# Influence of Particle Size Distribution in Cermet Nanocomposites on Magnetoresistance Sensitivity

C. J. O'Connor, V. O. Golub, A. Ya. Vovk, A. F. Kravets, and A. M. Pogoriliy

***Abstract*—Tunneling magnetoresistance (TMR) and magnetic properties of $(Co_{50}Fe_{50})_x$-$(Al_2O_3)_{1-x}$ granular films prepared by dual electron beam evaporation were studied. The influence of the particle size distribution (PSD) on magneto-transport properties is reported. It was found that TMR versus M behavior does not follow $(M/M_s)^2$ law ($M$ and $M_s$ are magnetization and saturation magnetization, respectively), predicted for uniform size spherical particles with uncorrelated magnetic moments. Deviation from quadratic law is determined by PSD and is maximal for the films with the broadest distribution of superparamagnetic particles. For these films, the highest values of TMR and field sensitivity were also observed. The effect of PSD could be used for enhancement of TMR sensitivity in granular nanocomposites.**

***Index Terms*—Cermet, magnetoresistance, tunneling.**

## I. Introduction

CERMET films containing immiscible nanopaticles of metal and insulator have been the objects of intensive investigations during the past few decades (see, e.g., [1]–[7] and references therein). The study of cermets is interesting both for practical applications and for scientific reasons. Specific features of cermets are attributed to heterogeneity and nanoscale size that give a combination of the properties that are normally not associated with homogeneous materials. The ability to vary the particle size and the volume fraction of metallic component without any structural or chemical restrictions allows the systematic study of the properties in a wide composition range. Thus, cermets are an attractive model system for investigation of the basic features of magnetic nanoparticles, different types of magnetic interactions, and transport in disordered media. A number of applications, like magnetic reading heads and magnetic field sensors, are based on the tunneling magnetoresistance (TMR) effect, which is a unique property observed for magnetic metal-based cermets [1]. For these applications, both high values of TMR and high field sensitivity ($dMR/dH$) are necessary. Recently, it was shown that formation of specific granular structures (e.g., discontinuous metal insulator multilayers [8], [9]) leads to increase in $dMR/dH$ in low- and medium-field ranges. However, the influence of the topology on magnetic and MR properties of ordinary granular composites is not completely clear. In this paper, we report the influence of particle size distribution on magneto-transport properties of cermet films.

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## II. Film Preparation and Characterization

Thin $(Co_{50}Fe_{50})_x(Al_2O_3)_{1-x}$ cermet films ($x$ is magnetic metal volume fraction) with a thickness of about 400 nm were deposited on glass substrates at room temperature using the dual electron beam evaporation technique. The composition of the films was determined using energy dispersive analysis of x-ray (EDAX) and from the reference film thickness. The structural investigations included X-ray diffractometry and high resolution transmission electron microscopy (HRTEM) studies. The details of film preparation, composition, and structural characterization, as well as magnetoresistance measurement procedures, were reported elsewhere [6].

## III. Magnetic Measurements

Magnetic susceptibility [$M(T)$] was studied in the temperature range from 5 to 300 K at 50 Oe, and magnetization hysteresis loops [$M(H)$] were measured at different temperatures and in the fields ($H$) up to 50 kOe using a Quantum Design MPMS SQUID magnetometer. It was found from $M(H)$ curves that only part of magnetic material forms magnetic clusters. The rest is dispersed in non or weak magnetic inclusions within the oxide matrix. Previously, it was reported [3] that insulating matrix in Co–$Al_2O_3$ films prepared by electron beam evaporation can contain up to 5 vol.% of atomically dispersed Co. Analysis of our data shows that an $Al_2O_3$ matrix of the investigated films contains $\sim 3-4$ vol.% dispersed CoFe. As a result, the true concentration of the magnetic clusters ($x_M$) is somewhat lower than the one ($x_E$) determined from EDAX and reference film thickness measurements (see Table I). The PSD (see Fig. 1) was obtained from the fit of $M(H)$ curves at 10, 100, and 300 K. A sum of weighted Langevin functions was used. This approximation was done because the affects of magnetocrystalline and shape anisotropy [10], as well as dipole-dipole interactions between particles [11], can be neglected in particle size calculations in the case of broad distribution [12]. The calculations show that the PSD became broader and the average particle size ($d_{\rm avr}$) increased with $x$ (see Table I, column $M(H)$). These data are in a good agreement with our direct HRTEM observations that show spherical CoFe particles with bcc structure embedded in an amorphous matrix [6]. For the film with $x_E=0.16$, the particles of various sizes in the range from 1 nm to 3 nm were observed. The $d_{\rm avr}$ was also determined from field cooled-zero-field cooled susceptibility measurements [see

TABLE I
COMPOSITION, PARTICLE SIZE DISTRIBUTION, AND TMR OF THE FILMS AT 300 K

| $x_E$ Vol. % | $x_M$ Vol. % | $d_{avr}$, nm M(H) | $d_{avr}$, nm M(T) | TMR at 300K and 50kOe, % |
|---|---|---|---|---|
| 50 | 47 | - | - | -0.2 |
| 45 | 41.5 | - | - | -0.3 |
| 39 | 35 | - | - | -0.3 |
| 31 | 27 | >4.0 | 6.5 | -0.5 |
| 24 | 19 | >3.0 | 3.8 | -1.5 |
| 16 | 12 | 1.9 | 2.3 | -7.9 |
| 12 | 8 | 1.4 | 1.5 | -9.3 |
| 9 | 5 | 1.2 | 1.2 | -8.6 |
| 7 | 3 | 1.1 | 1.0 | -4.0 |

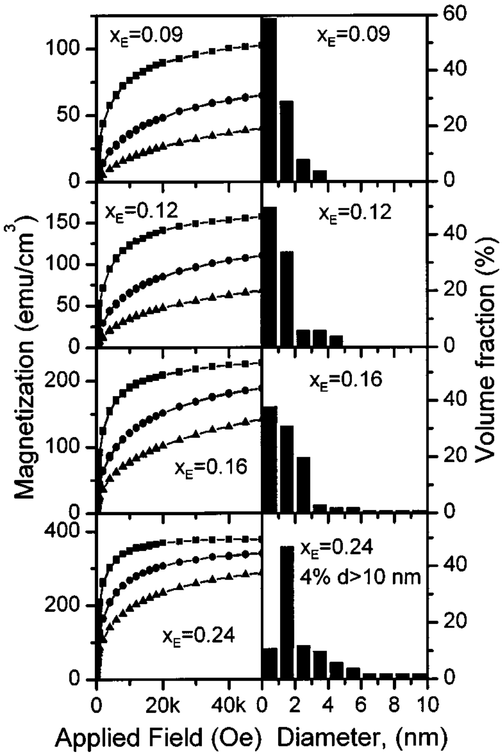


Fig. 1. Magnetization as a function of applied field at 10 K (■), 100 K (●) and 300 K (▲) (left side) and corresponding particle size distribution (right side) for $(Co_{50}Fe_{50})_x(Al_2O_3)_{1-x}$ cermet films of different composition.

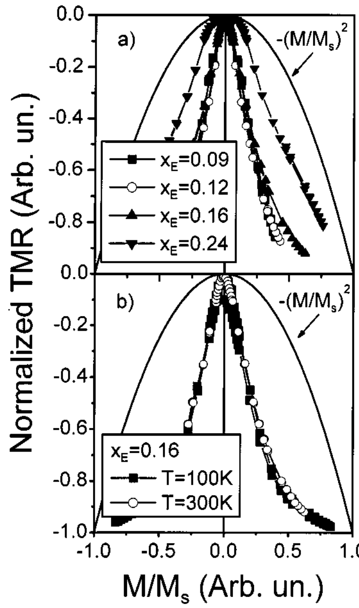


Fig. 2. Normalized TMR as a function of normalized magnetization for $(Co_{50}Fe_{50})_x(Al_2O_3)_{1-x}$ cermet films of (a) different composition and (b) at different temperatures.

Table I, column $M(T)$]. It was supposed that above the bifurcation temperature, a Currie-Weiss behavior is held. It was found that the average particle size varies from $\sim$ 1 nm ($x_E = 0.07$) to $\sim$ 3 nm ($x_E = 0.24$). The results of the calculations are summarized in Table I. It is to be pointed out that both calculations gave practically the same values of the $d_{\rm avr}$.

## IV. MAGNETORESISTANCE

The dependencies of TMR on composition, temperature and applied magnetic fields were studied earlier [6], [7]. The maximal TMR value $\sim 10\%$ and $\sim 16\%$ in fields 50 kOe were found at 300 and 10 K, respectively, for compositions $x_E < 16$. For the higher $x$, the MR dramatically decreases to the vanishing values (see Table I). This effect was attributed to formation of the percolation cluster of magnetic material.

It is known that the magnetoresistance in cermet films is due to charge transport caused by tunneling through the insulator barriers [4]. The tunneling conductance depends on the cosine of the angle between the directions of magnetization of neighbor ferromagnetic particles ($\theta$), like $G \sim 1 + P^2 \cos\theta$, where $P$ is the effective spin polarization of the ferromagnetic-barrier couple. It increases with alignment of the particles magnetic moments in applied magnetic field. The average over $\theta$ gives $\langle\cos\theta\rangle = m^2$, where $m$ is the relative magnetization of the system. For small $P$ values and in the case of uniform size of superparamagnetic particles with uncorrelated moments, the TMR ratio will be proportional to $m^2 \quad (m = M/M_s)$ [4]. In general, proposed model does not take into account such important features as tunneling barrier height and nonuniformity, crystal potential, spin waves, electron-electron correlations, elastic and inelastic tunneling between particles of different size, and the Coulomb blockade effect. In a real granular system, a broad PSD can be formed due to coalescence processes and conductance due to high order tunneling take place [5]. However, a detailed description of MR versus M dependence for this case has yet to be presented. Recently, giant magnetoresistance (GMR) in granular metals containing interacting magnetic particles with dispersed sizes and shapes was studied numerically [11]. It was shown that size dispersion alone causes enhancement of GMR while dipolar interactions have degrading effect. However, both factors are responsible for minor deviations from MR versus $(M/M_s)^2$ behavior in low fields. In this work, dramatic swerve from quadratic law for cermet granular films with PSD was observed. Fig. 2 shows experimental curves of normalized TMR (TMR/$TMR_s$, where $TMR_s$ is magnetoresistance at saturaded state) for the films of different compositions (a) and at different temperatures (b) as a function of normalized magnetization of the films and theoretical -$(M/M_s)^2$ behavior as well. The resistance of the films decreases in the lower fields much faster than for the case of the system contained uniform particles of average size determined for given PSD. The strongest deviations were found for $x_E < 20$. For the films of composition near

$x_E \sim 0.16$, the highest values of $dMR/dH$ up to 2.5%/kOe at 300 K and $\sim 8\%$/kOe at 100 K were observed. In these films, the broadest variation of magnetic properties of superparamagnetic particles was found.

The MR versus M behavior cannot be described in the terms of dipole-dipole interactions [11] as it does not obey the quadratic law even at the lowest $x$, where these interactions are negligible. Moreover, the MR versus M dependence approaches the quadratic one only for the samples with high $x$. Analysis of the data presented on Fig. 2 showed that the particles of the larger size give a higher contribution to TMR than can be expected from their concentration. It is curiously as the distance between them is large and that direct tunneling is rather unlikely. Such a behavior of TMR could be expected for high-order elastic tunneling between large particles through intervening small ones. It is known [13] that high-order tunneling can have inelastic and elastic characteristics. In an inelastic process, two different electrons tunnel into two different junctions: One jumps into a central electrode (particle) above the Fermi level, whereas another jumps out the particle from below the level. This cotunneling process involves creation of an electron-hole excitation on the central particle. In contrast, in elastic tunneling, no such excitation is formed. This mechanism implies that the same electron tunnels through both junctions. The intermediate electron state on the central particle during tunneling is virtual, and the tunneling rate depends on the character of the real electron motion within this particle. It was shown that inelastic tunneling suppresses magnetoresistance due to spin-flip processes at the central particle [14]. Contrarily, the elastic tunneling in some cases can lead to the increase in tunneling probability [13]. However, elastic tunneling strongly depends on temperature, and its contribution is sufficient only at low temperatures and low bias voltages. In our case, the temperatures are high [100 and 300 K; see Fig. 2(b)], and elastic tunneling should be suppressed. At the present moment, there is no definitive theory for a quantitative explanation of such tunneling at the higher temperatures, i.e., the conservation of the electron spin during high-order tunneling process.

## V. Conclusions

Thin $(\mathrm{Co_{50}Fe_{50}})_x(\mathrm{Al_2O_3})_{1-x}$ cermet films with broad PSD were prepared using a dual electron beam co-evaporation technique. Their magnetic and transport properties were studied. Dramatic deviation from quadratic law caused by PSD was found for TMR versus M behavior. Size dispersion is responsible for enhancement of TMR value and increases field sensitivity of granular nanocomposites. As a result, high field sensitivity can be achieved, even in the films containing a large number of small superparamagnetic granules. Thus, size distribution plays a crucial role in the transport properties of such systems.

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