Classical theory of giant magnetoresistance in granular metals

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(Received 1 March 1994)

I have extended the one-dimensional spin-diffusion theory of Valet and Fert [Phys. Rev. B **48**, 7099 (1993)] [which treats perpendicular giant magnetoresistance (GMR) in magnetic multilayers] to three dimensions in order to deal with the problem of GMR in granular magnetic metals. These materials are composed of tiny precipitates of magnetic granules suspended within an immiscible nonmagnetic metallic matrix. It is assumed that the granules are spherical in shape, and have identical radii. The differential equations are solved separately for three different physical situations: (1) Bulk spin-dependent scattering in the nonmagnetic matrix; (2) Bulk spin-dependent scattering within the magnetic spheres; and (3) surface spin-dependent scattering at the ferromagnetic-normal interface. The magnetically induced electrical interaction between spheres, which gives rise to observed GMR, is treated in lowest order only. It is found to act via an induced electric-dipole field for cases (1) and (3), and an induced spin-diffusion field for case (2). Using these solutions, and macroscopic parameters from experiment, I have attempted to explain the observed singular dependence of the GMR on annealing temperature.

I. INTRODUCTION

Berkowitz *et al.*[1] and Xiao, Jiang, and Chien[2] were the first to demonstrate that granular films, composed of tiny precipitates of magnetic material (e.g., Co) embedded in a nonmagnetic matrix (e.g., Ag), have very large magnetoresistances when properly annealed. Such granular materials are often formed by codepositing two immiscible metallic elements, one of which is magnetic, to form a metastable solid solution. Subsequent annealing at an elevated temperature increases the size of the ferromagnetic granules by recrystallization. The presumed spherical nature of very tiny precipitates is caused by surface tension, and has been verified in some instances by transmission electron micrography.

Recently, Valet and Fert[3] published a classical theory of magnetoresistance, and applied it to the problem of perpendicular giant magnetoresistance (GMR) in magnetic multilayers. Valet and Fert postulate the existence of a spin-diffusion length Λ, which they define as the distance a spin-polarized conduction electron travels before it undergoes a spin-flip collision. As a consequence of spin diffusion, Fert and Valet demonstrate the occurrence of spin accumulation about the interfaces of the multilayers in the perpendicular magnetoresistance geometry. The spin-accumulation layers become regions of high magnetoresistivity. Adjacent interfaces can interfere with, and partly balance each other's spin-accumulation layers. In addition to this process, Fert and Valet also consider interfacial spin scattering. In the present paper, I extend the concepts of Fert and Valet to three dimensions, and apply these equations to the problem of granular magnetic materials.

Throughout this paper, I adopt a simplified picture of the structure of these granular alloys. In reality this is more a *model* than a physical representation. I assume that the ferromagnetic particles are spherical in shape. It is further assumed that all the particle spheres have the *same* radius a which is dependent on the annealing temperature T_A, and which monotonically increases with T_A. In this model, all the ferromagnetic material is presumed to exist as precipitates, and not as an alloy with the host matrix: the two components are presumed completely immiscible. As a consequence of this assumption, the volume fraction of the magnetic material f remains independent of T_A, while the particle radius a increases monotonically with T_A. This model presents many of the essential features found in real granular materials. However, the above assumptions break down beyond the percolation threshold when the particles overlap—perhaps in Co_xCu_{1-x} when $x > 0.3$.

In Fig. 1 we display the dependence of the magnetoresistance $\Delta\rho/\rho$ on the annealing temperature T_A, for various granular materials and various sample preparations. These results are taken from the papers of different investigators, and are assembled here in order to display the magnitude of the effect and to demonstrate the ubiquity of an optimum annealing temperature. The percentage GMR is usually small for both low- and high-temperature anneals, and can display a pronounced maximum at some intermediate temperature. These heterogeneous alloys have been manufactured by various techniques: Figs. 1(a) and 1(b) are of magnetron sputtered thin films; Figs. 1(d) and 1(e) are of melt-spun alloys; Fig. 1(c) is from a discontinuous multilayer, and Fig. 1(f) is taken from a granular film synthesized by annealed multilayers. The references and compositions are to be found in the figure caption.

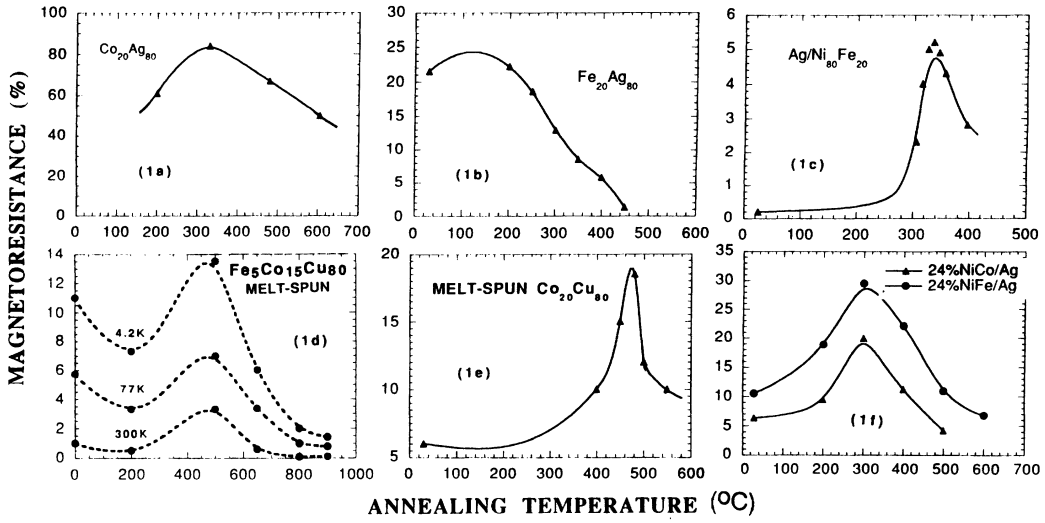

FIG. 1. Percent magnetoresistance ($\Delta\rho/\rho$) versus annealing temperature (T_A) for various granular samples. Data taken from the literature: (a) Ref. 5; (b) Ref. 6; (c) T. L. Hylton *et al.*, Science **261**, 1021 (1993); (d) M. Rubinstein and B. N. Das (unpublished); (e) B. Dieny *et al.*, JMMM (to be published); (f) X. Bian *et al.*, J. Appl. Phys. (to be published).

II. PREVIOUS THEORIES

In addition to the macroscopic theory of Valet and Fert, there exists several other treatments for dealing with GMR in granular alloys.

To explain the dependence of granular GMR on the average radius a of Co particles in a Cu matrix, Berkowitz *et al.*[1] adopted a spin-dependent scattering model at the interfaces between the particles and the matrix. This quantum-mechanical scattering theory was further elaborated by Zhang and Levy.[4] This theory predicts an inverse dependence of GMR on particle size, $\Delta\rho/\rho = a^{-1}$, a purely monotonic decrease of the GMR with particle size, and therefore with annealing temperature. The authors explain the maximum in $\Delta\rho/\rho$ observed by experimenters by assuming that small particles become superparamagnetic when the radius a becomes small.

A different explanation of the dependence of GMR on particle size has been alluded to by Xiao, Jiang, and Chien.[2] These authors state that a length scale of the order of the conduction-electron mean free path is present, and when the particle size becomes large compared to the mean free path, the GMR is degraded. (Provided such a picture is applicable, we shall argue in this paper that the correct scaling length is, in fact the conduction-electron spin-diffusion length.)

However, Xiong *et al.*[5] and Wang and Xiao[6] maintain that the maximum in $\Delta\rho/\rho$ vs T_A has no fundamental significance, since *both* $\Delta\rho$ and ρ decrease sharply with T_A yielding an accidental maximum at some temperature.

In the next few sections, we shall develop a classical theory for granular GMR, following closely the formulation of Valet and Fert for multilayers. By "classical" we mean, simply, that the theory is not explicitly quantum mechanical, and uses macroscopic parameters such as conductivities and spin-diffusion lengths as input parameters.

III. MACROSCOPIC THEORY

In order to deal with spherical particles, we must first extend the one-dimensional differential equations of Valet and Fert[3] to three-dimensional equations by trivially replacing one-dimensional derivatives by gradients. Previous to Valet and Fert, the one-dimensional differential equations, applicable to multilayers, had been published by van Son, van Kempen, and Wyder[7] but Valet and Fert's treatment is much more complete.

We first characterize our medium by several macroscopic parameters. σ_+ and σ_- are the spin-up and spin-down conductivities, respectively, of the two spin subbands; these differ from each other only in a magnetic medium. ϕ_+ and ϕ_- are the potentials acting on the spin-up and spin-down conduction electrons. Λ is the spin-diffusion scattering length; this can be roughly defined as the length which a spin-polarized conduction electron travels before reaching spin equilibrium, and is given approximately by the formula $\Lambda=[(1/3)(v_F\lambda)\tau_{sf}]^{1/2}$, where v_F is the Fermi velocity, λ is the conduction-electron mean free path, and τ_{sf} is the conduction-electron "spin-flip" relaxation time.

The differential equations are

$$\nabla^2(\phi_+-\phi_-)=(\phi_+-\phi_-)/\Lambda^2\ ,$$
$$\nabla^2(\sigma_+\phi_++\sigma_-\phi_-)=0\ . \tag{1}$$

[These equations are the three-dimensional extensions of Eqs. (14) and (15) of Valet and Fert.] The first equation, known as the spin-diffusion equation, expresses the fact that the spin-up and the spin-down potential approach each other in a distance which is characterized by the spin-diffusion scattering length. This macroscopic transport equation has been derived from the Boltzmann equation by Valet and Fert. The second equation is simply an expression of conservation of charge, assuming the validity of Ohm's law.

$$j_+ = \sigma_+ \nabla \phi_+ ,$$
$$j_- = \sigma_- \nabla \phi_- . \tag{2}$$

In order to apply these equations to a granular material, we must first solve the problem of an isolated magnetic sphere embedded in a nonmagnetic metallic matrix. The solution of this preliminary problem reveals no magnetoresistance, since we *postulate* that σ_+ and σ_- are not dependent on the external magnetic field. It is only when interaction between the magnetic spheres are admitted that we find giant magnetoresistance.

The explicit dependence of conductivity on magnetic field is the source of ordinary magnetoresistance[8] (OMR). We are, however, interested in the investigation of the GMR, phenomenon, which can be *defined* as resistivity which is caused by the interaction of heterogeneous magnetic structures. Experimentally, GMR differs from OMR in that it is independent of the direction of the applied magnetic field with respect to the current, and in that its value is always negative,[9] i.e., the resistance uniformly decreases with applied magnetic field.

All differential equations require boundary conditions. In the case of the lone sphere, there are three boundary conditions to satisfy: one at infinity, one at the surface of the sphere, and one at the sphere's origin (see Fig. 2). Using spherical coordinates, with the sphere of radius a centered at $r=0$, and an electric field of magnitude E imposed parallel to the z axis, these boundary conditions are

$$\phi_\pm^{>}(\infty) \rightarrow Er\cos(\theta) ,$$
$$\phi_\pm^{<}(0) \neq \infty ,$$
$$\sigma_\pm^{<} \nabla \phi_\pm^{<}(a) = \sigma_\pm^{>} \nabla \phi_\pm^{>}(a) , \tag{3}$$
$$\phi_\pm^{>}(a) - \phi_\pm^{<}(a) = r_s J_\pm(a) ,$$

where r_s is the spin-dependent boundary resistance per unit surface of the F/N interfaces, and $\phi_\pm^{<}$ and $\phi_\pm^{>}$ are the spin-dependent potentials inside and outside the ferromagnetic sphere, respectively. [The boundary conditions are the three-dimensional analog of Valet and Fert's Eqs. (19) and (20).] Equations (1), (2), and (3) can be solved in all their generality, with no simplifying assumptions. It is, however, much more instructive to solve certain specific cases, since the physical consequences are more illuminating severally than conjoined.

These equations, *absent the spin-diffusion term and spin-dependent boundary resistance term,* have an illustrious history. The electrical conductivity of a matrix of spherical particles in a conducting medium was derived to lowest order by James Clerk Maxwell,[10] while Lord Rayleigh[11] deduced the form of the higher-order interaction terms. In addition to the theory of electrical conduction, these solutions have formed the backbone of the theory of dielectric constants, magnetic permeability, and thermal conductivity in inhomogeneous media, all of which have identical mathematical formulations.

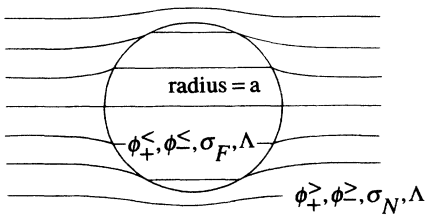

FIG. 2. Magnetic sphere of radius a and conductivity σ_F suspended in a normal metal with conductivity σ_N and spin-diffusion length Λ. Lines of electric current are shown schematically.

IV. MAGNETORESISTANCE VIA SURFACE SPIN-DEPENDENT SCATTERING

First, we consider the case where bulk spin-dependent scattering can be neglected with respect to the surface spin-dependent scattering. In addition we make the simplifying assumption that the ferromagnetic material contains majority spin conduction electrons *exclusively,* i.e., we assume that only the spin-up conduction band intersects with the Fermi surface. (This assumption agrees with the recent conclusions of Stearns.[12]) As a consequence of these physical assumptions we have

$$\sigma_+^{<} = \sigma_F ; \quad \sigma_-^{<} = 0 ,$$
$$\sigma_\pm^{>} = \sigma_N ; \quad \Lambda = \infty , \tag{4}$$

where σ_N and σ_F are the conductivities of the nonmagnetic host matrix and its ferromagnetic inclusions, respectively. With these assumptions, the differential equations, Eqs. (1), reduce to a set of four Laplace equations. Because of the boundary conditions at $r=0$ and $r=\infty$, the only terms that survive are those associated with the $n=1$ Legendre polynominal. The solutions of the differential equations are

$$\phi_+^{<} = B^+ r ,$$
$$\phi_-^{<} = B^- r ,$$
$$\phi_+^{>} = Er + D^+/r^2 , \tag{5}$$
$$\phi_-^{>} = Er + D^-/r^2 ,$$

where E is the external electric field, and the $\cos(\theta)$ factor is understood to multiply each term. B_+, B_-, D_+, and D_- are to be determined by the four boundary conditions at the interface, and are given by

$$B^+ a = Ea + D^+/a^2 - B^+ r_s \sigma_F ,$$
$$B^- a = Ea + D^-/a^2 ,$$
$$B^+ \sigma_F = (E - 2D^+/a^3)\sigma_N , \tag{6}$$
$$0 = E - 2D^-/a^3 ,$$

which have as their solution

$$B^+ = \frac{3Ea\sigma_N}{2a\sigma_N + a\sigma_F + 2r_s\sigma_N\sigma_F} ; \quad B^- = \frac{3E}{2} ,$$
$$D^+ = \frac{Ea^3(a\sigma_N - a\sigma_F + r_s\sigma_N\sigma_F)}{2a\sigma_N + a\sigma_F + 2r_s\sigma_N\sigma_F} ; \quad D^- = \frac{Ea^3}{2} . \tag{7}$$

The effective conductivity σ_{eff} of a matrix of *noninteract-*

ing spheres with volumetric filling factor f is given by the *average* current divided by the external field E:

$$\sigma_{\text{eff}}=(1-f)\sigma_N+f\sigma_F B^+/E \\ =(1-f)\sigma_N+\frac{3f\sigma_N}{1+2(\sigma_N/\sigma_F)+2r_s\sigma_N/a} . \tag{8}$$

In order to calculate the magnetoresistance, we must consider the mutual interaction between spheres. This interaction is calculated only to lowest order, using the Lorentz field approximation.

Consider a single-domain magnetic sphere whose localized magnetic moment is pointed "up." Since we have postulated the absence of a minority-spin conduction subband, only spin-up conduction electrons are contained within the sphere.

Next consider the surrounding spheres. Their localized spins may be either up *or* down. In a completely magnetized sample, all these neighboring spins are pointed up, while in a demagnetized sample, there are equal numbers of up and down spins. We are not concerned with the process of aligning spins, at present, since the magnetoresistance is expressed as the difference in the resistance of the completely magnetized state and the completely demagnetized state, without regard for the magnetization process itself. From the point of view of the central spin-up sphere, we see from Eq. (6) that the effective *electric* dipole moment of a neighboring spin-up sphere is D^+, while the effective *electric* dipole moment of a neighboring spin-down sphere is D^-. The fact that $D^+\neq D^-$ is responsible for the phenomenon of magnetoresistance. The Lorentz electric field acting on the central sphere is therefore

$$E^M=(4\pi/3)\frac{D^+}{(4\pi/3)a^3}f \text{ , magnetized ,} \\ E^D=(4\pi/3)\left[\frac{D^+/2+D^-/2}{(4\pi/3)a^3}\right]f \text{ , demagnetized ,} \tag{9}$$

where f is the volumetric filling factor, and a is the radius of the spheres. These dipolar fields assist the external field, thereby increasing the effective conductivity. Defining $E^P=\alpha E$, $E^M=\beta^M E$, and $E^D=\beta^D E$ where β^M and β^D are obtained from Eq. (9) and α is obtained from Eq. (7), we obtain for the conductivity of the magnetized state, the conductivity of the demagnetized state, and the percent magnetoconductivity

$$\sigma_{\text{eff}}^M=(1-f)(1+\beta^M)\sigma_N+f(\alpha+\beta^M)\sigma_F , \\ \sigma_{\text{eff}}^D=(1-f)(1+\beta^D)\sigma_N+f(\alpha+\beta^D)\sigma_F , \\ \frac{\Delta\sigma}{\sigma_N}=(\beta^M-\beta^D)\left[1-f\left[1-\frac{\sigma_F}{\sigma_N}\right]\right] . \tag{10}$$

To first order in f, we obtain

$$\frac{\Delta\sigma}{\sigma_N}\approx\frac{f}{4}\left[\frac{3\sigma_F/2\sigma_N}{1+\sigma_F/2\sigma_N+\sigma_F r_S/a}\right] . \tag{11}$$

V. MAGNETORESISTANCE VIA BULK SPIN-DEPENDENT SCATTERING

We next consider the case where the surface spin-dependent scattering can be neglected with respect to the bulk scattering. We assume, again, that the ferromagnetic metal contains only a majority conduction-electron spin band. The conductivity for spin-up conduction electrons within the ferromagnetic spheres is σ_F, while the spin-down conductivity is defined as zero. The spin-diffusion length inside the ferromagnet is taken as infinite, while the spin-diffusion length in the normal metal is Λ. The differential equations and boundary conditions become

$$\nabla^2(\phi_+^>-\phi_-^>)=(\phi_+^>-\phi_-^>)/\Lambda^2 ; \quad \nabla^2(\phi_+^>+\phi_-^>)=0 , \\ \nabla^2(\phi_+^<)=0 ; \quad \nabla^2(\phi_-^<)=0 , \\ \phi_+^<(a)=\phi_+^>(a) ; \quad \phi_-^<(a)=\phi_-^>(a) , \\ \sigma_F\nabla\phi_+^<(a)=\sigma_N\nabla\phi_+^>(a) ; \quad 0=\sigma_N\nabla\phi_-^>(a) , \\ \phi_\pm^>(\infty)\rightarrow Er\cos(\theta) ; \quad \phi_\pm^<(0)=\text{finite} . \tag{12}$$

These differential equations have as their solutions

$$\phi_+^<=B^+r , \\ \phi_-^<=B^-r , \\ \phi_+^>=Er+CH(r/\Lambda)+D/r^2 , \\ \phi_-^>=Er-CH(r/\Lambda)+D/r^2 , \tag{13}$$

where $H(x)=\exp(-x)(x^{-1}+x^{-2})$ is a Hankel function of the imaginary argument, and B^+, B^-, C, and D are constants to be determined by the boundary conditions. We obtain

$$B^+a=Ea+CH(a/\Lambda)+D/a^2 , \\ B^-a=Ea-CH(a/\Lambda)+D/a^2 , \\ \sigma^<B^+=\sigma^>[E+CH'(a/\Lambda)-2D/a^3] , \\ 0=E-CH(a/\Lambda)-2D/a^3 ,$$

$$H(r/\Lambda)=\exp(-r/\Lambda)[(r/\Lambda)^{-1}+(r/\Lambda)^{-2}] , \\ H'(r/\Lambda)=-\frac{1}{\Lambda}\exp(-r/\Lambda)[(r/\Lambda)^{-1}+2(r/\Lambda)^{-2} \\ +2(r/\Lambda)^{-3}] , \tag{14}$$

$$B^+=3E\sigma^N/(\sigma^F+2\sigma^N) , \\ B^-=3E/2 , \\ C=-\frac{3}{4}a^3\frac{E}{\Lambda^2}\sigma^F/(\sigma^F+2\sigma^N) , \\ D=-\frac{1}{4}a^3E(\sigma^F-4\sigma^N)/(\sigma_F+2\sigma^N) .$$

In terms of these parameters, the average current $\langle J\rangle$ and the conductivity of a matrix of *noninteracting* spheres $\sigma_{\text{eff}}=\langle J\rangle/E$ is given by

$$\langle J\rangle=(1-f)\sigma_N E+f\sigma_F\left\{\frac{3\sigma_N}{\sigma_F+2\sigma_N}\right\}E\ , \tag{15}$$

$$\frac{\sigma_{\rm eff}}{\sigma_N}=1+f\left\{\frac{2(\sigma_F-\sigma_N)}{\sigma_F+2\sigma_N}\right\}\ . \tag{16}$$

In addition to the applied electric field, Eq. (13) shows that two kinds of induced fields are present in the normal metal: a dipolar field $E^{\rm dip}$ caused by the induced electric dipole moment on the ferromagnetic spheres, and a spin-diffusion field $E^{\rm dif}$ originating at the spin-accumulation layer surrounding the spheres. These two electric fields constitute the electrical interaction between the magnetic spheres at low filling factors; at higher filling factors, higher-order (e.g., octupole) induced moments become important,[13] but we shall restrict our analysis to the lowest order. In a manner of speaking, even Eq. (16) can be said to involve interactions since it contains the filling factor f, but we shall resist this interpretation.

We must next evaluate the z component of $E_{\pm}^{\rm dip}$ and $E_{\pm}^{\rm dif}$, the dipolar and spin-diffusion electric fields which originate from the induced charges surrounding each sphere. (The $\pm$ subscript refers to the sign of the conduction-electron subband on which the field acts.) The electric interaction fields between magnetic spheres are given by evaluating the z gradient of the spin-dependent potentials, $\phi_{\pm}^{>}$ in Eq. (13), and then summing the contributions due to all the neighboring spheres. The net dipolar field acting on a sphere can be evaluated as due to polarization charges on the surface of a fictitious cavity, as demonstrated by Lorentz[14]

$$E^{\rm dip}=+\frac{4\pi}{3}\frac{6}{4\pi a^3}D=3f\left\{\frac{4\sigma_F-\sigma_N}{\sigma_F+2\sigma_N}\right\}\ . \tag{17}$$

The net spin-diffusion $E^{\rm dif}$ is evaluated by differentiating the spin-diffusion potential [including the implied $\cos(\theta)$ term] to obtain the z component of the field emanating from each of the spherical particles. This field component is then averaged over θ, multiplied by the density of spheres, $3f/4\pi a^3$, and integrated from a to ∞ using $4\pi r^2 dr$ as the volume element to obtain an approximation. The result is

$$E^{\rm dif}=\frac{9f}{4\pi}E\left\{\frac{\sigma_F}{\sigma_F+2\sigma_N}\right\}e^{-a/\Lambda}(a/\Lambda)^2\left\{\frac{2}{3}\frac{a}{\Lambda}-1\right\}\ . \tag{18}$$

In this case, *unlike* that of surface scattering, the dipolar electric field is *independent* of the magnetization of the spheres. $E^{\rm dip}$ contributes a term to the electrical conductivity, but does not contribute to the magnetoresistance. On the other hand, the diffusion field $E^{\rm dif}$ of each sphere changes sign whenever the sphere's magnetization is reversed. In an unmagnetized sample, the average field is zero; in a completely magnetized sample the field is equal to $2E^{\rm dif}$. This result gives rise to magnetoresistance.

We neglect the dipolar term for simplicity. Since $\langle J\rangle_{\rm demag}=\sigma_{\rm eff}E$ and $\langle J\rangle_{\rm mag}=\sigma_{\rm eff}(E+2E^{\rm dif})$, we obtain the result $\Delta\sigma/\sigma=2E^{\rm dif}/E$, and

$$\frac{\Delta\sigma}{\sigma}=\frac{9f}{2\pi}\left\{\frac{\sigma_F}{\sigma_F+2\sigma_N}\right\}e^{-a/\Lambda}(a/\Lambda)^2\left\{\frac{2}{3}\frac{a}{\Lambda}-1\right\}\ , \tag{19}$$

which is a function of the ratio or the particle radius to the spin-diffusion length a/Λ.

VI. BULK SPIN-DEPENDENT SCATTERING WITHIN THE FERROMAGNET

We next consider the case where spin-dependent scattering is experienced *solely* within the bulk of the ferromagnetic spheres. Temporarily assuming that the conduction-electron spin subbands in the ferromagnet are not completely polarized, the differential equations and boundary conditions become

$$\begin{aligned}
&\nabla^2(\phi_+^<-\phi_-^<)=(\phi_+^<-\phi_-^<)/\Lambda^2;\ \ \nabla^2(\phi_+^>-\phi_-^>)=0\ ,\\
&\nabla\phi^2(\sigma_+^<\phi_+^<+\sigma_-^<\phi_-^<)=0\ ,\\
&j_+^<=\sigma_+^<\nabla\phi_+^<\ ;\ \ j_-^<=\sigma_-^<\nabla\phi_-^<\ ,\\
&j_+^>=\sigma_N\nabla\phi_+^>\ ;\ \ j_-^>=\sigma_N\nabla\phi_-^>\ ,\\
&\phi_+^<(a)=\phi_+^>(a)\ ;\ \ \phi_-^<(a)=\phi_-^>(a)\ ,\\
&\phi_\pm^>(\infty)\to Er\cos(\theta)\ ;\ \ \phi_\pm^<(0)=\text{finite}\ .
\end{aligned} \tag{20}$$

The solution of these equations are

$$\begin{aligned}
&\phi_+^<(r)=Br+A\sigma_-^<J(r/\Lambda)/(\sigma_+^<+\sigma'_-)\ ,\\
&\phi_-^<(r)=Br-A\sigma_+^<J(r/\Lambda)/(\sigma_+^<+\sigma'_-)\ ,\\
&\phi_+^>(r)=Er+C/r^2\ ,\\
&\phi_-^>(r)=Er+D/r^2\ ,\\
&J(r/\Lambda)=\sinh(r/\Lambda)/(r/\Lambda)^2-\cosh(r/\Lambda)/(r/\Lambda)\ ,
\end{aligned} \tag{21}$$

where $J(x)$, the first-order Bessel function, is finite at $r=0$. The first-order Legendre polynomial $P_1(\theta)=\cos(\theta)$ is understood to multiply each potential.

Although it is not necessary, we now choose $\sigma_-^<=0$. This choice expresses the notion that the conduction electrons are strongly spin polarized in the ferromagnetic metal. The boundary conditions become

$$\begin{aligned}
&Ba=Ea+C/a^2\ ,\\
&Ba-AJ(a/\Lambda)=Ea+D/a^2\ ,\\
&B=\frac{\sigma^N}{\sigma^F}(E-2C/a^3)\ ,\\
&0=E-2D/a^3\ .
\end{aligned} \tag{22}$$

These four equations have the solution

$$A=\frac{-3aE}{2[1+2(\sigma_N/\sigma_F)]J(a/\Lambda)}\ ;\ \ B=\frac{3E(\sigma_N/\sigma_F)}{1+2(\sigma_N/\sigma_F)}\ ; \tag{23}$$

$$C=\frac{a^3E(-1+\sigma_N/\sigma_F)}{1+2(\sigma_N/\sigma_F)}\ ;\ \ D=\frac{a^3E}{2}\ .$$

In the completely magnetized state, the induced Lorentz field is

$$E^{\text{mag}}=(4\pi/3)(C/2+D/2)f/(4\pi a^3/3)\ ;$$

and in the completely demagnetized state the induced Lorentz field is

$$E^{\text{dem}}=(4\pi/3)(C/2-D/2)f/(4\pi a^3/3)\ .$$

Since $\langle J\rangle\approx\sigma_N(E+E_L)$, where $E_L=E^{\text{dem}}$ or E^{mag}, depending on the magnetic state, we obtain as an approximate expression for the magnetoconductance

$$\frac{\Delta\sigma}{\sigma_{\text{eff}}}=\frac{fC}{Ea^3}=\frac{\sigma_N-\sigma_F}{2\sigma_N+\sigma_F}\ . \tag{24}$$

We find that making the above assumptions—zero minority band conductivity, spin diffusion confined to the ferromagnetic spheres, use of the Lorentz field to approximate the interaction field—leads to a magnetoresistance which is independent of a, the radius of the spheres, and of Λ, the spin-diffusion constant.

VII. MAGNETORESISTANCE IN MULTILAYERS: VALET AND FERT'S CALCULATION

Valet and Fert[3] found an *exact* solution of the one-dimensional spin-diffusion equation (but not the corresponding Boltzmann equation) appropriate to the calculation of the magnetoresistance of a one-dimensional multilayer. This is in contrast to the *approximate* solution of the three-dimensional spin-diffusion equation appropriate to a granular material, above. As an example of Valet and Fert's calculation, we choose, for simplicity, a multilayer with zero interfacial scattering whose magnetic layers are the same thickness t as the nonmagnetic layers. We also choose the spin-diffusion length Λ to be equal in the two types of layers. We choose the resistivity of the ferromagnetic metal to equal to that the normal metal ρ, and we assume that the ferromagnetic metal is completely spin polarized (i.e., only a spin-up conduction band exists). According to Valet and Fert the value of the magnetoresistance *per layer*—defined as the difference in resistance of the ferromagnetically aligned multilayer and the antiferromagnetically aligned multilayer—is given by the formula

$$(\Delta\rho/\rho)_{\text{per layer}}=(2\coth[u])^{-1}-(\tanh[u]+\coth[u])^{-1}\ , \tag{25}$$

where $u=t/2\Lambda$. This function has a maximum with respect to u at $u=1$. The total magnetoresistance is the above equation multiplied by the number of layers in the multilayer T/t, where T is the total thickness of the multilayer. When plotted versus t, with Λ fixed, this function has no maximum, and falls off with increasing t when $t>\Lambda$.

Because of geometrical considerations, the physics of multilayers is a bit different from that of granular solids. Consider, for example, the forces determining the ground-state magnetic configuration. In a multilayer, dipole fields do not join two neighboring layers, except for edge effects. On the other hand, the interaction via indirect exchange fields is more important for the determination of the magnetic configuration in multilayers than in granular materials.

In comparing the GMR in granular materials to that in multilayers, a few generalities may be safely made. The magnetoresistance of granular materials is intermediate between that of the current perpendicular-to-plane and current in-plane multilayer geometries. Whereas the direction of the electric field is trivially solved in the multilayers, it is a distinctly nontrivial problem in granular compounds.

VIII. EXPERIMENTS IN GRANULAR METALS

Figure 1 of this paper displays the percent magnetoresistance in granular materials as a function of annealing temperature, (and, *ispo facto,* as a function of grain size) with data taken from several recent papers. An (almost) universal characteristic of these curves is the appearance of a maximum, which occurs at some annealing temperature characteristic of the constituent elements and initial preparation of the sample. It has seemed clear from the first observations[1,2] that this phenomenon required an explanation.

First, let us enquire in what manner the percent GMR would vary with the size of the precipitates (the radius a of the magnetic spheres) if we *assume* that the conductivities σ_F and σ_N are independent of a.

The formulas for $\Delta\sigma/\sigma$ the change in conductivity from the demagnetized to the fully magnetized state, and for σ_{eff}, the conductivity at zero magnetic field, have been derived in previous sections of this paper for surface spin scattering and for two types of bulk spin scattering. The percent magnetoresistance $\Delta\rho/\rho$ is obtained from these expressions using $\Delta\rho/\rho=-\Delta\sigma/\sigma_{\text{eff}}$. In Figs. 3(a)–3(c) we show how the percent magnetoresistance varies with sphere radius a for the three different types of scattering, assuming that the conductivities do not vary with a. The filling factor f is assumed to remain independent of a, reflecting the immiscible nature of the elements.

In Fig. 3(a) we make the additional assumption that the spin-dependent surface resistance per unit surface r_s is also independent of the particle radius a. The magnetoresistance as a function of particle size is seen to be zero in the limit of small particles, and to attain a constant positive value when $r_s\gg a$.

The percent magnetoresistance for a granular compound whose conduction electrons undergo spin scattering solely within the magnetic granules is shown in Fig. 3(b). The magnetoresistance is independent of the granule size. Again, this statement assumes f, σ_N, σ_F, and Λ to be independent of granule size.

Finally, the percent magnetoresistance for a granular compound whose conduction electron undergo spin scattering solely within normal metallic matrix is shown as a function of particle size in Fig. 3(c), again assuming constancy of σ_F, σ_N, f, and Λ with particle size. In this case the percent GMR is zero when $a=0$, attains a weak minimum when $a=\Lambda$, and then a strong maximum when $a=4L$. As a approaches infinity, the GMR tend to zero,

again. This result has an interesting physical interpretation. When the particle size is very large, the distance between ferromagnetic particles is large, and the interaction is small, hence a small magnetoresistance. When the size of the granules is very small, the granules more and more appear to be alloyed with the normal host metal, as the spin-polarization cloud surrounding each of the particles begin to overlap. Since the essence of GMR is heterogeneity, magnetoresistance disappears at small distances (sizes) also. And, since Λ is the only characteristic length in the problem, the GMR is a maximum when a equals a small integer times the quantity Λ.

Figure 3(c) bears a resemblance to the experimental results shown in Fig. 1. Alas, however, this appears to be merely coincidental. Figures 3(a)–3(c) all assume that the conductivities and spin-diffusion lengths are all independent of granule size. But quite the opposite is true. Both the conductivities and the diffusion lengths are strongly dependent on a, the radius of the spheroids composing the granular material.

By now, several workers have measured the effect of annealing on the resistivity (as well as the percent magnetoresistance), and have found that the resistivity decreases by an order of magnitude on going from an unannealed to a well-annealed sample. For example, Xiong *et al.*[5] have studied the magnetotransport properties of granular Co-Ag, simultaneously measuring the resistivity ρ, the magnetoresistivity $\Delta\rho$, and the Hall resistivity ρ_{xy}. From these measurements, together with TEM measurements of the particle size, they have concluded $\Delta\rho \propto a^{-1}$, in agreement with the quantum transport theory of Zhang and Levy.[4] In fact, *both* ρ and $\Delta\rho$ decrease with increasing particle size (or equivalently with annealing temperature) in a similarly precipitous manner. The picture which emerges from these experiments is that the mean free path of the conduction electrons λ_{mfp} is limited by the interparticle distance, and is very nearly equal to the particle radius a to a fairly good approximation.

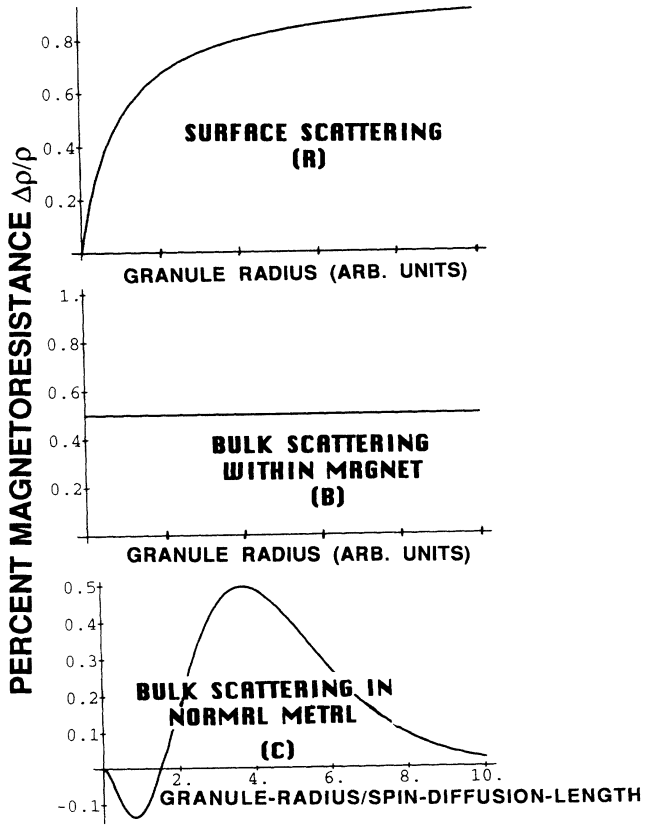

FIG. 3. Percent magnetoresistance ($\Delta\rho/\rho$) versus particle radius (a) for granular sample composed of magnetic spheres of radius a packed with filling factor f, for (a) surface interface spin scattering of conduction electrons between magnetic and normal metals, (b) bulk spin scattering of conduction electrons in the magnetic metal only, and (c) bulk spin scattering of conduction electrons in the normal metal only. It is assumed in these figures that the conductivities do not vary with a.

$$\lambda_{\text{mfp}} \approx a \ . \tag{26}$$

Thus, we are forced to abandon any pretense that the macroscopic parameters in our differential equations are independent of particle size. Since the conductivity and the spin-diffusion length are proportional to the mean free path, we can assume

$$\begin{aligned} \Lambda &\propto a \ , \\ \sigma_N &\propto a \ , \\ \sigma_F &\propto a \ , \end{aligned} \tag{27}$$

where Λ is greater than λ_{mfp} by an order of magnitude, since only a small fraction of electron collisions are spin-flip events. In addition, we can no longer assume that the surface resistance r_s is independent of particles size.

r_s, the ferromagnet/normal interface resistance defined by Eq. (3), can be given by the formula[7]

$$r_s = \frac{\Lambda_s}{\sigma_F} \ , \tag{28}$$

where Λ_s is the interface spin-scattering length. From a quantum-mechanical transition probability calculation, Zhang and Levy have derived

$$\Lambda_S \propto a \ . \tag{29}$$

This proportionality comes about because the number of "surface impurities" responsible for producing conduction-electron spin-flips is proportional to the surface area of the particles, if interface spin-scattering dominates. The surface-to-volume ratio of a sphere is equal to $3/a$. From Eqs. (27), (28), and (29), we deduce that r_s is actually *independent* of the particle size.

Using the above assumptions that σ_F, σ_N, and Λ are all proportional to a, and that value of r_s is also independent of a, we are now in a position to determine the dependence of ρ, $\Delta\rho$, and $\Delta\rho/\rho$ on the particle size, and inferentially on the annealing temperature. We find that, for all three mechanisms which we have discussed—surface scattering, bulk normal metal scattering, and bulk magnetic scattering—the following equations hold within our level of approximation:

$$\rho \propto a^{-1},$$
$$\Delta\rho \propto a^{-1}, \tag{30}$$
$(\Delta\rho/\rho)$ independent of a .

In Fig. 4 we show the transport data published by Wang and Xiao[6] on granular $Fe_{20}Ag_{80}$. Both ρ and $\Delta\rho$ decrease monotonically with T_A (and with a). $\Delta\rho/\rho$ increases slightly at low T_A and then decreases at high T_A, giving rise to a broad maximum. Equation (30) predicts that both ρ and $\Delta\rho$ decrease with a. Indeed, TEM measurements on these samples reveal that $\Delta\rho$ varies inversely with the Fe particle radius. But the behavior of $\Delta\rho/\rho$ cannot be explained by Eq. (30), since the $\Delta\rho/\rho$ data has a maximum between $T_A=100\,^{\circ}\mathrm{C}$ and $T_A=200\,^{\circ}\mathrm{C}$, and decreases substantially thereafter, while the theory predicts that $\Delta\rho/\rho$ should remain independent of T_A.

The order of magnitude predicted by these models can be estimated using the surface scattering mechanism, alone. We assume the following as rough values for the parameters which can be associated with granular $Fe_{20}Ag_{80}$: $\sigma_F/\sigma_N=0.2$ (clean limit) or $\sigma_F/\sigma_N=1$ (dirty limit), $\Lambda_s=a$ for the interface spin-scattering length, and $f=0.2$. We obtain $\Delta\rho/\rho=0.7\%$ (clean limit) and $\Delta\rho/\rho=3\%$ (dirty limit). This can be compared to an optimum annealing value of 22% at 4.2 K and 7% at 300 K. The data, without units, is displayed in Fig. 4.

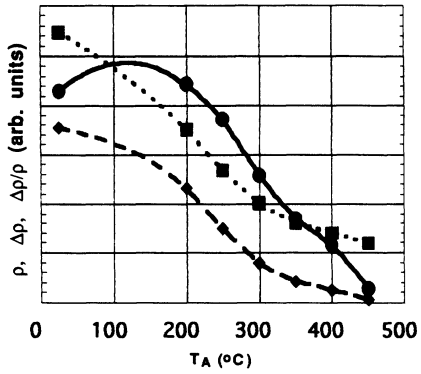

FIG. 4. Percent magnetoresistance (circles), resistivity (squares), and net change in resistivity between $H=0$ and 8 T (diamonds) versus annealing temperature. Data is taken from Ref. 6 for a sputtered $Fe_{20}Ag_{80}$ film.

IX. DISCUSSION

Equation (30) predicts that both ρ and $\Delta\rho$ vary with particle size as a^{-1}. In Fig. 4, $\Delta\rho$ is seen, experimentally, to vary inversely with particle size. ρ, on the other hand, varies first faster, and then slower, than a^{-1}. Considering its limitations, these facts do not contradict the theory, but point out its limitations. However, Eq. (30) *incorrectly* predicts that $\Delta\rho/\rho$ will be *independent* of particle size or of annealing temperature when the curves in Fig. 1 show that a maximum in $\Delta\rho/\rho$ vs T_A will occur, and that some materials can possess a very sharp maximum, indeed. Below, I will briefly discuss two ideas which can remedy this fault in the theory.

The first idea—suggested previously by Berkowitz *et al.*[1] and worked out by Zhang and Levy[4]—simply states that very small particles require enormous magnetic fields to achieve saturation. Since the measurements are taken at a finite field < (which varies from experimenter to experimenter), we may expect that the measured magnetoresistance can differ increasingly from the true saturated magnetoresistance as the particles sizes become smaller and smaller. Naturally, each experiment must be separately analyzed for this effect.

The second idea to which I allude is a mathematical extension of the theory to include higher-order multipoles in our expansion of the electric-field interaction between magnetic particles. I will only give a historical introduction to this method, briefly explaining how its introduction *might* alter the conclusions about granular magnetoresistance which were reached in Eq. (30). I am forced to delay discussion of its consequences, if any, to a later paper.

We have seen that an applied electric field can induce a spin-dependent electric dipole moment on a magnetic sphere imbedded within a normal metal. The dipole-dipole interaction have been seen to be one of the sources of magnetoresistance. As the spheres approach each other, these dipolar fields grow larger, causing additional rearrangements of electric charges on the spheres, creating octupole moments, etc. In 1892 Lord Rayleigh[11] devised a method to calculate the conductivity of a simple cubic lattice of conducting spheres in a conducting matrix by taking into account these induced octupole moments. Higher-order multipole effects have been recently calculated by McPhedran and McKenzie,[13] yielding excellent agreement with measurements on arrays of perfectly conducting spheres, even when they are close to touching. It is my hope that a similar inclusion of higher multipoles will yield results in the *magnetoresistance* which are in better agreement with experiment than the simple dipole expansion used above.

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