Tunneling magnetoresistance in ensembles of ferromagnetic granules with exchange interaction and random easy axes of magnetic anisotropy

Y M Beltukov\textsuperscript{1,\#}, V I Kozub\textsuperscript{1}, A V Shumilin\textsuperscript{1,\#} and N P Stepina\textsuperscript{2,\#}

\textsuperscript{1} Ioffe Institute, 194021 St. Petersburg, Russia
\textsuperscript{2} Rzhanov Institute of Semiconductor Physics, 630090 Novosibirsk, Russia

E-mail: avshumilin@mail.ioffe.ru

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Abstract
We study the tunneling magnetoresistance in the ensembles of ferromagnetic granules with random easy axes of magnetic anisotropy taking into account the exchange interaction between granules. It is shown that due to the exchange interaction magnetoresistance is effectively decoupled from magnetization, i.e. the strongest negative magnetoresistance can be observed at the field where magnetization is almost saturated. Under some conditions, the sign of magnetoresistance can be reversed and tunneling magnetoresistance can become positive at certain magnetic fields. Our theory agrees with measurements of magnetoresistance in ensembles of Fe granules in SiC\textsubscript{x}N\textsubscript{y} matrix.

Keywords: granular ferromagnets, tunneling magnetoresistance, hopping transport, magnetic nanoparticles

(Some figures may appear in colour only in the online journal)

1. Introduction

The nanostructured ferromagnetic materials and, in particular, ensembles of ferromagnetic nanoparticles (granules) attract significant attention due to the diversity of their magnetic and conductive properties. It leads to numerous interesting physical phenomena including superparamagnetism and spin-glass-like behavior [1] and possible applications in electronics, for example in magnetic memory and magneto-optics [2, 3].

The tunneling magnetoresistance (TMR) is the essential property of ensembles of ferromagnetic granules and relates their conductive and magnetic properties [4]. In hopping regime, the tunneling rates \( w_{ij} \) depend on the mutual orientation of the magnetic moments of granules \( i \) and \( j \)

\[
w_{ij} = w_{ij}^{(0)} (1 + P^2 \cos \theta_{ij}).
\]  

(1)

Here \( \theta_{ij} \) is the angle between the magnetization of the granules, \( P \) is the degree of electron spin polarization in the ferromagnetic granules. \( w_{ij}^{(0)} \) does not depend on orientations of magnetic moments. When the magnetic field leads to overall sample magnetization, the angles \( \theta_{ij} \) decrease leading to negative magnetoresistance (MR).

Equation (1) is significantly modified in the variable range hopping regime of conductivity (VRH). In this case the hopping occurs between the distant granules and includes co-tunneling through some number of Coulomb blockaded intermediate granules [5]. Each co-tunneling process between intermediate granules \( n, m \) adds a factor \( (1 + P^2 \cos \theta_{nm}) \) to the hopping probability [6–8]. Therefore, the hopping probability
in VRH regime can be estimated as

\[ w_{ij} = w_{ij}^{(0)} (1 + P^2 \cos \theta_{ij})^{N_{\text{int}}+1}, \]

where \( N_{\text{int}} \) is the average number of intermediate granules in the hopping process. TMR in granules is discussed in the review [9].

It is tempting to think that the magnetic field where TMR is observed is the field where the dependence of magnetization on magnetic field is the strongest. It can be shown that in an ensemble of independent identical granules the explicit relation exists between MR and magnetization [4, 10–12]. In this case \( \Delta R/R \propto \langle \cos \theta_{ij} \rangle = \langle \cos \theta_i \rangle^2 \propto M_{\text{tot}}^2 \). Here \( \theta_i \) is the angle between the magnetization of \( i \)th granule and the external magnetic field. The averaged value of \( \langle \cos \theta_i \rangle \) is proportional to the total sample magnetization \( M_{\text{tot}} \). \( \Delta R/R \) is the relative correction to the resistance in the magnetic field. This relation between the MR and magnetization was reported in some experimental studies [13–16]. When the distribution of granule sizes is taken into account some deviations from \( \Delta R/R \propto M_{\text{tot}}^2 \) law are possible [10, 11]. However, these deviations are not extremely strong and are most relevant at high magnetic field.

However, in a number of different granular ferromagnetic materials negative MR continue to grow even when magnetization is almost saturated. Similar behavior was observed in granular magnetic oxides [17–20], nanoclusters of ferromagnetic metals [21–23] and in some other materials [24–26] including Mn-doped semiconductors [27]. Usually, this MR is ascribed to existence of ‘anti-boundaries’, the spins aligned against the main magnetization direction on the edge of granules [17, 24, 28, 29]. Although in principle such spins can exist, their existence does not follow from any general law. It is doubtful that these reversed spins appear in all the materials where the discussed MR was measured.

Here we show that TMR itself can sometimes lead to MR that is most strong in the fields when magnetization seems to saturate. It occurs due to the interplay of exchange interaction and magnetic anisotropy of the granules. When the exchange interaction is taken into account, the relation \( \langle \cos \theta_{ij} \rangle = \langle \cos \theta_i \rangle^2 \) is broken. It can lead to quite sophisticated dependencies of TMR on the applied magnetic field including the reversed sign of MR in certain magnetic fields. The discussed behavior of MR is observed when the anisotropy energy is large compared to the exchange energy. In this case the exchange energy significantly suppresses the TMR at the fields \( H \) when the dependence \( M_{\text{tot}}(H) \) is the strongest.

Note, that both the exchange interaction and anisotropy naturally appear in an array of ferromagnetic granules and exist in almost any material. It is important for our theory that the easy axes of anisotropy of different granules are random. It may occur due to non-spherical shape of granules [30] or due to different crystallographic orientations in different granules.

We compare our theoretical results with recent experiments obtained by us in the SiC-N_{x}:Fe granular system. In this material we observed the behavior of MR similar to that for several other mentioned granulated films: negative MR linearly increases with magnetic field and does not saturate up to the high field while magnetization tends to saturate in small magnetic fields. The narrow peak of MR is observed with their width roughly match the range of strong magnetization change. The MR for samples with conductance being in hopping regime increase with decreasing the temperature demonstrating the increase of the hopping length at low temperatures.

The article is organized as follows. In section 2 we discuss our model in details. In section 3 we show the results of numeric simulation of our model. In section 4 we provide a simplified mean-field description of the model and compare it with numerical results. In section 5 we compare our theory with our recent experimental results on MR of Fe nanoclusters in SiCN matrix.

2. Model of ferromagnetic granular material

In this section we discuss the model that is applied to describe transport and magnetization of an array of ferromagnetic granules. The model includes all the effects crucial to our theory, i.e. the magnetic nature of the granules, anisotropy with random easy axis and exchange interaction. However, we tried to keep the model as simple as possible and did not include into the model several less significant features such as the distribution of granule sizes.

We describe the array of granules arranged on a square lattice (figure 1). Each granule has random easy axis of magnetic anisotropy \( \mathbf{a} \). The energy \( K \) associated with the anisotropy is considered to be the same for all the granules. The magnetic moments \( \mathbf{M}_i = M_\mathbf{s}_i \) of all the granules have the same absolute value but different directions. Neighbor granules have ferromagnetic exchange interaction. The total energy of the system is as follows:

\[
E = -K \sum_i (\mathbf{s}_i \cdot \mathbf{a}_i)^2 - M \sum_i \mathbf{s}_i \cdot \mathbf{H} - J \sum_{ij} \mathbf{s}_i \cdot \mathbf{s}_j.
\]

Here \( \mathbf{s}_i \) is the unit vector in the direction of magnetization of the granule \( i \). \( J \) is the exchange energy. This model of granular ferromagnetic system was discussed in [31–33], with dipole–dipole interaction sometimes added to the energy \( E \). In [34–37] a similar model without ferromagnetic exchange coupling but with dipole–dipole interaction was treated numerically.

The magnetization of the system is equal to \( \mathbf{M}_{\text{tot}} = M \sum_i \mathbf{s}_i \). In a macroscopic system it is always directed along the magnetic field and is equal to \( N_g M_\mathbf{g} \langle \cos \theta_\mathbf{g} \rangle \) where \( N_g \) is the number of granules and \( \theta_\mathbf{g} \) is the angle between magnetic field and \( \mathbf{s}_i \). Transport in a hopping system can be described by Miller–Abrahams resistor network [38, 39]. The conductivities of its resistors are proportional to the hopping rates \( w_{ij} \). According to equation (2), the effect of the applied magnetic field on these conductivities is controlled by \( \cos \theta_{ij} \). Usually in a hopping system the distribution of the conductivities is exponentially broad and only rare critical resistors of the percolation theory are relevant to the macroscopic resistivity of the sample. Therefore, to calculate the MR one should average \( \cos \theta_{ij} \) over all the critical resistors \( ij \). However, we presume
that the distribution of $\cos \theta_{ij}$ is almost the same for critical and non-critical resistors. Therefore, we average $\cos \theta_{ij}$ over all the neighbor pairs of granules to calculate TMR.

$$\frac{\Delta R}{R(0)} = -\mathcal{P} \left( \langle \cos \theta_{ij} \rangle (H) - \langle \cos \theta_{ij} \rangle (0) \right).$$  \hspace{1cm} (4)$$

Here $\Delta R = R(H) - R(0)$ is the correction to the system resistivity due to the magnetic field. It is considered to be small compared to $R(0)$. $\mathcal{P} = P^2(N_{\text{int}} + 1)$ is the constant that controls the magnitude of the effect of granule magnetizations on conductivities. It is related to the polarization of material $P$ and the average number $N_{\text{int}}$ of intermediate granules in the hop. In VRH regime $N_{\text{int}}$ grows with decreasing temperature.

3. Numeric simulation

There are different methods to simulate a thermal equilibrium of magnetic granules. The direct method is to simulate the granular magnetic system using the stochastic Landau–Lifshitz equation [40, 41]. However, it requires a simulation with a small timestep. Another issue of this method is exponentially slow relaxation between two minima separated by the barrier caused by anisotropy.

In this paper we study a thermal equilibrium of a system rather than the magnetization dynamics. In this case we can use the Metropolis algorithm [42] to describe a system at a given temperature $T$. We simulate a periodic sample with $N = 30 \times 30$ granules. For each step we make a random perturbation $\xi_i^{(n)}$ of each magnetization direction using the following Markov scheme:

$$s_i^{(n+1)} = s_i^{(n)} + \frac{\xi_i^{(n)}}{|\xi_i^{(n)}|}.$$  \hspace{1cm} (5)$$

This equation ensures that $s_i^{(n+1)}$ is unit vector. Following the Metropolis algorithm, the new state $s_i^{(n+1)}$ is accepted with probability

$$p = \min \left[ \exp \left( \frac{E^{(n+1)} - E^{(n)}}{T} \right), 1 \right].$$ \hspace{1cm} (6)$$

Otherwise, the state remains unchanged: $s_i^{(n+1)} = s_i^{(n)}$. In equation (6) $E^{(n)}$ is the energy of the system on step $n$ and $E^{(n+1)}$ is the energy of the perturbed state.

The random perturbation $\xi_i^{(n)}$ was chosen as an uncorrelated random Gaussian vector

$$\langle \xi_{ij}^{(n)} \xi_{j\beta}^{(m)} \rangle = \sigma^2 \delta_{nn} \delta_{ij} \delta_{\alpha\beta}.$$ \hspace{1cm} (7)$$

The variance $\sigma^2$ was chosen to meet the balance between a large step and a reasonable acceptance rate: $\sigma^2 = 0.16 T / N$.

For a small temperature $T$, there is a barrier for magnetization of each granule due to anisotropy $K$. To increase the probability to hop over this barrier we make the following trick. With a small probability $p_u$ for each granule $i$ at each step $n$ we take the magnetization direction $s_i^{(n+1)}$ from uniform distribution on unit sphere instead of the perturbation described by equation (5). In this case the probability to get over the barrier is proportional to $p_u T$ instead of a small activation exponent $-T/K$. Numerical simulation shows that $p_u = 1/N$ is close to an optimal value.

To obtain a system at different temperatures we start from some temperature $T = T_0 \sim K$, which is comparable with a typical barrier energy of the system. Then we gradually decrease the temperature of the system using the robust logarithmic annealing protocol: $T^{(n)} = T_0 / \ln(n + b)$ where $n$ is the step number [43]. For system under consideration, the parameter $b = 10^{-3}$ was small enough to obtain a thermal equilibrium for $T \gg 0.01 K$.

The results of the simulation are presented in figure 2 for different values of the temperature $T$ in the case of small exchange energy $J = 0.025 K$. The magnetization grows very fast at $MH \sim J$. However, its growth slows down when $MH \geq 0.2 K$. One can say that the magnetization reaches quasi-saturation. However, MR has a significant increase up to $MH \sim 1.5 K$. Thus, there is a strong MR with a quasi-saturated magnetization, which will be explained using the mean-field analysis. Also in figure 2 we compare the results of numeric simulation with the mean-field analysis that is discussed in the next section.

4. Mean-field analysis

In this section we describe the mean-field treatment of the model (3). The mean-field theory contains some additional simplification compared to the direct numeric simulation described in section 3. However, it can be more easily solved numerically and allows analytical solution in some limiting cases. It is compared with numeric simulation in figure 2.

Let us consider the granule $i$ connected to four other granules (figure 3). The exchange energy related to the granule $i$ is equal to $-J s_i \cdot \sum_j s_j$. Here index $j$ enumerates granules connected to granule $i$ with the exchange interaction. Within the framework of the mean-field approximation we substitute $\sum_j s_j = 4 \mathbf{S}$ where $\mathbf{S}$ is the averaged magnetization of the granules. It is directed along the axis of the magnetic field and is equal to $\langle \cos \theta \rangle$. It should be found self-consistently. The exchange interaction is reduced to the increased
Here the integration is taken overall possible unit vectors

The comparison between numeric simulation (solid lines) and mean-field theory (dashed lines) for

Here the integration is taken overall possible unit vectors $\mathbf{s}$. The energy $E_i(s_i)$ has two minima near these directions, that control the integrals in equation (8)

Here $\mathbf{e}_H$ is the unit vector in the direction of the magnetic field. The first term in rhs of equation (9) describes the thermodynamic distribution of magnetization between minima of $E_i(s_i)$. The second term shows that the minima are

shifted by the effective magnetic field. This term is proportional to $MH_{\text{MF}}/K$. The terms proportional to $(MH_{\text{MF}}/K)^3$ are neglected in equation (9).

The mean-field magnetization can be obtained by averaging of single granule magnetization $\mathbf{M}$ over the easy axis directions of all granules. This averaging yields

Here $\text{Li}_i(z)$ is the polylogarithm. Note that $H_{\text{MF}}$ depends on $\mathbf{M}$. Therefore, equation (10) is the equation that should be solved to find $\mathbf{M}$. It can be done numerically.

In figure 2(a) we compare $\mathbf{M}$ calculated in the mean-field theory with the results obtained by the numeric simulation. The results of the simulation are shown with solid lines and the results of mean-field theory with dashed lines. The colors of the lines correspond to different temperatures. To improve the agreement between mean-field approximation and Monte-Carlo simulation we multiply the magnetization $\mathbf{M}$ with the mean-field theory, by a phenomenological constant $\alpha$. For small ratios $T/K$ and $J/K$ this constant is close to 1. The value $\alpha = 0.93$ is used in figure 2.

The mean-field approximation (10) describes the magnetization of the system relatively well. However, in the mean-field theory the magnetization is discontinued at $H = 0$. Its behavior in the results of simulation is more smooth.

To describe the relative directions of magnetizations of neighbor granules, it is important to take into account, that even without averaged magnetization $\mathbf{M}$, the exchange interaction tends to align nearby granules. Therefore, we modify the mean-field scheme as shown in figure 4. We explicitly consider the directions of easy axes and magnetizations of the two granules $i$ and $j$ and treat their 6 neighbors with mean-field approximation.

The energy in this model is equal to

$$E_{ij}(s_i, s_j) = -K(a_i s_i)^2 - K(a_j s_j)^2 - M H_{\text{MF}}(s_i + s_j) - J s_i s_j.$$ (11)
The mean-field $H_{MF}$ in this scheme is equal to $H_{MF} = H + 3J\pi$, because each of the granules $i$ and $j$ is connected with three granules treated with mean-field approximation. The value $\overline{\pi}$ is taken from the solution of equation (10).

In the general case the thermodynamic average of $\cos \theta_{ij}$ is given as follows

$$\langle \cos \theta_{ij} \rangle = \frac{\int (s_s') \exp \left( -E_{ij}(s_s') \right) ds_s' ds_s''}{\int \exp \left( -E_{ij}(s_s') \right) ds_s' ds_s''}. \quad (12)$$

With the approximation that is applied in equation (9) the integration can be reduced to the summation over four minima of the energy $E_{ij}$. Here we give the expression for $\cos \theta_{ij}$ in the first minimum.

$$\cos \theta_{ij}^{(1)} = a_i \cdot a_j + \frac{J}{K} (1 - (a_i \cdot a_j)^2) + \frac{MH_{MF}}{2K} (1 - a_i \cdot a_j) (a_i + a_j). \quad (13)$$

The energy of this minimum is equal to

$$E_{ij}^{(1)} = -(a_i \cdot a_j)MH_{MF} - J(a_i \cdot a_j) \left( -\frac{1}{4K} (MH_{MF} + Ja_j - a_i (a_i \cdot (MH_{MF} + Ja_j)))^2 \right) \left( -\frac{1}{4K} (MH_{MF} + Ja_j - a_i (a_j \cdot (MH_{MF} + Ja_j)))^2 \right). \quad (14)$$

The corresponding expressions for other minima differ from (13) and (14) by the inversion of the sign of either one of $a_i$ and $a_j$ or both their signs.

The expression for the averaged $\cos \theta_{ij}$ that controls the MR is as follows

$$\langle \cos \theta_{ij} \rangle = \left\langle \frac{\sum_n \cos \theta_{ij}^{(n)} e^{-E_{ij}^{(n)}/T}}{\sum_n e^{-E_{ij}^{(n)}/T}} \right\rangle_a. \quad (15)$$

Here index $n$ enumerate local minima. The averaging $\langle \rangle_a$ is made over all possible directions of the easy axes $a_i$ and $a_j$.

In the general case expressions (13)-(15) can be used to calculate $\langle \cos \theta_{ij} \rangle$ numerically. In figure 2(b) we compare $\langle \cos \theta_{ij} \rangle$ calculated with mean-field theory with numeric simulations described in section 3. Similarly to figure 2(a), the results of the mean-field theory are multiplied by the constant $\alpha = 0.93$ in figure 2(b). With this modification, the mean-field theory is in reasonable agreement with numerical simulations. It means that mean-field approach can give a qualitative understanding of the behavior of the model (3).

In certain limiting cases, it is possible to obtain an analytical expression for the mean-field approximation. When the magnetic energy $MH$ is larger than the exchange energy $J$ and much larger than temperature, it is possible to take into account only the first minimum ($n = 1$) in equation (15). In this case $\langle \cos \theta_{ij} \rangle$ is described by the averaging of equation (13) over the directions of easy axes. This averaging leads to the following result

$$\langle \cos \theta_{ij} \rangle = \frac{1}{4} + \frac{1}{3K} MH_{MF} + \frac{2J}{3K}. \quad (16)$$

Within the same assumptions, equation (10) for the averaged magnetization can be reduced to $\overline{\pi} = 1/2 + MH/3K + 2J/3K$. It leads to

$$\langle \cos \theta_{ij} \rangle = \frac{1}{4} + \frac{MH}{3K} + \frac{4J}{3K}. \quad (17)$$

Only the terms linear over $1/K$ are kept in equation (17).

We compare expression (17) with the numeric solution of the mean-field model in figure 5 for $J = 0.01K$. At sufficiently high field equation (17) agrees with the numeric solution. It is interesting that at low field the sign of MR can be different and depends on temperature. To analyze these small fields it is useful to neglect $\alpha MH/K$ terms and keep only zeroth-order terms over $MH/K$.

When the terms $\alpha MH/K$ are neglected in equations (13)-(15), $\langle \cos \theta_{ij} \rangle$ is controlled by the relations $MH/J$ and $MH/T$. If we also neglect $J/K$ terms, it allows to reduce equation (15) to

$$\langle \cos \theta_{ij} \rangle = \left\langle \frac{\cosh \left( \frac{MH_{MF}(a_i \cdot a_j)}{T} \right)}{\cosh \left( \frac{MH_{MF}(a_i \cdot a_j)}{T} \right)} \exp \left( \frac{J a_i a_j}{T} \right) - \cosh \left( \frac{MH_{MF}(a_i \cdot a_j)}{T} \right) \exp \left( -\frac{J a_i a_j}{T} \right) \right\rangle_a. \quad (18)$$
This equation corresponds to the situation when the magnetization of a granule is always directed along the easy axis. However, one of the two directions along this axis is selected with respect to the temperature, magnetic field and exchange interaction. Equation (18) contains three energy scales: $MH_{MF}$, $J$ and $T$. When one of these energies is much larger than the other two, it is possible to find $\langle \cos \theta_{ij} \rangle$ explicitly. At large temperature $T \gg J, MH_{MF}$ the directions of magnetizations are random and $\langle \cos \theta_{ij} \rangle = 0$. At sufficiently large magnetic field $MH_{MF} \gg J, T$ magnetization projection on the magnetic field is always positive, in this case $\langle \cos \theta_{ij} \rangle = 1/4$. When the exchange energy is the largest $J \gg MH_{MF}, T$, the magnetization of neighbor granules tries to be directed along each other. Sometimes it leads to the negative projection of the magnetization on the magnetic field. In this case $\langle \cos \theta_{ij} \rangle = 1/2$.

To discuss the low-field limit of $\langle \cos \theta_{ij} \rangle$ at the arbitrary relation $J/T$ we consider $H_{MF}' = 0$. Although this value does not appear in the solution of equation (10) at sufficiently low temperatures (due to the discontinues magnetization in zero field), it gives qualitatively correct results. In this case equation (15) for $\langle \cos \theta_{ij} \rangle$ can be reduced to the explicit expression

$$\langle \cos \theta_{ij} \rangle = \frac{1}{2} + \frac{2J}{3K} - \frac{\pi^2 T^2}{24 J^2} + \frac{T}{J} \log \left(1 + e^{-2J/T}\right)$$

$$+ \frac{T^2}{2K} L_2 \left(-e^{-2J/T}\right).$$

The term $J/K$ is also included into equation (19). In figure 6 we compare equation (19) with numeric solution of the mean-field model. A quantitative agreement is achieved.

To understand the sign of the low-field MR one should compare $\langle \cos \theta_{ij} \rangle$ described with equation (19) with the value 1/4 that corresponds to the limit $K \gg MH \gg J, T$. At sufficiently high temperature, zero-field value of $\langle \cos \theta_{ij} \rangle$ is small and tends to zero when $K \gg T \gg J$. External magnetic field increases it to the value 1/4 and leads to negative MR, that is usual for TMR. However, at low temperatures, zero-field value for $\langle \cos \theta_{ij} \rangle$ is larger than 1/4. It is equal to 1/2 in the limit $K \gg J \gg T$. External magnetic field decreases it leading to the positive MR at low fields. Therefore, in certain cases TMR changes its sign and leads to positive MR. Negative MR is re-established at higher fields as shown in figure 5.

5. Comparison with experiment

In this section we compare the theoretical results with our recent experiments in SiC$_x$N$_{1-x}$-Fe granular ferrimagnetic films. The SiC$_x$N$_{1-x}$-Fe films were synthesized using the CVD technique on high resistance Si(001) substrates by the thermal decomposition of the following gaseous mixture: 1,1,1,3,3,3-hexamethyldisilazane, ferrocene and helium. The deposition procedure was carried out in the horizontal flow-type reactor at 1000°C and described in details elsewhere [44]. Using STEM and HRTEM it was shown, that in the sample under study, Fe is concentrated in large granules (∼ 30 nm) homogeneously distributed in the matrix and in small nanoclusters (5–10 nm) between them. Large granules consist of several α-Fe clusters with different crystallographic orientations of [100] axes. Additional information on the sample properties including magnetization curves and ESR spectra can be found in [45]. Here we provide the MR data for one typical sample (figure 7(a)) and compare our experimental results with theoretical data.

In figure 7(b) the temperature dependence of conductivity for the sample is shown as $G(T^{-1/2})$ plot. The conductivity drops ∼ 40 times when the temperature decreases from 140 K to 4 K. It shows that the sample is dielectric and its low-temperature conductivity is due to hopping. However, the range of the conductivity change is not enough to reliably identify the regime of hopping (neighbor hopping or VRH). We compare the measured temperature dependence of conductivity with Efros–Shklovskii law that is expected for VRH in metallic granules in the low temperature limit [solid red line in figure 7(b)]. The agreement can be achieved over one order of magnitude of the conductivity change. If the dependence is compared with Arrhenius law that is expected for neighbor hopping the agreement is significantly worse. Note, however, that the regime of hopping (nearest neighbor of VRH) is not
essential for the following analysis and leads only to the different definition of $P$: $P = P^2$ in the nearest neighbor hopping regime and $P = P^3(N_{\text{int}} + 1)$ in VRH regime.

The MR curve for this sample is shown in figure 7(a). The shape of MR is quite similar to the one obtained in different granular magnetic films (see the introduction for details). The curve includes two regions: low-field ($H \lesssim 0.25$ T) and high-field ($H \sim 1$ T) regions. The range of magnetic fields for low-field region approximately corresponds to the field range of the fast change of magnetization before it reaches the quasi-saturation, see figure 7(c). In high magnetic fields MR is linear and there is no sign of saturation of the negative MR up to the fields $\gtrsim 2$ T. This behavior of MR is similar to that predicted by our theory. We compare the obtained experimental data with the mean-field model in figure 8.

In figure 8(a) we show that the quantitative agreement between our theory (with certain parameters) and experimental data can be reached for MR. Blue points show the measured MR while red line corresponds to the mean-field calculation. The parameters taken were as follows: $K = 2.3 M \cdot 1 T$, $J = 0.19 M \cdot 1 T$, $T = 0.13 M \cdot 1 T$, $P = 0.033$. Here $M \cdot 1 T$ is the magnetic energy of the granule in the magnetic field 1 T. Therefore, the exchange energy corresponds to the field 0.19 T that is similar to the field where magnetization reaches quasi-saturation. Anisotropy energy corresponds to the field $\sim 2.3$ T. It means that the negative MR can persist up to the fields equal to several Tesla.

In figure 8(b) we compare magnetization calculated with the mean-field theory with the same parameters and magnetization measured in the SiC$_3$N$_4$:Fe film. The mean-field theory cannot capture the hysteresis of magnetization and reduces the low-field behavior of magnetization to the discontinuity at $H = 0$. Also, it overestimates the slow increase of magnetization with magnetic field after the quasi-saturation is reached. However, this increase can be observed in the experimental data. Therefore, the quantitative agreement exists between theory and experiment. We believe that qualitative agreement for both the magnetization and MR can be reached by taking into account the distributions of granule sizes, anisotropy energies $K$ and exchange energies $J$. However, the discussion of these distributions is out of the scope of this article.

6. Discussion

We have shown that the exchange interaction between ferromagnetic granules and the magnetic anisotropy with random axis leads to quite sophisticated dependence on TMR on the applied magnetic field. TMR is effectively decoupled from the magnetization and can even change its sign becoming positive at low field. The decoupling of MR from magnetization was observed in a number of different granular magnetic materials [17–27]. Most often it was ascribed to the so-called ‘anti-boundaries’. However, the existence of these ‘anti-boundaries’ was not proved independently in most of the discussed materials. We believe that at least in some materials the discussed shape of MR can be related not to ‘anti-boundaries’ but to the interplay of anisotropy and exchange interaction.

We obtain our results with two methods: with Monte-Carlo numeric simulations and with the mean-field theory. The mean-field theory contains more simplifications that the simulation, however, it was shown that the results of mean-field model are in semi-quantitative agreement with simulation. In some limiting cases the equations of the mean-field model can be solved analytically. In other cases they should be solved numerically, however, their numerical solution is much easier than the Monte-Carlo simulation. Sometimes the mean-field theory allows the analysis of MR of the granular systems with parameters that make the converging of the Monte-Carlo calculation problematic.

The shape of MR depends on the relation between the anisotropy energy, exchange energy and temperature. Therefore, these parameters of the sample of granular ferromagnetic material can be estimated with the measurements of MR. Such an estimate is made for the SiC$_3$N$_4$:Fe sample in this work. However, some care should be taken when comparing the experimental results with the present theory. The model described in this study deals with almost identical granules.
with the same magnetization, anisotropy energy and exchange interaction between neighbors. Only the easy axes of magnetization are different. However, in real systems there is always some distribution of sizes, exchange energies, etc. For example, our SEM and HREM data [45] show the difference in granule sizes and shape and their random distribution in the film. This distribution can modify the quantitative results of TMR in anisotropic granules with exchange interaction. Also, to calculate the MR, $⟨\cos \theta_{ij}⟩$ should be averaged not over all the pairs but only over the pairs that are critical for conductivity. Our approach is applicable when there is no correlation between $\cos \theta_{ij}$ and conductivity of the effective resistor connecting granules $i$ and $j$. There is a reason for such a correlation to exist. High conductivity of the resistor $ij$ can be related to large overlap integral between granules $i$ and $j$ that will lead to strong exchange interaction between the granules. Therefore, $\cos \theta_{ij}$ for the granules that are connected with a resistor with high conductivity tends to be larger than the average $⟨\cos \theta_{ij}⟩$. All these details are not included into the present theory. Nevertheless, even our simplified model shows the possibility to describe decoupling of magnetization and MR in granular arrays without any MR mechanisms beside TMR.

In conclusion, we have shown that TMR in a system of anisotropic ferromagnetic granules with exchange interaction and random easy axes can be decoupled from magnetization. In some cases TMR changes its sign at low magnetic field and becomes positive. We obtained our results with two methods: Monte Carlo simulations and mean-field theory. Our theoretical results agree with our measurements of MR in Fe nanocrystals in SiCN matrix.

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ORCID iDs

Y M Beltukov https://orcid.org/0000-0002-3648-0312
A V Shumilin https://orcid.org/0000-0003-4841-1805
N P Stepina https://orcid.org/0000-0001-6111-5774

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