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The Model for Linear Magnetoresistance of Two-Dimensional Metal-Semiconductor Composites with Interfacial Shells *

XU Jie(徐洁)[1,2**], WANG Guo-Dong(王国栋)[3], LI Shan-Dong(李山东)[1,4], LI Qiang(李强)[1], GAO Xiao-Yang(高小洋)[1]

[1] *College of Physics, Qingdao University, Qingdao 266071*
[2] *College of Chemica, Science and Engineering, Qingdao University, Qingdao 266071*
[3] *College of Information Engineering, Qingdao University, Qingdao 266071*
[4] *Laboratory of Fiber Materials and Modern Textile, Growing Base for State Key Laboratory, Qingdao University, Qingdao 266071*

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A metal-semiconductor composite with the interfacial shells is investigated theoretically for the large linear magnetoresistance effect of high doping $Ag_{2+\delta}Se$ and $Ag_{2+\delta}Te$ materials. The magnetoresistance (MR) of composites is a function of the magnetic field, temperature, the conductivities of two phases without magnetic field, and the thickness and conductivity of the interfacial shells. The MR increases with the increase of the magnetic field and with the decrease of temperature, and no saturation is found even under the high magnetic field. Moreover, it is interestingly found that the interfacial shell is an important factor for the MR of the composites. The MR increases with the thickness and the conductivity of the interfacial shells. Lastly, the theoretical results on the MR are compared with the experimental data. It is found that the value of the MR of the composite with the interfacial shell is larger than that without the interfacial shell.

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Recently, there has been growing interest in the non-magnetic compounds such as silver chalcogenides due to their large linear magnetoresistance (LMR) effect.[1–4] These materials have outstanding properties, such that the value of the MR can be compared with the colossal MR materials at room temperature and the linear field dependence of the MR is still observed under the high magnetic field of 60 T. Except for silver chalcogenides, the extraordinary MR effect is found in the other doped non-magnetic semiconductors such as the symmetric Au-InSb van der Pauw disk,[5] MnAs-GaAs composite[6] and the self-doped Si semiconductor.[7] Generally, the MR of the pure non-magnetic semiconductors will be saturated quickly in the low magnetic field due to the Hall-effect. However, the materials have a large LMR effect when the semiconductors are doped. Driven by the experimental results, theoretical models are proposed to interpret the unusual effect. Two major theories for the LMR effect at present are the quantum and classical models. Abrikosov[8] predicted a quantum linear MR effect under the extreme condition $\hbar\omega_c \gg E_F$ (ω_c is the cyclotron frequency and E_F is the Fermi energy). This condition limits the model as being only appropriate for a small number of specific materials, such as the narrow bandgap semiconductor graphene with very small effective mass and topological insulator materials. The classical models are proposed based on the resister network model,[9,10] the finite element method,[11,12] and the effective medium theory.[13–15] We proposed the classical models which can be compared with the experimental data quantitatively.[16,17] The above effective medium models were modified by considering the inhomogeneousness of the doped semiconductors, which assume that materials are composites consisting of homogeneous semiconductor matrices and the precipitated metal particles. In fact, due to the different work functions of two components, there exists the semiconductor's band bending on the interface between metal and semiconductor. Thus the MR of the composite may be sensitive to the property of the interface.

Although there are some studies on the LMR of non-magnetic semiconductors, to the best of our knowledge, interfacial shells on the MR of materials are rarely reported. To investigate such an effect, we propose a model for the MR of the metal-semiconductor composite with interfacial shells, emphasize on the dependence of the MR on the conductivity and the thickness of the interfacial shells, and compare the model predictions with the experimental data.

Beck *et al.*[18] found that the maximum silver excess which can be solved homogeneously in the Ag_2Se semiconductor is 0.79×10^{-4} at $T = 20$℃. A sample is described as 'heterogeneous' when the silver excess in the semiconductor is higher than the maximum equilibrium, although we cannot find the maximum equilibrium value in an Ag_2Te semiconductor, while the value should be similar to 0.79×10^{-4}. Moreover, some micrographs[2,18,19] showed that the excessive silver forms precipitates in the Ag_2Se and Ag_2Te semiconductor matrix. The electron-microscope photo of the sample was shown in Fig. 1(a) in Ref. [18]. Based on

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**Corresponding author. Email: xujie@qdu.edu.cn
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these micrographs, we consider that the high-doping $Ag_{2+\delta}Se$ and $Ag_{2+\delta}Te$ materials consist of two phases: an assembly of metal particles embedded in a homogeneous semiconductor matrix. In the whole 2D composite, the metal particles and matrix particles are considered as circular particles. The interfacial shells between the metal particles and the semiconductor matrix are introduced in our model, which reflect the effect of contact between two components. When metal and semiconductor are contacted, there is an interface between metal and semiconductor due to the lattice mismatch, in which the electric properties for different types of contact are not the same. The metal-semiconductor contact is divided into two classes: one is the metal and n-type semiconductor contact, and the other is the metal and p-type semiconductor contact. For the metal and n-type semiconductor contact, when the work function of metal is more than semiconductor, $W_{\rm m} > W_{\rm s}$, a positive space-charge region will be formed, which is a barrier layer with high resistance. On the contrary, when $W_m < W_{\rm s}$, a negative space-charge region will be formed, which is an inverse barrier layer with high conductivity. For the metal and p-type semiconductor contact, it will also be the inverse barrier layer or the barrier layer for $W_{\rm m} > W_{\rm s}$ or $W_{\rm m} < W_{\rm s}$, respectively. Considering the Ag-doping $Ag_{2+\delta}Se$ and $Ag_{2+\delta}Te$ material, the Ag element will enter into the crystal lattice of the semiconductor when $\delta < 0.79 \times 10^{-4}$ is at room temperature, then there will be a form of Ag aggregation in the semiconductor when $\delta > 0.79 \times 10^{-4}$. Thus for high-doping $Ag_{2+\delta}Se$ and $Ag_{2+\delta}Te$ materials, a metal and n-type semiconductor contact can be regarded, which forms an inverse barrier layer with high conductivity due to $W_{\rm m} < W_{\rm s}$. Therefore, we regard the interface shell as the inverse barrier layer between Ag particles and Ag_2Se/Ag_2Te semiconductor with high conductivity in this model (the conductivity of interface shell is more than semiconductor, $\sigma_{\rm is} > \sigma_{\rm s}$). The interfacial shell and the metal particle are combined as a complex particle, seeing Fig. 1(b).[20] The complex particle is composed of a circular metal particle of conductivity $\sigma_{\rm m}$ with a radius R and a circular interfacial shell of conductivity σ_{is} with a thickness t.

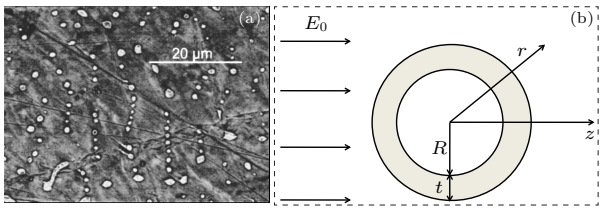

Fig. 1. (a) The electronic microscope photo of $Ag_{2.33}Se$,[18] and (b) the model of the complex particle.[20]

Applying the complex particle in a uniform electric field, based on the effective medium theory, the conductivity of the complex particle is[20]

$$\sigma_{\rm c} = \sigma_{\rm m} \frac{2\sigma_{\rm m} + \sigma_{\rm is} + 2k(\sigma_{\rm is} - \sigma_{\rm m})}{2\sigma_{\rm m} + \sigma_{\rm is} - k(\sigma_{\rm is} - \sigma_{\rm m})}, \qquad (1)$$

where $k = [R/(R+t)]^2$ for 2D thin films. Thus the volume fraction of the complex particles in the matrix (f/k) will substitute the original volume fraction of the metal particles f in the equation for the conductivity of two-phase composites. According to Bruggu-man's effective media theory,[21] we obtain the equation for the conductivity of the composites,

$$\left(1 - \frac{f}{k}\right) \frac{\sigma_{\rm e} - \sigma_{\rm s}}{2\sigma_{\rm e} + \sigma_{\rm s}} + \frac{f}{k} \frac{\sigma_{\rm e} - \sigma_{\rm c}}{2\sigma_{\rm e} + \sigma_{\rm c}} = 0. \qquad (2)$$

The effective conductivity of the composites is

$$\sigma_{\rm e} = \sigma_{\rm s} \cdot \frac{1 + 2(f/k)[(\sigma_{\rm c} - \sigma_{\rm s})/(\sigma_{\rm c} + 2\sigma_{\rm s})]}{1 - (f/k)[(\sigma_{\rm c} - \sigma_{\rm s})/(\sigma_{\rm c} + 2\sigma_{\rm s})]}, \qquad (3)$$

where $\sigma_{\rm c}$ is the conductivity of the complex particles (component 1), $\sigma_{\rm s}$ is the conductivity of the semiconductor matrix (component 2), (f/k) is the area fraction of the complex particles. Equation (3) has been widely used for the effective conductivity of the two-component composite thin film with the interfacial shells. Now if the composite is applied into a uniform magnetic field, the conductivity of each component becomes the 2D tensor and has the following form[13]

$$\hat{\sigma}_i = \begin{pmatrix} \sigma_{ixx} & -\sigma_{ixy} \\ \sigma_{ixy} & \sigma_{ixx} \end{pmatrix}, \quad (i = \text{c or s}) \qquad (4)$$

where σ_{ixx} and σ_{ixy} are the diagonal and transverse conductivities of component i, respectively. For metals and semiconductors,[13]

$$\sigma_{ixx} = \frac{\sigma_{i0}}{1 + (\mu_i H)^2},$$
$$\sigma_{ixy} = \frac{\mu_i H \sigma_{i0}}{1 + (\mu_i H)^2}, \quad (i = \text{c or s}) \qquad (5)$$

where H is the magnetic field, and μ_i is the mobility of carriers in component i.

The effective conductivity tensor of the composite thin film in the transversal magnetic field can be written as

$$\hat{\sigma_{\rm e}} = \begin{pmatrix} \sigma_{exx} & \sigma_{exy} \\ -\sigma_{exy} & \sigma_{exx} \end{pmatrix}, \qquad (6)$$

where σ_{exx} is the effective diagonal conductivity corresponding to the effective Ohmic conductivity, and σ_{exy} is the effective transverse conductivity corresponding to the effective Hall conductivity.

If we consider the presence of a uniform transversal magnetic field, the conductivities of the composite will be tensors and Eq. (3) will become invalid. Fortunately, Milton found a general transformation between the 2D electrical conductivity tensors of the thin film with magnetic field and the conductivity scalar without the magnetic field,[22] which can help us to discuss the MR effect of the composite. The 2D electrical conductivity tensors of the two components (Eq. (4)), $\hat{\sigma}'_i$, can be mapped into a new, isomorphic, well defined conductivity tensor without the magnetic field,

$$\hat{\sigma}'_i = \hat{\sigma}'_{ixx}\hat{I} = \begin{pmatrix} \hat{\sigma}'_{ixx} & 0 \\ 0 & \hat{\sigma}'_{ixx} \end{pmatrix}.$$

The relationship between the primal and transformed conductivities is

$$\hat{\sigma}'_{ixx} = \frac{\sigma_{ixx}}{c - \sigma_{ixy}}, \quad (i = \text{c or s}) \tag{7}$$

where c is the parameters determined by

$$c = \{\Delta_\text{s} - \Delta_\text{c} \pm [(\Delta_\text{s} - \Delta_\text{c})^2 + 4(\sigma_{sxx} - \sigma_{cxx}) \cdot (\sigma_{sxx}\Delta_\text{c} - \sigma_{cxx}\Delta_\text{s})]^{1/2}\}/[2(\sigma_{sxy} - \sigma_{cxy})], \tag{8}$$

where $\Delta_\text{s} = \sigma_{ixx}^2 + \sigma_{ixy}^2$ (i=c or s), and the positive square root should be used if $\sigma_{cxy} < \sigma_{sxy}$ and the negative one if $\sigma_{cxy} > \sigma_{sxy}$. Through the above transformation, we can obtain the conductivity scalars of two components, which can be used to calculate the effective conductivity of the composite based on Eq. (3), i.e.,

$$\sigma'_\text{e} = \sigma'_\text{s} \cdot \frac{1 + 2(f/a)[(\sigma'_\text{c} - \sigma'_\text{s})/(\sigma'_\text{c} + 2\sigma'_\text{s})]}{1 - (f/a)[(\sigma'_\text{c} - \sigma'_\text{s})/(\sigma'_\text{c} + 2\sigma'_\text{s})]}. \tag{9}$$

Then the effective conductivity scalar of the composite needs to be transformed into the primal tensor form with the magnetic field. The relationship between $\hat{\sigma}'_\text{e}$ and $\hat{\sigma}_\text{e}$ is

$$\sigma_{exx}\frac{b + c}{1 + {\sigma'_{exx}}^2}\sigma'_{exx}, \quad \sigma_{exy} = c{\sigma'_{exx}}^2 - b1 + {\sigma'_{exx}}^2, \tag{10}$$

with

$$\hat{\sigma}'_\text{e} = \sigma'_\text{e}\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \hat{\sigma}_\text{e} = \begin{pmatrix} \sigma'_{exx} & \sigma'_{exy} \\ -\sigma'_{exx} & \sigma'_{exx} \end{pmatrix}$$

where b and c are the parameters related to σ_{ixx} and σ_{ixy}, respectively, with c decided by Eq. (8) and b expressed as

$$c = \{\Delta_2 - \Delta_1 \pm [(\Delta_2 - \Delta_1)^2 + 4(\sigma_{2xy} - \sigma_{1xy}) \cdot (\sigma_{2xy}\Delta_1 - \sigma_{1xy}\Delta_2)]^{1/2}\}/[2(\sigma_{2xy} - \sigma_{1xy})], \tag{11}$$

where $\Delta_\text{s} = \sigma_{ixx}^2 + \sigma_{ixy}^2$ (i=c or s), and the positive and negative square roots are adopted in the cases of $\sigma_{1xy} < \sigma_{2xy}$ and $\sigma_{1xy} > \sigma_{2xy}$, respectively.

The MR of the composition is defined by $MR = \rho_{exx}(H) - \rho_{exx}(0)/\rho_{exx}(0)$, where $\rho_{exx}(0)$ and $\rho_{exx}(H)$ are the effective Ohmic resistivity of the composition without and with the magnetic field, respectively. When the magnetic field is zero, the resistivity $\rho_{exx}(0) = 1/\sigma_{exx}(0)$; and when the composite is in the magnetic field, the relation between the resistivity and the conductivity is $\rho_{exx}(H) = \sigma_{exx}/\sigma_{exx}^2 + \sigma_{exy}^2$. Thus we obtain

$$\text{MR} = \frac{\sigma_{exx}(0)\sigma_{exx}(H)}{\sigma_{exx}^2(H)\sigma_{exy}^2(H)} - 1, \tag{12}$$

where $\sigma_{exx}(0)$ is determined by Eq. (1), $\sigma_{exx}(H)$ and $\sigma_{exy}(H)$ are determined by Eqs. (9) and (10), respectively. The proposed model indicates that the MR is a function of the magnetic field, the volume fraction of the complex particles, and the conductivity and carriers' mobility of the two components without the magnetic field.

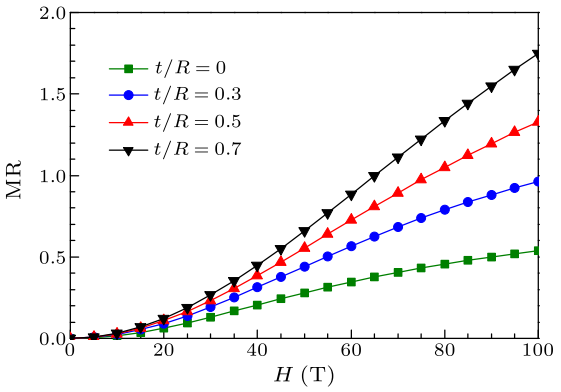

Fig. 2. The MR of the composite thin film as a function of magnetic field H for different thicknesses of the interfacial shells ($t/R = 0$, 0.3, 0.5 and 0.7).

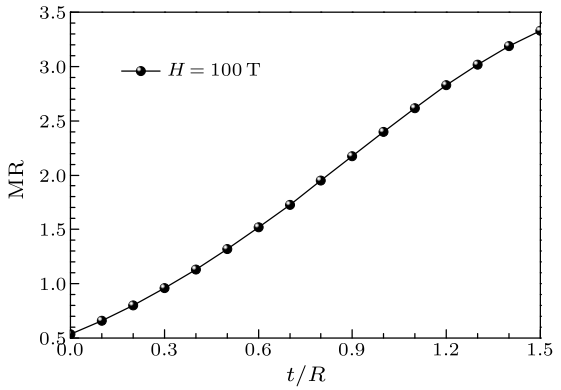

Fig. 3. The MR of the composite thin film as functions of the ratio of the thickness of the interfacial shells and the radius of the metal particles.

The MR of the composite thin film is a function of the conductivity of the complex particles based on our model. Next, the influence of the interfacial shells on the MR is discussed in detail. The magnetic field H dependence of the MR is shown in Fig. 2 for the different thicknesses of the interfacial shell t ($t = 0$, $0.3R$, $0.5R$, and $0.7R$), where we take the physical parameters of the model as follows: the area fraction of metal particles $f = 0.1$, the ratio of the conductivity of two components without the magnetic field $\alpha = \sigma_\text{m}/\sigma_\text{s} = 1000$, the ratio of the conductivity of interfacial shell to semiconductor without magnetic field $\beta = \sigma_\text{is}/\sigma_\text{s} = 500$, and the ratio of the carriers mobility in the two components $\gamma = \mu_\text{c}/\mu_\text{s} = 0.01$. The results indicate that the MR increases with H and no saturation is found in the high magnetic field, even at $H = 100\,\text{T}$, which is in agreement with many experimental results of the doped semiconductors.[1–5] Moreover, it is also found that there is a remarkable influence of the thickness of the interfacial shells on the MR of the composite. Figure 3 shows the thickness of the interfacial shell dependence of the MR when

$H = 100\,\mathrm{T}$. When we take $t/R = 0$, there is not the interfacial shell between the metal particles and the semiconductor matrix, and the MR of the composite is about 0.65 at $H = 100\,\mathrm{T}$. The MR increases with the thickness of the interfacial shell, and the value of the MR reaches about 3.3 when $t/R = 1.5$, about five times greater than the value of the MR without interfacial shell. Some researchers[9] attributed the large LMR to the disorder of the current flow in the materials. Due to the fact that the electric properties of metal and semiconductor are different, Ag aggregation in a semiconductor will increase the disorder of the current flow, which will increase the MR of materials. The interface shell between metal and semiconductor will increase the disorder of the current flow and MR of the composite further. The thickness of the interfacial shells is an important parameter for the MR of the composite.

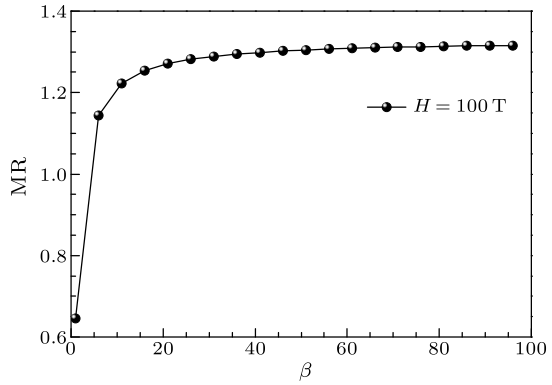

Fig. 4. The MR of the composite thin film as a function of the ratio of the conductivities of the interfacial shells and the semiconductor.

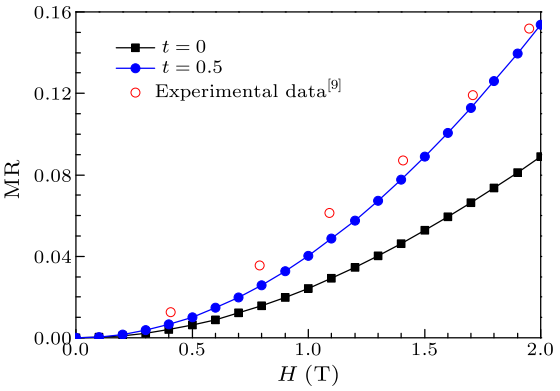

Fig. 5. The MR of the composite thin film as a function of magnetic field H at room temperature for the $Ag_{2.4}Te$ sample.

The properties of the interfacial shells include not only the thickness but also the conductivity. The dependence of the MR on the ratio of two components conductivities β is shown in Fig. 4, where we take $f = 0.1$, $t/R = 0.5$, $\alpha = \sigma_\mathrm{m}/\sigma_\mathrm{s} = 1000$, $\gamma = \mu_\mathrm{c}/\mu_\mathrm{s} = 0.01$ and $H = 100\,\mathrm{T}$, respectively. When $\beta = 1$, the interfacial shells are not considered in the composite thin film, and the value of the MR is about 0.65. The MR of the composite increases rapidly with the conductivity of the interfacial shells; then the MR tends to be saturated when β continues to increase to more than 20. The results indicate that the disparity between the conductivity of the interfacial shells and the semiconductor matrix will induce the larger MR, although the increase of the MR will be saturated for $\beta > 20$.

The above results indicate that the property of the interfacial shells is an important factor for the MR of the composite. Generally, a larger thickness and an appropriate value of β for the sample induce a larger MR value.

Figures 5 and 6 compare the model predictions with the experimental data for the $Ag_{2.4}Te$ and $Ag_{2.3}Se$ samples. We consider 1 mol $Ag_{2+\delta}Se$ or $Ag_{2+\delta}Te$ as a composite consisting of 1 mol pure semiconductor and δ mol silver metal. The area fraction of silver metal can be calculated approximately by

$$f = \frac{V_\mathrm{m}}{V_\mathrm{s} + V_\mathrm{m}} = \frac{\delta M_\mathrm{m}/\rho_\mathrm{m}}{M_\mathrm{s}/\rho_\mathrm{s} + \delta M_\mathrm{m}/\rho_\mathrm{m}},$$

where M_s and ρ_s are the molecular weight and the density of the semiconductor, M_m and ρ_m are the atomic weight and the density of the silver metal, respectively. We take $M_\mathrm{s} = 294.7$ and $\rho_\mathrm{s} = 7.96\,\mathrm{kg/m^{-3}}$ for Ag_2Se, $M_\mathrm{s} = 343.3$ and $\rho_\mathrm{s} = 8.5\,\mathrm{kg/m^{-3}}$ for Ag_2Te, $M_\mathrm{m} = 107.87$ and $\rho_\mathrm{m} = 10.502\,\mathrm{kg/m^{-3}}$ for silver metal. The conductivities without magnetic field and mobility of carriers of two components are taken as $\sigma_{\mathrm{s}0} = 5 \times 10^4\,(\Omega{\cdot}\mathrm{m})^{-1}$ and $\mu_\mathrm{s} = 2500\,\mathrm{cm^2/Vs}$ for Ag_2Se;[12] $\sigma_{\mathrm{s}0} = 2.78 \times 10^4\,(\Omega{\cdot}\mathrm{m})^{-1}$ and $\mu_\mathrm{s} = 2400\,\mathrm{cm^2/Vs}$ for Ag_2Te;[17] $\sigma_{\mathrm{m}0} = 6.62 \times 10^7\,(\Omega{\cdot}\mathrm{m})^{-1}$ and $\mu_\mathrm{m} = 70\,\mathrm{cm^2/Vs}$ for silver metal[12] at room temperature, respectively. Furthermore, the parameters σ_{i0} and μ_i in the model are affected by temperature T. The relations among σ_{i0}, μ_i and T for the semiconductor and for the metal are $\sigma_{\mathrm{s}0} \propto T^{-3/2}$, $\mu_\mathrm{s} \propto T^{-3/2}$, $\sigma_{\mathrm{m}0} \propto T^{-1}$ and $\mu_\mathrm{m} \propto T^{-1}$, respectively, which can determine the values of parameters at different temperatures. For all samples, it is found that the calculated MR by the model is well consistent with the experimental results when $t/R = 0.5$ and $\beta = 100$ for all the samples.

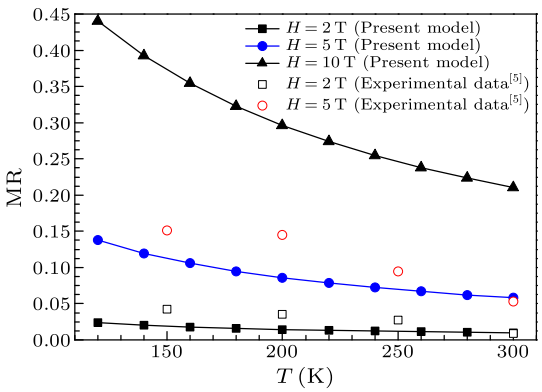

Fig. 6. The MR of the composite thin film as a function of temperature at different magnetic fields for the $Ag_{2.3}Se$ sample.

Figure 5 shows the magnetic field dependence of

the MR for the $Ag_{2.4}Te$ sample. As is illustrated, the MR increases with the magnetic field H, while it does not exhibit saturation characteristics with the magnetic field. It is unsurprising that the value of the MR of the composite with the interfacial shells is larger than that without the interfacial shells. The results for MR predicted by the model with the interfacial shells are in good agreement with the experimental data.[2]

The dependence of the MR on temperature T in different magnetic fields is shown in Fig. 6, where we take the $Ag_{2.3}Se$ thin film as an example. It is also found that the MR increases with the magnetic field at the same temperature. The MR decreases with the increasing temperature and the linear dependence of the MR on temperature is found from 200 K to 300 K, which can be mainly attributed to the decrease of the mobility of two components with the increase of temperature. Xu *et al.* reported that the MRs of the $Ag_{2+\delta}Se$ and $Ag_{2+\delta}Te$ bulks decrease linearly with the increasing temperature from 200 K to 300 K.[1] Their conclusion is consistent with our result. The theoretical predictions are in good agreement with the experimental data of the $Ag_{2.3}Se$ thin film.[5]

In summary, a model for the linear MR of metal-semiconductor thin films with interfacial shells has been proposed. The materials are considered as an assembly of the composite particles (the metal particles are packaged by the inverse barrier interfacial shells with high conductivity) embedded in a homogeneous semiconductor matrix. The model addresses the dependences of the MR on magnetic field, temperature, the conductivities of two phases without magnetic field, and the thickness and conductivity of the interfacial shells. It is revealed that the properties of the interfacial shells are important for improving the MR of the composite. Lastly, the theoretical results on the MR are in good agreement with the experimental data.

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