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Tunable electron transport with intergranular separation in FePt-C nanogranular films

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Abstract

We report electron transport mechanism in FePt-C granular films as a function of temperature by varying intergranular separation. FePt-C nanogranular films were prepared by sputtering on MgO substrates. From magnetic measurement of the sample, a coercivity of about 3 T was found in the perpendicular direction. Above 25 K, the electrical resistivity of the films were found to obey Mott variable range hopping, Efros-Shklovskii variable range hopping and extended critical regime depending on the intergranular separation. However, at lower temperatures it deviates from the above behaviour showing an increase in conductance. Reduced activation energy calculated from resistivity data of these films shows metal-insulator transition. The metallic nature observed at low temperature was attributed to the intergranular ferromagnetic type ordering between granules that enhances the transport of electrons. Intergranular separation, thus, can be used as a tool to engineer the electron transport mechanism to different hopping regimes or extended critical regime in these films.

1. Introduction

$L_1_0$ ordered FePt granular films are found to have huge magnetocrystalline anisotropy $\approx 7 \times 10^6$ Jm$^{-3}$ suitable for ultra-high density magnetic storage devices. Critical grain size of FePt granular films for super-paramagnetism at room temperature is as low as 3 nm and $L_1_0$ ordering of granules was observed for a granular size of above 7 nm [2]. A large number of studies are available in literature on these systems, observing its potential in magnetic recording media [3–6], varying the matrix material [7, 8], annealing temperature [9] and substrates to avoid the diffusion of the matrix material and to raise the columnar growth of FePt. Recently, Electric field induced spin control in the perpendicular magnetic anisotropy films [10–14] has become an intriguing technique in spintronic devices, as it consumes ultra-low power and avoids joule heating occurring in current induced spin control. However, electron transport in these systems has not been studied enough.

Electron transport in granular films has attracted large attention [15] due to its tunability in granule size, dimensionality and separation between them. Percentage of the matrix material in the granular structure decides the electron transport mechanism to be dielectric or metallic [16]. In more amounts of insulator segregants, the metal grains are well separated with hopping mechanism in the dielectric regime and in interconnected metal grains, percolated conductance is observed which is metallic. In the insulating regime, transport mechanism at low temperature (temperatures at which thermal energy is less than the average difference in site energies) is described as variable range hopping (VRH). Here, the electrons find the site having energy close to the initial site which is not necessarily nearest neighbour site [17]. VRH has been debated over years with two prevalent mechanisms in action; Mott variable range hopping (MVRH) considering constant density of states around Fermi level and Efros-Shklovskii variable range hopping (E-S VRH) considering electron-electron interaction, leading to a reduction in density of states at Fermi level, known as coulomb gap [18]. General expression for hopping mechanism in resistivity is,
Table 1. Details of the FePt granular film samples are shown. Films deposited by co-sputtering are of 8 nm thick. The last two samples are about 5 nm of Carbon capped over FePt films of 8 nm and 20 nm.

| Sample name  | Vol % of C | Deposition temperature(K) | Method of deposition | Thickness (nm) |
|--------------|------------|---------------------------|---------------------|---------------|
| FePtC20850   | 20         | 850                       | co-sputtering       | 8             |
| FePtC20750   | 20         | 750                       | co-sputtering       | 8             |
| FePtC15750   | 15         | 750                       | co-sputtering       | 8             |
| FePtC12600   | 12.5       | 600                       | co-sputtering       | 8             |
| FePtCap600   | —          | 600                       | C as capping        | FePt-8,C-5    |
| FePtCap600b* | —          | 600                       | C as capping        | FePt-20,C-5   |

\[
\rho(T) = \rho_0 \exp \left[ \left( \frac{T_0}{T} \right)^x \right]
\]  

where \( \rho_0 \) and \( T_0 \) are constants and values of \( x \) are, 1/4 and 1/3 for MVRH in 3-dimensional and 2-dimensional materials respectively and 1/2 for E-S VRH irrespective of dimensions.

Though magnetic properties of FePt granular films have been studied well, its electrical properties have not been given enough attention. Resistance and magnetoresistance in FePt-Ag multilayer films have been reported [19], in which a dip in resistance was observed at very low temperatures (< 11 K). Carbon is an excellent segregant for FePt granular films, as it retains higher coercivity and smaller grain size distribution of FePt compared to other insulators such as B, SiO\(_2\), TiO\(_2\), etc., and also enhance \( L_{106} \) ordering [8]. Hence, in this paper, the effect of intergranular separation on resistivity of FePt-C granular films is presented.

2. Experiment

FePt-C granular films were prepared on MgO substrates by co-sputtering FePt and Carbon. Another set of samples were deposited by sputtering FePt first and then Carbon. These samples were deposited at substrate temperatures of 600 °C, 750 °C and 850 °C. The samples of three different granular distributions (12.5, 15 and 20 Volume % of Carbon) were used for this study. Sample names, deposition methods and parameters are listed in table 1.

The microstructures of these films were investigated with Transmission Electron Microscope (TEM) using an FEI, TitanG2 80-200. M-H curves were obtained using Quantum Design SQUID VSM. Electrical resistivity measurements were done using conventional two probe method in cryogenic probe station with Keithley 2635 source meter and Lakeshore temperature controller in a temperature range from 6 to 290 K. I-V measurements were collected after settling the sample stage to the set temperature with standard deviation <0.1 K. Resistance was deduced at each temperature from the slope of the I-V measurement.

3. Results and discussions

FePt granules in the FePt-C film with 20 volume % of Carbon deposited at 850 °C substrate temperature as shown in figure 1(a), are well separated with 1-3 nanometers (nm) of distance between them, whereas, the film having 20 volume % of Carbon deposited at 750 °C shows granules closer to each other while some of them are connected to each other as shown in figure 1(b). The film deposited with 15 volume % of Carbon deposited at 750 °C shows the granules mostly connected to each other as shown in figure 1(c). And as shown in figure 1(d), the film having 12.5 volume % of carbon deposited at 600 °C shows well connected grains. Hence films with different average intergranular separation were produced. From figures 1(a) to (d) it is observed that the average distance between the granules decreases. The FePt films of thickness 8 nm and 20 nm capped with 5 nm Carbon at 600 °C are shown in figures 1(e) and (f). The thick film consists of grains of the order of 100 nm and nanograins in between those big grains which can be observed with reference to the given scale bar. The large grains of the last two samples result from the deposition of FePt single layer followed by C capping, unlike the co-sputtering of C, which acts as a grain refinement material during deposition.

It has been reported that films with granular size less than 4 nm shows a complete disorder in \( L_{106} \) structure and for the size greater than 7 nm a complete order [2]. Since our films are having granular size above 6nm as observed from the TEM micrographs, \( L_{106} \) ordering should be present in our samples as reported [20].

Perpendicular magnetic anisotropy is the direct consequence of \( L_{106} \) ordering. Magnetic measurement (M-H curve) at room temperature confirms the perpendicular magnetic anisotropy with coercivity of about 3T as shown in figure 2. Slope in the M-H curve of perpendicular to the plane measurement and hysteresis in the in-plane measurement indicates that a perpendicular anisotropic film has been obtained with small in-plane anisotropy.
Figure 3 shows the resistivity of the FePt-C films as a function of temperature. Throughout the temperature regimes, negative temperature coefficient of resistance was observed. The data above 25 K were fitted well to general expression for hopping mechanism (equation (1)), as generally observed in granular films. Gradient search method was adopted to fit the data. Table 2 represents the fitting parameters. The exponent values (x) are 0.576 and 0.313 for the films of 20 volume % of Carbon deposited at 850 °C (Figure 3(a)) and 750 °C (Figure 3(b)) respectively. The 20 nm thick sample (figure 3(f)) gives the exponent value of 0.221. Other samples don’t fit to the general hopping expression. Hence, the granules which were well distributed and separated with a bigger gap shows ES-VRH mechanism. The exponent obtained is in good agreement with the value, 0.55 deduced by Mobius and Ritcher [21] by means of computer, in contrast to the analytical result (0.5) of Efros and Shklovskii. Whereas, the granules which are closer show 2d-MVRH and 3d-MVRH mechanisms for 8 nm and
20 nm thick samples respectively. Noticeably, a change in thickness of the film from 8 nm to 20 nm modifies electron conduction from 2d to 3d. This can be inferred from the exponent values of 2d and 3d MVRH while fitting to VRH expression.

The cross over between MVRH and ES-VRH has been reported in granular films as a function of temperature [22, 23]. In a similar note, controlling the intergranular distance electron transport can be tuned to different regimes. Correlating the microstructure and the resistivity graph, we show that the gap between the granules play the role of modifying the electron transport to different hopping mechanisms in FePt-C granular films.

At lower temperatures (<25 K), resistivity of the samples deviates to a lower value compared to the extrapolated data of hopping mechanism fitted at higher temperatures. Paolo Allia et al. [19, 24] and Paolo Tiberto et al. [25] have shown that FePt-Ag multilayered granular films have a drop in metallic resistivity behavior below 11 K which is similar to our observations. This suggests that the drop in the resistivity at low temperature arises from the electron transport across FePt granules irrespective of the matrix material.

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**Table 2.** Relevant fitting parameters of hopping transport at temperatures of the films between 25 and 290 K.

| Sample          | $T_0$(K) | $x$   | $R$ - square |
|-----------------|----------|-------|--------------|
| FePtC20850      | 152.48   | 0.576 | 0.9999       |
| FePtC20750      | 597.44   | 0.313 | 0.9998       |
| FePtCcap600b    | 2.83E+5  | 0.221 | 0.9998       |

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*Figure 3.* Resistivity of FePt films of FePtC20850, FePtC20750, FePtC15750, FePtC12600, FePtCcap600 and FePtCcap600b displayed in log-log scale with fit to general hopping expression above 25 K. The dashed line is the extrapolation of fit to temperatures below 25 K.
According to Anderson criteria, around the critical regime of metal-insulator transition of a disordered system, resistivity follows a power law behaviour \[ \rho \propto T^{-\beta} \], where \( \beta \) is independent of temperature. In these systems reduced activation energy \( (W) \) is used as a tool to describe the type of conduction mechanism \[27, 28\] and is defined as

\[
W(T) = -\frac{\ln(\rho)}{\ln(T)}
\]  

(2)

Subsequently, deriving the \( W(T) \) of power-law expression yields \( W(T) = \beta \) and metal-insulator transition is nothing but a sign conversion in the slope of \( W \). If the slope of \( W \) is negative, the conductance is insulating and if the slope is positive it is metallic. Figures 4(a) to (f) show \( W \) versus T for FePt films of FePtC20850, FePtC20750, FePtC15750, FePtC12600, FePtCcap600, FePtCcap600b respectively. In figures 4(a), (b) and (f), there is a slope change from positive to negative at 21 K and in figures 4(c) and (e) at 32 K, indicating metal-insulator transition according to the above described criteria. It shows that the hopping transport suppresses the metallicity and reduces the critical region to a lower temperature. A linear fit was shown with red lines to the linear portion of the \( W \) versus T graph in figure 4. The fit gives the slopes, \(-0.56, -0.35, 0.09, -0.13, -0.28\), respectively from figures 4(a), (b), (c), (e) and (f). There is no shift in the slope from positive to negative value in (d). The negative of the slopes of figures (a), (b) and (f) are found comparable to the exponents of the hopping mechanisms seen from the resistivity fit. The slope above critical regime is found deviating depending on the intergranular separation. Therefore, assuming the straight correlation with the microstructure, we state that electron transport mechanism in these films can be tuned by varying the intergranular separation at temperatures above critical region.

From our analysis in figure 3, we observed an enhanced conductance at lower temperatures with respect to the fit. This enhancement at lower temperatures can be attributed to the spin-dependent electron transport.
through granular channel having magnetic intergranular interaction of ferromagnetic type. Lee et al have explained the spin dependent electron transport in similar systems with the huge magnetoresistance at low temperature \[29\]. The magnetoresistance drastically reduces with increase in temperature as the tunneling activation energy of this system is small. To check that the enhanced conductance at low temperatures is due to spin dependent transport a sample was chosen which show variable range hopping above 25 K in zero field. The measured resistivity data of FePtCcap600b at zero field and at 5 Tesla have been shown in figure 5. We observed that at low temperatures the resistance with an applied field of 5 T does not reduce with decrease in temperature as much as at zero field cooled resistance (figure 5). Moreover, resistivity at 5 T fits to the variable range hopping throughout the temperatures measured with the exponent 0.27 which is close to the 3D-Mott VRH. In the presence of the saturating magnetic field, the scattering due to spin dependent transport does not change with the temperature. Hence, the enhancement in conduction at low temperatures in the absence of the field can be assumed to be due to spin dependent transport, which changes with temperature as the magnetic moments align due to ferromagnetic interaction. Total conductivity of electrons due to the spin dependent transport through the granules can be expressed as \[30\]-\[32\],

\[
G = G_0(1 + B \langle \cos \alpha \rangle)
\]

(3)

where, \(B\) is a constant, \(G_0\) is the conductivity independent of spin orientation (here, it is the hopping mechanism) and the fraction, \(G/G_0\) is decided by the angle \(\alpha\) between spin orientation of adjacent granules. \(G/G_0 (1 + B \langle \cos \alpha \rangle)\) versus temperature of the sample having 20 volume % of Carbon annealed at 850 \(^\circ\)C has been shown in figure 5(a). The data is obtained by dividing the measured conductance \(G(T)\) with the VRH fitted data extrapolated to low temperature \(G_0(T)\).

For a homogeneous system, \( \langle \cos \alpha \rangle \) can be assumed from the model for superparamagnetic particle ensemble as reported to calculate temperature dependent magnetoresistance in magnetic granular films with perpendicular magnetic anisotropy \[32\]. The model here considers how thermal energy affects the ordering of granules in the presence of anisotropy energy and FM interaction energy between granules. Though, the system studied here is not super-paramagnetic, the model accounts the thermal effect on the ordered granules itself and shows how the conductivity or resistivity can be modified towards low temperatures in these systems. If \(\theta_1\) and \(\theta_2\) represent polar angles and \(\phi_1\) and \(\phi_2\) represent azimuthal angles of spin orientation of adjacent granules, then \(\cos \alpha\) can be expressed as

\[
\cos \alpha = \cos \theta_1 \cos \theta_2 + \sin \theta_1 \sin \theta_2 \cos (\phi_1 - \phi_2)
\]

(4)

Magnetic energy of the pair of granules can be written as

\[
U = -KV (\cos^2 \theta_1 + \cos^2 \theta_2) - J \cos \alpha
\]

(5)

where, \(K\), \(V\) and \(J\) are anisotropy constant, volume of granule and exchange integral respectively. The first term in the above expression is the anisotropy energy and the second term is the energy due to intergranular interaction. \( \langle \cos \alpha \rangle \) can be calculated by taking Boltzmann average of \(\cos \alpha\).
Figure 6. Depicting (a) the fractional increase in conductivity obtained from the resistance data of the sample having 20 vol% of Carbon annealed at 850 °C divided by the VRH fit extrapolated to low temperature and (b) simulation of the model showing \( \langle \cos \alpha \rangle \) as a function of temperature at different values of exchange integral.

\[
\langle \cos \alpha \rangle = \frac{\int_{\theta_1} \int_{\theta_2} \int_{\phi_1} \int_{\phi_2} [\cos \theta_1 \cos \theta_2 + \sin \theta_1 \sin \theta_2 \cos(\phi_1 - \phi_2)] e^{-U/K_B T} \sin \theta_1 \sin \theta_2 \sin \phi_1 \sin \phi_2 \, d\phi_1 \, d\phi_2 \, d\theta_1 \, d\theta_2}{\int_{\theta_1} \int_{\theta_2} \int_{\phi_1} \int_{\phi_2} e^{-U/K_B T} \sin \theta_1 \sin \theta_2 \sin \phi_1 \sin \phi_2 \, d\phi_1 \, d\phi_2 \, d\theta_1 \, d\theta_2}
\]

Figure 6(b) shows numerical simulation of the temperature dependence in \( \langle \cos \alpha \rangle \) at different exchange integral(J) values. The exchange integral enhances the conductance at low temperature. Hence, intergranular interaction can be a key factor in modifying the conductance as thermal energy reduces.

4. Conclusion

In summary, electrical properties were correlated with the microstructure of the FePt-C granular films. They can be tuned among different hopping regime by varying the intergranular separation. However, additional conductance was observed in the samples at temperatures below 25 K. Exhibited metallicity in these films at low temperatures is attributed to the intergranular magnetic ordering of these films which are affected by thermal energy. Finally, enhanced conductance at low temperature was compared with the model for spin-dependent transport in granular systems with temperature. Hence conductance in FePt granular films can be tuned by the intergranular separation and this can be implemented as spin current channel in spintronic devices.

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