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Effect of interparticle interaction on the magnetic relaxation in NiO nanorods

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Temperature variation (5 K–300 K) of the AC magnetic susceptibilities (χ' and χ'') at frequency f=0.1, 1, 50, 100, 500, 1000, 2000, 5000, and 10, 000 Hz are reported in 5 nm diameter nanorods of NiO, with and without oleic acid (OA) coating. Using the peak in χ' as the blocking temperature T_B, it is observed that T_B increases with increasing f. The data for the two samples fit the Vogel-Fulcher law: $f=f_0 \exp[-E_a/k(T_B-T_0)]$, with $f_0=9.2\times 10^{11}$ Hz, $E_a/k=1085$ K, and $T_0=165$ K (0 K) for the uncoated (coated) particles. This shows that T_0 provides a good measure of the effects of interparticle interactions on magnetic relaxation and that these interactions are essentially eliminated with the OA coating. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2165787]

I. INTRODUCTION

A new frontier in the physics of materials is understanding the properties of materials in reduced dimensions [thin films, wires, nanoparticles (NPs)] and exploiting these properties for applications.[1–4] Bulk antiferromagnets (AFs) that are normally compensated with an equal number of up and down magnetic moments below the Néel temperature T_N can acquire substantial magnetization in reduced dimensions because of the uncompensated moments, especially at the surfaces.[5] Bulk NiO is a compensated AF with $T_N\approx 523$ K,[6] which is well above room temperature. This fact makes it a material of special interest for room temperature applications in spin-valve devices that require an antiferromagnet/ferromagnet interface.[1,2] Several studies have reported the presence of size-dependent uncompensated moments in NiO NPs, the origin of which is not yet completely understood.[7–9]

One of the important issues in NP magnetism is the effect of interparticle interaction on the various measured properties.[10–16] In this article, we follow up on our recent work in the oleic acid (OA)-coated NiO NP to reduce the interparticle interaction and compare the measured properties of the coated versus uncoated particles so as to determine the effects of interparticle interactions.[17] Here we report the results on the variations of the measured blocking temperature T_B with the measuring frequency f for the coated and uncoated 5-nm nanorods of NiO. The analysis of the results shows that the data fit well with the Vogel-Fulcher law:[11,18]

$$f = f_0 \exp[-E_a/k(T_B - T_0)], \tag{1}$$

with $T_0=0$ K for the coated and $T_0=162$ K for the uncoated particles. The details of these results, their discussion, and analysis are presented.

II. EXPERIMENTAL SETUP

In a recent paper,[17] we have described the procedures for the synthesis as well as the transmission electron microscopy characterization of the samples of the NiO nanorods (average diameter $\approx$5 nm, average aspect ratio $\approx$12) investigated here. Therefore, these details are omitted here. Measurements of the ac magnetic susceptibilities χ' and χ'' were carried out using commercial superconducting quantum interference device magnetometers in 7 Oe amplitude of the ac field but zero dc magnetic field. The data above 1 kHz were taken at Boise State University, whereas the data at the lower frequencies were taken at West Virginia University.

III. RESULTS AND DISCUSSION

Figure 1 shows the plots of χ' and χ'' against temperature for the uncoated sample. A similar plot for the coated 5 nm NiO nanorods is shown in Fig. 2. It is noted that χ' peaks at a temperature higher than that for χ'' for both samples. A closer examination of the data shows that the position of the peak in χ'' nearly coincides with the peak position in $d(\chi')/dT$. This is similar to the observation reported in other NP systems.[11,15,19] For both χ' and χ'', the peak positions shift to higher temperatures with increasing frequency f, as expected from Eq. (1) and as observed in other NP systems also. However, there are two noteworthy observations for the coated NP in Fig. 2. First, for lower frequencies, the data in χ'' are too scattered without any district peak, and second, there is a second peak at lower temperatures in both χ' and χ'', which does not shift with changing f. Since the second lower temperature peak is not present in the uncoated NP (Fig. 1), this second peak in the coated particles might be due to the oleic acid coating. The scatter in the χ'' data at lower f might be due to smaller magnitudes of χ''.

From Eq. (1), it follows that

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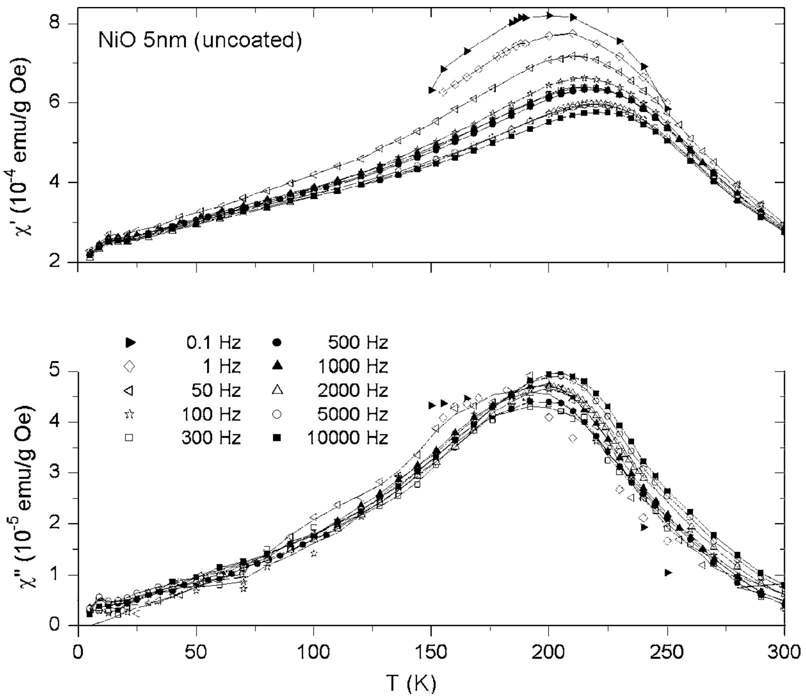

FIG. 1. Temperature variation of χ' and χ'' in uncoated NiO NP at different frequencies is shown. The lines through the points are for visual aid.

$$\ln f = \ln f_0 - (E_a/k)/(T_B - T_0). \qquad (2)$$

For $T_0=0$, Eq. (1) reduces to the Néel-Arrhenius (NA) relation, with

$$\ln f = \ln f_0 - (E_a/k)/T_B. \qquad (3)$$

To check the validity of the NA relation for our system, we first plot $\ln f$ vs $1/T_B$ in Fig. 3 for both the uncoated and coated NP using the peak position in χ' as T_B. The magnitudes of T_B estimated from electron magnetic resonance (EMR) experiments at $f=9.28$ GHz (see Ref. 17) are also included in Fig. 3 after taking into account the effects of the applied static field (which is needed to observe EMR) on T_B. In Ref. 17, it was found that $T_B=118$ K for the coated and $T_B \approx 260$ K for the uncoated NiO, measured in $H \approx 2$ kOe. Using the data of T_B vs H from Ref. 17, it is estimated that in $H=0$, $T_B(0) \approx 220$ K (325 K) for the coated (uncoated) NiO. In Fig. 3, these $T_B(0)$ values at $f=9.28$ GHz are also plotted. The data fit straight lines as expected from Eq. (3), with the following magnitudes for the attempt frequency f_0 and the energy barrier E_a/k: $f_0=5\times 10^{12}$ Hz and $(E_a/k) \approx 1230$ (50) K for the coated NP and $f_0 \approx 10^{28}$ Hz and $(E_a/k) \approx 12\,840$ (1360) K for the uncoated NP.

For the coated NP, these magnitudes of f_0 and E_a/k are in line with the theoretical estimates for AF nanoparticles and the observed values for other systems.[20–22] However, for the uncoated NP, the magnitudes of f_0 and E_a/k just mentioned are too large and unphysical, leading us to conclude that the NA relation [Eq. (3)] is not valid for the uncoated

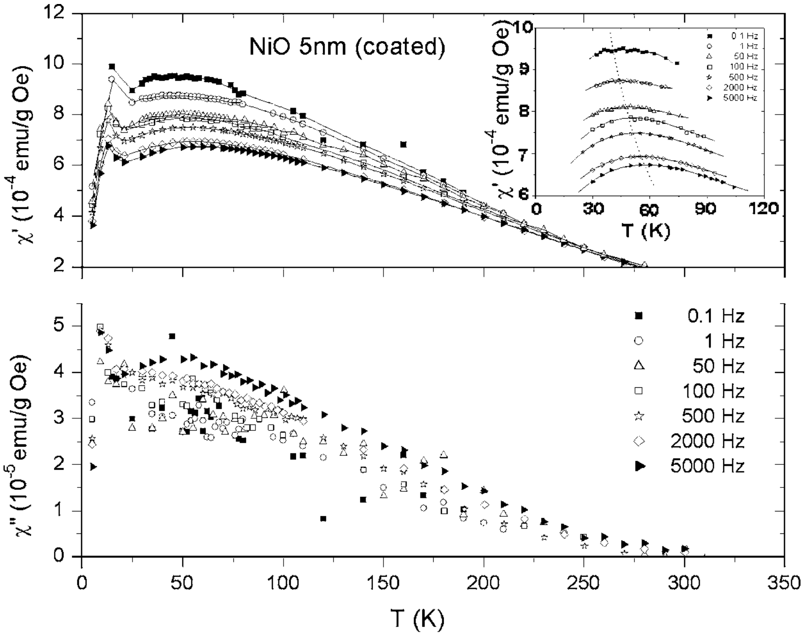

FIG. 2. Same as Fig. 1 except for the coated NiO NP. The inset shows the expanded view for χ', where the solid lines are polynomial fits to the data for accurately determining the peak positions and the dotted line shows the shift in the peak position with f.

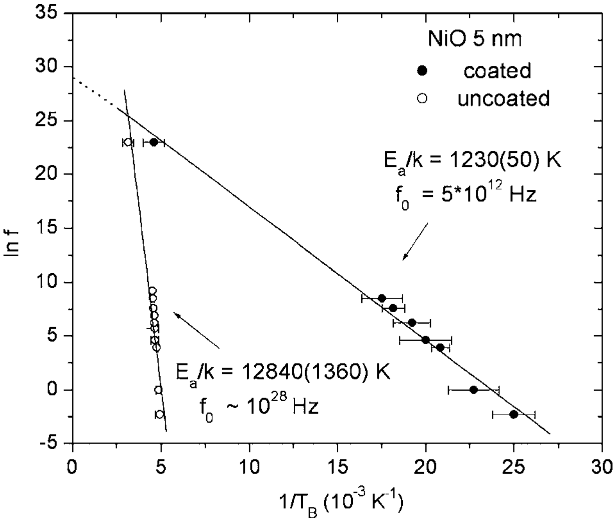

FIG. 3. Plot of $\ln f$ against $1/T_B$ for the coated and uncoated NiO NP. The solid lines are fits to Eq. (3) with parameters given in the plot. The size of error bars for the uncoated sample is within the size of the data points.

NP. In a recent paper,[17] we compared the variations of T_B in these two coated and uncoated NiO NPs with applied dc field H. The relative variation of T_B with H in the coated NP was found to be more pronounced compared to that observed in the uncoated NP. These differences were interpreted in terms of significant interparticle interaction in uncoated vis á vis the coated particles. In the latter, the OA coating significantly reduces the interparticle interaction.

The differences observed in the variation of $\ln f$ vs $1/T_B$ and in the magnitudes of T_B for the coated and uncoated NP (Figs. 1 to 3) are similarly interpreted in terms of the presence of considerable interparticle interaction present in the uncoated NP and its absence in the coated NP. We further show that our data is well described by the Vogel-Fulcher law [Eq. (1)] with $T_0 = 162$ K for the uncoated NP and $T_0 = 0$ K for the coated NP, so that T_0 represents a measure of the interparticle interaction on magnetic relaxation. In Fig. 4, $\ln f$ vs T_B is plotted for both samples, where the solid line represents the Vogel-Fulcher law [Eq. (1)] with the just-described magnitudes of T_0 noted on the graph. The fit of the data to Eq. (3) is very good, except for a slight difference at $f = 9.28 \times 10^9$ Hz for the uncoated NP. This departure may be related to the fact that the Vogel-Fulcher law is valid only for weak interparticle interaction, with departures expected at higher f for strong interparticle interaction.[11,18]

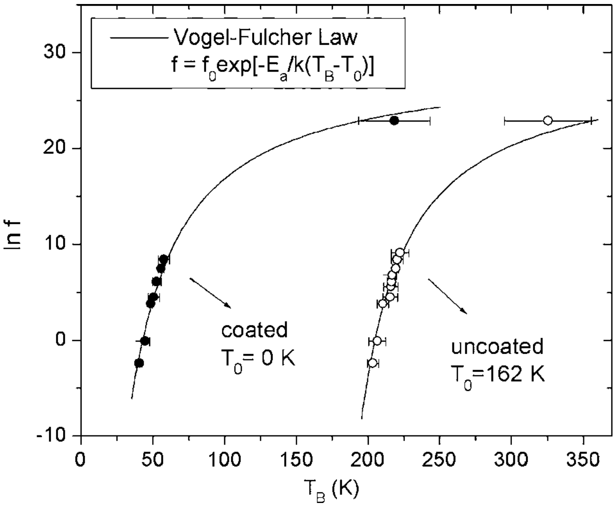

FIG. 4. Plot of $\ln f$ against T_B for the coated and uncoated NiO NP. The solid lines are fits to Eq. (1) with $T_0 = 162$ K for the uncoated and $T_0 = 0$ K for the coated NiO NP. The magnitudes of E_a/k and f_0 used in the fit are the same as in Fig. 3 for the coated particles.

From Eq. (2), it also follows that $T_B = T_0 + (E_a/k)/\ln(f_0/f)$, so that interactions represented by T_0 enhance T_B and the theoretical fits in Fig. 4 for the coated and uncoated particles are simply shifted by T_0. This enhancement of T_B by the interparticle interaction is in agreement with more explicit calculations for the dipolar interparticle interaction.[12] Another view of this enhancement of T_B is that the interactions enhance interparticle correlation and thus effectively increase the particle volume V.[14,16,23] This enhances T_B since $E_a = KV$ (K=the anisotropy constant) and T_B is proportional to V.

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