Magnetic behavior of nanoparticles in patterned thin films

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The magnetic behavior of truncated conical nanoparticles in patterned thin films is investigated as a function of their size and shape. Using a scaling technique, phase diagrams giving the relative stability of characteristic internal magnetic structures of the particles are obtained. The role of the uniaxial anisotropy in determining the magnetic properties of such systems is discussed, and a simple method for establishing its strength is proposed.

Nanolithography techniques combined with material deposition and pattern transfer processes have made possible the production of regular arrays of magnetic particles with individual linear dimensions in the submicron range. A relatively high degree of precision has been achieved in the growth process of these structures, allowing the control of both the particle size and the spacing between them. The samples thus produced exhibit rather interesting properties, and show great potential for technological applications.\textsuperscript{10}

Electrodeposition, and evaporation followed by lift-off are the two methods that have been most widely used in the preparation of this sort of arrays or patterned thin films.\textsuperscript{3,4} The first is suitable for producing cylindrically shaped particles in which the ratio $R$ between their height $H$ and diameter $D$ of the bottom basis (aspect ratio) is bigger than 1. On the other hand, evaporation and lift-off process is more suitable for the production of periodic arrays covering large areas, which may reach several square cm\textsuperscript{1}. Taking under consideration its low cost of production, this method is seen as quite convenient for commercial applications. Particles produced by evaporation have the shape of truncated cone\textsuperscript{1} which can be characterized by three parameters, namely $H$, $D$, and the ratio $\zeta = (D-d)/2H$, where $d$ is the diameter of the top basis.

It is generally accepted that particles of ferromagnetic materials with linear dimensions of the order of $10^2$ nm or less are too small to accommodate a domain wall. As a consequence, one could envisage the production of arrays of single domain ferromagnetic particles, which could be used in the design of spintronic devices or as magnetic media for high density recording\textsuperscript{11,12}. In recent years, several groups have concentrated their attention on the magnetic behavior of nanosized particles\textsuperscript{3,5,10}. It has been found that the internal magnetic structure of sufficiently small particles is indeed rather close to that of a ferromagnet monodomain. Larger particles, however, may also exhibit vortex-like configurations, depending on their size and shape. Clearly, the determination of the conditions under which these distinct magnetic configurations or phases are found is of great relevance for practical applications. However, from the experimental point of view, this is not a simple task since transitions between magnetic configurations as the shape or size of the particle changes are not sharp\textsuperscript{9,10}. Hence, theoretical investigations of the magnetic behavior of such nanoparticles become highly desirable.

In this paper, we focus our attention on patterned thin films produced by evaporation followed by lift-off. The systems we have in mind are Ni films, which have been extensively studied in recent years\textsuperscript{3,11,12}. We aim at determining, for various shapes and sizes of the particles, the configuration of lowest energy out of three characteristic ones. These configurations are: (I) ferromagnetic with the magnetization parallel to the cone basis (in-plane); (II) ferromagnetic with the magnetization perpendicular to the cone basis (out-of-plane); (III) vortex-state with the magnetic moments laying parallel to the cone basis. We shall deal with the situation in which the distance $L$ between the particles in arrays is large enough (i.e. $L > D$) for the interaction between them to be safely neglected\textsuperscript{6}. In such cases, the magnetic structure of each particle is determined solely by the internal interactions and its magnetic moments are subjected to, namely the short-ranged exchange interaction, the classical dipolar interaction (which is responsible for the shape anisotropy), and the magneto-crystalline anisotropy. It is worth mentioning that transmission electron microscopy has clearly established that both Ni and Co particles in samples prepared by evaporation are polycrystalline, with elongated columnar structures\textsuperscript{13}. Micromagnetic calculations of the remanence in those particles indicate that the inclusion of an out-of-plane crystalline anisotropy, in addition to the magnetocrystalline anisotropy, is necessary in order to reproduce the experimental data\textsuperscript{14}. The importance of the uniaxial anisotropy in stabilizing configuration II is discussed towards the end of this article.

Thus, we consider truncated conical Ni nanoparticles consisting of $N$ magnetic moments occupying sites of an underlaying fcc structure. We assume that the cone axis (growth direction) is in the (111) direction\textsuperscript{3}. For fixed
The phase diagram giving the relative stability of the three configurations described above is obtained by comparing the corresponding total energies at each point in the $D\times H$ plane. For each configuration $\{\vec{m}_i\}$ of the magnetic moments, it is given by

$$E_{tot}(\{\vec{m}_i\}, \{J_{ij}\}, K) = \frac{1}{2} \sum_{i \neq j} [E_{ij} - J_{ij} \vec{m}_i \cdot \vec{m}_j] + U_K,$$

where $E_{ij}$ is the dipolar interaction between magnetic moments $\vec{m}_i$ and $\vec{m}_j$, $J_{ij}$ is the exchange coupling, and $U_K$ is the crystalline uniaxial anisotropy term given by $U_K = -K \sum_i \cos^2 \theta_i$. Here, $\theta_i$ is the angle between $\vec{m}_i$ and the cone axis. For the systems under consideration, we take the lattice parameter $a_0 = 3.52 \text{ Å}$, $\mu_i = \mu = 0.62 \mu_B$, and $J_{i,j} = J = 0.9 \text{ mK for first nearest-neighbors}$, and zero otherwise. Estimates of the anisotropy constant $K$ fall in the range $1.8\times10^5 - 2.6\times10^5 \text{ erg/cm}^2$ [3,11], so we take for the moment the value $2 \times 10^5$ [3].

The calculation of the above expression is a well-defined problem. However, with the present day standard computational facilities, it becomes rather time-consuming as soon as the number $N$ of magnetic moments assumes values comparable to those in real systems, i.e. $10^8$ or even $10^9$. As a consequence, the computational effort necessary for obtaining a phase diagram in which particles in such size range are included turns out to be prohibitively large. Nevertheless, it can be significantly reduced with the use of a scaling technique recently proposed and used to study the magnetic properties of cylindrically shaped particles [13]. Such technique involves two steps: (a) calculations are performed for systems with the exchange interaction $J$ scaled down by a factor $x < 1$; (b) the $H$ and $D$ axes in the resulting phase diagrams are scaled up a factor $(1/x)^\eta$, where $\eta$ is a positive constant (see below).

In which follows, we first demonstrate that the scaling technique can be also used to obtain the phase diagram of truncated conical particles, even in the presence of a strong uniaxial anisotropy, and show how $\eta$ is obtained. Having done this, we use such technique to obtain phase diagrams for different values of $\zeta$. Results are compared to existing experimental data. Finally, we comment on the effects of the uniaxial anisotropy on the magnetic behavior of those particles.

Fig.(1) shows results for cones with $\zeta = 0.3$, and scaling factors $x = 0.010$ (squares), 0.015 (circles), and 0.020 (triangles). Dashed line corresponds to the maximum height $H_{max} = D/2 \zeta$ one particle can reach for each diameter $D$. As in the case of cylinders [13], the curves in the three phase diagrams have similar shapes, with a triple point that moves along a common line as $x$ is varied. From the plotting of the height $H_t$ at the triple point as a function of $x$, we find that these two quantities are related by the equation $H_t \approx A x^{\eta}$, with $A \approx 74.4 \text{ nm}$ and $\eta \approx 0.55$. The uncertainty in the values of both $A$ and $\eta$ is smaller than 2%. Within the numerical accuracy of the present calculations, the value of $\eta$ that we obtained in this case coincides with that for cylindrically shaped particles [13]. It means that $\zeta$ is either independent of $\zeta$ or has a rather weak dependence on such parameter. This point has been confirmed by the calculations we have performed for other values of $\zeta$. Having determined the value of $\eta$, we then scale up the three diagrams by dividing the $D$ and $H$ axes by the corresponding $x$ to power $\eta$. Results are shown in Fig.(2), where all points fall on a single set of lines, representing the phase diagram for the full strength of the exchange interaction ($x = 1$). We point out that the slope of the line separating phases I and II gives the critical value ($R_c$) of the aspect ratio $R = H/D$, which depends on the strength of the uniaxial anisotropy. We shall return to this point below.

![Phase diagrams](image)

**FIG. 1.** Phase diagrams giving the relative stability of the in-plane ferromagnetic (I), out-of-plane ferromagnetic (II), and vortex-like (III) configurations of truncated conical Ni particles with $\zeta = 0.3$, for scaling factor $x = 0.01$ (squares), 0.015 (circles), and 0.02 (triangles). Dashed line gives the maximum height $H_{max}$ of the particles as a function of the diameter $D$ of their basis, for the specified value of $\zeta$. The inset illustrates the geometry of the particle under consideration.

We are now in position to examine the phase diagrams for different values of $\zeta$ and full strength of the exchange coupling. Fig.(3) shows results for $\zeta = 0$ (squares), 0.2 (triangles), and 0.4 (circles). Dashed and dotted lines correspond to $H_{max}(D)$ for $\zeta$ equals to 0.2 and 0.4, respectively. We notice that as $\zeta$ increases from zero, that is to say, as the shape of the particles deviates from that of a cylinder, changes in the lines separating the three phases become less pronounced. In fact, the lines for $\zeta = 0.3$ would appear in between those for 0.2 and 0.4, and have been eliminated from the figure for the sake of clarity. These results are confirmed by the experimental data of Ross et al. [13] for Ni samples. These authors have investigated the magnetic behavior of patterned thin films consisting of truncated conical particles with $75 \AA \leq D \leq 122 \AA$, $0.43 \leq R \leq 1.21$, and $0.14 \leq \zeta \leq 0.35$. They concluded that, independently of the value of $\zeta$, transitions from configuration I to II occurs at a critical aspect ratio $R_c = 0.65$, which is in good...
agreement with the phase diagrams in Fig.(3). Indeed, according to our results, for $\zeta > 0$, transitions between these two phases occur for $R_c \approx 0.61$.

Although the boundaries between phases in the diagrams in Fig.(3) do not change much as $\zeta$ is varied, the extent of region II (corresponding to the out-of-plane configuration) is reduced as $\zeta$ increases. Such region is bounded from above by the line $H_{\text{max}} = D/2\zeta$, which means that, for a fixed value of the uniaxial anisotropy constant $K$, a critical value $\zeta_c = 1/2R_c$ exists, above which phase II cannot be observed. However, we expect the uniaxial anisotropy to favor the out-of-plane configuration. Thus, $R_c$ should decrease as $K$ increases. This is confirmed by the curves in Fig. (4), where $R_c$ is plotted as a function of $K$ for different values of $\zeta$, namely zero (squares), 0.2 (triangles), and 0.4 (full circles). We find that for the whole range of values of $\zeta$ considered here, the relation between $R_c$ and $K$ is not much sensitive to that parameter. Indeed, for $\zeta \geq 0.2$ and $R_c < 0.7$, the curves can be regarded as independent of $\zeta$. Apart from confirming the intuitive idea that a strong uniaxial anisotropy is necessary for stabilizing the out-of-plane configuration in particles with large $\zeta$, Fig.(4) provides us with an extremely simple and practical method for determining the value of $K$ in truncated conical particles. The reason is the fact that the computational effort necessary for obtaining such curve is minimum. In fact, since the border line between phases I and II, for fixed values of $K$ and $\zeta$, is a straight line, its slope (that is to say, $R_c$) can be obtained from calculations for small particles. Thus, as a general procedure, the strength $K$ of the uniaxial anisotropy necessary for obtaining the whole phase diagram can be immediately determined from a similar graph and the measured value of critical aspect ratio $R_c$, without much computational effort.

In conclusion, we have investigated the magnetic behavior of truncated conical particles, which exhibit a strong uniaxial anisotropy along the cone axis. We have shown that even in such cases, the phase diagrams giving the relative stability of three characteristic arrangements of the magnetic moments within each particle can be obtained using a scaling method recently proposed. We have found that the stability of the ferromagnetic out-of-plane configuration strongly depends on the strength $K$ of the uniaxial anisotropy, particularly when the shape of the particles deviates significantly from that of a cylinder (i.e. for large $\zeta$). On the basis of our results, we have proposed a simple method for determining the value of $K$ from the measured critical aspect ratio $R_c$. We believe the present work represents a relevant contribution to the understanding of the magnetic properties of nanoparticles, and expect it to help the development of new magnetic devices.
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