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Spin-pair correlation driven the colossal magnetoresistance effect in multiferroics $CdCr_2S_4$

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To understand the anomalous conductivity and colossal magnetoresistance effect of multiferroics $CdCr_2S_4$ around magnetic transition temperature T_C, we propose the spin-pair correlation dependence of magnetic polarons model. In $CdCr_2S_4$, system shows the spontaneous magnetic order at T_C and the magnetic order promotes the delocalization of magnetic polarons. According to the proposed model of a dual-conduction behavior, the normal and delocalized magnetic polarons coexist below T_C due to the gradual delocalization process of magnetic polarons. Compared to the conductivity of normal magnetic polarons, the conductivity from the delocalized magnetic polarons is dominant. It is suggested that the spin-pair correlation modifies the hopping activation energy of delocalized polarons to realize the anomalous conductivity and colossal magnetoresistance effect. In addition, the applied magnetic field, which promotes the magnetic order and delocalization of magnetic polarons, also leads to the increase of conductivity via spin-pair correlation. It is found that the obtained conductivity and colossal magnetoresistance are in agreement with the experimental results.

Keywords: magnetic order, magnetic polaron, delocalization, spin-pair correlation, colossal magnetoresistance.

1. Introduction

The magnetic chromium spinels with the general formula ACr_2X_4 have become the focus of theoretical and experimental interest in recent years, because they exhibit many exotic magnetic and electrical properties [1–4]. Many relevant researches of these compounds reveal the strong interplay among the spin, the charge and the lattice, which is regarded as the source of the interesting physical phenomena [3,5–7]. The strong interplay among different properties offer extra degree of freedom and make ACr_2X_4 the potential candidates for applications in magnetic data storage and high-frequency magnetic devices [8,9].

$CdCr_2S_4$ is typical ferromagnetic semiconductor, which shows ferromagnetic phase transition at the Curie temperature $T_C = 85$ K, has captured the attention of researchers since 1960th due to the various interests. Early studies mainly focused on the anomalous expansion coefficient, the strong blue shift of absorption edge, the unusual phonon shift and so on [10–12]. It has been found that these phenomena are relevant to the paramagnetic-ferromagnetic (PM–FM) transition of system. The ion Cr^{3+} with a spin $S = 3/2$, which has a half-filled t_{2g} ground state and a saturated moment μ_s is equal to $3\mu_B$, is responsible for the magnetic properties and the PM–FM transition originates from the superexchange interaction of the 90° Cr^{3+}–S^{2-}–Cr^{3+} path [13,14]. The magnetic properties of $CdCr_2S_4$ can be characterized as a typical Heisenberg ferromagnet.

Besides, it was claimed that $CdCr_2S_4$ exhibits evident multiferroic behavior, as well as colossal magnetoresistance (CMR) and colossal magnetocapacitance (CMC) effects [1,15]. The similar effects have also been found in other members of ACr_2X_4 [2–4]. All these make thio-spinel compounds regain much attentions. Researches reveal that multiferroic materials $CdCr_2S_4$ shows the simultaneous occurrence of magnetic order and relaxor ferroelectricity as well as many interesting phenomena below T_C [15]. It has been firmly testified the magnetic ion Cr^{3+} plays the key role in the exotic properties of the system via the off-center displacement of Cr^{3+}. Guided by previous conclusions, it is suggested that the relaxor ferroelectricity in $CdCr_2S_4$ originates from an off-center displacement of the magnetic ion Cr^{3+} [5]. This off-center displacement of Cr^{3+} also resulted in a peculiar coupling between the magnetic properties and relaxor ferroelectricity. The study of electron paramagnetic resonance (EPR) spectra and x-ray diffraction measurement reveal the spin-lattice coupling happen accompanying with the ferromagnetic phase transition.

Moreover, this coupling is also impacted by the external magnetic field and pressure [16,17]. We regard this spin-lattice coupling as the major contribution of magnetoelectric (ME) coupling between the magnetic order and relaxor ferroelectricity. Based on the ME coupling theory, many phenomena, such as the anomaly of dielectric constant and heat capacity, magnetocaloric effect and magnetic entropy change etc., can be explained reasonably [7,16,18–21].

The CMR effect in ACr_2X_4-type compounds are also becoming one of the most attractive issues in recent years. In $CdCr_2S_4$, dc conductivity $\sigma_d(T)$ exhibits an evident anomaly and the CMR effect occurs around its magnetic transition temperature [15]. In general, the CMR effect is mainly reported in manganites, which is thought to be relevant to the double exchange (DE) mechanism [22,23]. However, DE mechanism can be excluded in ACr_2X_4 due to the magnetic ion Cr^{3+} having a half-filled t_{2g} state level [2]. It is plausible that an alternative mechanism is responsible for the CMR effects of ACr_2X_4-type system. Studies reveal that a kind of spin-pair correlation dependent magnetic polarons exist in ACr_2X_4 [3,24–27]. The conduction process of the system mainly attributes to the thermal hopping of magnetic polarons. With the decrease of temperature, the magnetic polarons delocalize gradually and grow in size. Furthermore, the spontaneous ferromagnetic order of system below T_C promotes a more evident delocalization process. The delocalized magnetic polarons possess a much higher hopping probability and lead to a much stronger conduction ability. More importantly, the conduction of delocalized magnetic polarons are believed to be completely different from the itinerant behavior which has not been found from experimental data for $CdCr_2S_4$ [28]. The spin-pair correlation, which reflects the degree of magnetic order and the delocalization of magnetic polarons, can be used to represent the change of hopping probability of delocalized magnetic polarons. For comparison, the spectroscopic signatures of (localized) polarons can be found going with transport process and show metallic *T*-dependence of resistivity and Drude-like conductivity in the CMR-system La:$SrMnO_3$ [29]. Accordingly, when temperature is lower or magnetic field stronger, the rate of thermal hopping of delocalized magnetic polarons enhance due to the spin-pair correlation and the anomalous conductivity as well as the CMR effect ensue.

In this article, we concentrate on the temperature and magnetic field dependent conduction properties as well as the CMR effect in $CdCr_2S_4$. An appropriate relation of the proportion and the modified hopping activation energy for delocalized magnetic polarons are applied to describe the conduction behavior. We consider that the gradually delocalized magnetic polarons and the decreasing hopping activation energy are the origin of anomalous conductivity and the CMR effect. Briefly this article is arranged as follows: model and methods which describe the temperature and magnetic field dependent conduction properties are given in Sec. 2, the numerical results and discussion are given in Sec. 3, and finally the conclusions are proposed in Sec. 4.

2. Model and methods

In $CdCr_2S_4$, electron polarizes the magnetic moment of magnetic ion Cr^{3+} around itself via exchange interaction forming a small ferromagnetic spin cluster and localizes into this cluster to form a magnetic polaron. The energy of magnetic polarons in the framework of mean-field approach can be expressed as [30,31]

$$H = t\left(\pi\frac{d}{a}\right)^2 + JS^2\frac{4\pi}{3}\left(\frac{a}{d}\right)^3, \tag{1}$$

where the first term is the kinetic energy of electron localized in the sphere cluster of radius a and the second term is the energy loss of the antiferromagnetic exchange due to the FM order of local spin S inside the magnetic polaron. J is antiferromagnetic exchange, t is the bandwidth, d is the inter-site distance.

The average conductivity of magnetic polaron can be expressed as [30,32]

$$\sigma = \frac{\sigma_0}{T}\exp\left(-\frac{U_0}{k_BT}\right), \tag{2}$$

where U_0 is the hopping activation energy of electron localized in magnetic polaron and proportional to $e^2/\varepsilon a$ with ε being the static dielectric constant, σ_0 is the zero temperature conductivity, k_B is the Boltzmann constant. We find the conductivity increases monotonously with temperature, which is the typical conduction behavior of magnetic polarons.

However, this single relation can not describe the evolution of conductivity of $CdCr_2S_4$ for the wide temperature range. The reason lies in the occurrence of spontaneous magnetic order at T_C which promotes the delocalization of magnetic polarons. Knowledge of magnetic order is important for understanding electronic transport properties. Accordingly, studying the magnetic properties of system are essential.

The magnetic properties of system originate from the magnetic ion Cr^{3+} and can be described by a Heisenberg model. The Hamiltonian reads [13]

$$H = -\sum_{\langle i,j\rangle} J_1\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{[i,k]} J_2\mathbf{S}_i\cdot\mathbf{S}_k - \sum_i \mathbf{h}\cdot\mathbf{S}_i, \tag{3}$$

where $\mathbf{S}_i$ is the Heisenberg spin of Cr^{3+} at site i, J_1 and J_2 represent the nearest and next-nearest neighbor exchange integral, respectively. The denotation $\langle i,j\rangle$ and $[i,k]$ mean that the sum of nearest and next-nearest neighbor are involved, respectively. $\mathbf{h}$ is the external magnetic field.

When the magnetic field **h** is parallel to the z axis, the Hamiltonian on the single ion Cr^{3+} can be expressed as

$$H_i = H_x S_i^x + H_z S_i^z. \tag{4}$$

Based on the mean field theory, H_x and H_z are given as

$$H_x = 6J_1\langle S^x\rangle + 30J_2\langle S^x\rangle, \tag{5}$$

$$H_z = 6J_1\langle S^z\rangle + 30J_2\langle S^z\rangle + h. \tag{6}$$

Here, x and z represent the spin components of Cr^{3+}. For each magnetic ion, the number of nearest and next-nearest neighbor magnetic ion are 6 and 30, respectively. According to the statistical theory, the thermo-average of spin components can be expressed as

$$\langle S^x\rangle = \frac{H_x}{2\sqrt{H_x^2+H_z^2}} \frac{\sum_{i=1}^{2}(2i-1)\sinh\left(\frac{2i-1}{2k_BT}\sqrt{H_x^2+H_z^2}\right)}{\sum_{i=1}^{2}\cosh\left(\frac{2i-1}{2k_BT}\sqrt{H_x^2+H_z^2}\right)}, \tag{7}$$

$$\langle S^z\rangle = \frac{H_z}{H_x}\langle S^x\rangle. \tag{8}$$

The spin-pair correlation of nearest neighbor can be decoupled as

$$\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle = \langle S^x\rangle^2 + \langle S^z\rangle^2. \tag{9}$$

In general, magnetic polarons only survive over a limited range of temperature and magnetic field. The PM state has been deemed to be the surviving environment of magnetic polarons. In $CdCr_2S_4$, the PM–FM transition happens at T_C and external magnetic field can also promotes the magnetic order. The magnetic order makes the magnetic polarons delocalized. The delocalization process refers to magnetic polarons confining from a spin cluster before the delocalization process to several of overlapped spin clusters, which enables the delocalized polarons to hop much easier inside several of spin clusters [33]. This easy hopping embodies the activation energy being modified by the magnetic order inside the several of spin clusters. Furthermore, we should realize that the enhancement of magnetic order is a gradual process rather than an abrupt one for the effect of temperature and magnetic field, which brings up the gradual delocalization process of magnetic polarons. Accordingly, the activation energy of hopping process U_0 of delocalized magnetic polarons can be modified as

$$U = U_0 + g\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle, \tag{10}$$

where the spin-pair correlation $\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle$ reflects the magnetic order degree of system, g is viewed as the correlation strength between the magnetic moment and polarons.

Besides, it is also considered that the delocalization of magnetic polarons are stochastic, and the number of delocalized magnetic polarons are dependent on system magnetization M. This dependence is assumed to be linear and approached by $m = M/M_{\text{sat}}$ with M_{sat} the saturated magnetism (equivalent to $3\mu_B/Cr^{3+}$) [31]. Thus, the normal and delocalized magnetic polarons will coexist in the same phase until the total magnetic order achieves. Under this kind of hypothesis, the conductivity of system can be amended as

$$\sigma = \left(1-\frac{M}{M_{\text{sat}}}\right)\frac{\sigma_0}{T}\exp\left(-\frac{U_0}{k_BT}\right) + \\ + \frac{M}{M_{\text{sat}}}\frac{\sigma_0}{T}\exp\left(-\frac{U_0 + g\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle}{k_BT}\right). \tag{11}$$

The first term of Eq. (11) denotes the conductivity from the contribution of normal magnetic polarons, and the second term from the delocalized magnetic polarons.

3. Results and discussion

In $CdCr_2S_4$, we choose $J_1 = 12.53k_B$ and $J_2 = 0.25k_B$ to produce the ferromagnetic order at $T_C = 85$ K [13,19]. We define the dimensionless magnetization as $m(h) = M(h)/M_{\text{sat}}$, where $M(h)$ represents the real magnetization and M_{sat} is the saturation magnetization corresponding to $3\mu_B/Cr^{3+}$ for $CdCr_2S_4$. Based on the definition, the dimensionless magnetization $m(h)$ is proportional to the magnetization $M(h)$. Temperature and magnetic field dependence of dimensionless magnetism $m(h)$ are shown in Fig. 1(a). It is found that the non-zero dimensionless magnetization arises at T_C indicating the occurrence of PM–FM transition, and the magnetization enhances with the increase of external magnetic field. In order to show clearly the low fields of magnetization of $CdCr_2S_4$, we exhibit the relation between the dimensionless magnetization $m(h)$ and $\ln h$ in Fig. 1(b). When the temperature are below T_C (the red curve), system shows the spontaneous order in extremely low field and the degree of magnetic order enhances gradually with the decrease of temperature. Meantime, the applied magnetic fields promote the degree of magnetic order until the total magnetic order completes. All these clearly suggest the effect of temperature and magnetic field on the magnetization of system. Corresponding to the magnetization, the spin-pair correlation has the similar tendency for the onset of temperature and magnetic field (seeing Figs. 1(a) and 2(a)). The fluctuation of spin-pair correlation, defined as $\Delta\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle\left(\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle_{(h,T)} - \right.$ $\left. - \langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle_{(0,T)}\right)$, reaches its peak around T_C embodying the sudden occurrence of magnetic order in $CdCr_2S_4$. Meanwhile the external magnetic field also promotes the fluctuation degree of spin-pair correlation via the growing of magnetic order (seeing Fig. 2(b)).

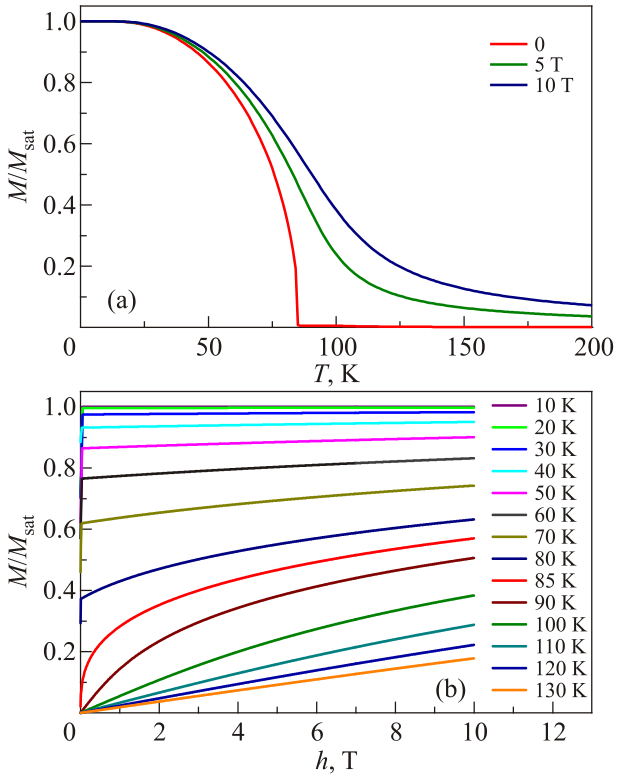

Fig. 1. (Color online) (a) Temperature dependences of dimensionless magnetism for different magnetic field. (b) Magnetic field dependences of dimensionless magnetism for different temperatures.

Through the linear fit for the relevant experimental results, we yield a hopping activation energy of magnetic polarons $U_0 = 0.13$ eV. Base on the fit data, the temperature and magnetic field dependences of conductivity are plotted in Fig. 3(a). With the decrease of temperature, the conductivity σ decreases exponentially above the magnetic transition temperature T_C, which conforms to the conduction character of magnetic polarons. When the temperature is below T_C, the conductivity exhibits sudden increase (seeing the red curve of Fig. 3(a)). It is found the conductivity behavior is in good agreement with the experimental data of Ref. 15, in which the curve at 3 mHz gives a good approximation of the dc conductivity. Below T_C, the coexistence of the normal and delocalized magnetic polarons happens and this coexistence will survive until the total magnetic order performs. Therefore, two types of magnetic polarons contribute to the conductivity of system, in which the delocalized one is dominant due to the hopping activation energy modified by the spin-pair correlation. Anomalous conductivity can attribute to the gradual delocalization process of magnetic polarons due to the occurrence of magnetic order at T_C. In addition, a continuous conduction behavior around T_C exhibits in Fig. 3(a), which also results from the gradual delocalization process of magnetic polarons. Due to the gradual delocalization, the coexistence of normal and delocalized magnetic polarons should appear, then the continuous transition of conductivity happens. Besides, the applied magnetic field also enhances the magnetic order and promotes the delocalization, which makes the anomalous con-

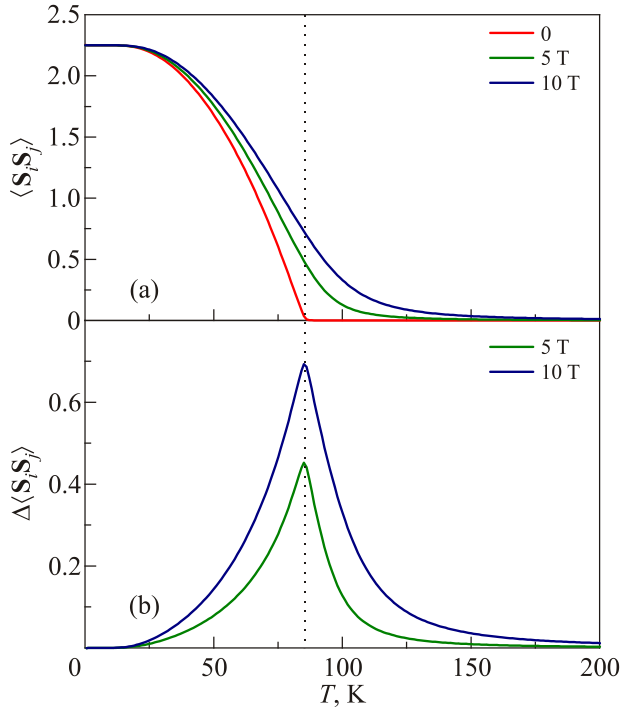

Fig. 2. (Color online) Temperature dependences of (a) spin-pair correlation $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$, (b) the fluctuation of spin-pair correlation $\Delta\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ for different magnetic field. The dotted line refers to the magnetic transition temperature T_C.

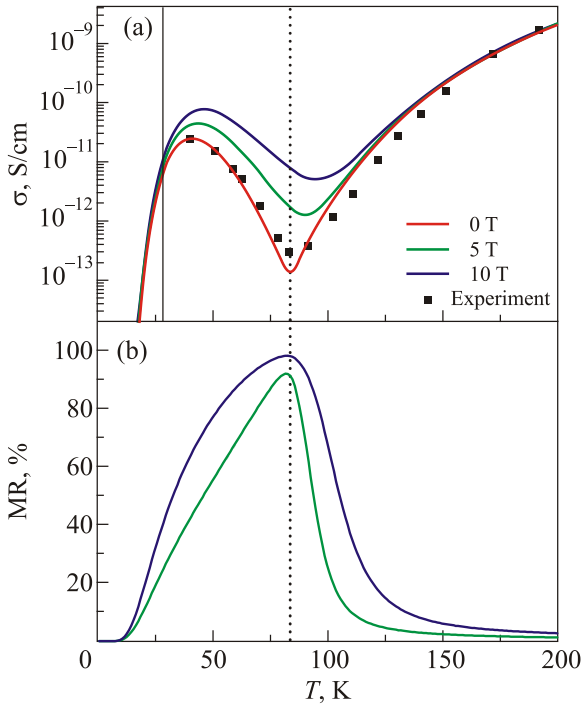

Fig. 3. (Color online) Temperature dependences of conductivity σ (a) and the CMR effect (b) for different magnetic field. The closed squares are the experimental data and the dotted line refers to the magnetic transition temperature T_C.

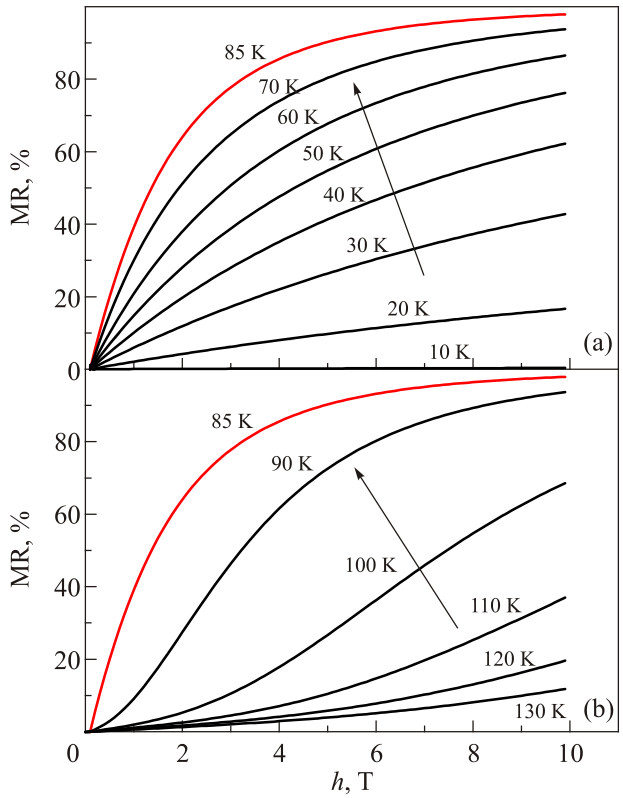

Fig. 4. The magnetic field dependences of CMR effect for different temperatures: (a) below T_C, and (b) above T_C.

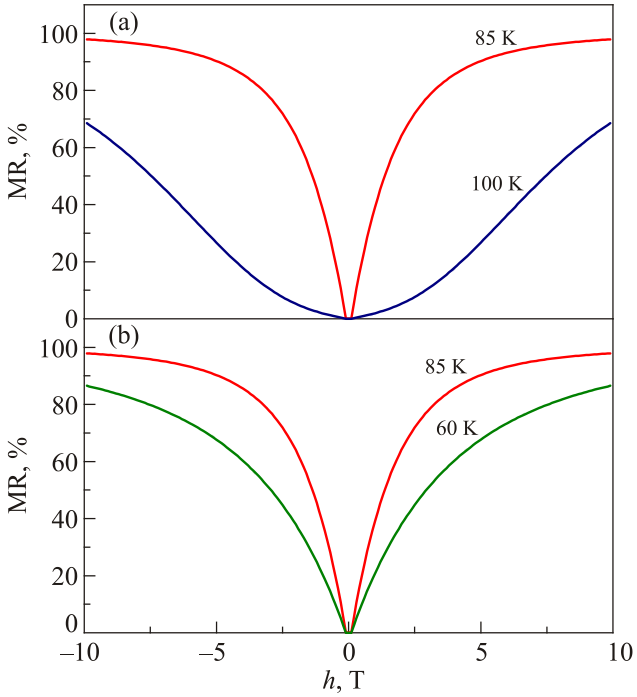

Fig. 5. The evolution behavior of CMR effect on the magnetic field: (a) above T_C, and (b) below T_C.

ductivity shift to a higher temperature. As a result, the spin-pair correlation is the main reason to cause the anomalous conductivity.

The MR, defined as MR = $(\rho(0,T)-\rho(h,T))/\rho(0,T)$ (ρ is the resistivity of system and the inverse of conductivity), are shown in Figs. 3(b) and 4. At T_C, the magnetic transition temperature, the maximum value of MR happens. When the temperature deviates from T_C, the MR effect diminishes gradually. The most remarkable MR effect is observed at magnetic transition temperature T_C, which is very similar to the behavior of the fluctuation of spin-pair correlation $\Delta\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ (Fig. 2(b)). All these confirm that the spin-pair correlation can play a key role in the anomalous conductivity. Besides, the magnetic field also promotes the spin-pair correlation to realize the increase of MR effect. However, this MR effect does not always increase, but tend to saturation with the enhanced magnetic field (seeing Fig. 4).

Although, the promotion of magnetic field on MR effect is obvious, the manner of promotion is quite special at different temperature range. Figure 5 shows the evolution trend MR effect below and above the magnetic transition temperature T_C. The smooth magnetic field dependence of MR effect above T_C, while this dependence is sharp below T_C. The discrepancy results from the difference of spin-pair correlation between two types of temperature ranges. Above T_C, only magnetic field driven spin-pair correlation contributes to the MR effect. Below T_C, both the spontaneous magnetic order and magnetic field simultaneously promote the spin-pair correlation, which leads to the sharp MR effect. Undoubtedly, the quantitatively coincident results with the experimental data approve our conception [2,3,15].

4. Conclusions

In this article, the conductivity and CMR effect of $CdCr_2S_4$ have been studied. It is found that the spontaneous magnetic order at T_C promotes the formation of overlapped spin cluster and destroys the survival environment of magnetic polarons to make the magnetic polarons delocalized gradually. Thus the normal and delocalized magnetic polarons coexist in the same phase until the entire magnetic order has been established. Herein, the delocalized magnetic polarons locate in the overlapped spin clusters distinguished from the normal magnetic polarons in single spin cluster, which enables an electron to hop more easily from one cluster to another inside the overlapped spin clusters and enhances the conductivity of system. The hopping activation energy, which is modified by spin-pair correlation, can depict quantitatively this facilitation of hopping conductivity. Furthermore, the applied magnetic field promotes the magnetic order to enhance the conduction ability of system. The anomalous conductivity and CMR effect in $CdCr_2S_4$ result from the spin-pair correlation driven delocalization process of magnetic polarons. Accordingly, small-radius magnetic polarons describe the dielectric state, and the overlap of large-radius magnetic polarons with the formation of a narrow conduction band

can describe the metallization of the sample after the ferromagnetic transition. The presence of colossal changes in the magnetoresistance and the magnetic capacity also follows from this physical picture.

In this article, we mainly focus on the spin-pair correlation dependent dc conductivity. We also notice that the magnetic polarons hopping may lead to variable range hopping. Although the present work only confirms the key role of spin-pair correlation for the anomalous dc-conductivity, we believe that it may play an important role on ac conductivity of system. The relevant work is undertaking and will deliver in the following time.

Acknowledgments

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Кореляція спінових пар, що обумовлена колосальним магніторезистивним ефектом в мультифероїках $CdCr_2S_4$

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Для пояснення аномальної провідності та колосального магніторезистивного ефекту в мультифероїках $CdCr_2S_4$ поблизу температури магнітного переходу T_C запропоновано модель магнітних поляронів з кореляційною залежністю спінових пар. В системі $CdCr_2S_4$ показано наявність спонтанного магнітного порядку в T_C, що обумовлює делокалізацію магнітних поляронів. Згідно з запропонованою моделлю, нормальні й делокалізовані магнітні полярони співіснують нижче T_C завдяки плавним делокалізаційним процесам. В порівнянні з провідністю нормальних магнітних поляронів провідність

через делокалізацію магнітних поляронів є домінуючою. Передбачується, що кореляція спінових пар модифікує енергію стрибкової активації делокалізованих поляронів, що призводить до аномальної провідності й колосального магніторезистивного ефекту. Додатково прикладене магнітне поле, яке сприяє формуванню магнітного порядку та делокалізації магнітних поляронів, призводить також до зростання провідності через кореляцію спінових пар. Одержані провідність та колосальний магнітоопір узгоджуються з експериментальними результатами.

Ключові слова: магнітний порядок, магнітний полярон, делокалізація, кореляція спінових пар, колосальний магнітоопір.

Корреляция спиновых пар, обусловленная колоссальным магниторезистивным эффектом в мультиферроиках $CdCr_2S_4$

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Для объяснения аномальной проводимости и колоссального магниторезистивного эффекта в мультиферроиках $CdCr_2S_4$ вблизи температуры магнитного перехода T_C предложена модель магнитных поляронов с корреляционной зависимостью спиновых пар. В системе $CdCr_2S_4$ показано наличие спонтанного магнитного порядка в T_C, который обусловливает делокализацию магнитных поляронов. Согласно предложенной модели, нормальные и делокализованные магнитные поляроны сосуществуют ниже T_C благодаря плавным делокализационным процессам. По сравнению с проводимостью нормальных магнитных поляронов проводимость из-за делокализации магнитных поляронов является доминирующей. Предполагается, что корреляция спиновых пар модифицирует энергию прыжковой активации делокализованных поляронов, что приводит к аномальной проводимости и колоссальному магниторезистивному эффекту. Дополнительно приложенное магнитное поле, которое способствует формированию магнитного порядка и делокализации магнитных поляронов, приводит также к возрастанию проводимости вследствие корреляции спиновых пар. Полученные проводимость и колоссальное магнитосопротивление согласуются с экспериментальными результатами.

Ключевые слова: магнитный порядок, магнитный полярон, делокализация, корреляция спиновых пар, колоссальное магнитосопротивление.