Random walk simulation of magnetotransport in magnetic granular systems

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(Received 10 June 2003; published 2 October 2003)

An integral series for the conductivity of magnetic inhomogeneous systems was previously derived from the real-space Kubo formula and semiclassical Boltzmann equation independently. In this work, we show that the integral series can be evaluated by using a random-walk sampling technique. The approach is applied first to a simple nonmagnetic single-granule system, and then to the giant magnetoresistance (GMR) effect of magnetic granular systems. It is well suitable for the granular systems in which the granular size is smaller than the electron mean free path, where the macroscopic approach becomes invalid. The GMR is found to always increase with decreasing size of granules. Optimum GMR could be obtained by a proper adjustment of the spin-dependent scattering strengths due to interfacial roughness and bulk impurities.

DOI: 10.1103/PhysRevB.68.132401 PACS number(s): 75.47.De, 72.10.Bg, 72.25.Ba

Since the discovery of the giant magnetoresistance (GMR) effects in magnetic layered structures[1–4] and magnetic granular solids[5,6] a decade ago, there have been many continuous research activities motivated to understand the transport properties of magnetic inhomogeneous systems. A recent study of the promising technology applications of spintronics devices[7] has renewed much interest in the field. A problem of theoretical importance in this area is how to describe the electronic transport in systems where the inhomogeneous characteristic length is comparable to or smaller than the electron mean free path. In such systems, the electric current density at a site is determined by the integrated effect of the spatially varying impurity scattering and electric field in the vicinity, which are generally related by a nonlocal conductivity tensor.

Among the GMR systems, the magnetic layered structures are relatively easy to investigate theoretically because of the translational invariance in the plane of the layers. By symmetry, in the case with electric currents in the plane of the layers (CIP), the electric field is uniform. In the case with electric currents perpendicular to the plane of the layers (CPP), the current density is uniform. These conditions greatly reduce the complexity of determining the transport properties. Semiclassical theories[8–12] based on the Boltzmann equation and quantum theories[13–15] using the Kubo formula have been well established for the layered structures. The magnetic granular systems are more challenging in the sense that neither the electric field nor the current density is uniform. Existing theories are usually based upon certain unjustified approximations. In earlier works,[5,16] it was assumed that the transport in the granular systems is very close to that in the layered structures with CPP, which essentially neglects the spatial variance of the current density. In later works,[17–20] the macroscopic approximation is adopted straightforwardly, and the electric field and current density are determined self-consistently. However, the approximation is questionable if the size of the granules is smaller than the electron mean free path.

As an effort toward a general transport theory for three-dimensionally magnetic inhomogeneous systems, Camblong and Levy[14] derived an integral series for the two-point conductivity tensor starting from the real-space Kubo formula. The same result was recovered by use of an extended Boltzmann equation.[18,21] However, no efficient approach has so far been developed to tackle the series. Such an approach is highly desirable. In this work, we propose an algorithm to evaluate the two-point conductivity formula. It is found that the integral series can be computed by Monte Carlo (MC) simulation as a random-walk problem. A simple system with a single nonmagnetic granule embedded in a nonmagnetic matrix is studied, and the result is compared with that obtained under the macroscopic approximation. The average conductivity and GMR amplitude of magnetic granular solids are then calculated.

Let us now write down the nonlocal formula for the current density:[14,18,21]

$$\mathbf{J}_s(\mathbf{r}) = \int d^3r' \overleftrightarrow{\sigma}_s(\mathbf{r},\mathbf{r}') \cdot \mathbf{E}(\mathbf{r}'). \tag{1}$$

The two-point conductivity tensor is given by

$$\begin{aligned}\overleftrightarrow{\sigma}_s(\mathbf{r},\mathbf{r}') = 3C_D \Big[& \hat{\mathbf{n}}_{\mathbf{r}\mathbf{r}'} \hat{\mathbf{n}}_{\mathbf{r}\mathbf{r}'} D_s(\mathbf{r},\mathbf{r}') \\ & + \int d^3r_1 \hat{\mathbf{n}}_{\mathbf{r}\mathbf{r}_1} \hat{\mathbf{n}}_{\mathbf{r}_1\mathbf{r}'} D_s(\mathbf{r},\mathbf{r}_1) \lambda_s^{-1}(\mathbf{r}_1) D_s(\mathbf{r}_1,\mathbf{r}') \\ & + \int d^3r_1 \int d^3r_2 \hat{\mathbf{n}}_{\mathbf{r}\mathbf{r}_1} \hat{\mathbf{n}}_{\mathbf{r}_2\mathbf{r}'} D_s(\mathbf{r},\mathbf{r}_1) \lambda_s^{-1}(\mathbf{r}_1) \\ & \times D_s(\mathbf{r}_1,\mathbf{r}_2) \lambda_s^{-1}(\mathbf{r}_2) D_s(\mathbf{r}_2,\mathbf{r}') + \cdots \Big],\end{aligned} \tag{2}$$

where $C_D = ne^2/2mv_F$, $\hat{\mathbf{n}}_{\mathbf{r}\mathbf{r}'} = (\mathbf{r}-\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|$ is a unit vector in the direction of $\mathbf{r}-\mathbf{r}'$, $s=+$ and $-$ represent the electron spins parallel and antiparallel to a fixed reference direction, respectively, and

$$D_s(\mathbf{r},\mathbf{r}') = \frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|^2} \exp[-\xi_s(\mathbf{r},\mathbf{r}')]. \tag{3}$$

Here, $\xi_s(\mathbf{r},\mathbf{r}') = \int_{\mathbf{r}\to\mathbf{r}'} dr''/\lambda_s(\mathbf{r}'')$ is the effective distance between $\mathbf{r}$ and $\mathbf{r}'$ measured by the electron mean free path. We will confine ourselves to collinear magnetization con-

figurations, where the magnetic moments of the ferromagnetic entities can only be parallel or antiparallel to the reference direction. In this case, the electrons in the two spin channels are independent, and the total current density is a sum over the two spin channels.

In general, both the charge density and internal electric field $\mathbf{E}(\mathbf{r})$ are nonuniform. Self-consistent calculation of $\mathbf{E}(\mathbf{r})$ and the redistributed charge density in terms of the Maxwell equations and the equation of motion for the electrons could be very complicated. Fortunately, in Ref. 21, by taking into consideration of the continuity condition of the electric currents, it is shown that the current density depends only on the electric voltage drop applied between the two ends of the system, being irrelevant to the specific distribution of the electric field. One can choose a convenient electric field in calculations, which will not change the measurable physical results. It is formally proved[18] that there always exists an *optimized electric field*, which eliminates the contributions to the electric currents from all the terms except for the first in Eq. (2). For layered structures, it is possible to work with the optimized electric field directly. In fact, for the CIP case, the optimized electric field is simply uniform. For the CPP case, the optimized electric field is found to be in inverse proportion to the local electron mean free path.[22] However, for the granular systems under consideration, it is difficult to determine the optimized electric field. The present approach is to choose a uniform electric field $\mathbf{E}(\mathbf{r})=\mathbf{E}$. The cost is that we have to sum the integral series of Eq. (2). Intuitively, the convergence speed of the integral series will depend on the deviation of the chosen electric field from the optimized electric field.

We set the z axis in the direction of the electric field. By substituting Eq. (2) into Eq. (1), the z component of the current density is obtained as

$$J_s^z(\mathbf{r})=3C_DE\lim_{N\to\infty}\int d^3r_1\cdots\int d^3r_N \times\left\langle \hat{n}^z_{\mathbf{r}_0\mathbf{r}_1}\sum_{i=1}^{N}\hat{n}^z_{\mathbf{r}_{i-1}\mathbf{r}_i}\lambda_s(\mathbf{r}_i)\right\rangle\prod_{i=1}^{N}\rho_s(\mathbf{r}_i|\mathbf{r}_{i-1}), \quad (4)$$

where, for convenience, $\mathbf{r}$ is denoted as $\mathbf{r}_0$ on the right-hand side, and

$$\rho_s(\mathbf{r}_i|\mathbf{r}_{i-1})=\frac{1}{4\pi\lambda_s(\mathbf{r}_i)|\mathbf{r}_i-\mathbf{r}_{i-1}|^2}\exp[-\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)]. \quad (5)$$

Using a spherical coordinate system with the origin at $\mathbf{r}_{i-1}$ and designating $(r_{i,i-1},\theta_{i,i-1},\varphi_{i,i-1})$ as the coordinates of $\mathbf{r}_i$ in the coordinate system, we can obtain

$$\int d^3r_i\rho_s(\mathbf{r}_i|\mathbf{r}_{i-1})=\frac{1}{4\pi}\int_{-1}^{1}d(\cos\theta_{i,i-1}) \times\int_0^{2\pi}d\varphi_{i,i-1}\int_0^{\infty}d\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i) \times\exp[-\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)]=1, \quad (6)$$

where the relation $\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)\lambda_s^{-1}(\mathbf{r}_i)dr_{i,i-1}=d\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)$ has been used. With identity (6), Eq. (4) can be readily derived back to Eqs. (1) and (2). The identity also indicates that $\rho_s(\mathbf{r}_i|\mathbf{r}_{i-1})$ can be regarded as a conditional probability density function (PDF) governing the distribution of $\mathbf{r}_i$ for a given $\mathbf{r}_{i-1}$. As a consequence, Eq. (4) can be sampled as a random-walk problem by using the MC technique. First, for a given $\mathbf{r}_0$, $\mathbf{r}_1$ is chosen according to PDF $\rho_s(\mathbf{r}_1|\mathbf{r}_0)$. Then $\mathbf{r}_2$ is chosen with respect to $\mathbf{r}_1$ according to PDF $\rho_s(\mathbf{r}_2|\mathbf{r}_1)$, and so on to $\mathbf{r}_i$ with $i=3,4,\ldots,N$, which are chosen in the same way. More specifically, in terms of the spherical coordinates defined for Eq. (6), to obtain $\mathbf{r}_i$ for given $\mathbf{r}_{i-1}$, one can choose $\cos\theta_{i,i-1}\in[-1,1)$ and $\varphi_{i,i-1}\in[0,2\pi)$ randomly. For the radial coordinate, $\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)$ is first chosen according to PDF $\exp[-\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)]$, and $r_{i,i-1}$ is then determined inversely from $\xi_s(\mathbf{r}_{i-1},\mathbf{r}_i)$ by geometric calculation. Many such repeated sampling procedures will generate an ensemble of coordinates $(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$ for each spin channel. The current density is obtained as an average over the ensemble

$$J_s^z(\mathbf{r})=3C_DE\left\langle \hat{n}^z_{\mathbf{r}_0\mathbf{r}_1}\sum_{i=1}^{N}\hat{n}^z_{\mathbf{r}_{i-1}\mathbf{r}_i}\lambda_s(\mathbf{r}_i)\right\rangle_s. \quad (7)$$

Let us consider first a single nonmagnetic granule with electron mean free path λ embedded in a nonmagnetic medium with mean free path λ_0. The granule is assumed to be spherical with radius R_0, and its center is taken as the origin of our coordinate system. To model the interface roughness scattering, a thin mixed layer of thickness t and mean free path λ_I is assumed at the interface between the granule and the medium. For simplicity, we will take the limit $t/R_0\to 0$ but keep t/λ_I as a constant. In this simple system, the transport properties are spin-independent and the spin index will be omitted for a while. Since it is difficult to take the limit $N\to\infty$ in Eq. (4) exactly, we wish to see the convergence of the integral series with increasing N. The current density $J^z(\mathbf{r})$ is calculated for some points on the x and z axes with N up to 20, as shown systematically in Fig. 1. Here, $\sigma_0=2C_D\lambda_0$ is the conductivity of the medium. For each point, 2×10^6 realizations of $(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$ are sampled. We see that the dominant contribution to the current density mainly comes from the first several terms. Most curves become rather flat at $N\simeq 20$, indicating that contributions from the higher-order terms are insignificant. In order to balance between accuracy and computation time, $N=20$ will be used hereafter.

In several previous papers,[17–20] the single-granule problem has been studied based upon the macroscopic approach, in which a local dependence of the current density on the electric field was assumed. We wish to make a comparison between the present theory and the macroscopic approach. Since the detailed solution in the macroscopic approach has been documented in the cited references, without repeating it we present its result directly. To measure the contribution of a single granule to the system resistivity, it is convenient to introduce an equivalent volume for the granule:

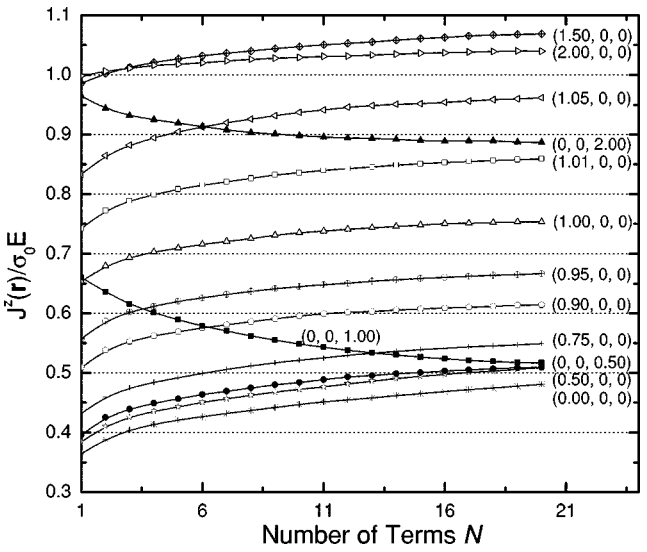

FIG. 1. Calculated current density $J^z(\mathbf{r})$ normalized by $\sigma_0 E$ as a function of N. The parameters used are $R_0=\lambda_0$, $\lambda=\lambda_0/3$, and $t/\lambda_I=0.05$. The corresponding position $\mathbf{r}$, for which the current density $J^z(\mathbf{r})$ is calculated, is normalized by λ_0 and shown in the form $\mathbf{r}/\lambda_0=(x/\lambda_0,y/\lambda_0,z/\lambda_0)$ to the right of each curve.

$$\Omega_{\rm eq}=\frac{1}{\sigma_0 E}\int d^3r[\sigma_0 E-J^z(\mathbf{r})]. \tag{8}$$

By definition, $V\sigma_0 E-\int J^z(\mathbf{r})d^3r=\Omega_{\rm eq}\sigma_0 E$ with V the total volume of the system. It indicates that the change in the total currents due to the addition of the granule in the medium is equal to that due to removal of a volume $\Omega_{\rm eq}$ from the original medium. The more resistive the granule, the greater the $\Omega_{\rm eq}$. A helpful identity $V^{-1}\int d^3r J^z(\mathbf{r})/\lambda(\mathbf{r})=C_D E$ can be derived by substituting Eq. (2) into Eq. (1), dividing both sides of Eq. (1) by $\lambda(\mathbf{r})$ and integrating over $\mathbf{r}$. To calculate the equivalent volume, Eq. (8), we only need to determine $J^z(\mathbf{r})$ inside the granule (including the interfacial mixed layer), and the integral of $J^z(\mathbf{r})$ in the medium can be obtained by using the above identity. In Fig. 2, the calculated

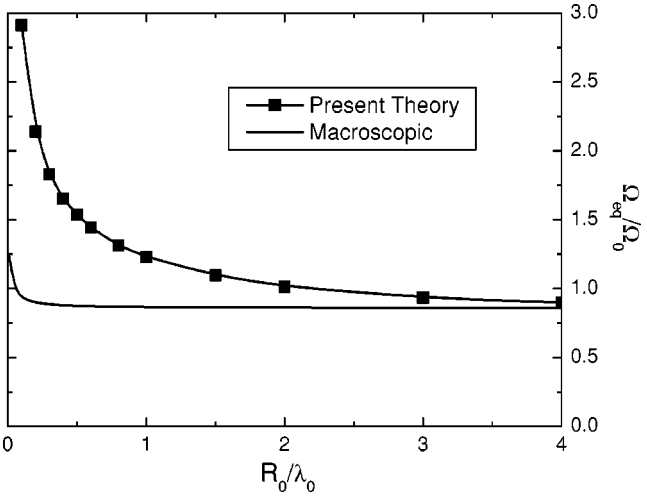

FIG. 2. Normalized granular equivalent volume as a function of R_0/λ_0. Here, $\lambda=\lambda_0/3$ and $t/\lambda_I=0.05$.

equivalent volume normalized by the granular volume $\Omega_0=4\pi R_0^3/3$ is plotted as a function of R_0/λ_0 against the result from the macroscopic approximation. 4×10^6 realizations of $(\mathbf{r}_0,\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$ with $\mathbf{r}$ $(=\mathbf{r}_0)$ randomly distributed inside the granule are sampled for each value of R_0. The two results are close for $R_0\gg\lambda_0$. However, for $R_0\lesssim\lambda_0$, the macroscopic theory underestimates $\Omega_{\rm eq}$ by tens of percent.

Now we turn to the GMR effect of magnetic granular systems. Multiple granules can be included directly into our numerical approach. The main difference is that, depending on the positions $\mathbf{r}$ and $\mathbf{r}'$, more than one granule may now affect the line segment integral in $\xi_s(\mathbf{r},\mathbf{r}')$ together. The centers of the granules are assumed to form a cubic lattice simply for the convenience of numerical calculations. For the present nonquantum problem, it can be expected that the assumption of a periodic arrangement instead of a random distribution of the granules will not change the result significantly. By introducing a volumetric filling factor f of the ferromagnetic material, the lattice constant can be obtained as $a_0=(4\pi/3f)^{1/3}R_0$. We designate $\lambda_{\uparrow(\downarrow)}$ to be the electron mean free path inside a granule for spin parallel (antiparallel) to the local magnetic moment, and $\lambda_{I\uparrow(\downarrow)}$ to be the mean free path in the interfacial mixed layers. A single spin asymmetric factor $\Lambda=\lambda_\downarrow/\lambda_\uparrow=\lambda_{I\downarrow}/\lambda_{I\uparrow}$ is assumed.

In order to determine the GMR amplitude, we calculate separately the average conductivities $\sigma_{\rm FM}$ and $\sigma_{\rm AF}$ for ferromagnetic and antiferromagnetic magnetization configurations. Then the GMR amplitude is given by

$$\mathrm{GMR}=\frac{\sigma_{\rm FM}-\sigma_{\rm AF}}{\sigma_{\rm FM}}. \tag{9}$$

In the calculation, $\mathbf{r}$ $(=\mathbf{r}_0)$ in Eq. (7) is moved randomly in two neighboring cells of the cubic lattice formed by the granules. Obviously, this will yield the same average conductivity as moving $\mathbf{r}$ throughout the whole sample. The calculated GMR amplitude and the conductivity for different values of $t/\lambda_{I\downarrow}$ and $\lambda_\downarrow/\lambda_0$ are plotted in Fig. 3 as functions of the radius. Here, the volumetric filling factor f is fixed at 0.3. In all cases, with decreasing granular radius, the GMR amplitude increases and the conductivity decreases. The GMR has a nontrivial dependence on the strength of spin-dependent scattering. For example, for $r<0.5$, among the three curves with $t/\lambda_{I\downarrow}=0.0,0.05,0.1$ and with $\lambda_\downarrow/\lambda_0=1$, $t/\lambda_{I\downarrow}=0.05$ corresponds to the greatest GMR, an implication that there exists an optimum strength of interfacial scattering for the GMR. Similarly among the three curves with $\lambda_\downarrow/\lambda_0=0.2,1.0,5.0$ and with $t/\lambda_{I\downarrow}=0.05$, we find that there exists an optimum strength of spin-dependent bulk impurity scattering as well. Furthermore, the optimum strengths are different for different granular sizes.

In earlier papers,[5,16] the transport in granular systems was assumed to be close to the CPP case in the layered structures, where the resistances from different sources are additive. In that theory, the GMR must increase monotonically with the strength of spin-dependent scattering. Gu *et al.*[19] pointed out that the transport in granular systems is between the CPP and CIP cases. The present theory supports this viewpoint. It has

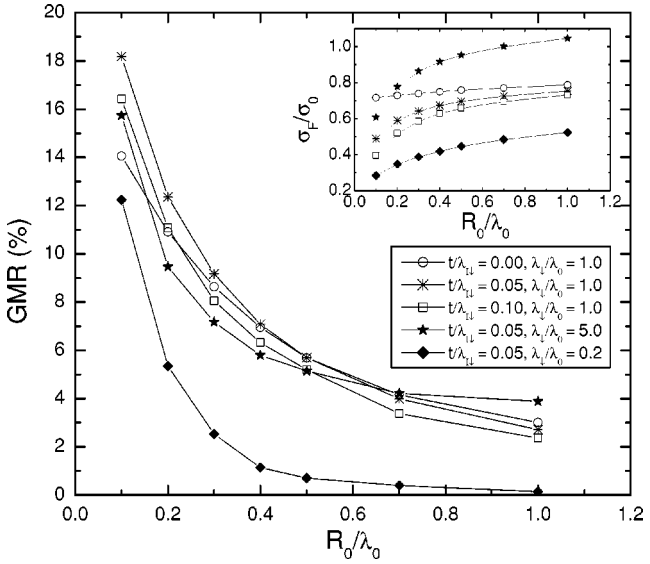

FIG. 3. GMR amplitude and conductivity (inset) calculated for some $t/\lambda_{I\downarrow}$ and $\lambda_\downarrow/\lambda_0$ as functions of granular radius at $\Lambda = 6$.

been found that, for the magnetic layered structures with CIP, there exists indeed an optimum strength of spin-dependent scattering for the GMR.[23]

In experiments,[5,6] the GMR is observed to first increase and then decrease with decreasing annealing temperature, which is believed to decrease the granular size. In the present theory, however, the GMR always increases with decreasing granular size. A reasonable explanation for this difference is that, even though decreasing the granular size is favorable for the GMR, the magnetic moments of smaller granules is more difficult to be saturated, which disfavors the GMR.[16] Additionally, the number of ferromagnetic granules may decrease at lower annealing temperatures due to incomplete phase separation between the nonmagnetic matrix and the ferromagnetic material.[5,6] It is also interesting to compare our calculated GMR with that obtained by using the macroscopic approximation.[18–20] It is found that the two approaches give qualitatively different dependences of GMR on granular size in the limit of small granules. The GMR obtained in the macroscopic approximation[18–20] first increases and then decreases, exhibiting a maximum at a certain granular size smaller than the electron mean free path. At first glance, the nonmonotonic dependence obtained there is qualitatively consistent with the experimental data. In reality, however, it is only a coincidence, because the macroscopic approximation fails to work for a granular size smaller than the mean free path. Indeed, one can easily verify that the macroscopic approximation always yields a vanishing GMR in the CIP case of the layered structures. This unreasonable result stems from the fact that there will be no CIP GMR if the layer thickness is assumed to be much greater than the mean free path, which is just the appropriate range of the macroscopic approach. As a result, the prediction of a decreased GMR with decreasing granular size is a failure of the macroscopic approximation in the small granule region, even though the argument is valid that the transport in the granular systems becomes close to the CIP case of the layered structures with decreasing granular sizes.[19] The present theory leads to much higher GMR amplitudes than the macroscopic theory does, being more comparable to the experimental data.[5,6]

In summary, we have developed a numerical approach to compute the integral series of the two-point conductivity formula for magnetic inhomogeneous systems. This work fulfills a unified practicable description of the GMR effect in magnetic inhomogeneous systems.

D.Y.X. is grateful for support from the State Key Program for Basic Research of China. D.N.S. was supported by ACS-PRF Grant No. 36965-AC5, Research Corporation Grant No. CC5643, and the NSF through Grant No. DMR-00116566.

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