Low-temperature scaling of the susceptibility of Ni films

X. H. Song,[1] X.-G. Zhang,[2] J. Fan,[1] Y. R. Jin,[1] S. K. Su,[1] and D. L. Zhang[1,*]

[1]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Science, Beijing 100080, China*

[2]*Center for Nanophase Materials Sciences and Computer Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6493, USA*

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Measurement of low field ac susceptibility of Ni thin films over the temperature range of 5–300 K reveals a surprising power law scaling. The temperature-dependent part of the normalized susceptibility, $\chi_{\parallel}/M_S - \chi_{\rm rot}/M_S$, where $\chi_{\parallel}$ is the initial susceptibility for in-plane magnetization, $\chi_{\rm rot}$ is the domain rotation contribution, and M_S is the saturation magnetization, scales with the nonlinear reduced temperature as t^{-2} over the entire temperature range, where $t=(T-T_C)/(T+T_C)$ and T_C is the Curie temperature. Thickness and reduced temperature dependences are completely decoupled. This result implies that domain wall motion does not contribute to the low field susceptibility.

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Ferromagnetism of Ni thin films has been studied extensively[1] because it allows to study a wide array of physical phenomena with simple and relatively easy to control parameters. In particular, both temperature scaling of the magnetic properties[1,2] near the Curie temperature T_C and finite size scaling[2–4] with respect to the film thickness d have been studied. For thin films, the scaling law generally predicts a separation between the dependence on the film thickness d and on the reduced temperature $t_L=T/T_C-1$, for example, for the magnetic susceptibility $\chi=dM/dH$, through the following form:

$$\chi(T,d) = d^{\omega} f(d^{\theta} t_L), \tag{1}$$

where $f(z)$ is a function with the leading term $f_0 z^{-\gamma}$ for small z. This reflects the fact that T and d are usually not independent variables in scaling.[4] The so-called generalized Curie-Weiss law[5,6] extends the power law scaling well above T_C by replacing the linear reduced temperature t_L by a nonlinear reduced temperature $t_{NL}=1-T_C/T$. Using the nonlinear reduced temperature, the power law scaling was shown to be accurate over the entire paramagnetic regime.

The extension of the scaling law to low temperatures was never investigated because of the general belief that the presence of magnetic domains should invalidate the scaling argument. Thermally activated domain wall motion is expected to contribute significantly to the temperature dependence of magnetic properties. Configuration details, such as sample geometry, impurity and defect content, and thermal and magnetic history, can strongly influence domain wall motion and through it influence the magnetic properties. Adding to the complication is that two different mechanisms, magnetic moment rotation and domain wall motion, in addition to the spin-spin correlation which is believed to dominate near T_C, may contribute to the magnetic susceptibility. In a typical measurement, these contributions are not separated.[7]

In the present work, we report the measurement of the low field susceptibility of Ni films as a function of temperature from 5 to 300 K and film thickness from 5 to 440 nm. Within this thickness range, the ground state moment is parallel to the film. Measuring the initial susceptibility for the in-plane and out-of-plane magnetization directions separately allows clear separation of the contributions from moment rotation, domain wall motion, and spin-spin correlation. While the out-of-plane susceptibility is nearly temperature independent, the in-plane susceptibility shows a surprising power law scaling for the entire temperature range. In addition, thickness and temperature dependences are complete decoupled in the form of Eq. (1), with the exponent $\theta=0$, i.e., that the function is independent of the thickness. This scaling result implies that even in the ferromagnetic regime, there is no domain wall motion contribution to the low field susceptibility.

Pure (99.99%) Ni films were deposited onto Si(111) substrates by dc magnetron sputtering at room temperature. The film thicknesses were measured by Dektak 8 (Veeco Instruments, Inc.). The structure of selected samples was checked by x-ray diffraction analysis. The results show obvious (111) oriented characteristics at the thin end of the thickness range, and the texture weakens with the increase of film thickness. The lattice parameters are very close to the bulk crystal ($\approx$0.1%). The full width at half maximum of the main (111) peak is about 0.40, from which the grain size can be estimated to be around 30 nm. This value is close to the bulk mean free path deduced from the thickness dependence of residual resistivity, 30–50 nm. The temperature dependences of resistivity for all the thicknesses are nearly parallel, showing similar electron-phonon scattering and phonon spectrum for different samples. From this, we conclude that there is no significant variation in the lattice structure among the samples.

The dc magnetization and ac susceptibility were measured using ac/dc magnetometry system with ac susceptibility sensitivity of 2×10^{-8} emu. The M-H curves for all samples were taken at selected temperatures between 5 and 300 K. All samples except the 5 nm sample showed anisotropy indicating in-plane spin alignment. Figure 1 shows the curves for 5 and 284 nm samples. The saturation magnetization was fitted to a universal scaling curve $M_S(T)/M_S(0)$ as a function of T/T_C (Fig. 2) with T_C as the fitting parameter for each sample, yielding a thickness dependent T_C (Fig. 2, inset).

The low field susceptibility was measured by ac method with excitation fields of 1, 5, 10, and 15 Oe and frequencies

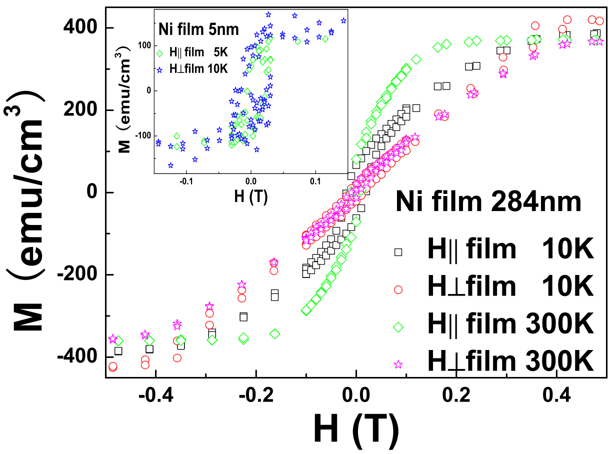

FIG. 1. (Color online) M-H for 5 nm (inset) and 284 nm samples for both $H\|$film and $H\perp$film.

of 633 and 1333 Hz. The signal is too small to measure for field far below 1 Oe under which initial susceptibility measurements are usually carried out for bulk samples. However, two samples (122 and 72 nm) were checked at 1 and 15 Oe, which yielded consistent results. This means that for our thin film samples the present excitation fields are well within the linear reversible range, and the observed behaviors represent the intrinsic dynamical properties of the films in the low field limit. The contribution from the Si substrate to the susceptibility is subtracted from a separate measurement of the substrate without the Ni film. For the perpendicular field direction, the correction due to the demagnetization factor N is accounted for using $\chi=\chi_{\mathrm{meas}}/(1-N\chi_{\mathrm{meas}})$, where χ_{meas} is the measured value for the susceptibility and we used $N=1$.

We first contrast the difference in the temperature dependence between the initial susceptibility for in-plane and out-of-plane magnetization directions. Figure 3 shows the comparison for a 151 nm thick sample. To eliminate the uncertainty in thickness measurement, and also to show the real contribution of average spins, the data are normalized by $M_S(T)$. The normalized in-plane susceptibility, $\chi_{\|}/M_S$, rises

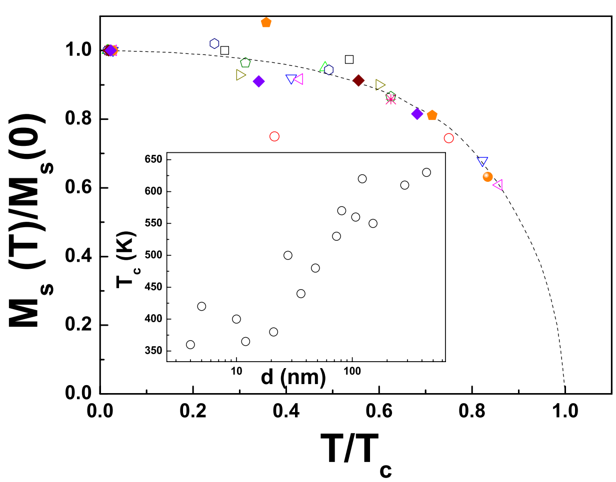

FIG. 2. (Color online) Reduced saturation magnetization as a function of reduced temperature T/T_C for Ni films. Dashed curve is bulk Ni. Inset: T_C versus film thickness.

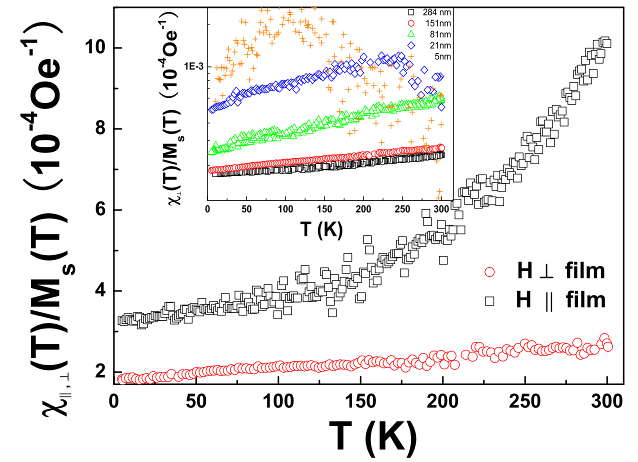

FIG. 3. (Color online) Normalized low field anisotropic susceptibility versus temperature for a Ni film of thickness 151 nm. Inset: Normalized low field out-of-plane susceptibility versus temperature for Ni films of different thicknesses.

rapidly with the temperature, while the out-of-plane susceptibility, $\chi_{\perp}/M_S$, depends only weakly on the temperature. The weak temperature dependence of the out-of-plane susceptibility is observed for all the samples except for the very thin samples, as shown in the inset of Fig. 3. Because of the in-plane spin alignment in the thicker films, at least in the initial stage of vertical magnetization, only domain rotation contributes to the susceptibility. The weak temperature dependence means that the rotation is not thermally activated.

For the in-plane magnetization, the temperature dependence is much stronger. It is expected that both the domain rotation and the domain wall motion should contribute to the in-plane magnetization. Many works[8–11] show that at low temperature and low field, the velocity of the domain wall motion is zero and the wall motion is thermally activated. Indeed, the in-plane low field susceptibility at 10 K as a function of d tracks closely the susceptibility for perpendicular magnetization (Fig. 4), with a slight anisotropy that shows the susceptibility for in-plane magnetization a little larger than for out-of-plane magnetization. Based on this ob-

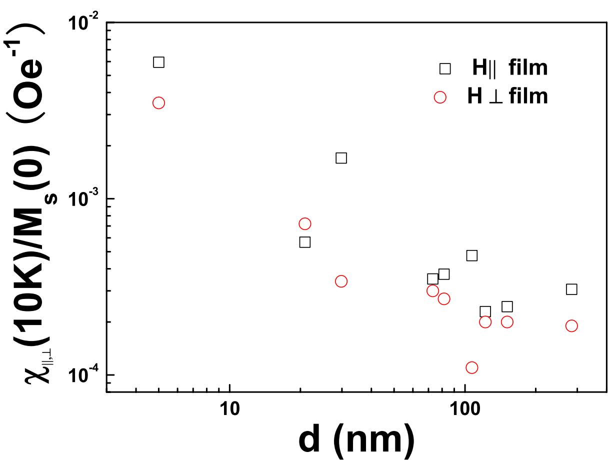

FIG. 4. (Color online) The normalized initial susceptibility at $T=10$ K for $H\|$film (black squares) and $H\perp$film (red circles).

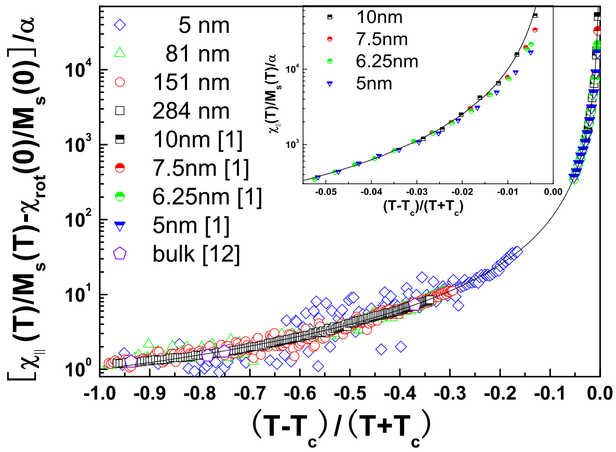

FIG. 5. (Color online) Temperature-dependent part of the normalized susceptibility, $\chi_{\parallel}(T)/M_S(T)-\chi_{\rm rot}(0)/M_S(0)$, for Ni films of different thicknesses, as a function of $t=(T-T_C)/(T+T_C)$. The data by Baberschke (Ref. 1) and for the bulk sample by Onnes and Perrier (Ref. 12) are also shown. Inset shows the data by Baberschke (Ref. 1) closer to the critical temperature. Solid curve is fitted using Eq. (2).

servation, we can define a domain rotation contribution to the normalized susceptibility at the low temperature limit as $\chi_{\rm rot}(0)/M_S(0)$, for both in-plane and out-of-plane susceptibilities. As we will see below, the value of $\chi_{\rm rot}(0)$ will be extracted from the in-plane susceptibility data by fitting to Eq. (2).

After subtracting the domain rotation contribution $\chi_{\rm rot}(0)/M_S(0)$ from the normalized in-plane susceptibility at all temperatures, all data fall on a universal curve described by

$$\frac{\chi_{\parallel}(T)}{M_S(T)}-\frac{\chi_{\rm rot}(0)}{M_S(0)}=\alpha\left(\frac{T-T_C}{T+T_C}\right)^{-2}, \qquad (2)$$

with $\chi_{\rm rot}(0)$ and α being the only fitting parameters for each sample. The result is plotted in Fig. 5 along with the data by Baberschke[1] closer to the critical temperature, and the data for bulk sample by Onnes and Perrier,[12] both of which also fall on the same universal curve. Using $t_{NL}=(T-T_C)/T$ instead of $t=(T-T_C)/(T+T_C)$ as the scaling variable would be equivalent to keeping only the T^2 term in the polynomial expansion of Eq. (2), leading to a poor fit valid only near $T=T_C$.

The scaling of t^{-2} seems to suggest $\beta+\gamma=2$, for the critical scalings $M_S\propto|T-T_C|^{\beta}$ and $\chi\propto|T-T_C|^{-\gamma}$. This is quite different than the known values[13] of the critical exponents for Ni, $\beta=0.395$ and $\gamma=1.345$. We note that the range of fitting for the critical exponents is typically within $|\epsilon|=|T/T_C-1|<0.01$, whereas our fit covers the entire temperature range from $T=0$ to T_C. Also note that the data by Baberschke,[1] magnified in the inset of Fig. 5 indeed deviates slightly from t^{-2} scaling in the range $|t|<0.01$. The scaling close to the critical temperature has been studied extensively in the literature. We instead focus on the scaling behavior outside the critical scaling region.

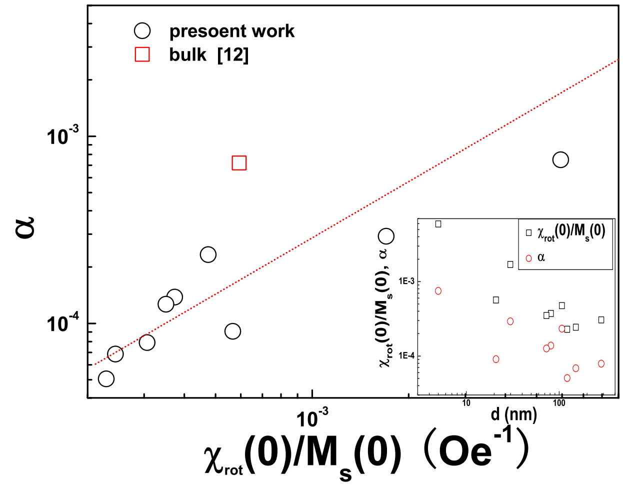

FIG. 6. (Color online) Correlation between $\chi_{\rm rot}(0)/M_S(0)$ and α. The line shows linear correlation. Inset: Thickness dependence of both quantities.

The uniform scaling of Eq. (2) independent of film thickness (and possibly other film qualities such as roughness and bulk defects) is quite surprising. This is clearly not anticipated by models based on thermally activated domain wall motion. It strongly suggests that the low field in-plane susceptibility is a function of the correlation length over the entire temperature range. Such a result calls for a reexamination of current models for micromagnetics. In contrast to the tight temperature scaling, the thickness dependence of the susceptibility, as shown in Fig. 4, displays a lot of scatter. This may be due to the variations in the film roughness or due to the error bars in the subtraction of the substrate susceptibility. Despite the scatter in the data, Eq. (2) represents a clear separation of the temperature and thickness dependence.

The correlation between the two temperature-independent coefficients in Eq. (2), $\chi_{\rm rot}(0)/M_S(0)$ and α, is shown in Fig. 6. The correlation is not far from a linear relationship, indicated by the line in Fig. 6. The strong correlation between the two quantities suggests that both may be viewed as parametric functions of a common variable, e.g., the film thickness. The thickness dependence is shown in the inset of Fig. 6. Again, similar to Fig. 4, there is considerable scatter in the thickness dependence. In addition, the position of the bulk data in Fig. 6 is closer to the values of the thinnest films rather than the thicker films. These observations suggest that both $\chi_{\rm rot}(0)/M_S(0)$ and α depend not just on the film thickness. They may also depend on other factors, such as lattice strain, disorder, and impurities.

Our result can be summarized as the following. The low temperature susceptibility of Ni films can be clearly separated into two terms, one mostly independent of the temperature, and the other scaling as t^{-2} uniformly between $T=0$ and close to T_C. Some striking conclusions can be drawn from these results. First, because the domain size strongly depends on the temperature, close to T_C the domains are very small and domain wall motion does not contribute to the susceptibility. If at lower temperatures a significant contribution arises from domain wall motion, a crossover temperature should be visible when examined from the scaling behavior. No such crossover temperature is observed in our data. Therefore, we conclude that there is no domain wall motion contribution to the low field susceptibility for the entire temperature range, and that the temperature scaling of the susceptibility is through the temperature dependence of the spin correlation length. Second, in general the spin correlation length depends on both the temperature and the film thickness. The correlation between $\chi_{\mathrm{rot}}(0)/M_S(0)$ and α approximately follows a linear relationship, but the term with α has a temperature dependence while the other term does not. This suggests that the correlation length depends weakly on the film thickness except through the change of T_C, and the corollary that there is a separate thickness dependence of the susceptibility outside the dependence through T_C and not due to the spin correlation. This is an important conclusion and must be examined with further experiments.

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*Author to whom correspondence should be addressed; zhangdl@aphy.iphy.ac.cn

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