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Analysis of the Magnetic Anisotropy in Advanced Metal Particles for Recording Media

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The magnetic anisotropy of metal particles with high coercivity, useful in the recording technology, is investigated with measurements of the anisotropy distribution, as obtained from the second derivative of the perpendicular magnetization, and of the transverse susceptibility. Also the temperature dependence of the magnetic properties is studied at the same aim. The relevance of shape anisotropy, and therefore of the particle morphology, in determining the magnetic behavior of the advanced metal particles is expressed. The effect of the particle–particle interactions on anisotropy is studied in aligned samples with different packing densities. The average anisotropy field does not change with packing, which on the contrary affects the width of the anisotropy distribution, which is larger in packed samples, because of the diversified contributions of the interacting particles.

1. Introduction

The evolution of magnetic recording technology to pursue the request of ever increasing information density makes necessary a continuous increase of coercivity and magnetization of particulate media materials, and a decrease of the recorded layer thickness, which means a corresponding decrease of the particle size. The advanced metal particles (AMP) we study in this work have properties which satisfy such requirements: the static coercive field is about 2500 Oe, the specific magnetization 147 emu/g and the acicular particles have a mean length of 85 nm.

Generally, the commercial method of preparation of metal particles starts from acicular particles of goethite, α-FeOOH, which are subjected to the dehydration. The particles are converted to hematite, α-Fe_2O_3, and subsequently to α-Fe metal particles through some reduction steps at various temperatures in presence of hydrogen gas. Finally they are subjected to a passivation treatment in dilute air whose purpose is to coat the particles with a layer of iron oxide and to protect them from further oxidation. In the processes, the particles are treated with aluminum, cobalt, yttrium, neodymium or other elements to prevent sintering, and contemporarily to avoid an excessive deterioration of the particle morphology [1, 2]. A regular and smooth acicular shape is relevant also to the magnetic properties.

As a result of these treatments, the particles have a rather complicated structure with a combination of various elements and compounds, particularly in the surface layer. Also the morphology and the internal structure of the inner metal core cannot be completely known and described. Consequently, even the intrinsic magnetic behavior of these metal particles is not completely understood and needs further study and investigation. The aim of this work is to analyse the magnetic anisotropy of the advanced metal particles, also in order to interpret the origin of their superior macroscopic magnetic properties, particularly of the high coercive field.

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2. Experimental Results and Discussion

With the above introduced advanced metal particles we have prepared aligned assemblies, similar to the coatings of particulate recording media, with a relatively high packing fraction ($p \approx 0.35$). The properties of some AMP samples are reported in Table 1 (samples A, B and C). For sake of comparison, also the data of a sample (D) of previous generation metal particles (MP), which were available in 1990, are reported.

The magnetic anisotropy has been investigated both with a method which deduces the anisotropy field distribution from the measurement of the magnetization curve from saturation to remanence in a direction perpendicular to the easy axis [3] and with the standard method of the transverse susceptibility [4].

In the first method, the hysteresis loop is measured by a VSM in the plane of the sample perpendicularly to the easy magnetization direction. The distribution of anisotropy fields is obtained by

$$P(H_a) = -H\frac{d^2m}{dH^2}\,, \tag{1}$$

where m is the reduced magnetization M/M_S between saturation and remanence. Such curve, when fitted by a suitable cumulative function with high correlation, allows to calculate the second derivative and find the anisotropy distribution through relationship (1). The field value where the curve has its peak is the most probable, or mean, anisotropy field $H_{a,m}$ of the system.

We have measured the transverse susceptibility $\chi(H)$ superimposing an ac field produced by a solenoid which generates a peak field of 5 Oe at a frequency of 50 kHz perpendicularly on the dc field of an electromagnet. Two balance pickup coils situated axially within the solenoid detect the signal amplitude which depends on the susceptibility of the sample at the center of one of them. In order to eliminate any possible influence of the hysteresis of the electromagnet core on the pickup signal, the sample is removed for each value of the dc field and the difference between the pickup signal with the sample and without it is recorded. The obtained $\chi(H)$ curves are practically symmetric with respect to H. The mean anisotropy field of a particle assembly is given by the field where the maximum of the transverse susceptibility $\chi(H)$ occurs [5].

In Fig. 1 the curves $P(H_a)$ and $\chi(H)$ of the AMP sample B are reported. The peaks of the two curves occur at the same field value (~4800 Oe); i.e., the mean anisotropy field $H_{a,m}$ obtained from $P(H_a)$ coincides with that one obtained with the transverse susceptibility measurement. Such value coincides also with the anisotropy field measured with the torque curves. This coincidence occurs for all the particles analysed in

Table 1
Magnetic properties of metal particle samples

sample	σ_s (emu/g)	H_c (Oe)	M_r/M_s	$H_{a,m}$ (Oe)
A (AMP)	145	2450	0.87	4780
B (AMP)	147	2500	0.88	4800
C (AMP)	148	2510	0.87	5000
D (MP)	125	1520	0.87	3700

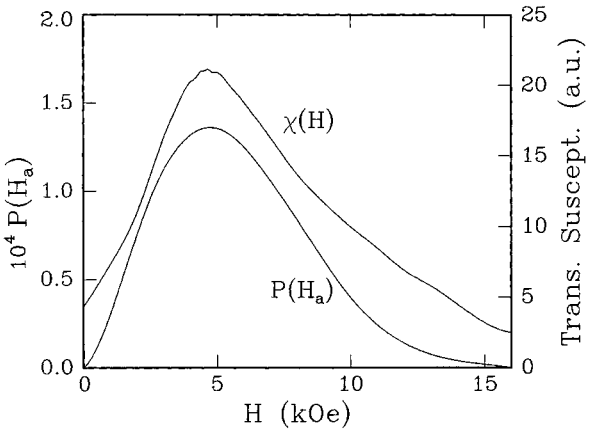

Fig. 1. Anisotropy field distribution $P(H_a)$ and transverse susceptibility $\chi(H)$ of advanced metal particles (sample B)

this work. The obtained $H_{a,m}$ values are given in Table 1. As the two methods give the same results, we will privilege the $P(H_a)$ measurement, which furnishes also the distribution of anisotropy fields. Since the results of the anisotropy measurements can be affected by the texture, the compared samples in this work have been prepared with the same alignment degree ($M_r/M_s \approx 0.87$, Table 1). The good orientation degree justifies the coincidence of the mean anisotropy field obtained with the different measurement methods [6].

From the TEM observation these AMP result to have a mean length of 85 nm and an aspect ratio of about (3–3.2):1, as reported also in [7]. Such shape gives rise to a difference between the demagnetizing factors in hard and easy direction $N_a - N_c =$ 4.2–4.4, and then to a shape anisotropy field $H_a = (N_a - N_c)\, M = 4800$–$5000$ Oe, with $M_s = 1140$ emu/cm^3 (from $\sigma_s = 147$ emu/g). The values are in very good agreement with the measured mean anisotropy fields (Table 1). This occurrence seems to suggest that the magnetic anisotropy in the advanced metal particles is that of shape only.

Because of the large M_s, it has been hypothesized that the AMP contain a relevant percentage of Co (up to 30%). The formation of FeCo bulk alloy, according to the Slater-Pauling curve, determines an increase of the saturation magnetization. In presence of a so high Co amount, some relevant quantity of CoO should exist in these particles because of the passivation treatment. The interaction between the Co and CoO phases could, with any probability, result in exchange anisotropy at low temperatures [8, 9]. We cooled the AMP samples in the presence of a strong applied field, but we could not observe any shift of the hysteresis loop, which is the main manifestation of the presence of exchange anisotropy. Besides, the bulk presence of Co would introduce a magnetocrystalline anisotropy, which should result in a dependence of the anisotropy, and consequently of the coercivity, on the temperature [10]. On the contrary, we observed that H_c does not vary with T from $-200\,^\circ$C to room temperature (Fig. 2). This lets us believe that Co, if present in the AMP, is confined to the surface layer, where it contributes to save the regular acicular shape of the inner iron core. The observed invariance of H_c with T, together with that one of M_s (Fig. 2) which makes the shape anisotropy independent of T, confirms that the magnetic anisotropy of AMP is that of shape.

The origin of the high σ_s of AMP must be found in the superior morphology of these advanced metal particles. The improvements which have been recently introduced in

the industrial preparation of the AMP have also the effect of preserving a regular and smooth acicular shape particularly of the ferromagnetic core [1, 2]. In previous generation MP, the process of conversion from goethite to passivated iron particles strongly induced the arising of surface and bulk irregularities, with pores, dendrites, presence of magnetic subunits inside the particles. These are sources of exchange interaction interruptions, internal demagnetization and surface canting, which contribute to depress the technical saturation magnetization. This occurs at a much smaller extent to advanced metal particles, whose good morphology maintains the magnetization larger than in MP.

The large coercivity of AMP with respect to previous MP cannot be simply imputed to the effect of the increase of magnetization on shape anisotropy, since σ_s increases by about 18% (from ≈125 to ≈147 emu/g), whilst H_c increases by more than 65% (from ≈1500 to ≈2500 Oe). The morphology of the AMP can lead us to understand the origin of the large coercivity. In particles with a good morhology and structure the magnetization reversal mode of coherent rotation is favoured [11], and the role that coherent rotation has in the magnetization reversal of the advanced metal particles has been underlined [12]. The larger coherency of the reversal implies a larger coercive field, closer to the anisotropy field. Actually, if we compare the coercive field value with the mean anisotropy field (Table 1), we see that $H_c/H_{a,m} = 0.50–0.52$ in AMP and $H_c/H_{a,m} = 0.40$ in MP. The evolution of reversal mode, together with the enhancement of shape anisotropy, give rise to the larger coercivity of the advanced metal particles.

Shape magnetic anisotropy arises from the configuration of the magnetostatic energy, whose minima result in easy magnetization axes, as an effect of the particle shape only. Recording media cannot be considered as composed of independent particles, and in closed particle aggregates the magnetostatic field and energy are changed, with consequent effect on shape anisotropy. We investigated such effect in AMP media preparing coatings of advanced metal particles with different packing densities p in the range 0.005–0.3. The properties of the different samples are reported in Table 2. The alignment degree is the same ($M_r/M_s = 0.88$) in all samples.

The magnetostatic interactions have been evaluated with the measurement of the deviation

$$\Delta m^* = m_i(H) - \tfrac{1}{2}[m_u(H) + m_l(H)]\,, \tag{2}$$

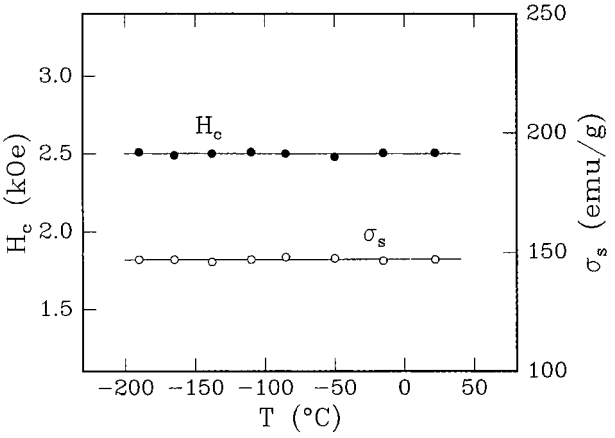

Fig. 2. Temperature dependence of coercivity H_c and saturation specific magnetization σ_s

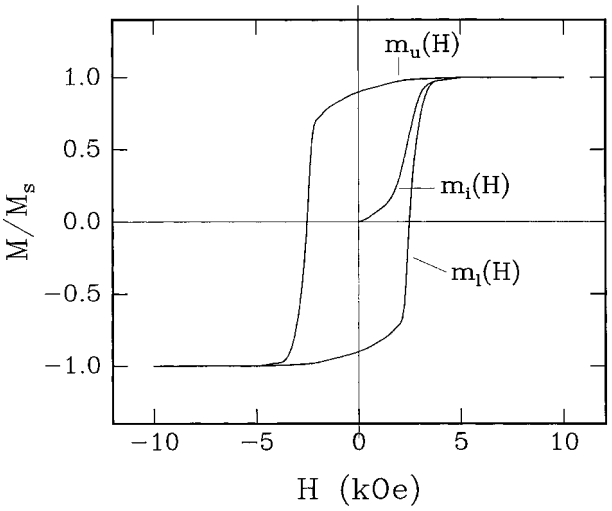

Fig. 3. Hysteresis cycle of advanced metal particle sample ($p = 0.3$)

where $m_i(H)$ is the initial magnetization curve, $m_u(H)$ and $m_l(H)$ respectively the upper and lower branches of the hysteresis loop [13]; all the magnetization values are reduced with respect to M_s. In Fig. 3 the hysteresis cycle of the sample at $p = 0.3$ is represented. In absence of interactions $\Delta m^* = 0$. The deviation from such value indicates the presence and strength of interactions. The obtained Δm^* deviations of the different packing AMP samples as a function of the applied field H are reported in Fig. 4. The Δm^* deviations are negative and their absolute values increase with the increase of packing. This indicates a regular increase of interactions with packing, even if the difference of intensity suggested by the Δm^* evolution is much smaller than what is said by the p variation: the maximum absolute value of Δm^*, which occurs at H close to coercivity, reported in Table 2, ranges from 0.07 to 0.12.

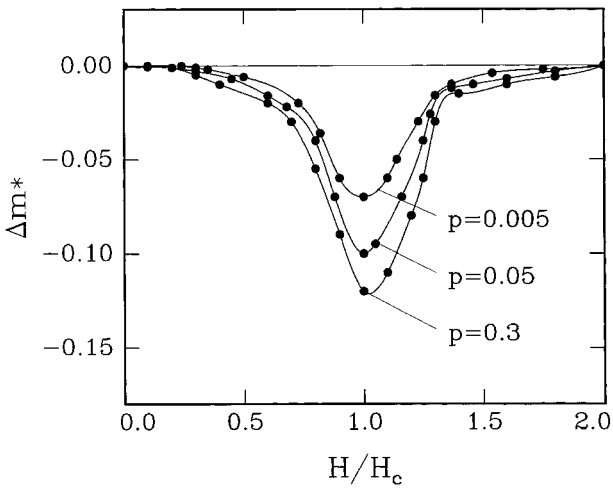

Fig. 4. Δm^* deviations following Thamm and Hesse [13] (for details see the text) of advanced metal particle samples with different packing densities

Table 2
Properties of advanced metal particle samples with different packing densities p

p	Δm^*_{max} (Oe)	H_c (Oe)	$H_{a,m}$ (Oe)	standard deviation
0.3	0.12	2490	4800	2600
0.05	0.10	2520	4800	2250
0.005	0.07	2550	4800	1900

Figure 5 shows the measured anisotropy distributions $P(H_a)$ of the different packing samples. The mean anisotropy field $H_{a,m}$, the peak field of the distribution, is the same for all the samples ($H_{a,m}$ = 4800 Oe), while H_c decreases when p and Δm^* increase. In this material, the mean anisotropy field is not affected by the interactions, but the coercive field lowers for the increasing intensity of the interaction field which adds to the external field. On the contrary the measured distribution is affected by the interactions, and it is larger in the samples with higher p and stronger interactions (Fig. 5 and standard deviation values in Table 2). This enlargement is imputable to the presence of clusters of coupled particles in the closely packed aggregates. The clusters, where the particles are arranged in closed flux structures, have their own anisotropy, smaller than that of the single particles. The results of the anisotropy measurements are conditioned by the overall anisotropy of the sample, which includes the anisotropy of the single particles, that of the clusters, the one of the whole specimen, and so on. Of course this is not the intrinsic anisotropy of the particles, but we can consider it as the effective anisotropy of the sample, which practically affects the macroscopic magnetic behavior, besides the anisotropy measurement results. The contemporary presence of the above described anisotropies enlarges the anisotropy distribution of the packed samples, where the clusters are more frequent, particularly towards the small anisotropy field values, as it is observed.

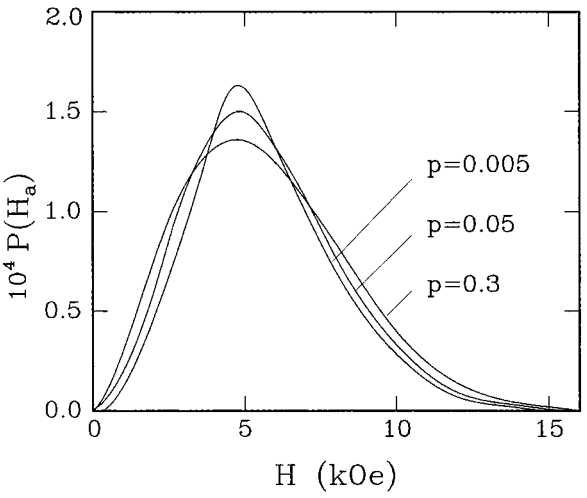

Fig. 5. Anisotropy field distributions of advanced metal particle samples with different packing densities

3. Conclusion

The magnetic anisotropy of advanced metal particles with large magnetization and coercivity has been studied with various experimental methods. The different measurement techniques give the same value of average anisotropy field, and the anisotropy distribution is also analysed.

From the geometrical and magnetic properties of the particles and from the observed independence of H_c of temperature, it is possible to conclude that the anisotropy is shape anisotropy only. The larger magnetization of the advanced metal particles with respect to previous metal particles is imputable to the superior morphology of the AMP, probably due to the recently introduced improvements in the technique of preparation of the material. The consequent evolution of the magnetization reversal mode justifies the high value of coercive field.

The width of the anisotropy distribution is affected by the interparticle interactions, and it is larger in packed assemblies. The measurements of anisotropy unavoidably report the effective anisotropy of the sample, which includes that of possible clusters of particles, which are more frequent in the closely packed samples. This gives rise to a spreading of the spectrum of anisotropy fields, which results in a wider anisotropy field distribution.

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