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Journal of Magnetism and Magnetic Materials 226–230 (2001) 626–629

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

Percolation and magnetism: interplay and relevance

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Abstract

Magnetism of artificial structures, multilayer arrays and granular systems has lately attracted wide attention. On the contrary, the concept of percolation, first introduced in 1957, has scarcely been related to the former, in spite of the fact that it has been applied to many problems in completely different fields. In physics, percolation has been useful to study both macroscopic and microscopic phenomena, such as the electrical conduction of a mixture and the localization and hopping of electrons, respectively. However, recent experimental studies of magnetic granular solids have clearly established a connection between percolation and magnetism. In fact, magnetic anomalies were observed in a percolating system where the coercivity H_c of Ni films exhibits a non-monotonic behavior, related to topological morphology changes, which coincides with the onset of percolation. In this contribution we propose a theoretical model that describes the behavior of the coercivity H_c, of granular Ni films, as a percolation phenomenon. We show that the non-monotonic dependence of H_c, as a function of the occupation probability p, is due to the interplay of percolation and magnetism. Moreover, we also explain the dependence of H_c as a function of external magnetic field orientation.

Keywords: Percolation; Coercivity; Thin films

In physics, percolation has been useful in the study of macroscopic level phenomena, such as the electrical conduction of a mixture, superconductivity, as well as topics at a microscopic level, as classical localization and electron hopping. In particular, granular systems inherently prone to percolation, have experimentally shown magnetic anomalies when the grains are magnetic. Panina et al. [1] found that the effective magnetic permeability of composite materials containing small Fe particles (of 1–2 μm size) tends to zero near the percolation threshold. Xiao and Chien [2] have observed that across the whole volume fraction range, in granular Fe–(SiO_2) solids, magnetic coercivity experiences dramatic variations due to the change of grain size and percolation effects. In thin films the relation between magnetism and percolation has been investigated: for example, the transition from superparamagnetism to ferromagnetism of submonolayer Fe on W (1 1 0). Also, recently Gor'kov and Kresin [3] studied the transition from paramagnetic to conducting ferromagnetic phases of manganites by percolation theory. Moreover, Choi et al. [4] observed that the coercivity H_c of Ni films, whose magnetic and structural properties are found in the literature, exhibits a non-monotonic behavior, related to topological morphology changes, which coincides with the onset of percolation. However, no theoretical ideas have been advanced which directly connect coercivity and percolation.

In this paper, we study the behavior of H_c of a percolating system and its dependence on the direction of the applied field H. We show that H_c, as a function of site occupation probability p, has a non-monotonic dependence due to the fractional variation of the cluster sizes and their topology, as they form during the film deposition process. Moreover, it is shown that this behavior is independent of the growth direction of the magnetic film. It is also established that the symmetry of H_c changes when $p \approx p_{cr}$.

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0304-8853/01/$ - see front matter
PII: S0304-8853(01)00022-1

Our model focuses upon the morphological changes that take place during the growth of a thin film. In the early stage the growth process is characterized by a homogeneous distribution of the small islands. When additional material is deposited these islands grow in size, due to the large mobility of the small droplets, until coalescence sufficient for percolation is achieved. Based on this the coercivity $H_c(p)$, as a function of occupation probability p, can be understood assuming that the magnetization reversal mechanism depends on cluster size and topology:

(i) If the particles are smaller than a certain critical size z_{cr}, of the order of a domain wall, H_c is large, due to the fact that for such particles wall nucleation is energetically unfavorable. Consequently, the magnetization reversal proceeds, for instance, through coherent rotation of the spins [5,6].
(ii) For simply connected particles, whose sizes are greater than z_{cr}, the coercive field H_c is smaller than the former since now a domain wall [7] can nucleate. Thus, the magnetization reversal is achieved by processes that require less energy, predominantly by domain wall nucleation and displacement.
(iii) However, larger clusters may be multiply connected and the domain walls might get pinned by the voids, especially if these are of considerable size. Thus, when the fraction of voids is large enough the inversion mechanism depends both on cluster size and topology, since domain wall formation is likely but their propagation is hindered by pinning to the voids that are present.

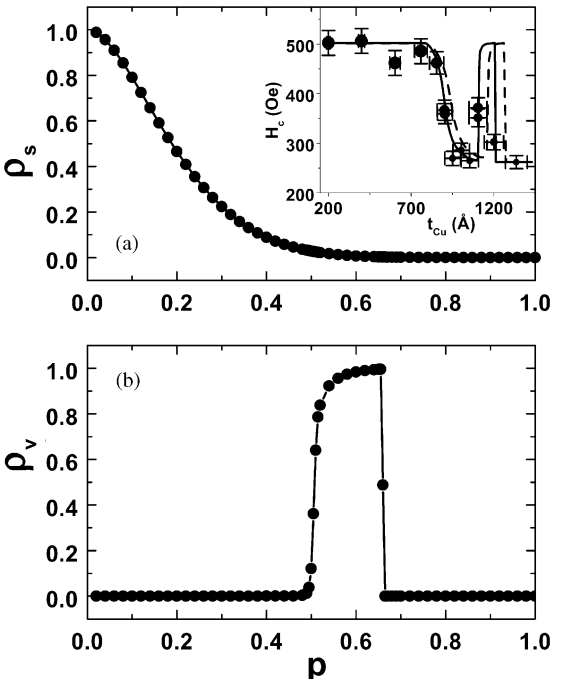

Fig. 1. (a) Small cluster fractional area as a function of p, which is related to the Cu thickness. (b) Fractional area of large clusters with voids as a function of p. The insert is the coercivity H_c (Oe) as a function of Cu thickness t_{Cu} (Å) observed experimentally at 5 K. The continuous (dashed) line is obtained using our model for a 1000×1000 triangular (square) lattice. The circles are experimental data from Choi et al.

To determine the coercivity we introduce the reduced magnetization $M_\mu(H)$, which corresponds to the quotient between the actual and the saturation magnetization of a single cluster. The indices $\mu = s$, b, and v label clusters of types (i)–(iii), as described above. Thus, the analytic expression for the film magnetization $M(H)$ is

$$M(H) = \rho_s M_s(H) + \rho_v M_v(H) + (1 - \rho_s - \rho_v) M_b(H), \quad (1)$$

where ρ_μ is the fractional area occupied by each type of clusters. Consequently, we obtain the coercivity H_c solving the equation $M(H_c) = 0$. A relevant issue in this context is the fact that, in our formalism, we have ignored the dipolar interaction between Ni clusters. In granular systems these interactions are small, but may play a role when computing the full magnetization loop. This is supported by the experiments reported in Ref. [2], which confirm that the only consequence of increasing the cluster concentration is a slight increase of the squareness of the hysteresis loop. Moreover, experiments also confirm that the temperature dependence of H_c is the one predicted for non-interacting particles by Kneller and Lubensky [8], given by $H_c \propto (1 - \sqrt{t})$, where t is a reduced temperature. But, in this paper we focus our efforts on the evaluation of H_c. Thus, the dipolar interaction of each cluster with all the rest, which have an average magnetic moment rigorously equal to zero, vanishes identically when $M(H_c) = 0$ is satisfied.

The fractional areas ρ_s and ρ_v, of the different type clusters, are obtained by using the Monte Carlo technique. The criterion we used in our algorithm to classify a cluster as belonging to type (iii) as defined above (large cluster with voids) is: a cluster belongs to class (iii) if f, defined as the ratio between the number of empty sites inside a cluster and the number of occupied sites in the same cluster, is larger than a critical value f_{cr}.

In Fig. 1, we display $\rho_s(p)$ and $\rho_v(p)$ for a 1000×1000 triangular lattice. As expected, the fractional area of small clusters $\rho_s \approx 1$ for $p \ll 1$, and decreases to $\rho_s \approx 0$ as p grows. On the other hand, the fractional area $\rho_v \approx 0$ for $p \ll 1$ and grows very fast when the percolation threshold is approached. Finally, ρ_v undergoes an abrupt drop when the dominant contribution is due to the formation

of large clusters without voids. The kinks of ρ_v arise from finite size effects.

The coercivity as a function of p is displayed in the inset of Fig. 1(a), where we have applied the model to the description of the H_c behavior of a Ni film deposited on Cu, as reported in Ref. [4]. This functional dependence was obtained using the following simple assumption that only two types of islands are important, as far as magnetic properties are concerned. They only differ in their switching mechanisms; the clusters of types (i) and (iii), with broader loops, switch by magnetization rotation, while the larger type (ii) ones, with narrower loops, only switch after they nucleate a domain wall. We have adopted the linear relation $t_{Cu} = 780\,p + 650$ between p and the Cu film thickness t_{Cu}, measured in Å, to fit the experimental data of Choi et al. [4]. This expression was derived fitting two values: (i) the minimum of H_c; and (ii) the value for which H_c versus p, in the inset of Fig. 1, ceases to be flat as a function of t_{Cu}. $M_s(H) = M_v(H)$ and $M_b(H)$ in the region of interest were fitted by hyperbolic tangents.

We stress the fact that the main features of $H_c = H_c(p)$ do not depend on the detailed form of $M_s(H)$ and $M_b(H)$. In fact, we found no significant changes in the behavior of H_c for two extreme cases: a rectangular hysteresis loop, and assuming linear fits of $M_s(H)$ $[M_b(H)]$ which go through H_s $[H_b]$ and M_r. Always (i) if $p \ll p_{cr}$ then H_c versus p is flat, since a single switching mechanism dominates and thus $H_c = H_s$; (ii) independent of the form of $M_s(H)$ and $M_b(H)$ a minimum is obtained around the percolation threshold p_{cr}; and (iii) a maximum of H_c appears for $p > p_{cr}$, which decays to the minimum value of H_c as p grows. We employed the value $f_{cr} = 0.5$ in the inset Fig. 1(a); changing the latter only narrows or enhances the width of the $p > p_{cr}$ maximum. Moreover, the fact that the H_c versus t_{Cu} displayed in the insert has the same general shape both for the triangular and square lattices is a strong indication that this behavior is universal, i.e. does not depend on the growth direction of the thin film.

On the other hand, the angular dependence of the coercivity is also modified by percolation. For the small clusters we consider a single domain thin film, with its surface oriented in the [1 1 1] direction. Thus, only three terms contribute to the total energy (per unit volume): the magnetocrystalline anisotropy E_K, the Zeeman term and the magnetostatic energy. E_K determines the easy magnetization directions, which lie along the diagonals of the cubic cell (in the [1 1 1], $[\bar{1}\,1\,1]$, $[1\,\bar{1}\,1]$, $[1\,1\,\bar{1}]$ directions).

The coercivity H_c is obtained setting the first and second derivatives of the total energy equal to 0. The results of these calculations are displayed in Fig. 2. The parameters used in the computations are the saturation magnetization $M_0 = 500\,\mathrm{G}$, the effective anisotropy constant $K = 2022\,\mathrm{erg/cm^3}$ and the demagnetization factor $N = 0.05$. Assuming a Gaussian distribution of the anisotropy axes of magnetic clusters around the mean orientation, it is found analytically that K is reduced in good agreement with the fits obtained from Fig. 2.

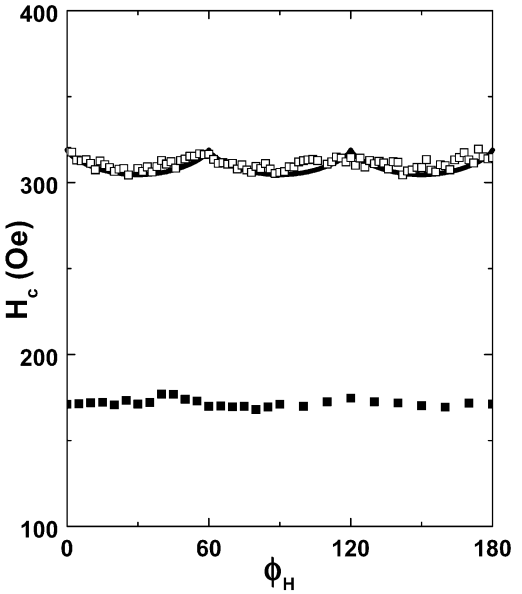

Fig. 2. Coercivity H_c as a function of the magnetic field direction ϕ_H. The continuous line is the theoretical six-fold fit of the experimental data of Pechan et al. (unpublished) depicted, for $p \ll p_{cr}$, by the open squares. The full squares illustrate the isotropic dependence of H_c versus ϕ_H for $p \approx p_{cr}$, which corresponds to $t_{Cu} = 100\,\mathrm{nm}$, measured at room temperature.

H_c has a six-fold symmetry, related to the fact that we are dealing with the (1 1 1) face. However, an additional condition is also required that the spins adopt a small out of the (1 1 1) plane component, since otherwise the six-fold symmetry is quenched. Thus, as the system is rotated around the [1 1 1] axis (perpendicular to the (1 1 1) plane) the spins are barely drawn towards the easy [1 1 1] axis. All this is in satisfactory agreement with experiment.

On the other hand, when the percolation threshold p_{cr} is reached, the system behavior is dominated by the large clusters. In that case, the dipolar interaction becomes larger and has the effect of aligning in plane the total magnetization of the cluster. This favors isotropic in plane magnetization, which was verified experimentally as shown in Fig. 2. A very small orientation dependence of the coercive field is observed beyond the percolation limit which may be understood as follows: reversal in that limit takes place by domain wall nucleation followed by wall motion. Nucleation may be sensitive to orientation, but since it takes place in only a small fraction of a cluster, and since the rate of wall motion is presumably much less sensitive to orientation, the overall anisotropic effect is very small.

In conclusion, we have developed a model which displays and provides an understanding for the relation

between coercivity H_c and percolation. This relation is a consequence of morphological changes in the magnetic system and was recently observed experimentally [4]. We have also shown that, when a FCC film is grown in the [1 1 1] orientation, a six-fold symmetry of H_c for $p < p_{cr}$ exists, which crosses over to an isotropic behavior for $p \approx p_{cr}$.

MK and JML acknowledge support by the *Fondo Nacional de Investigaciones Científicas y Tecnológicas* (FONDECYT, Chile) under Grant No. 8990005. IKS acknowledges partial support by AFOSR-MURI at UCSD and HS by the Center for Magnetic Recording Research at UCSD.

References

[1] L.V. Panina, A.S. Antonov, A.K. Sarychev, V.P. Paramonov, E.V. Timasheva, A.N. Lagarikov, J. Appl. Phys. 76 (1994) 6365.
[2] G. Xiao, C.L. Chien, Appl. Phys. Lett. 51 (1987) 1280.
[3] L.P. Gor′kov, V.Z. Kresin, JETP Lett. 67 (1998) 985.
[4] J.M. Choi, S. Kim, I.K. Schuller, S.M. Paik, C.N. Whang, J. Magn. Magn. Mat. 191 (1999) 54.
[5] E. Stoner, E.P. Wohlfarth, Philos. Trans. R. Soc. London Ser. A 240 (1948) 74.
[6] E.H. Frei, S. Shtrikman, D. Treves, Phys. Rev. 106 (1957) 446.
[7] H. Suhl, H.N. Bertram, J. Appl. Phys. 82 (1997) 6128.
[8] E.F. Kneller, F.E. Luborsky, J. Appl. Phys. 34 (1963) 656.