Polarized neutron scattering and sub-Kelvin magnetization measurements in two-dimensional gadolinium stearate Langmuir-Blodgett films

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The role of dipolar interactions and anisotropy are important to obtain, otherwise forbidden, ferromagnetic ordering at a finite temperature for ions arranged in two-dimensional (2D) arrays (monolayers). We performed magnetization measurements with conventional low temperature magnetometry and with polarized neutron scattering on a multilayer stack of noninteracting monolayers of gadolinium ions formed by the Langmuir-Blodgett technique. The spontaneous magnetization could not be detected in the heterogeneous magnetic phase observed here and the saturation value of the net magnetization was found to depend on the sample temperature and applied magnetic field. The net magnetization rises exponentially with a lowering temperature and then reaches saturation following a $T\ln(\beta T)$ dependence. The $T\ln(\beta T)$ dependence of the magnetization has been predicted from the spin-wave theory of a 2D in-plane spin system with the ferromagnetic interaction above $\beta T=1$. The experimental findings reported here could be parametrized by extending the fit of this expression to a temperature domain of $\beta T<1$. Complete understanding of our observations will require further theoretical development.

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I. INTRODUCTION

Ferromagnetic materials confined in ultrathin 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.[1–4] A theorem[3] showed, following spin-wave theory, that long-range ferromagnetic order and hence spontaneous magnetization cannot exist at a finite temperature in two-dimensional systems provided spin-spin interactions are isotropic and short range.[4] 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 the variety of ordering effects has been observed.[5–10] These measurements have also demonstrated the existence of a spontaneous magnetization and have revealed hysteresis curves in two-dimensional[5] and one-dimensional[9] systems, where magnetic ions are arranged in a grid or in a line within a monolayer. One can argue that the apparent contradiction between theory and experiment basically arises due to the fact that the ferromagnetic monolayers are not an ideal two-dimensional (2D) systems primarily because monolayers are intrinsically anisotropic. Moreover dipole-dipole long-range interaction exists among ions of a monolayer and an additional complication in magnetic ordering arises through substrate-monolayer interactions.[5,6,9] A theoretical formalism[7,8,11,12] and computer simulations[6,10] have been developed to include anisotropy and dipolar interactions to explain the apparent contradiction between theory and experiment in ferromagnetic ordering of low-dimensional systems.

In metal-organic structures formed by Langmuir-Blodgett (LB) techniques,[13–15] metal ions are separated by approximately 5 Å within a monolayer to form a distorted hexagonal 2D lattice and the monolayers are separated from each other by 50 Å by organic chains (refer to Fig. 1). It has been demonstrated recently[15] that 2D short-range ferromagnetic order with no spontaneous magnetization occur in the gadolinium stearate (GdSt) LB films as a magnetic field is applied along any in-plane (xy plane) direction, whereas the structure remains paramagnetic down to 2 K if the field is applied along the out-of plane direction (z direction). It was also observed that magnetization values obtained with field along the z direction are higher than that obtained in any in-plane direction at any temperature indicating that the z direction is the easy axis.[15]

The presence of a large multilayer stack of noninteracting monolayers of gadolinium has enabled us to carry out conventional quantitative magnetization measurements at sub-Kelvin temperatures and polarized neutron scattering measurements.[16,17] The results of these measurements presented here indicate that the ordered phase of the in-plane spins is inhomogeneous and the monolayers remain magneti-

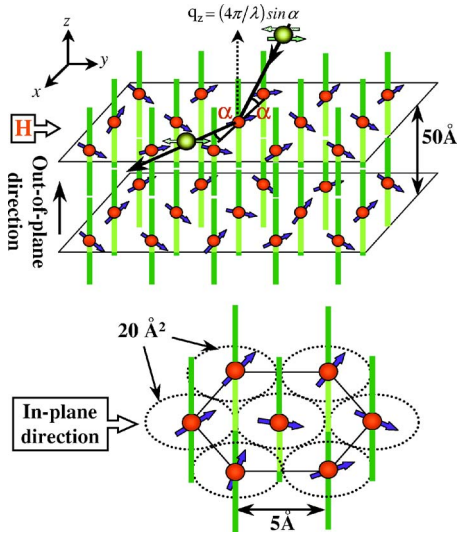

FIG. 1. (Color online) Schematic diagram of the out-of-plane and in-plane structure of the gadolinium stearate Langmuir-Blodgett film is shown with the scattering geometry employed for the polarized neutron reflectivity measurements. The *x*-*z* plane is the scattering plane and the magnetic field is applied along the +*y* direction. λ and α are the wavelengths of the radiation and the angle of incidence, respectively.

cally uncorrelated even when the net magnetization reaches a saturation value. The nature of the ordering of in-plane spins was found to have three distinct characteristics depending on the external field and sample temperature. For a fixed field at a higher temperature region net magnetization increases exponentially with the lowering of the sample temperature. In the next lower temperature region the net magnetization saturates at different values depending on the applied field. The experimental data could be parametrized by the expression $T\ln(\beta T)$. The theoretically predicted lowest temperature region giving $T^{3/2}$ behavior could not be probed in this study as a temperature zone (<30 mK) was not available in our setup.

In the next two sections we will discuss the experimental details and the results of the polarized neutron reflectivity and sub-Kelvin magnetization studies. Finally we shall present a possible explanation of our experimental observations.

II. EXPERIMENTAL DETAILS

Gadolinium stearate LB films having 50 monolayers of gadolinium ions were deposited on 1 mm thick Si(001) substrates using an alternating trough (KSV5000, Finland) from a monolayer of stearic acid on a Milli-Q (Millipore, USA) water subphase containing gadolinium ions. The details about the deposition process have been discussed earlier.[15] The electron density profile obtained from the analysis[15] of the x-ray reflectivity profile confirmed that the number ratio between the gadolinium ions and the stearic acid tails in the film is 1:2. This is similar to the observation of divalent Gd ions in the ferromagnetic GdO, instead of trivalent nonmetallic sesquioxide Gd_2O_3.[18] The schematic of the out-of-plane and in-plane structure of the GdSt LB films is shown in Fig. 1.

Neutron reflectivity measurements were carried out in the CRISP reflectometer at the Rutherford Appleton Laboratory (RAL), UK, using a cold, polychromatic neutron beam[19,20] and in the ADAM beamline[21,22] of the Institut Laue-Langevin, Grenoble, France, using a monochromatic cold neutron beam. In the CRISP reflectometer the sample was placed in a helium cryostat and an incident wavelength range 1.2–6.5 Å of the incident neutron flux has been used. Three different glancing angles 0.25°, 0.65°, and 1.5° were used to collect the specular reflectivity data at a fixed temperature 4.2 K, where the field was allowed to vary from 0 to 15 kOe. In the ADAM beamline, the specular reflectivity data were collected using the monochromatic neutron beam at different sample temperatures from room temperature to 1.75 K and also for different field values from 0 to 20 kOe.

The polarization of the incident neutrons were either parallel (+) or antiparallel (−) to the applied (along +*y* axis) field *H* in all these measurements. In the ADAM beamline we carried out a spin-flip (SF) analysis of a reflected neutron as a function of *H* by measuring reflectivity profiles R^{+-} ($\equiv R^{-+}$) as a function of $q_z[=(4\pi/\lambda)\sin\alpha]$, α being the angle of reflection as shown in Fig. 1.[16,17,23,24] The SF intensity profile is of a purely magnetic origin and provides information about the component of the average moment (μ_x) in the in-plane direction (+*x* in Fig. 1) perpendicular to the applied field. In this geometry the effective scattering lengths b_{eff} become equal to $A\mu_x$ and $b_{\text{coh}} \pm A\mu_y$ with $A = 0.2695 \times 10^{-4}$ Å$/\mu_B$ for SF ($R^{+-} \equiv R^{-+}$) and non-spin-flip (NSF) (R^{++} or R^{--}) reflectivity data, respectively. We have used well-known conventions of polarized neutron reflectivity (R^{--}) where first and second superscripts indicate polarization of incident and scattered neutrons, respectively.

The dc magnetization measurements at sub-Kelvin temperature were carried out in a Faraday force magnetometer as a function of magnetic field and temperature.[25] A cylindrical magnet has been used to apply the field up to 100 kOe and the sample was mounted on an ultrapure Ag plate vertically to get the in-plane magnetization information of the sample. The sample temperature was varied from 1.5 K down to 30 mK. The field-cooled (FC) and zero-field-cooled (ZFC) data have been taken by cooling the sample from 1.5 K to the base temperature 30 mK in the presence (in the case of FC) or absence (in case of ZFC) of applied field and all the data were collected during a heating of the sample. Experiments during cooling of the sample also have been done but no temperature hysteresis was observed.

III. RESULTS AND DISCUSSION

A typical spin-polarized neutron reflectivity data taken at 2 K by applying a magnetic field of 13 kOe are shown in Fig. 2(a). In the left inset of Fig. 2(a) we have shown reflectivity profiles of parallel (+) and anti parallel (−) incident neutrons at the first peak position. The average of (+) and (−) profiles represent the nonmagnetic contribution to the reflectivity. In the right inset of Fig. 2(a) we have shown a peak-normalized transverse diffuse neutron scattering inten-

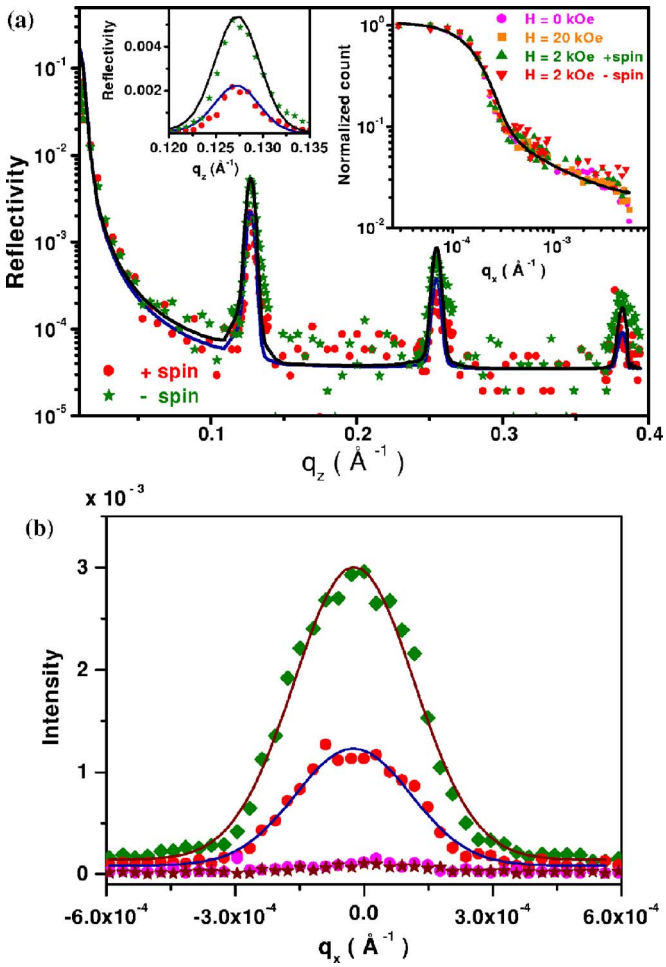

FIG. 2. (Color online) (a) Neutron reflectivity data (symbols) at $H=13$ kOe and at $T=2$ K for the neutron spin along (+) and opposite (−) to the magnetic field direction with the corresponding fit (line). In the left inset the first Bragg peak is shown in (+) and (−) channels in an expanded scale. Right inset: Transverse diffuse neutron scattering profiles (symbols) measured at 2 K with unpolarized and polarized neutron beams. The solid line is a fitted hypergeometric curve as described in the text. (b) Transverse diffuse neutron scattering profiles for + (diamonds) and − (circles) spin state of the incident neutrons at $T=2$ K and $H=13$ kOe. Transverse diffuse neutron scattering profiles in the SF channel for the applied fields $H=2$ kOe (triangles) and 13 kOe (stars) at $T=2$ K are also shown. The solid lines are a guide to the eye.

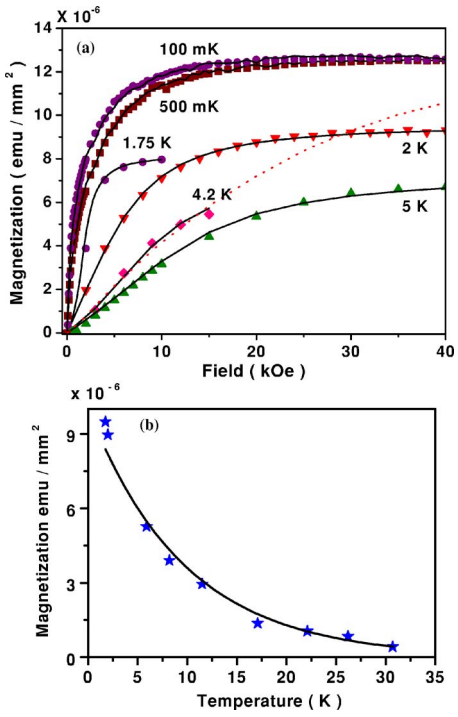

FIG. 3. (Color online) (a) In-plane magnetization curves obtained as a function of the field (H) using neutron reflectivity measured at 4.2 K (diamond) and 1.75 K (star) compared with conventional magnetization data (Ref. 15) measured at 2 K (down triangle) and 5 K (up triangle). Solid lines are the fits with a modified Brillouin function (Ref. 15). Magnetization measured at 100 and 500 mK is shown for the first (symbols) and second (line) cycle of the hysteresis loop. The red (dotted) line is the magnetization curve when the applied field is in the out-of-plane (z) direction. (b) The magnetization obtained from neutron reflectivity measurements as a function of temperature (symbols) and fit with Eq. (1) (line).

sity profile at the first Bragg peak. Transverse diffuse scattering intensity provides us with information regarding the nature of surface roughness through the height-height correlation function and neutron diffuse scattering can, in principle, determine magnetic and structural contributions in roughness. The diffuse scattering line shape profile could be fitted with a hypergeometric Kummar function confirming that the in-plane correlation is logarithmic in nature and that the interfaces are conformal.[26,27] 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 that the magnetic contribution in roughness is negligible here.[26,27] This again confirms that the GdSt LB films represent a collection of isolated 2D spin membranes of gadolinium ions. In Fig. 2(b) we have shown two sets of NSF (R^{++} and R^{--}) and SF (R^{+-}) transverse data of the first Bragg peak collected at $T=2$ K by applying a field of 2 and 13 kOe. Negligible intensity in the SF data clearly indicate that the $\langle \mu_x \rangle$ component is not detectable and all the reflectivity profiles (collected without analyzing the spin of scattered neutrons) can be analyzed with $b_{\text{eff}}=b_{\text{coh}}\pm A\mu_y$, where μ_y represent the average moment per gadolinium ion along the field direction ($+y$ axis). Here the + or − sign is used for the analysis of reflectivity data R^+ and R^- for incident neutrons having polarization parallel (+) and antiparallel (−) to H, respectively.

The systematic analysis of all the reflectivity profiles provides us with values of μ_y as a function of H at 4.2 K and at 1.75 K obtained in the CRISP and ADAM spectrometers, respectively. The results are shown in Fig. 3(a) with the results obtained from earlier magnetization measurements[15] carried out at 2 and 5 K. Results of these two independent measurements show that the obtained average saturated moment per the gadolinium ion is consistent with earlier data and much less than the expected value of $7\mu_B$. The μ_y values obtained from neutron reflectivity data and magnetometry data obtained earlier[15] clearly show that the saturation mo-

ment increases with the lowering temperature. In Fig. 3(a) we have also presented 100 and 500 mK temperatures data collected in the present study. The saturation value of the net magnetization at 100 and 500 mK are found to reach the same value of 12.7×10^{-6} emu/mm$^2\cong5.4\mu_B$/Gd atom; much lower than the expected $7.0\mu_B$/Gd atom for a homogeneous phase. In Fig. 3(a) we have also plotted magnetization data collected at the 5 K temperature earlier[15] along the growth direction (+z direction) that exhibit paramagnetism. The magnetization value of in-plane data is always lower than that of the growth direction (+z direction) for the same temperature at each applied field, indicating that it is hard to keep the spins in the xy plane. But the exchange interaction resulting in a saturation of a moment becomes evident only when spins are kept in the xy plane.[15] The absence of hysteresis and remanence ($M=0$ at $H=0$) is apparent in magnetization data although the saturation magnetization is observed in all in-plane data. It should be mentioned here that absence of hysteresis with the saturation of moment at high field has been observed in conventional ferromagnets[1,2] along hard-axis and in soft magnetic materials.[28,29]

We performed systematic M vs T measurements for investigating the nature of magnetic ordering of in-plane spins forming a heterogeneous phase in the gadolinium monolayer. M vs T was first measured using the neutron reflectivity technique at a fixed field $H=13$ kOe and values of the component of average moment μ_y obtained from the analysis of the reflectivity profiles at different temperatures is shown in Fig. 3(b) along with a fit by an exponential function. The magnetization extracted from the reflectivity data shows that the magnetization and hence the percentage of an ordered majority phase increases exponentially with the decrease in temperature. 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 correlation length ($l^\star$) of the ordered phase that minimizes the zero-field energy[6] and can be written as

$$M \propto Hl^\star \quad \text{and} \quad \chi \propto l^\star \quad \text{with } l^\star \propto \exp(-\gamma T), \qquad (1)$$

where γ is a constant. It is expected that at low enough temperatures, correlation length $l^\star$ reaches saturation either because $l^\star$ becomes comparable to the sample size or due to a freezing of the walls of in-plane domains. This explains our observation of exponential dependence on magnetization with temperature [Fig. 3(b)].

We carried out M vs T measurements using the Faraday balance[25] to investigate the nature of ordering below the temperature where correlation length $l^\star$ saturates. In Fig. 4(a) we have shown the magnetization data taken with different applied fields as a function of temperature. The higher temperature region of the M vs T curves taken with fields of 0.25, 0.5, and 1.0 kOe exhibited exponential behavior as observed in neutron measurements and were fitted with Eq. (1). It is also observed that at a lower field, the magnetization at a fixed temperature is nearly proportional to the applied field (5.03×10^{-7} at 0.25 kOe and 1.29×10^{-6} emu/mm^2 at 0.5 kOe and at a temperature 0.9 K) as predicted by Eq. (1). The values of γ obtained from the fitting were found to increase with the reduction of H and at 0.25 kOe, it is found to

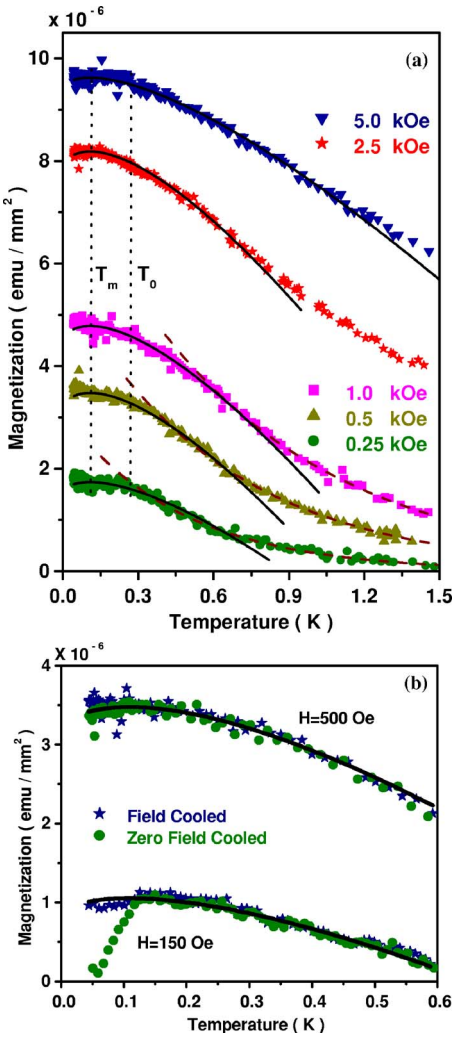

FIG. 4. (Color online) (a) Sub-Kelvin magnetization results with various applied fields (symbols) fitted with Eq. (5) (black line) and with Eq. (1) [wine colored (dashed lines)]. Dotted lines indicate the temperatures T_m and T_0 (refer to text). (b) ZFC [green (circles)] and FC [blue (stars)] along with the fit (line) for FC measurements.

be 2.162 K^{-1}. But below a certain temperature (T_w) corresponding to each field (~450 and 600 mK for the applied fields 0.5 and 1.0 kOe, respectively) the growth of $l^\star$ stops possibly due to the freezing of the domain walls of the ordered phase. Below this temperature measured data show a much slower increase in M with lowering temperature than predicted by Eq. (1). This transition temperature (T_w) shifts toward higher temperature as the field increases and we do not have an appreciable temperature region of the M vs T curves at $H=2.5$ and 5.0 kOe [refer to Fig. 4(a)] that shows exponential behavior. We have discussed in the next section the fitted curves [wine colored (solid) lines], shown in Fig. 4(a).

All these data give us different maximum magnetization (M_m) values at $T_m=0.108$ K, for details refer to the next section, depending upon the H values used to take the data. We obtained M_m values as 1.05×10^{-6}, 1.74×10^{-6}, 3.51×10^{-6}, 4.81×10^{-6}, 8.18×10^{-6}, and 9.64×10^{-6} emu/mm^2 with 0.15, 0.25, 0.5, 1.0, 2.5, and 5.0 kOe magnetic fields,

respectively. These maximum values of net magnetization indicate that the percentage of the ordered phase is increasing from 7.1% to 74.8% as we approach the maximum saturation value of the net magnetization obtained of 12.7 $\times 10^{-6}$ emu/mm^2 ($\cong 5.4\mu_B$/Gdatom) as shown in Fig. 3(a). As any in-plane direction is a hard axis for all these films, one needs to apply a field to keep spins in the xy plane. We obtained lower M_m values for lower field [Fig. 4(a)] and higher temperature [Fig. 3(a)] as spins get oriented along the out-of-plane direction, which is the easy axis. In Fig. 4(b) we have shown ZFC and FC magnetization data taken with 0.15 and 0.5 kOe field. We observe a temperature of 125 mK below which there is branching in the ZFC and FC data. These data are consistent with the fact that one needs a field to keep the spins in the xy plane and this branching is not observed when 0.5 kOe or a higher field was used.

IV. A POSSIBLE EXPLANATION

The experimental observations clearly show a strong crystalline anisotropy that keeps spins along the out-of-plane ($\pm z$) direction making it the easy axis of magnetization. But the exchange interaction was found[15] to take place only among in-plane spins. Negligible intensity in SF neutron reflectivity, which is proportional[21] to μ_x^2, even at low fields, clearly indicate that the spins remain confined in the yz plane, where y is the applied field direction. The magnetization data could not be analyzed using the Kostrerliz-Thouless (KT) theory for the xy model[30,31] as spins do not remain in the xy plane. Hence applied field, exchange, anisotropy, and dipolar interaction plays the crucial role in establishing the short-range ferromagnetic ordering of in-plane spins, observed here.

We can use a 2D array of magnetic ions with lattice parameter "a" of spins S to explain our observation with a Hamiltonian,

$$\mathcal{H} = \mathcal{H}_{ex} + \mathcal{H}_d + \mathcal{H}_k. \tag{2}$$

The strength of the three terms arise from exchange, dipolar, and magnetocrystalline anisotropic interactions, respectively, and have been approximated by expressing [6,7] these terms in equivalent magnetic field units as

$$2\mu_B H_{ex} = JS, \quad 2\mu_B H_d = 4\pi\alpha g S, \quad 2\mu_B H_k = 6KS. \tag{3}$$

In the above expression $\alpha(\sim 1)$ depends on the lattice type and g is equal[6] to $(2\mu_B)^2/a^3$. H_{ex}, H_d, and H_k are the exchange, dipolar, and anisotropy fields, respectively, μ_B is the Bohr magneton, J and K are constants corresponding to the exchange and anisotropy, respectively. A particular form for the anisotropy was used[7] so that one can take the difference between the dipolar energy for perpendicular (z) and that for in-plane (xy) magnetization and can combine this with an anisotropy term to define[6,7] an effective anisotropy coefficient K_{eff}. The magnetization reduction due to thermally activated spin waves was calculated with this Hamiltonian and the axis of easy magnetization is determined by the sign of the effective anisotropy field ($H_k^{\text{eff}} = H_k - H_d$), which is defined[7] as

$$H_k^{\text{eff}} = \frac{1}{2\mu_B}(6K_{\text{eff}}S) \quad \text{with } K_{\text{eff}} = K - \frac{2\pi\alpha g}{3}. \tag{4}$$

For $H_k^{\text{eff}} > 0$ and $H_k^{\text{eff}} < 0$, the magnetization lies perpendicular to the plane (z) and in the in-plane (xy), respectively. The stability criteria for the homogeneously magnetized state are automatically satisfied for the perpendicular easy axis magnetization state as $H_k^{\text{eff}} > 0$. One obtains a nonzero temperature for long-range ferromagnetic ordering as a gap of width $\Delta_z = 2\mu_B H_k^{\text{eff}}$ opens up at the bottom of the spin-wave spectrum for an easy-magnetization axis perpendicular to the film plane. However, the situation is quite different for spins oriented in a 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 [11] to be responsible for creating a pseudogap $\Delta_{xy} = (\pi S g/2)\sqrt{(6|K_{\text{eff}}|/J)}$ in the spin-wave spectrum that may give rise to ferromagnetic order in 2D in-plane spins. The stability criteria for the homogeneously magnetized state for obtaining the in-plane spins are $|K_{\text{eff}}| > K_c = \pi^2 g^2/(6J)$. The temperature dependence of the magnetization $M(T)$ above a transition temperature $T_c (= 6S|K_{\text{eff}}|/k_B)$ takes the form

$$M(T) = M_0[1 - AT\ln(\beta T)] \tag{5}$$

for the ordering of in-plane spins [with $\beta = k_B/\Delta_{xy} = 2\sqrt{6|K_{\text{eff}}|J/(\pi g T_c)}$]. Here $A = k_B/(4\pi JS^2)$ and M_0 is the saturation value of the net magnetization at $T = 1/\beta$ that depends on the field applied to carry out measurements.[12] For $0 < |K_{\text{eff}}| < \pi^2 g^2/(6J)$ in-plane spins cannot stabilize in a homogeneous phase as the magnetocrystalline anisotropy becomes large enough to pull some of the spins in the out-of-plane direction and create a ripplelike instability.[6,7] This results in a formation of ferromagnetic domains and the net magnetization $M(T)$ is then a sum of magnetization of each of these ordered domains. Equation (5) remains valid to describe the growth of net magnetization $M(T)$ to reach a saturation value in this inhomogeneous phase, if we assume that the size of the domains is not increasing in this temperature domain ($T_c < T < T_w$).

This assumption is valid as the domain walls freeze below a certain temperature ($T < T_w$) for each field ($\sim$0.45 and 0.6 K for the applied fields 0.5 and 1.0 kOe, respectively). Hence the net magnetization $M(T)$ curves in this lower temperature range could be analyzed by Eq. (5). We extracted the value of exchange J as 8.76×10^{-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 β as 3.4 for all the data and hence $|K_{\text{eff}}|$ was calculated to be 1.7×10^{-19} erg (or $H_k^{\text{eff}} = 0.19$ kOe) for the 0.25 kOe data giving $T_c = 26$ mK. In this calculation g was 6.88×10^{-18} erg, assuming that one gadolinium atom occupies 2.5 Å $\times$ 20 Å^2, as obtained from neutron and x-ray analysis (refer to Fig. 1). These values confirmed that $0 < |K_{\text{eff}}| < K_c (= 8.89 \times 10^{-17}$ erg) and the ferromagnetic ordered phase here is not homogeneous. It is known that Eq. (5) describes the temperature dependence of the magnetization for ferromagnetic ordering of both in-plane and out-of-plane spins for the temperature region $T > T_c$. The temperature region of validity of Eq. (5) auto-

matically satisfies the condition $\beta T > 1$ as $\beta = 1/T_c$ for the out-of-plane spin ordering. But certain combinations of H_{ex}, H_d, and H_k^{eff} can make the condition $\beta T < 1$ even in the temperature region $T > T_c$ for in-plane ordering of spins as $\beta = 2\sqrt{6|K_{\text{eff}}|}J/(\pi g T_c)$. Here unusually low values of H_{ex} and H_k^{eff} with a rather large value of H_d (=16.3 kOe) obtained from fitting parameters A and β in Eq. (5) 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. 4(a) attain respected saturation values M_0 at temperature $T_0 = 1/\beta (\simeq 0.29$ K) and a maximum magnetization (M_m) at temperature $T_m = 1/(e\beta) (\simeq 0.108$ K). We obtained M_0 values as 0.9×10^{-6}, 1.6×10^{-6}, 3.2×10^{-6}, 4.5×10^{-6}, 7.9×10^{-6}, and 9.5×10^{-6} emu/mm^2 with 0.15, 0.25, 0.5, 1.0, 2.5, and 5.0 kOe magnetic fields, respectively. The experimental uncertainties below 100 mK prohibit us from commenting on the nature of magnetization below this T_m but Eq. (5) with $\beta T < 1$, all the data fit quite well with same A and β.

We could not take the data in the temperature range below critical temperature $T_c \simeq 26$ mK. Below T_c spin-wave theory predicts[7] an enhancement of $M(T)$ as $M_0[1 - CT^{\nu}]$ for in-plane ordering where C depends on Δ_{xy} and ν is expected[6,7,11] to be $\frac{3}{2}$.

V. 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 a gadolinium stearate LB film. The in-plane ordering observed here shows that even at 100 mK a spontaneous magnetization could not be detected. The magnetic ordering observed here can be grouped in three temperature zones as $T < T_c$ (could not be accessed so far); $T_c < T < T_w$ and $T > T_w$. In the temperature zone above the temperature T_w, where domain walls freeze, the magnetization is found to increase exponentially with the lowering in temperature. The exponential increase of magnetization indicate the formation of a heterogeneous phase in the in-plane Gd lattice where the physical extent of the ferromagnetic domains increases exponentially with temperature lowering. In the temperature zone of $T_c < T < T_w$ the sizes of the domains stop growing due to a freezing of the domain walls and the ordering within each of these ferromagnetic domains ultimately saturate. The experimental data could be parametrized by the expression following $T \ln(\beta T)$ even for $\beta T \leq 1$. We believe that these experimental results will initiate further theoretical development.

[1] A. Aharoni, *Introduction to the Theory of Ferromagnetism* (Oxford University Press, Oxford, 2000).

[2] C. M. Schneider and J. Kirschner, in *Handbook of Surface Science*, edited by K. Horn and M. Scheffler (Elsevier, Amsterdam, 2000), p. 511.

[3] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966); **17**, 1307 (1966).

[4] P. Bruno, Phys. Rev. Lett. **87**, 137203 (2001).

[5] H. J. Elmers, G. Liu, and U. Gradmann, Phys. Rev. Lett. **63**, 566 (1989); H. J. Elmers, Int. J. Mod. Phys. B **9**, 3115 (1995).

[6] K. De'Bell, A. B. Maclsaac, and J. P. Whitehead, Rev. Mod. Phys. **72**, 225 (2000).

[7] P. Bruno, Phys. Rev. B **43**, 6015 (1991).

[8] A. Kashuba and V. L. Pokrovsky, Phys. Rev. Lett. **70**, 3155 (1993).

[9] P. Gambardella, A. Dallmeyer, K. Maiti, M. C. Malagoli, W. Eberhardt, K. Kern, and C. Carbone, Nature (London) **416**, 301 (2002).

[10] A. B. MacIsaac, J. P. Whitehead, M. C. Robinson, and K. De'Bell, Phys. Rev. B **51**, 16033 (1995).

[11] S. V. Male'eV, Sov. Phys. JETP **43**, 1240 (1976).

[12] P. Bruno, Mater. Res. Soc. Symp. Proc. **231**, 299 (1992).

[13] M. K. Sanyal, M. K. Mukhopadhyay, M. Mukherjee, A. Datta, J. K. Basu, and J. Penfold, Phys. Rev. B **65**, 033409 (2002); M. K. Mukhopadhyay, M. K. Sanyal, A. Datta, M. Mukherjee, Th. Geue, J. Grenzer, and U. Pietsch, *ibid.* **70**, 245408 (2004).

[14] J. K. Basu and M. K. Sanyal, Phys. Rep. **363**, 1 (2002).

[15] M. K. Mukhopadhyay, M. K. Sanyal, M. D. Mukadam, S. M. Yusuf, and J. K. Basu, Phys. Rev. B **68**, 174427 (2003).

[16] C. G. Shull and J. S. Smart, Phys. Rev. **76**, 1256 (1949).

[17] G. P. Felcher, Phys. Rev. B **24**, R1595 (1981).

[18] C. Schüßler-Langeheine, H. Ott, A. Yu. Grigoriev, A. Möller, R. Meier, Z. Hu, C. Mazumdar, G. Kaindl, and E. Weschke, Phys. Rev. B **65**, 214410 (2002).

[19] J. Penfold and R. K. Thomas, J. Phys.: Condens. Matter **2**, 1369 (1990).

[20] J. P. Goff, P. P. Deen, R. C. C. Ward, M. R. Wells, S. Langridge, R. Dalgleish, S. Foster, and S. Gordeev, J. Magn. Magn. Mater. **240**, 592 (2002).

[21] H. Zabel, Physica B **198**, 156 (1994); H. Zabel and K. Theis-Bröhl, J. Phys.: Condens. Matter **15**, S505 (2003).

[22] V. Leiner, K. Westerholt, A. M. Blixt, H. Zabel, and B. Hjörvarsson, Phys. Rev. Lett. **91**, 037202 (2003).

[23] H. Kepa, J. Kutner-Pielaszek, A. Twardowski, C. F. Majkrzak, J. Sadowski, T. Story, and T. M. Giebultowicz, Phys. Rev. B **64**, 121302(R) (2001).

[24] A. Schreyer, Th. Zeidler, Ch. Morawe, N. Metoki, H. Zabel, J. F. Ankner, and C. F. Majkrzak, J. Appl. Phys. **73**, 7616 (1993); H. Zabel, R. Siebrecht, A. Schreyer, Physica B **276–278**, 17 (2000).

[25] T. Sakakibara, H. Mitamura, T. Tayama, H. Amitsuka, Jpn. J. Appl. Phys., Part 1 **33**, 5067 (1994).

[26] M. K. Sanyal, S. K. Sinha, K. G. Huang, and B. M. Ocko, Phys. Rev. Lett. **66**, 628 (1991); J. K. Basu, S. Hazra, and M. K. Sanyal, *ibid.* **82**, 4675 (1999).

[27] The hypergeometric Kummar function describes diffuse scattering profiles of interfaces having capillary waves that generate long-range logarithmic correlation (Ref. 26). The measured roughness and "true" roughness of the interfaces are calculated to be 2.3 and 5.4 Å, respectively, and we get an effective surface tension equal to 10 mN/m.

[28] R. C. O'Handley, *Modern Magnetic Materials, Principles and Applications* (John Wiley, New York, 2000).

[29] M. Haüssler, R. Zheng, J. W. Y. Lam, H. Tong, H. Dong, and B. Zhong Tang, J. Phys. Chem. **108**, 10645 (2004); K. Ounadjela, G. Suran, and F. Machizaud, Phys. Rev. B **40**, 578 (1989).

[30] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973); J. M. Kosterlitz, *ibid.* **7**, 1046 (1974).

[31] R. Gupta, J. DeLapp, G. G. Batrouni, G. C. Fox, C. F. Baillie, and J. Apostolakis, Phys. Rev. Lett. **61**, 1996 (1988).