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Spin-flip effects on the current-in-plane magnetotransport in magnetic multilayers with arbitrary magnetization alignments

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An extended Boltzmann equation approach, with nondiagonal components of the electron distribution function taken into account, is proposed to study spin-flip effects on the magnetoresistance (MR) in magnetic inhomogeneous systems with arbitrary magnetization alignments. The presence of spin-flip scattering is found to reduce the MR and to decrease deviation of the MR from linear dependence on \( \sin^2 (\theta/2) \) where \( \theta \) is the angle between the magnetizations of successive magnetic films. [S0163-1829(98)01942-0]

Giant magnetoresistance (MR) in magnetic multilayers and magnetic granular composites has attracted much interest for recent years. In most theoretical approaches to the giant MR, only the collinear magnetization configuration (CLMC) was considered, i.e., all the ferromagnetic (FM) regions such as FM layers or FM granules are assumed to have only two possible magnetization directions, parallel or antiparallel to a fixed spin quantization axis. However, such an assumption is untenable in real magnetic inhomogeneous systems. For a magnetic granular composite in the absence of external magnetic field, the magnetizations of FM granules are usually oriented randomly. In magnetic multilayers, even though there may be antiferromagnetic coupling between adjacent FM layers, the angle dependence of the giant MR has been experimentally investigated for both current in plane (CIP) of the layers and current perpendicular to plane (CPP) of the layers. In the CIP case, it is found that the magnitude of the MR is approximately proportional to \( \sin^2 (\theta/2) \), where \( \theta \) is the angle between the magnetizations in successive FM layers. In the CPP case, significant deviations from this \( \theta \) dependence were observed in some systems. As a result, the theoretical study of MR in arbitrary magnetization orientations is highly desirable.

The problem of angular dependence was first discussed by Levy, Zhang, and Fert for an infinite magnetic superlattice, where a linear variation of MR with \( \sin^2 (\theta/2) \) was obtained. Later, Vedyayev et al. applied the real-space Kubo formalism in a layered structure composed of two magnetic films in direct contact to obtain the linear variation of CIP MR with \( \sin^2 (\theta/2) \). Recently, Sheng et al. developed a reformed Boltzmann equation approach to this problem. They proposed that, for arbitrary magnetization arrangements, it is inadequate for the previous quasiclassical approach to view the electron distribution function as a classical two-component vector in the Boltzmann equation. Instead, the electron distribution function should be regarded as a spinor matrix that is usually off-diagonal in the spin space of conduction electrons. A similar quasiclassical treatment was used in a recent paper, in which the angular variation of CIP MR was studied in the quasiclassical and quantum limits. In these theoretical works, spin-flip and diffusion effects are not taken into account, since the spin-diffusion length is usually considered to be much longer than the thicknesses of the layers. Nevertheless, if the spin-diffusion length is comparable with the layer thickness, the spin-flip scattering will play an important role in the magnetotransport in magnetic multilayers. In this paper we further extend the Boltzmann equation approach with spinor distribution function to include the spin-flip effects from which the angular dependence of giant MR is studied in the presence of the spin-flip scattering. It is found that, in both local and homogeneous limits, the CIP MR is proportional to \( \sin^2 (\theta/2) \); in the intermediate region, however, there is a small deviation from the linear dependence. While reducing the magnitude of MR, the existence of spin-flip scattering suppresses the deviation of the MR from the linear \( \sin^2 (\theta/2) \) dependence.

In arbitrary magnetization configurations, the electron distribution function should be considered as a 2x2 matrix in the spin space, whose elements are determined by the choice of the quantization axis. The spin distribution function under a local quantization axis can be written in the form \( \tilde{f}_L (v, \mathbf{r}) = f_0 \tilde{1} + g_L (v, \mathbf{r}) \), where \( f_0 \) is the equilibrium distribution and \( g_L \) is the deviation from that equilibrium when an electric field is applied. Let us first consider the special case of CLMC, in which both the distribution function \( g_L \) and the electric field \( \mathbf{E}_L \) can be simultaneously diagonal in the spin space. In the presence of spin-flip scattering, the Boltzmann equation reads

\[
\mathbf{v} \cdot \nabla \tilde{g}_L + \tilde{\xi}_L (\mathbf{r}) \tilde{g}_L - \mathbf{E}_L (\mathbf{r}) \cdot \tilde{\sigma}_L \frac{\partial f_0}{\partial \varepsilon}. 
\]
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Here $\hat{\xi}_L$ and $\xi'\hat{1}$ are diagonal matrices that describe the inverse relaxation times. The presence of magnetic impurities can result in the spin-flip scattering. If the imurity magnetic fields on different sites are randomly oriented within an angle $\theta_i$ from the quantization axis direction, one has\(^{18}\) $\hat{\xi}_L = \tau^{-1} + (3 - x)/2\tau_{jy}$ and $\xi' = (1 - x)/2\tau_{sf}$, where $\tau_j$ is the non-spin-flip relation time of electrons with spin $j$, $\tau_{sf}$ is the spin-flip relation time, and $x = \sin(2\theta_j)/2\theta_j$. Thus, the next term of Eq. (1) is the Pauli matrix, and the action of $\hat{\sigma}_y\hat{g}_L(v, r)\hat{\sigma}_y$ is to exchange the two diagonal matrix elements of $\hat{g}_L(v, r)$.

We now extend Eq. (1) to the case of arbitrary magnetization alignments. In the local quantization axis, the diagonal $\hat{\xi}_L$ remains unchanged, but $\hat{g}_L$ and $\hat{E}_L$ are no longer diagonal. Taking into account that $\hat{g}_L$ and $\hat{E}_L$ cannot be commutative and that all the terms of the Boltzmann equation must be Hermitian, the second term in Eq. (1) needs to be rewritten as $\hat{\xi}_L\hat{g}_L(v, r)\hat{\xi}_L$, where $[\hat{g}_L\hat{E}_L]$ stands for an anticommutator.\(^{16}\) For arbitrary magnetization alignments, it is inconvenient to use position-dependent local quantization axes. Thus, the next step is to make a coordinate transformation from the reference frame of the local quantization axes to a new one with a fixed quantization axis. The unitary matrix of such a transformation is given by

$$
\hat{U}(r) = \begin{pmatrix}
\cos(\theta/2) & \sin(\theta/2)e^{-i\phi} \\
-\sin(\theta/2)e^{i\phi} & \cos(\theta/2)
\end{pmatrix},
$$

where $\theta(r)$ and $\phi(r)$ are the spherical polar angles subtended by the local quantization axis with respect to the new fixed one. Under this transformation, $\hat{g}_L(v, r)$ and $\hat{E}_L(v, r)$ in Eq. (1) will be replaced by $\hat{g}(v, r) = \hat{U}^\dagger(v, r)\hat{g}_L(v, r)\hat{U}(r)$, $\hat{\xi}_L = \hat{U}^\dagger(v, r)\hat{\xi}_L(v, r)\hat{U}(r)$, and $\hat{E}(r) = \hat{U}^\dagger(v, r)\hat{E}_L(v, r)\hat{U}(r)$, respectively. Besides, this transformation for Eq. (1) will produce new matrices $\hat{U}(v, r)\hat{\sigma}_y\hat{U}(r)$ so as to add difficulties in solving the equation. Taking into account the identity $\hat{\sigma}_y\hat{U} = \hat{U}^*\hat{\sigma}_y$, we find a simple treatment of replacing $g_L$ in the third term of Eq. (1) by its conjugate matrix $\hat{g}_L$. After considering the two amendments to the second and third terms of Eq. (1), we multiply both sides of it by $\hat{U}$ from the right and by $\hat{U}^\dagger$ from the left, yielding

$$
v \cdot \nabla \hat{g} + \frac{1}{2}[\hat{\xi}(r), \hat{g}] - \xi'(r)\hat{\sigma}_y\hat{g}^*\hat{\sigma}_y = ev \cdot \hat{E}(r)\frac{df_0}{de}.
$$

which is an extended Boltzmann equation in the reference frame of the fixed quantization axis. In the derivation of Eq. (3) we have used the relation $\hat{U}^\dagger\hat{\sigma}_y\hat{g}_L\hat{\sigma}_y\hat{U} = \hat{\sigma}_y(\hat{U}^\dagger\hat{g}_L\hat{U})^*\hat{\sigma}_y = \hat{\sigma}_y\hat{g}^*\hat{\sigma}_y$.

In order to solve Eq. (3), we rewrite it as a more compact form by introducing a fourth-rank scattering tensor $\tilde{T}_{\alpha\beta, \gamma\eta}(r)$. When such a tensor acts on an arbitrary $2 \times 2$ spinor matrix $\hat{A}$, a new spinor matrix $\hat{B}$ is obtained with matrix element $\hat{B}_{\alpha\beta} = \tilde{T}_{\alpha\beta, \gamma\eta}\hat{A}_{\gamma\eta}$ where summation over repeated greek indices is implied. This operation is equivalent to an action of a $4 \times 4$ matrix on a four-component column vector if the four vector elements of $\hat{A}$ are expressed as four components of a vector. For brevity, this operation is written as $\hat{B} = \tilde{T}\hat{A}$ thereafter, in which the tilde stands for the fourth-rank spinor tensor. The purpose of introducing $\tilde{T}$ is to replace the two scattering terms in Eq. (3) by $\tilde{T}\hat{g}$. It is easy to see that in the local quantization axis, $\tilde{T}_L$ has the following $4 \times 4$ matrix form:

$$
\tilde{T}_L(r) = \begin{pmatrix}
\xi_1(r) & 0 & 0 & -\xi'(r) \\
0 & \xi_22(r) & 0 & 0 \\
0 & 0 & \xi_33(r) & 0 \\
-\xi'(r) & 0 & 0 & \xi_1(r)
\end{pmatrix},
$$

with $\xi_22(r) = \xi_33(r) = (\xi_1(r) + \xi_1(r))/2 + \xi'(r)$. The corresponding column vector $\hat{A}$ has four components; they are $\hat{A}_{\uparrow\uparrow}$, $\hat{A}_{\uparrow\downarrow}$, $\hat{A}_{\downarrow\uparrow}$, and $\hat{A}_{\downarrow\downarrow}$ in proper order. As the local quantization axis is transformed to the fixed one, the transformation matrix $\tilde{V}$ is a $4 \times 4$ unitary matrix, whose matrix element is given by $\tilde{V}_{\alpha\beta, \gamma\eta} = \hat{U}_{\alpha\beta}^\dagger\hat{\tilde{U}}_{\gamma\eta}^\dagger$. Under this transformation, we have $\tilde{V} = \tilde{V}_L\tilde{V}$ and $\tilde{V}_L = \hat{U}\hat{A}$, where $\hat{A}$ stands for $\hat{g}$, $\hat{E}$, and $\hat{E}_L$. As a result, with the aid of the scattering tensor introduced above, Eq. (3) can be rewritten in a very compact form:

$$
v \cdot \nabla \hat{g}(v, r) + \tilde{T}(r)\hat{g}(v, r) = ev \cdot \hat{E}(r)\frac{df_0}{de}.
$$

Equation (5), together with Eq. (4), is one of the major results in this paper, which is suitable to arbitrary magnetization alignment and independent of the choice of the spin quantization axis. Furthermore, the spin-flip effects have also been included in it. It is easily shown that if the spin-flip scattering is neglected, Eq. (5) can reduce to Eq. (8) of Ref. 16. In the presence of the spin-flip scattering, both equations differ from each other but have the same form. It follows that their solutions also have the same form. Using the derivation similar to in Ref. 16, we obtain the two-point fourth-rank spinor conductivity tensor\(^{19}\)

$$
\frac{\nabla}{\nabla} \hat{\tilde{S}}(r, r') = \frac{3C_D}{4\pi \left|r - r'\right|^2}\hat{\tilde{S}}(r, r'),
$$

where $C_D = n\alpha^2/(2m\nu_F)$ is a constant, $n = (r - r')/(|r - r'|)$ is the unit vector in the direction of $r - r'$, and the spinor propagation factor $\hat{\tilde{S}}(r, r')$ is given by

$$
\hat{\tilde{S}}(r, r') = P_{r' \rightarrow r} \exp\left[-\frac{1}{\nu} \int_{\Gamma(r, r')} d\sigma/\hat{\tilde{T}}(r')\right],
$$

with $\Gamma(r, r')$ indicating the oriented straight path that starts at point $r'$ and ends up at point $r$, and $P_{r' \rightarrow r}$, the path ordering operator along $\Gamma(r, r')$, which reordered the noncommuting $4 \times 4$ matrices in the exponential series from $r'$ to $r$ and from right to left. The two-point conductivity obtained here seems to be in the same form as those obtained in Refs. 16 and 19, but $\hat{\tilde{S}}(r, r')$ in Eq. (6) is a $4 \times 4$ matrix and includes the spin-flip effects.

We now consider a magnetic multilayer whose layers are assumed to lie in the $x$-$y$ plane and to stack along the $z$ axis.
Owing to transitional invariance in the x-y plane, the two-point conductivity tensor depends only on z and z'. As a result, Eq. (6) can reduce to

$$\bar{\sigma}(z,z') = \frac{3C_D}{4\pi} \int_1^\infty dt \left( \frac{t^2-1}{t^3} \epsilon_1 \epsilon_2 + \frac{2}{t^3} \epsilon_3 \epsilon_3 \right) \bar{S}(z,z'),$$

(8)

with the propagation factor tensor

$$\bar{S}(z,z') = P_{z' \rightarrow z} \exp \left( -\frac{1}{v_F} \int_{z'}^z dz'' \bar{T}(z'') \right),$$

(9)

where $\epsilon_1$ and $\epsilon_2$ are the unit vectors in the plane of the layers and in the z direction, respectively, $t = v_F/|v_z|$, and $z''(z')$ is the smaller (larger) one of z and $z'$. In what follows we discuss the angular dependence of CIP MR in the presence of spin-flip scattering. Consider a FM/NM superlattice composed of FM and nonmagnetic (NM) layers. In the FM layers there are spin-dependent non-spin-flip scattering rate $\xi_i(F)(s=\uparrow$ and $\downarrow$) and spin-flip scattering rate $\xi'(F)$. In the NM layers, the non-spin-flip scattering rate $\xi(N)$ is assumed to be spin independent and the spin-flip rate $\xi'(N)$ may arise if magnetic impurities are present. The scattering at FM/NM interfaces is regarded as impurity scattering within thin mixing interlayers with non-spin-flip and spin-flip scattering rates $\xi_i(I)$ and $\xi'(I)$, respectively. The fixed spin-quantization axis is chosen along the external magnetic-field direction and so the magnetization direction of the nth FM layer (n being the integer) can be characterized by polar angles ($\theta_n$, $\phi_n$). In the absence of external magnetic field, it is assumed that $\theta_n = \phi_n = 0$ for odd n and $\theta_n = \phi_n = 0$ for even n. In the CIP geometry, the electric field $\bar{E}(z)$ is a constant independent of position, and so we can define an average conductivity tensor of the system

$$\bar{\sigma}(\theta) = \frac{1}{L} \int_0^L dz \int_{-\infty}^{\infty} dz' \bar{\sigma}^{\uparrow\downarrow}(z,z').$$

(10)

Here L is the periodic length of the FM/NM superlattice, which is determined by the magnetization orientations of the FM layers in the absence of external magnetic fields. Under the present assumption, a period includes two FM layers, two NM layers, and four interlayers. They will be labeled with subscript i with $i = 1, 2, \ldots, 8$ in order of the spatial arrangement of layers from left to right.

Taking into account the periodic conditions $\bar{S}(z, z') = \bar{S}(z + nL, z' + nL)$ and $\int_{-\infty}^{\infty} dz' \bar{S}(z, z') = \sum_{n=\infty}^{\infty} \int_{nL}^{(n+1)L} dz' \bar{S}(z, z')$, we perform integrals over $z'$ and $z$ in Eq. (10) and obtain the $\theta$-dependent average conductivity tensor

$$\bar{\sigma}(\theta) = \frac{3C_Dv_F^2}{2L} \int_1^\infty dt \left( \frac{1}{t^3} - \frac{1}{t^5} \right) \bar{F}(\theta),$$

(11)

with

$$\bar{F}(\theta) = \sum_{i=1}^8 \tilde{T}_i^{-1}(d_i^{-1} - \tilde{Q}_i) + \sum_{i,j} \tilde{Q}_i \prod_{k=j+1}^{i-1} \tilde{S}_k \tilde{Q}_j,$$

(12)

Here $d_i$ is the thickness of the ith layer, $\tilde{T}_i$ is the scattering matrix in layer $i$, $\tilde{Q}_i = \exp(-it\tilde{Q}_i/v_F)$, $\tilde{S}_i = \tilde{S}(L,0) = \prod_{n=1}^{i-1} \tilde{S}_i$ is the propagating factor of electrons passing through a period length, and $\tilde{Q}_i = \tilde{T}_i^{-1}(1 - \tilde{S}_i)$. The FM/NM interlayer includes only several atomic planes and its thickness is much shorter than either of FM or NM layers, but the scattering in the thin interlayer is very strong. Therefore, in Eq. (12), the contributions of the interlayers to the sum over $i$ and $j$ can be approximately neglected, while those to the products over $k$ of $\tilde{S}_k$ need to be taken into account.

The measured conductivity is given by $\sigma(\theta) = \bar{\sigma}_{\alpha\beta}\rho(\theta)$, and the MR is defined as $[\rho(0) - \rho(\theta)]/\rho(0)$ with $\rho(\theta) = 1/\sigma(\theta)$ being the $\theta$-dependent resistivity. Let us first see the homogeneous limit where the periodic length $L$ is much shorter than the electron mean free paths. In this

FIG. 1. Angular dependence of CIP MR for a FM/NM superlattice with $d_F = 2d_N$ and $d_i \ll d_N$. (a) The spin-flip scattering takes place only in the FM layers with $\lambda_{ij} = 2d_N$ (solid line), $7d_N$ (dashed line), and $50d_N$ (dotted line). (b) The spin-flip scattering takes place only in the interlayers with $\lambda_{ij}(I) = 2d_I$ (solid line), $7d_I$ (dashed line), and $50d_I$ (dotted line).
case, both $\tilde{S}_i$ and $\tilde{S}_L$ approach $\tilde{1}$, so that the third term on the right-hand side of Eq. (12) dominates $\tilde{F}(\theta)$, yielding $\tilde{F}_{aa,bb}(\theta) = tL/v_F Z(\theta)$ with

$$Z(\theta) = \frac{\tilde{\xi}_1 - \tilde{\xi}^2 + \sin^2(\theta/2)(\tilde{\xi}_1 - \tilde{\xi})}{\tilde{\xi}_1 + \tilde{\xi} + 2 \tilde{\xi}'},$$

where $\tilde{\xi}_i = (\Sigma_i \tilde{\xi}_i d_i)/L$ and $\tilde{\xi}' = (\Sigma_i \tilde{\xi}_i d_i)/L$ are the spatial average of the $\tilde{\xi}_i(z)$ and $\tilde{\xi}'(z)$, respectively. Substituting $\tilde{F}_{aa,bb}(\theta)$ into Eq. (11) and performing the integral over $t$, we obtain $\sigma(\theta) = C_D v_F / Z(\theta)$, so that the resistivity variation, $\Delta \rho(\theta) = \rho(\theta) - \rho(0)$, is proportional to $\sin^2(\theta/2)$ and the magnitude of $\Delta \sigma(\theta)$ has a linear dependence on $\sin^2(\theta/2)$. Another limiting case is that the thickness of the FM layer is much larger than its mean free path $v_F \xi^{-1}(F)$ with $v_F$ the Fermi velocity. For those terms of $\tilde{F}_{aa,bb}$, the first term of Eq. (12) is independent of $\theta$ and so has no contribution to $\Delta \tilde{F}_{aa,bb}(\theta) = \tilde{F}_{aa,bb}(\theta) - \tilde{F}_{aa,bb}(0)$ and $\Delta \sigma(\theta) = \sigma(\theta) - \sigma(0)$. In Eq. (12), the contribution to the sums over $i$ and $j$ comes mainly from those terms of $i$ and $j$ being the two adjacent FM layers. Taking into account the fact that the propagating tensor $\tilde{S}_F$ in the FM layers is very small in this limiting case, after a tedious and lengthy calculation, we find $\Delta \tilde{F}_{aa,bb}(\theta)$ and so $\Delta \sigma(\theta)$ to be just proportional to $\sin^2(\theta/2)$. Thus, the magnitude of MR is approximately proportional to $\sin^2(\theta/2)$. From the above discussion we see that in both limiting cases, the present theory yields linear dependence of the CIP MR on $\sin^2(\theta/2)$. This conclusion is the same as that in the absence of the spin-flip scattering.\(^{16}\) In the intermediate region, however, the situation is somewhat different. Numerical calculation indicates that there is a deviation from the linear $\sin^2(\theta/2)$ dependence, as shown in Fig. 1. In our calculation, the scattering rate in layer $i$ is taken to be $\xi(i) = 1/\tau_F(i) = (3 - x)/2 \tau_{sf}(i)$, and $\xi'(i) = (1 - x)/2 \tau_{sf}(i)$, with a unified $x = 2/3.\(^{18}\)$ Figure 1(a) [Fig. 1(b)] corresponds to the case where the spin-flip scattering takes place only in the FM layers (interlayers). The thickness of the FM layer is fixed to be twice as long as that of the NM layer, i.e., $d_F = 2d_N$, and the thickness of the interlayer, $d_I$, is much smaller than $d_N$. The spin-dependent mean free paths $\lambda_{i,j}(F) = v_F \tau_{sf}(i)$ are taken as $\lambda_{i,j}(F) = d_F$ in the FM layers, $\lambda(N) = 10d_N$ in the NM layers, and $\lambda(i) = 0.5d_I$ and $\lambda(i) = 5d_I$ in the interlayers. We find that as the spin diffusion length $\lambda_{i,j}(F)$ in layer $i$ is much greater than the thickness of the same layer, there is an evident deviation of the MR from the linear dependence on $\sin^2(\theta/2)$, as shown by dotted lines in Fig. 1. As the spin-flip scattering is increased and the spin diffusion length is shortened, the magnitude of MR reduces and the deviation of the MR from the linear dependence decreases as well. To obtain a larger MR, one should increase the spin-dependent scattering asymmetry either within FM layers or at interfaces or both of them. When $\lambda_{i,j}$ becomes smaller, the effective scattering rates for up and down spins are drawn closer by the spin-flip scattering. As a result, the spin-dependent scattering asymmetry is reduced and so is the giant MR. The suppression of the giant MR due to the spin-flip scattering is a general conclusion suitable for arbitrary magnetization alignments, while it was known only in the limit of transition from antiparallel to parallel configuration.\(^{18,20}\)

In summary, we have derived an extended Boltzmann equation with a quantization-axis transformation invariant and applied it to study effects of spin-flip and non-spin-flip scattering on the magnetotransport in magnetic inhomogeneous systems with arbitrary magnetization orientations. It is found that the CIP MR varies approximately linearly with $\sin^2(\theta/2)$, and the spin-flip scattering suppresses the MR and reduces the deviation from the linear behavior.

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\(^{11}\)M. Patel et al., J. Appl. Phys. 75, 6528 (1994).