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Finite-field magnetization of a two-dimensional Heisenberg ferromagnet

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Abstract. The magnetization, M, of a two-dimensional layer of the Heisenberg ferromagnet Pd(1.2 at% Fe) is examined. It is found that M increases as $\ln(H)$ over a wide range of magnetic field, H, and temperature. The temperature dependence of each parameter derived from such $\ln(H)$ fits to the data is compared with calculations concerning the two-dimensional classical Heisenberg model. The data do not follow all the predictions of the classical model.

1. Introduction

The uniform susceptibility of a two-dimensional Heisenberg (2DH) ferromagnet is predicted [1, 2] to vary with temperature (T) as $\chi(T) = f(T)\exp(B/T)$ for $T \ll B$ (for a square lattice with nearest neighbour interactions, $B = 4\pi JS^2$ where J is the exchange energy and S is the spin.) That is, there is no long-range order [3] and no phase transition [4] for $T > 0$. For a system with classical (vector) spins $f(T)$ is expected [1] to be independent of T whereas for the system with quantum spins calculations suggest [2] that $f(T) \propto T^2$. We have investigated [5] thin films of the Heisenberg [6] ferromagnetic alloy Pd(1.2 at% Fe) and found that they are excellent examples of the 2DH model. For a film which is 2 nm thick, measurements of $\chi(T)$ can be used to distinguish three regions of T. (1) There is a high-temperature region (but for T still well below the Curie temperature of bulk Pd(1.2 at% Fe)) in which $\chi(T)$ behaves as predicted for a 2DH ferromagnet. In this 2DH regime the dominant temperature dependence is exponential in $1/T$. Since it is quite difficult for an experiment to distinguish between possible forms for the prefactor of such an exponential, we have not attempted to distinguish between the different possibilities mentioned. However, we note that the value of B one derives by fitting $\chi(T)$ is 53 K for classical spins and 70 K for quantum spins. (2) Below a rather well defined temperature, $T^* = 6.3$ K, $\chi(T)$ begins to increase even more rapidly probably indicating a crossover to x–y behaviour as the dipole–dipole interactions force the spins to lie in the plane of the film. (3) Below a roughly defined temperature, $T' = 5$ K, we find magnetic field hysteresis effects and time relaxation phenomena. Below T' there is probably a zero-field magnetization either due to a phase transition or due to finite size effects in the plane of the film cutting off the growth of 2d ferromagnetic spin correlations.

We have also shown [5] that the magnetic field (H) dependence of the magnetization (M) at moderately strong fields, where there is curvature in $M(H)$ but no saturation, is consistent with theoretical predictions [7, 8] regarding the 2DH model with classical spins. As far as we know there are no finite-field predictions for the quantum spin model. The theoretical prediction for classical spins is that $M = A(T)\ln(H/H^*)$ where $A \propto T$ and

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$H^* \propto \exp(-B/T)$. In this note we present and discuss measurements of $M(H,T)$ taken over a much wider range of H and T than in [5]. We will see that M can always be fit to a $\ln(H)$ form at intermediate fields and that, in contrast to the case of $\chi(T)$, we can draw clear conclusions about the temperature dependences of the derived parameters. We find that H^* is proportional to $1/\chi$ as predicted but it turns out that the prefactor A is not proportional to T.

2. Experiment

The films to be discussed are the same as those in [5]. Briefly, the Pd(1.2 at% Fe) layer was grown by sputter deposition onto ambient temperature substrates from an alloy target of the same composition. The magnetic layer was grown on top of a seed layer of 300 Å of 99.99% pure Pd which was deposited at room temperature on Si (100) substrates. The magnetic layer was then covered with a 300 Å thick cap of pure Pd. X-ray diffraction and transmission electron microscopy (TEM) showed that the resulting film was completely crystalline and single phase with grain sizes of the order of 150–300 Å.

The magnetization was measured using a quantum design SQUID magnetometer with a superconducting magnet. In order to maximize the signal and minimize the background, four films were inserted into a long drinking straw which was then suspended from the rod supplied by the manufacturer. Measurements were made over the range 3 K $< T <$ 18 K. The signal from the Si substrates is dominated by a diamagnetic background but also has a small T-dependent paramagnetic component. A background run was made at 18 K where the signal from the films is negligible. A straight line was fit to the background data and this line (corrected for the known T-dependent paramagnetic part) was subtracted from all the data at lower T. The background subtraction was necessary for interpretation of data taken above 9 K but had essentially no effect on data taken below 7.5 K.

3. Results

Figure 1 shows $M(H)$ for these films over a wide range of T. It must be pointed out that the ranges of T and H in which we can make measurements are restricted on the high-H, high-T side because the sample magnetization becomes equal to or smaller than the background substrate magnetization and they are restricted on the low-H side because we are using a superconducting magnet to supply the field and we do not shield out the earth's field so any fields of the order of the earth's field (about 20 A m^{-1}) are not well defined. The lines through the data in figure 1 represent fits of M to a $\ln(H)$ form over the region for which that form is followed. Although the $\ln(H)$ form fits reasonably well over a wide range of T, we cannot make measurements over a wide enough range of H to verify that the $\ln(H)$ form is best. However, in addition to predicting the $\ln(H)$ form, the theory also predicts the dependence upon temperature of each parameter in the fit. We find that we can draw rather strong conclusions regarding the behaviour of these parameters as T is changed and we can easily determine if they follow the theory.

For instance, figure 2 shows the values of H^* (derived from figure 1) on a logarithmic scale as a function of T^{-1}. H^* represents the field below which M/H is independent of H and so must vary, roughly, as the inverse of $\chi(T)$. Clearly, the same crossover at 6.3 K which we saw in the examination of χ is evident also in H^* as well as the other change in behaviour at 5 K. In the inset to figure 2 we show χH^* as a function of T in the temperature

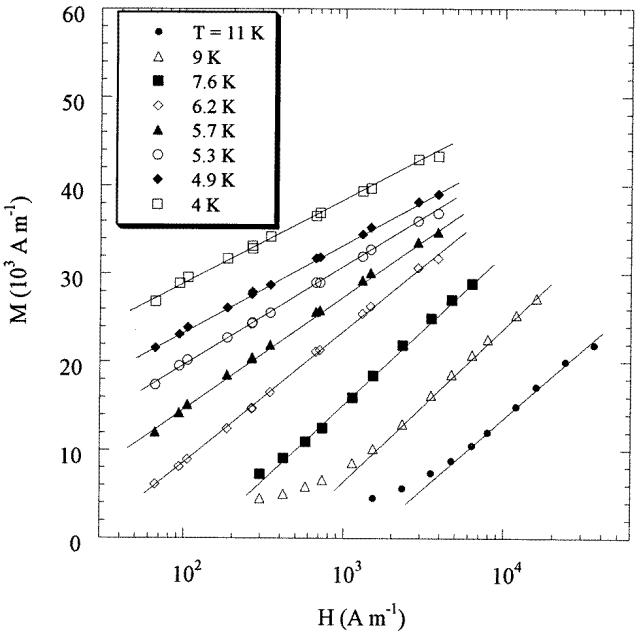

Figure 1. Magnetization as a function of H for a 2 nm thick Pd(1.2 at% Fe) film showing that M varies as $\ln(H)$ at intermediate magnetic fields for this two-dimensional Heisenberg ferromagnet.

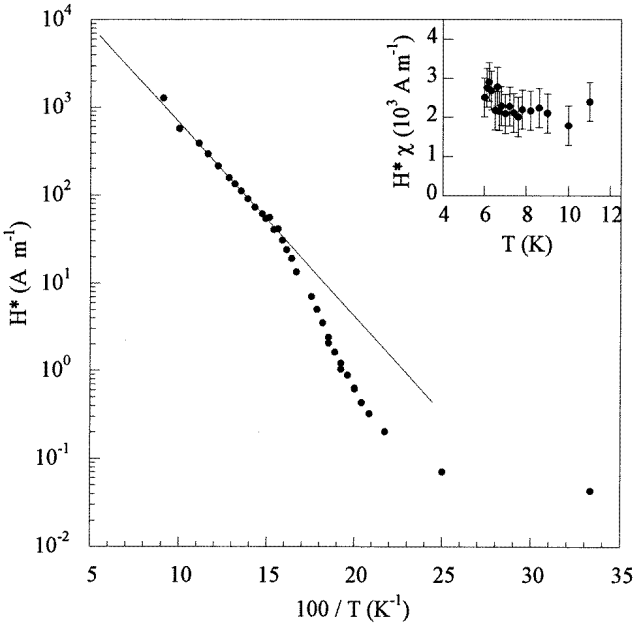

Figure 2. Temperature dependence of the magnetic field scale, H^*, derived from the $\ln(H)$ fits seen in figure 1. The crossover at about 6.3 K is clearly seen. Below about 5 K one finds magnetic field history and time dependences at low fields. The inset shows that H^* is inversely proportional to χ over the range of T for which both can be estimated.

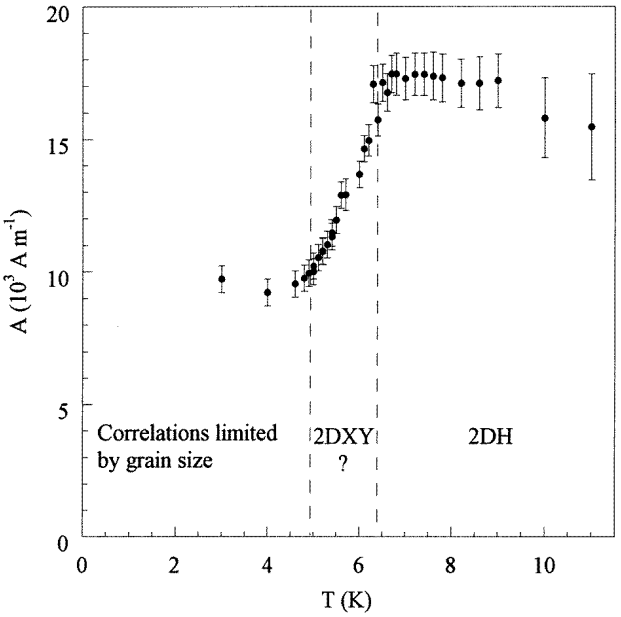

Figure 3. Temperature dependence of the prefactor A derived from figure 1. The T-dependence of A changes at the crossover temperature but never follows the predictions of [7].

range in which we can measure both quantities. One can see that χH^* is independent of T, as expected. It appears that the only new information we gain from an analysis of H^* is an estimate of $\chi(T)$ in the region of T ($T < 5.7$ K) in which we cannot measure it directly.

In figure 3 we show the T dependence of A, the prefactor to the $\ln(H)$ term determined from the fits in figure 1. Again we can distinguish the three regimes of T. The prediction of the classical 2DH model is that $A(T)$ is proportional to T. In contrast, our experiments show that: (1) in the high-temperature region ($T > 6.3$ K) where the 2DH prediction should hold, $A(T)$ is nearly independent of T; (2) for 5 K $> T >$ 6.3 K A decreases as T decreases. The decrease below the crossover may be approximately linear in T but it is clearly not directly proportional to T; and (3) below 5 K the parameter again changes its temperature dependence but it is unlikely to have any simple significance in this regime.

4. Discussion

One expects a thin film of a Heisenberg magnet to behave two-dimensionally when it is cooled below the Curie temperature of the 3D bulk magnet. As T is decreased further [1] the two-dimensional spin–spin correlation length, ξ, and χ grow exponentially until (a) the demagnetizing effects of the dipole–dipole coupling come into play and decrease the number of spin components to two (x–y model) or (b) the crystalline anisotropy comes into play and decreases the effective number of spin components to less than three. The system should then have a phase transition [1, 9] of either the Kosterlitz–Thouless type or to a state with long-range magnetic order. We estimate [5] the dipole–dipole coupling constant at $\delta \approx 0.1$ K and use measurements on bulk Pd(Fe) samples by various groups [10, 11] to estimate that the anisotropy energy per Fe is 0.01 K $< D <$ 0.08 K. Since the dipole

energy is probably somewhat larger than the anisotropy energy we will assume $\delta > D$ in the following discussion. However, the discussion would not be changed much by the reverse assumption.

Following an argument of Friedman *et al* [12] the dipolar coupling is expected to become important when the dipolar energy, $\delta\xi^2$, of a cluster of aligned spins of size ξ^2, is of order $k_B T$. From the quantum spin calculations of [2] there is an explicit form found for ξ so we can estimate this crossover temperature. One finds that $T_{crossover} = 4\pi J S^2/\ln(JS^2/\delta S C_\xi^2)$ where C_ξ was found [2] to be 0.07 and S is of order one [11]. We get $4\pi J S^2 = 70$ K from fitting $\chi(T)$ to the appropriate equation from [2] so that we expect to see the effects of dipole interactions below about 7.5 K. The main effect of the dipole term is a 'shape effect' tending to hold spins in the plane of the sample so that they are effectively x–y spins. In the same manner one can estimate that the temperature where the effects of the anisotropy become apparent is about 6 K. We think that it is most likely that the crossover at 6.3 K is due to dipole coupling and that the change at 5 K is the development of magnetic order due to the combined effects of dipole–dipole coupling and anisotropy.

We therefore suggest that the film is a 2D Heisenberg magnet below the bulk Curie temperature and above 6.3 K. In this temperature range we find that $A(T)$ is roughly independent of T. The disagreement between this result and the theory of Khokhlachev [7] may be due to the fact that we are dealing with a quantum system rather than a classical one. As the temperature is decreased below $T_{crossover}$, the spin system should be better described as XY spins and one finds that χ increases much faster, than in the 2DH regime, as the film approaches a ferromagnetic phase transition. In this crossover region there are no theoretical predictions for $M(H)$ so there is no reason to expect a simple T dependence of $A(T)$. For $T < 5$ K the appearance of magnetic hysteresis suggests that there is magnetic order. However, as noted in [5], the remanent magnetization, M_{rem}, decays logarithmically with time so we cannot easily examine $M_{rem}(T)$ as in [13]. In fact, the simplest conclusion is that there is no true long-range magnetic order even for $T < 5$ K. For instance, the correlation length may be limited by the microstructure of the film. In this case the magnetic hysteresis would be explained by blocking of what are basically superparamagnet regions. Finally, in trying to decide whether there is a true phase transition at 5 K we have examined $\chi(T)$ (actually we use H^* to estimate χ) for divergence near 5 K and $M(H)$ for power-law behaviour near 5 K. The uniform susceptibility (when determined in this fashion) certainly does not diverge but we do find that for all $T \leqslant 5$ K, $M \sim H^{0.1}$ and that for $T > 5$ K a power-law form clearly does not work. The problem we have with this fit is the same as for $\ln(H)$, M changes only slightly for the range of fields available to us so many functions might fit.

To summarize, we have seen that very thin films of the alloy Pd(1.2 at% Fe) behave as 2D Heisenberg magnets over a significant range of temperatures. At low-T the effects of the dipole and anisotropy energies come into play and the system crosses over to a different type of behaviour. In the 2DH regime M varies as $\ln(H)$ as predicted by theories of classical spins but the prefactor of the $\ln(H)$ form is independent of T rather than linear in T. The discrepancy may result from using a theory with classical spins rather than quantum spins.

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