretical development.

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Figure captions:

FIG. 1: (a) Schematic diagram of the out-of-plane and in-plane structure of the gadolinium stearate Langmuir Blodgett film is shown with the scattering geometry employed for the polarized neutron reflectivity measurements. $x - z$ plane is the scattering plane and the magnetic field is applied along the $+y$ direction. $\lambda$ and $\alpha$ are the wavelength of the radiation and angle of incidence respectively.

FIG. 2: (a) In-plane Magnetization curves obtained as a function of the field ($H$) using neutron reflectivity measured at 4.2K (diamond) and 1.75K (star) compared with conventional magnetization data[17] measured at 2K (down-triangle) and 5K (up-triangle). Solid lines are the fits with a modified Brillouin function[17]. Magnetization measured at 100mK and 500mK is shown for the 1st (symbols) and 2nd (line) cycle of the hysteresis loop. (b) Neutron reflectivity data (symbols) at $H = 13$ kOe and at $T=2K$ for the neutron spin along (+) and opposite (-) to the magnetic field direction with the corresponding fit (line). In the left inset the first Bragg peak is shown in (+) and (-) channels in an expanded scale. Right inset: Transverse diffuse neutron scattering profiles (symbols) measured at 2K with unpolarized and polarized neutron beams. The solid line is a fitted hypergeometric curve as described in the text.

FIG. 3: (a) Sub-Kelvin magnetization results with various applied fields (symbols) fitted with Eq. (4) (Black line) and with Eq. (5) (wine colored dashed lines). Dotted lines indicate the temperatures $T_m$ and $T_0$ (refer to text). Inset shows the magnetization obtained from neutron measurements as a function of temperature (symbols) and fit with Eq. (5) (line). (b) ZFC (green circles) and FC (blue stars) along with the fit (line) for FC measurements.
Out-of-plane direction

$q_z = \frac{4\pi}{\lambda}\sin\alpha$

$20 \text{ Å}^2$

5 Å

In-plane direction
(a) Graph showing the magnetization (emu/mm$^2$) as a function of temperature (K) for different magnetic fields (5.0 kOe, 2.5 kOe, 1.0 kOe, 0.5 kOe, 0.25 kOe). The graph includes two temperature points, $T_m$ and $T_0$. (b) Graph showing the magnetization (emu/mm$^2$) as a function of temperature (K) for magnetic fields of 500 Oe and 150 Oe. The graph indicates the magnetization in field-cooled and zero-field-cooled conditions.