[Article] Magnetic hysteresis in granular CuCo alloys

Original Citation:
P. Allia; M. Coisson; P. Tiberto; F. Vinai; M. Knobel (1999). *Magnetic hysteresis in granular CuCo alloys.* In: JOURNAL OF APPLIED PHYSICS, vol. 85, pp. 4343-4345. - ISSN 0021-8979

Publisher:
AIP

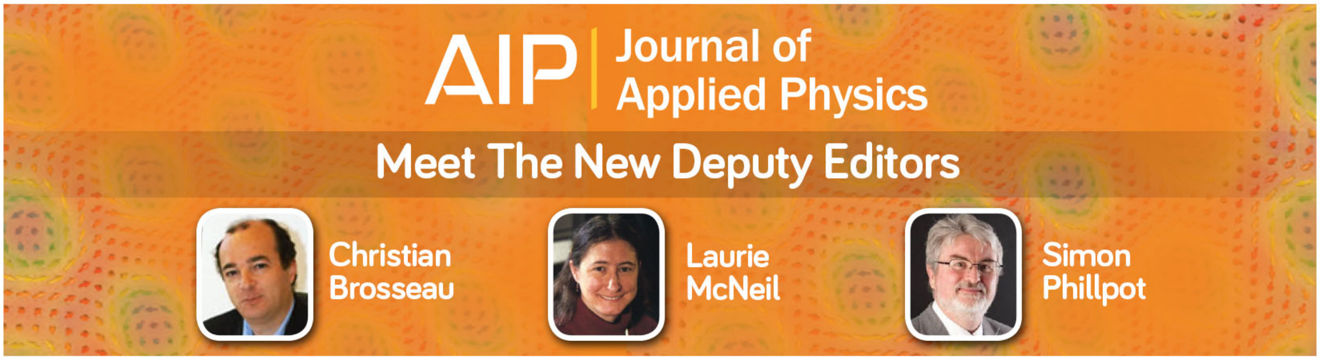
Published version:
DOI:10.1063/1.370362

AIP | Journal of Applied Physics

Magnetic hysteresis in granular CuCo alloys

P. Allia, M. Coisson, P. Tiberto, F. Vinai, and M. Knobel

Citation: Journal of Applied Physics **85**, 4343 (1999); doi: 10.1063/1.370362
View online: http://dx.doi.org/10.1063/1.370362
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/85/8?ver=pdfcov
Published by the AIP Publishing



JOURNAL OF APPLIED PHYSICS VOLUME 85, NUMBER 8 15 APRIL 1999

Magnetic hysteresis in granular CuCo alloys

P. Allia[a)]
Dipartimento di Fisica and INFM, Politecnico di Torino Corso Duca degli Abruzzi 42,
I-10129 Torino, Italy

M. Coisson, P. Tiberto, and F. Vinai
IEN Galileo Ferraris and INFM C.so M. D'Azeglio 42, I-10125 Torino, Italy

M. Knobel
Universade Estadual de Campinas, IFGW-LMBT Caixa Postal 6165, 18083-970 Campinas,
Sao Paulo, Brazil

Room-temperature hysteresis loops of granular $\mathrm{Cu}_{100-x}\mathrm{Co}_x$ alloys $(5 \leqslant x \leqslant 15)$ obtained by planar flow casting in air and submitted to proper annealing treatments have been measured up to a field of 10 kOe by means of a vibrating sample magnetometer. In major loops $(|H_{\mathrm{vert}}| = 10\,\mathrm{kOe})$, the reduced remanence-to-saturation ratio $m_r = M_r/M_s$ and the coercivity H_c measured on all studied materials appear to be related by an almost linear law of the type $m_r \approx 1/3\ (\mu H_c/kT)$, μ being the average magnetic moment on Co particles. A similar relation is also observed on minor symmetrical loops $(100\ \mathrm{Oe} \leqslant |H_{\mathrm{vert}}| \leqslant 9\ \mathrm{kOe})$. The observed results are accounted for by a model which considers the hysteresis as originating by magnetic interactions among nearly superparamagnetic Co particles.
[S0021-8979(99)51408-4]

Granular magnetic systems, where nanometer-sized, nearly superparamagnetic particles of a ferromagnetic metal are dispersed in a nonmagnetic metal are still attracting a considerable interest from the fundamentalist's viewpoint.[1–3] The $\mathrm{Cu}_{100-x}\mathrm{Co}_x$ system can be easily obtained in ribbon form over a wide composition range $(0 < x \leqslant 20)$ by planar flow casting. A wide variety of granular structures characterized by rather different average values of the particle sizes are obtained by varying Co content and/or annealing the as-cast material.[3,4] Generally speaking, the magnetization curves of all CuCo alloys exhibit nonzero coercivity and remanence values, which increase with increasing mean Co particle size. Magnetic hysteresis loops measured on Cu–Co systems appear to simultaneously exhibit superparamagnetic characters (such as the unsaturating behavior of the magnetization curves up to 5 T at room temperature)[4] and hysteretic features. The hysteretic behavior of a granular magnetic system could be related either to the presence of a fraction of blocked particles whose size exceeds the critical size for superparamagnetism at the measurement temperature,[5] or to the existence of interactions among superparamagnetic particles.[6] In this work, a detailed experimental study of the hysteretic properties of granular CuCo alloys is performed. The observed relationship between magnetic remanence and coercivity is definitely not compatible with a blocked-particle view. On the contrary, the experimental data are consistent with a simple model which considers the hysteresis as originated from magnetic interactions among nearly superparamagnetic Co particles.

Continuous ribbons of $\mathrm{Cu}_{100-x}\mathrm{Co}_x$ $(x = 5,\ 10,\ 15\ \mathrm{at.}\,\%)$ were obtained by planar flow casting in He atmosphere on a CuZr wheel. Different ribbon strips of the three compositions (width 5×10^{-3} m, thickness $4-6 \times 10^{-5}$ m) were submitted to direct current (dc) joule heating[7] in vacuum, in order to produce nanometer-sized Co particles. The room-temperature magnetic moment was measured using a vibrating-sample magnetometer (VSM) (LDJ, model 9500) under an applied field M varying between ± 10 kOe. Major symmetric hysteresis loops were obtained starting from the demagnetized state and cycling up to a maximum applied field $H_{\mathrm{vert}} = \pm 10\,\mathrm{kOe}$, with a field step of ± 100 Oe, while minor symmetric loops were measured with $|H_{\mathrm{vert}|}$ ranging in the interval 100 Oe–9 kOe, and suitably decreasing the field step in order to keep the number of experimental points constant. Major anhysteretic curves were measured starting from the demagnetized state up to $H_{\mathrm{vert}} = \pm 10\,\mathrm{kOe}$.

As known,[3] the anhysteretic magnetization curves are always well described by a superposition of a few Langevin functions characterized by different values of the magnetic moment. Such a fit allows one to obtain a value of the average magnetic moment in the system, as well as of the spread of the magnetic moment distribution.[3] The average particle size obtained using this procedure (3–4 nm) is consistent with results taken from different magnetic measurements on the same systems (e.g., field-cooled and zero-field cooled magnetization curves),[8] and from structural data analysis (e.g., small-angle neutron scattering),[9] when available. The saturation magnetization M_s cannot be determined from the largely unsaturating experimental anhysteretic curves, and was obtained in the present case by extrapolating the superposition of two Langevin fitting functions.

A first analysis of the hysteretic character of the magnetization curves may be performed by reporting for each sample the reduced magnetic remanence M_r/M_s (m_r) along with the coercivity H_c, both measured at room temperature on major hysteresis loops. The analysis was performed on about 40 different samples of the $\mathrm{Cu}_{100-x}\mathrm{Co}_x$ system $(x = 5,\ 10,\ 15\ \mathrm{at.}\,\%)$, both in as-cast condition and submitted to joule heating for 60 s under various electrical-current densities.

[a)] Electronic mail: allia@omega.ien.it

0021-8979/99/85(8)/4343/3/$15.00

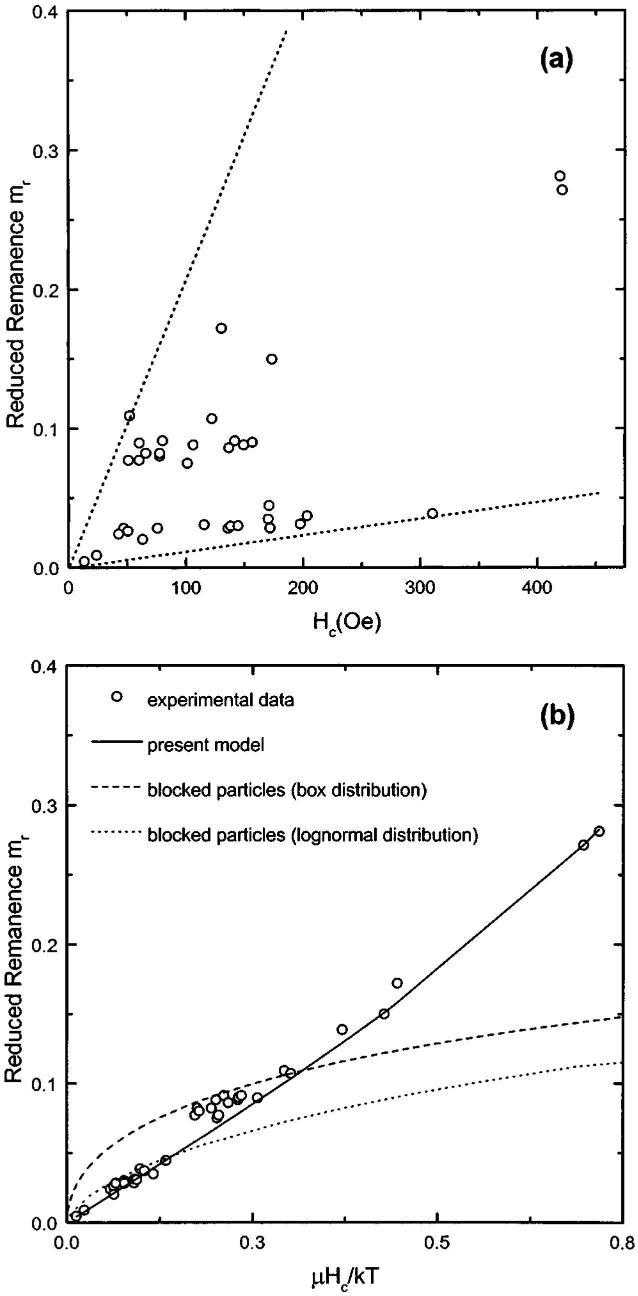


FIG. 1. (a) Reduced remanence m_r vs coercivity H_c for various $Cu_{100-x}Co_x$ alloys ($x=5$, 10, 15 at. %) submitted to different anneals; (b) same data as in (a) (open circles) plotted as functions of $\mu H_c/kT$ (μ: average particle moment); present model (full line); simulation for blocked particles with particle size distributed according to a box function (dashed line) and a log-normal curve (dotted line).

The $m_r(H_c)$ data is reported in Fig. 1(a). At a first sight, no evident relationship between the two quantities seems to exist, although all data are contained in the portion of the (m_r, H_c) plane defined by the two dotted lines. However, a well-defined universal curve emerges by plotting m_r as a function of the ratio $\mu H_c/kT$, μ being the average magnetic particle moment determined for each sample from the anhysteretic curve, k the Boltzmann constant, and T the absolute temperature [see Fig. 1(b)]. The behavior of m_r vs $\mu H_c/kT$ follows a nearly linear law with angular coefficient close to 1/3. Such a result is definitely incompatible with a model invoking the existence of blocked particles.[10] In that framework, the coercivity and reduced remanence are related to the presence of a tail towards large sizes of the distribution function $p(L)$ of particle size L, according to the expressions:

$$H_c = H_c^{\mathrm{MAX}} \int_{L_c}^{\infty} \left[1 - \left(\frac{L_c}{L} \right)^{1/2} \right] p(L)\, dL,$$
$$m_r = 0.5 \int_{L_c}^{\infty} p(L)\, dL, \tag{1}$$

H_c^{MAX} being equal to $2K/M_s$, where K is the dominant magnetic anisotropy constant of Co particles. The expression for H_c above the critical size for superparamagnetism L_c is well known.[5] The factor one-half appearing in the expression for m_r closely approximates the Stoner–Wohlfarth prediction[10] for an assembly of blocked particles with random anisotropy axes. We have simulated the relationship between m_r and H_c for a system of blocked particles using two different size distribution functions: a simple box distribution [which allows an analytical expression for $m_r(H_c)$ to be obtained] and a log-normal curve. The critical size L_c was set to 6.5 nm, appropriate to a face-centered-cubic (fcc) Co alloy with slightly elongated Co particles. For the box function, the minimum particle size was $L_{\min}=1.5$ nm, the upper limit $L_{\max}$ varying from 6.5 to 9.0 nm. The mean value of the log-normal size distribution was varied from 2 to 7 nm, keeping constant the standard deviation $\sigma_L = 1$ nm. Independently of the distribution function, the blocked-particle model fails to account for the experimental data, both simulations exhibiting a definite downward curvature [dashed and dotted lines in Fig. 1(b)]. Both curves reach the value $M_r/M_s = 0.5$ for $H_c \to \infty$, as predicted by any blocked-particle model.

A mean-field model relating the magnetic hysteresis in granular systems to the effect of long-ranged interactions among localized magnetic moments has been developed. Details will be given in a forthcoming publication; here an outline of the model follows. The magnetic interactions among moments (e.g., those of dipolar type) act to stabilize any magnetic equilibrium state against disturbances related to changes either in the external field H or in temperature T. As a consequence, the isothermal magnetization, measured starting from any nonzero initial value, is observed to follow magnetic-field changes with a certain lag with respect to the prediction for an ideal superparamagnet, therefore exhibiting a hysteretic behavior. This effect is accounted for by introducing a field-dependent memory function $\delta(H)$ in the argument of the Langevin function.

Considering for simplicity a single magnetic moment (extension to distributed moments is straightforward), the reduced hysteresis loop is simply written as:

$$m = L\left[\frac{\mu H}{kT} \pm \delta(H) \right], \tag{2}$$

where L is the Langevin function and the +/− signs refer to the upper/lower branch of the loop. The memory function for a loop starting from a vertex state $(H_{\mathrm{vert}}, m_{\mathrm{vert}})$ is defined as the product of two terms: (a) an interaction field, defined as the root-mean-square (rms) value $\langle H_i^2\rangle^{1/2}$ of the field produced by neighboring dipoles on the ith moment (as usual, this field is considered as a stochastic function of both time and site, with zero average value and nonzero variance); (b) the initial equilibrium magnetization state, expressed by the vertex value $|m_{\text{vert}}|$. Consequently,

$$\delta(H)=\frac{\mu}{kT}H_{\text{int}}. \tag{3}$$

When the coupling among equal magnetic moments randomly distributed in three-dimensional space is dominated by dipolar interaction, the field H_{int} depends on the degree of alignment of moments, being maximum for disordered moments $(m=0)$ and becoming zero when the moments are aligned by the external field $(|m|=1)$. This circumstance is accounted for by introducing a proper cutoff function in H_{int}, obtained by performing an explicit calculation of $\langle H_i^2\rangle^{1/2}$ for the case of pure dipolar interaction among uncorrelated magnetic moments. The result turns out to be:

$$\begin{aligned} H_{\text{int}}&=\tilde{H}_0[3(\langle u^2\rangle-\bar{u}^2)]^{1/2};\\ H_{\text{int}}(m=0)&=\tilde{H}_0;\\ H_{\text{int}}(m=1)&=0, \end{aligned} \tag{4}$$

where $\bar{u}=L(x)$ and $\langle u^2\rangle=1-2L(x)/x$. Introducing Eq. (3) in Eq. (2), one gets the following expression for the reduced remanence:

$$m_r=L[\delta(0)]\cong\frac{1}{3}|m_{\text{vert}}|\left(\frac{\mu}{kT}\right)\tilde{H}_0 \tag{5}$$

while the coercivity is found solving the implicit equation $H_c=(kT/\mu)\delta(H_c)$. One easily gets (to the second order in $\mu H_c/kT$):

$$m_r=\frac{\frac{1}{3}\frac{\mu H_c}{kT}}{1-\frac{3}{10}\left(\frac{\mu H_c}{kT}\right)^2}. \tag{6}$$

The parameter $x_c=\mu H_c/kT$ is often significantly smaller than unity at room temperature (representative values for μ and H_c are $\mu=4-6\times10^{-17}$ emu and $H_c=100-300$ Oe, so that in most cases $x_c\leqslant0.5$). As a consequence, the denominator only introduces a small upward curvature into the linear law. Equation (6), reported in Fig. 1(b) (full line), is in good agreement with the experimental data plotted as functions of the ratio $\mu H_c/kT$, μ being in this case the average particle moment.

The same behavior observed in the case of major hysteresis loops is also found studying minor loops. By changing the vertex field, both m_r and H_c are rescaled in the same way, so the same nearly linear relationship previously discussed is found when plotting m_r vs H_c. Figure 2 shows that the results obtained on two samples differing by composition and magnetic particle structure fall on the universal curve predicted by Eq. (6) (dotted line).

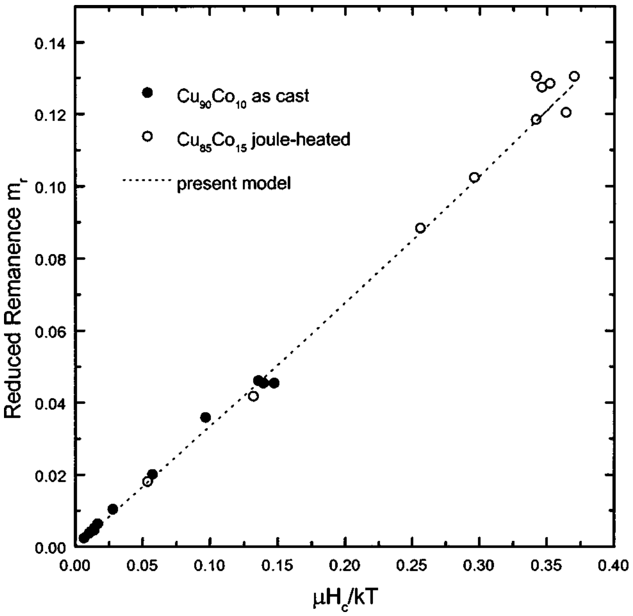


FIG. 2. m_r vs $\mu H_c/kT$ for as-cast $Cu_{90}Co_{10}$ (open circles) and joule-heated $Cu_{85}Co_{15}$ ($I=7.5$ A, $t=60$ s). Dashed line: present model.

In conclusion, this preliminary analysis indicates that the hysteretic behavior of CuCo granular systems is to be explained in terms of interacting magnetic particles rather than of blocked particles. Of course, the validity of the proposed approach must be accurately tested by applying Eq. (2) to the description of all hysteretic phenomena in granular alloys, in order to rule out the hypothesis of blocked-particle hysteresis. In particular, the theory must explain the overall shape of the whole hysteresis loop (and not only the m_r vs H_c relationship), as well as the changes in the hysteresis parameters (m_r, H_c) as functions of measurement temperature. The results appear to be particularly promising and will be the subject of a forthcoming work.

ACKNOWLEDGMENT

This work was partially supported by FAPESP-Sao Paulo (Brazil).

[1] J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).
[2] A. E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).
[3] P. Allia, M. Knobel, P. Tiberto, and F. Vinai, Phys. Rev. B **56**, 15398 (1995).
[4] B. J. Hickey, M. A. Howson, S. O. Musa, and N. Wiser, Phys. Rev. B **51**, 667 (1995).
[5] B. D. Cullity, *Introduction to Magnetic Materials* (Addison–Wesley, Reading, MA, 1972), p. 383.
[6] D. V. Berkov and S. V. Meshkov, IEEE Trans. Magn. **26**, 1804 (1990).
[7] P. Allia, M. Baricco, P. Tiberto, and F. Vinai, Rev. Sci. Instrum. **64**, 1053 (1993).
[8] E. Agostinelli, P. Allia, R. Caciuffo, D. Fiorani, D. Rinaldi, A. M. Testa, P. Tiberto, and F. Vinai, Mater. Sci. Forum **235–238**, 705 (1997).
[9] F. Vinai, P. Tiberto, A. Deriu, F. Malizia, M. Vittori-Antisari, M. Angiolini, and J. S. Pedersen, Mater. Sci. Forum **269–272**, 339 (1998).
[10] E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London Ser. A **240**, 599 (1948).