Estimation of the charge carrier localization length from Gaussian fluctuations in the magneto-thermopower of $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$

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The magneto-thermoelectric power (TEP) $\Delta S(T,H)$ of perovskite-type manganese oxide $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ is found to exhibit a sharp peak at some temperature $T^*=170$ K. By approximating the true shape of the measured magneto-TEP in the vicinity of T^* by a linear triangle of the form $\Delta S(T,H) \simeq S_p(H) \pm B^{\pm}(H)(T^*-T)$, we observe that $B^-(H) \simeq 2B^+(H)$. We adopt the electron localization scenario and introduce a Ginzburg-Landau (GL)-type theory which incorporates the two concurrent phase transitions, viz., the paramagnetic-ferromagnetic transition at the Curie point T_C and the ''metal-insulator'' (M-I) transition at T_{MI}. The latter is characterized by the divergence of the field-dependent charge carrier localization length $\xi(T,H)$ at some characteristic field H_0. Calculating the average and fluctuation contributions to the total magnetization and the transport entropy related magneto-TEP $\Delta S(T,H)$ within the GL theory, we obtain a simple relationship between T^* and the above two critical temperatures (T_C and T_{MI}). The observed slope ratio $B^-(H)/B^+(H)$ is found to be governed by the competition between the electron-spin exchange JS and the induced magnetic energy M_sH_0. The comparison of our data with the model predictions produce $T_C=195$ K, $JS=40$ meV, $M_0=0.4M_s$, and $\xi_0=5$ Å for the estimates of the Curie temperature, the exchange coupling constant, the critical magnetization, and the localization length, respectively. The magneto-TEP data obtained by other authors are discussed and found to be consistent with the model predictions as well. [S0163-1829(99)00734-1]

I. INTRODUCTION

The intriguing magnetotransport properties of manganite's family $R_{1-x}A_xMnO_3$ (where R=La,Y,Nd,Pr and A=Ca,Sr,Ba,Pb) with a Mn^{3+}/Mn^{4+} mixed valence keep attracting much attention of both experimentalists and theorists.[1–21] In the doping range $0.2<x<0.5$, these compounds are known to undergo a double phase transition from paramagnetic (PM) insulator (I) to ferromagnetic (FM) metal (M) state characterized by the Curie temperature T_C and the charge carrier localization temperature T_{MI}, respectively. The so-called giant magnetoresistivity (GMR) exhibits a sharp peak around T_{MI}, while below T_C the system acquires a spontaneous magnetization accompanied by giant magnetic entropy changes.[14] Despite a variety of theoretical scenarios attempting to describe this phenomenon, practically all of them adopt as a starting point the so-called double-exchange (DE) mechanism, which considers the exchange of electrons between neighboring Mn^{3+}/Mn^{4+} sites with strong on-site Hund's coupling. The estimated exchange energy[11] $JS=45$ meV (where $S=2$ is an effective spin on a Mn site), being much less than the Fermi energy E_F in these materials (typically, $E_F=0.15$ eV), favors a FM ground state. In turn, an applied magnetic field H enhances the FM order thus reducing the spin scattering and producing the observed negative GMR. The localization scenario,[13] in which Mn oxides are modeled as systems with both DE off-diagonal spin disorder and nonmagnetic diagonal disorder, predicts a divergence of the electronic localization length $\xi(M)$ at some M-I phase transition. In terms of the spontaneous magnetization M, it means that for $M<M_0$ the system is in a highly resistive (insulatorlike) phase, while for $M>M_0$ the system is in a low resistive (metalliclike) state. Within this scenario, the Curie point T_C is defined through the spontaneous magnetization M as $M(T_C,H)=0$, while the M-I transition temperature T_{MI} is such that $M(T_{MI},H)=M_0$ (with M_0 being a fraction of the saturated magnetization M_s). Furthermore, the influence of magnetic fluctuations on electron-spin scattering near T_{MI} is expected to be rather important, for they can easily tip a subtle balance between magnetic and electronic processes in favor of either charge localization or delocalization.[15] Besides, the observable difference between the two critical temperatures (usually attributed to the quality

of a particular sample used[5–8]) is ascribed to the random nonmagnetic scattering which is highly responsible for the magnitude of the observable GMR.[13]

The substitution on the La site was found to modify the phase diagram through cation size effects leading toward either charge-ordered (CO) or AFM instability.[17] In particular, Y substitution is responsible for three major modifications of the parent manganite: (i) it changes the Mn^{4+} content,[18] (ii) lowers the FM Curie temperature T_C,[19] and (iii) weakens the system's robustness against strong AFM fluctuations (which are developed locally within the ordered FM matrix).[15] The latter is considered[15,20] to be the most probable cause for strong magnetic localization of spin polarized carriers (polarons). According to Jaime *et al.*,[20] even in the FM metallic state (just below T_C) there still remain significant indications of spin scattering, and as a result the collapse of large polarons in the FM state reduces the effective exchange coupling via the DE mechanism, causing a ''bootstrap'' destruction of FM and concomitant M-I transition.

On the other hand, in view of its carrier charge (and density) sensitive nature, thermopower (TEP) $S(T,H)$ measurements could complement the traditional MR data and be used as a tool for probing the field-induced delocalization of the carriers. Indeed, it was found[16] that the TEP in manganites is very sensitive to the Mn^{4+} content. In particular, for our composition [with $S(T,0)=23\ \mu$V/K] the fractional carrier content is expected to be $x\simeq 0.2$. Field behavior of the TEP in manganites was found to essentially depend on the temperature range and the sample's quality (thin films, single crystals, or ceramics).[5–8,16,19–21] For example, below T_C the TEP of $La_{2/3}Ca_{1/2}MnO_3$ thin films was found[20] to *increase* with an applied magnetic field as in nonsaturated FM metal while ceramics usually exhibit a *decrease* of $S(T,H)$ with increasing the field (at least at high temperatures).[21]

Studying the observable magneto-TEP $\Delta S(T,H)=S(T,H)-S(T,0)$ has already proved to be useful for providing important insights into different aspects of high-T_c superconductors in the mixed state.[22–24] Besides, magneto-TEP can be directly linked to the transport entropy change in applied magnetic field. The recently observed[14] giant magnetic entropy change in manganites (produced by the abrupt reduction of the magnetization and attributed to an anomalous thermal expansion just at the Curie point) gives another reason to utilize the magneto-TEP data in order to get an additional information as for the underlying transport mechanisms in these materials.

In the present paper we discuss some typical results for magneto-TEP measurements on a manganite sample $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ at $H=1$ T field for a wide temperature interval (ranging from 20–300 K). By approximating the true shape of the measured magneto-TEP in the vicinity of the peak temperature T^* by a linear triangle of the form $\Delta S(T,H)\simeq S_p(H)\pm B^{\pm}(H)(T^*-T)$, we observe that $B^-(H)\simeq 2B^+(H)$. In an attempt to account for the observed behavior of the magneto-TEP, we adopt the main ideas of the microscopic localization theory[13] and construct a phenomenological free-energy functional of Ginzburg-Landau (GL) type which describes the magnetic field and temperature behavior of the spontaneous magnetization in the presence of strong localization effects near T^*. Calculating the background and fluctuation contributions to the total magnetization and the transport entropy-induced magneto-TEP $\Delta S(T,H)$ within the GL theory, we obtain a simple relationship between T^* and the above two critical temperatures (T_C and T_{MI}). We find also that the observed ratio $B^-(H)/B^+(H)$ asymmetry is governed by a universal parameter $z=JS/M_sH_0$ where JS is the electron-spin exchange and M_sH_0 is the localization related magnetic energy. By comparing our data with the model predictions, we deduce estimates for some important model parameters such as the Curie point T_C, the localization length ξ_0, the critical magnetization M_0, and the exchange energy J, all in good agreement with the existing microscopic localization theories. In addition, to further test the validity of the model, we discuss the field-induced behavior of the TEP maximum S_m for a similar material studied recently by Jaime *et al.*[21]

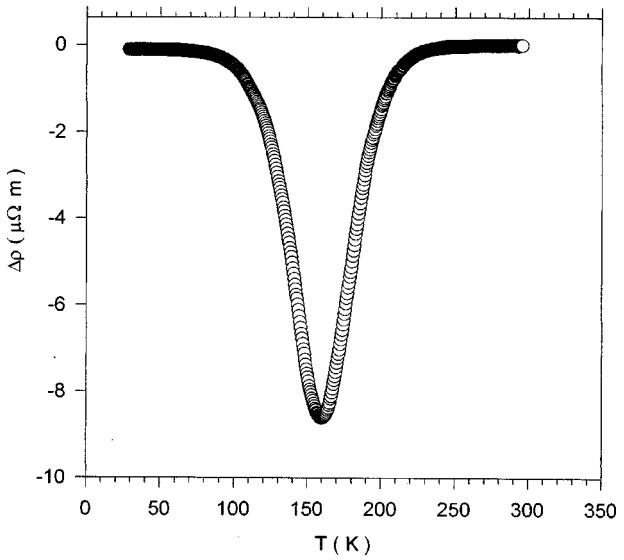

FIG. 1. Temperature behavior of the observed magnetoresistivity $\Delta\rho(T,H)=\rho(T,H)-\rho(T,0)$ in $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ at $H=1$ T.

II. EXPERIMENTAL RESULTS

$La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ samples were prepared from stoichiometric amounts of La_2O_3, Y_2O_3, $CaCO_3$, and MnO_2 powders. The mixture was heated in the air at 800 °C for 12 h to achieve the decarbonation. Then it was pressed at room temperature under 10^3 kG/cm^2 to obtain parallelipedic pellets. An annealing and sintering from 1350–800 °C was made slowly (during two days) to preserve the right phase stoichiometry. A small bar (length $l=10$ mm, cross section $\mathcal{S}=4$ mm^2) was cut from one pellet. The electrical resistivity $\rho(T,H)$ was measured using the conventional four-probe method. To avoid Joule and Peltier effects, a dc current $I=1$ mA was injected (as a one-second pulse) successively on both sides of the sample. The voltage drop V across the sample was measured with high accuracy by a KT256 nanovoltmeter. The magnetic field H of 1 T was applied normally to the current. Figure 1 presents the temperature dependence of the magnetoresistance (MR) $\Delta\rho(T,H)=\rho(T,H)-\rho(T,0)$ for a $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ sample at $H=1$ T field. As is seen, the negative MR $\Delta\rho(T,H)$ shows a peak (dip) at some temperature $T_0=160$ K (referred to as T_{MI}, in what

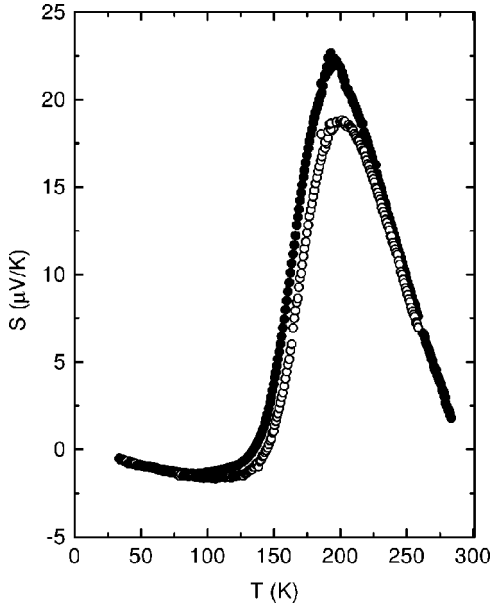

FIG. 2. Temperature behavior of the observed thermopower $S(T,H)$ in $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ at zero field (solid circles) and at $H=1$ T (open circles).

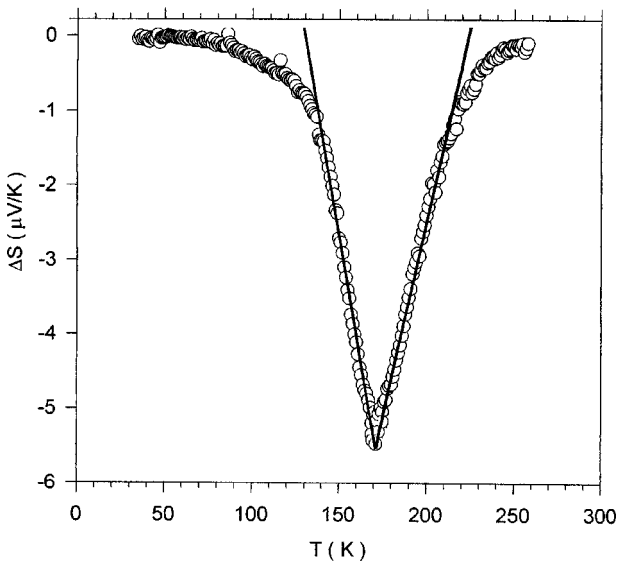

FIG. 3. Temperature behavior of the observed magneto-TEP $\Delta S(T,H)$ in $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ at $H=1$ T. The best fit to the data points according to Eq. (1) yields $S_p(H)=-5.49\pm0.01$ μV/K for the peak, and $B^-(H)=-0.14\pm0.01$ μV/K^2 and $B^+(H)=-0.08\pm0.01$ μV/K^2 for the slopes.

follows) where the GMR $\Delta\rho(T,H)/\rho(T,0)$ reaches 40%. The thermopower (TEP) S was measured using the differential method.[25] In order to generate a heat flow, a small heater film ($R=150$ Ω) was attached to one end of the sample. Two calibrated chromel-constantan thermocouples were used to measure the temperature difference between two points on the sample. The TEP $S(T,H)$ is deduced from the following equation: $S(T,H)=S_{Au}(T)-V_s(T,H)/\Delta T$, where $S_{Au}(T)$ is the TEP of the gold wires used to measure the voltage drop V_s at the hot junctions of both thermocouples. Figure 2 presents the temperature dependence of the observed TEP $S(T,H)$ for a $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ sample at zero and $H=1$ T field. Notice a maximum and a bell-like shape around $T=195$ K, typical for ceramics. As usual,[21] the applied field results in a decrease of the peak and its slight shift toward higher temperatures. As is seen, zero-field and field-induced contributions to the TEP merge both at low ($T<150$ K) and at high ($T>225$ K) temperatures marking the well-known scaling behavior for both temperature regions.[21] The corresponding magneto-TEP $\Delta S(T,H)=S(T,H)-S(T,0)$ for the same sample (at $H=1$ T) is shown in Fig. 3. Observe that it has an asymmetric Λ-like shape near some critical temperature $T^*>T_{MI}$ where it reaches its field-dependent peak (dip) value $S_p(H)$. Approximating the shape of the observed $\Delta S(T,H)$ by the asymmetric linear triangle of the form

$$\Delta S(T,H)\simeq S_p(H)\pm B^{\pm}(H)(T^*-T), \tag{1}$$

with positive slopes $B^-(H)$ and $B^+(H)$ defined for $T<T^*$ and $T>T^*$, respectively, we find (see Fig. 3) that $B^-(H)\simeq 2B^+(H)$ in the vicinity of T^*. Now, with all this information in mind, let us proceed to the interpretation of the experimental results.

III. DISCUSSION

A. Model

Since we are dealing with the magnetic-field induced changes of the TEP, it is reasonable to assume that the observed behavior can be attributed to the corresponding changes of transport magnetic entropy (and thus spontaneous magnetization) in the presence of strong electron-spin exchange and localization effects, near some critical temperature T^*. Later on, we will establish a simple (linear) relationship between the peak temperature T^* and the two critical temperatures T_C and T_{MI}, responsible respectively for PM-FM and M-I phase transitions. Based on the above considerations, we can write $\mathcal{F}=\mathcal{F}_M-\mathcal{F}_e$ for the balance of magnetic $\mathcal{F}_M$ and electronic $\mathcal{F}_e$ free energies participating in the transport processes under discussion. The observed magnetization M and the magneto-TEP behavior should result from the minimization of $\mathcal{F}$ (as, for example, is the case in superconductors where $\mathcal{F}$ measures the difference between the normal and condensate energies[22,23]). In our case, the above electronic contribution reads $\mathcal{F}_e=\mathcal{M}\mathcal{H}_e=\eta^2(n_eE_k+n_iV_{DE})$ and describes a coupling of spontaneous magnetization $\mathcal{M}=M_s\eta^2$ (where η is the order parameter and M_s the saturated magnetization) with (i) an effective DE energy $V_{DE}=-JS$ (where S is an effective spin on a Mn site, and J the exchange coupling constant), and (ii) the electronic (localization) energy $E_k(T,H)=\hbar^2/2m\xi^2(T,H)$ [where $\xi(T,H)$ is the localization length, and m an effective electron mass]; n_i and n_e stand for the number density of localized spins and conduction electrons, respectively. At the same time, the magnetic contribution $\mathcal{F}_M=\mathcal{M}(\mathcal{H}_{eff}-H)=M_s\eta^2(\gamma\eta^2-H)$ includes the effects due to the molecular field $\mathcal{H}_{eff}=\gamma\mathcal{M}/M_s$ (where $\gamma=3k_BT_C/2\mu_BS$ is the characteristic magnetic field with k_B the Boltzmann constant and μ_B the Bohr magneton) and an applied magnetic field H. After trivial rearrangements, the above functional $\mathcal{F}$ can be cast into a familiar GL-type form describing the second-order phase transition from PM (insulator) to FM (metal) state near T^*, namely

$$\mathcal{F}[\eta]=a\eta^2+\frac{\beta}{2}\eta^4-\zeta\eta^2. \tag{2}$$

Here $\zeta(H)=M_sH-n_iJS$ is the effective field-dependent chemical potential of quasiparticles; $a(T,H)=\alpha(H)(T-T^*)$ with $\alpha(H)=n_e\hbar^2/2m\xi_0^2(H)T^*$; $\beta=2\gamma M_s$, and we used the conventional expression $\xi^2(T,H)=\xi_0^2(H)/(1-T/T^*)$ for the correlation length. It is worthwhile to mention that a somewhat similar mean-field model has recently been suggested by Jaime *et al.*[21] to reproduce the essential features of a microscopic model and provide a comparison with their experimental data on the magnetization, susceptibility, and thermoelectric coefficient. Besides, to account for the field-induced localization effects, we assume after Sheng *et al.*[13] that $\xi_0(H)/\xi_0(0)=1/(1-H/H_0)$ with $H_0\simeq\gamma\propto M_0$.

B. Mean value of the magneto-TEP: $\Delta S_{av}(T,H)$

Given our previous experience with high-T_c superconductors, we can readily present the observed magneto-TEP in a two-term contribution form:[23]

$$\Delta S(T,H)=\Delta S_{av}(T,H)+\Delta S_{fl}(T,H), \tag{3}$$

where the average term $\Delta S_{av}(T,H)$ is nonzero only below T^* while the fluctuation term $\Delta S_{fl}(T,H)$ should contribute to the observable $\Delta S(T,H)$ both above and below T^*. In what follows, we shall discuss these two contributions separately within a mean-field theory approximation for GMR materials.

As usual, the equilibrium state of such a system is determined from the minimum-energy condition $\partial\mathcal{F}/\partial\eta=0$ which yields for $T<T^*$:

$$\eta_0^2=\frac{\alpha(H)(T^*-T)+\zeta(H)}{\beta}. \tag{4}$$

Substituting η_0 into Eq. (2) we obtain for the average free-energy density

$$\Omega_{av}(T,H)\equiv\mathcal{F}[\eta_0]=-\frac{[\alpha(H)(T^*-T)+\zeta(H)]^2}{2\beta}. \tag{5}$$

In turn, the magneto-TEP $\Delta S(T,H)$ can be related to the corresponding difference of transport entropies[22–24] $\Delta\sigma_{av}\equiv-\partial\Delta\Omega_{av}/\partial T$ as $\Delta S_{av}(T,H)=\Delta\sigma_{av}(T,H)/en_e$, where e and n_e are the charge and the number density of free carriers. Finally the mean value of the magneto-TEP reads (below T^*)

$$\Delta S_{av}(T,H)=S_{p,av}(H)-B_{av}(H)(T^*-T), \tag{6}$$

with

$$S_{p,av}(H)=-\frac{\alpha(0)\Delta\zeta(H)}{e\beta n_e}(1+z), \tag{7}$$

and

$$B_{av}(H)=\frac{2\alpha(0)\Delta\alpha(H)}{e\beta n_e}, \tag{8}$$

where $z=\Delta\alpha(H)\zeta(0)/\alpha(0)\Delta\zeta(H)$ with $\Delta\alpha(H)=\alpha(H)-\alpha(0)$ and $\Delta\zeta(H)=\zeta(H)-\zeta(0)$.

C. Mean-field Gaussian fluctuations of the magneto-TEP: $\Delta S_{fl}(T,H)$

The influence of fluctuations (both Gaussian and critical) on transport properties of high-T_c superconductors (including TEP, electrical, and thermal conductivity) was extensively studied and is very well documented (see, e.g., Refs. 26–32 and further references therein). In particular, it was found that the fluctuation-induced behavior may extend to temperatures more than 10 K higher than the critical temperature T_c. As for manganites, the fluctuation effects in these materials appear to be much less explored.[33] Nonetheless, according to the interpretation of the observed magneto-TEP we adopt in the present paper, influence of magnetic fluctuations on electron-spin scattering near T^* should be rather important. So, it seems appropriate to take a closer look at the region near T^* to discuss the fluctuations of the magneto-TEP $\Delta S_{fl}(T,H)$. Recall that according to the textbook theory of Gaussian fluctuations,[34] the fluctuations of any observable (such as heat capacity, magnetization, etc.) which is conjugated to the order parameter η can be presented in terms of the statistical average of the fluctuation amplitude $\langle(\delta\eta)^2\rangle$ with $\delta\eta=\eta-\eta_0$. Then the TEP above $(+)$ and below $(-)$ the critical point T^* have the form of

$$S_{fl}^{\pm}(T,H)=A\langle(\delta\eta)^2\rangle_{\pm}=\frac{A}{Z}\int d\eta(\delta\eta)^2e^{-\Sigma[\eta]}, \tag{9}$$

where $Z=\int d\eta e^{-\Sigma[\eta]}$ is the partition function with $\Sigma[\eta]\equiv(\mathcal{F}[\eta]-\mathcal{F}[\eta_0])/k_BT$, and A is a coefficient to be defined below. Expanding the free-energy density functional $\mathcal{F}[\eta]$,

$$\mathcal{F}[\eta]\approx\mathcal{F}[\eta_0]+\frac{1}{2}\left[\frac{\partial^2\mathcal{F}}{\partial\eta^2}\right]_{\eta=\eta_0}(\delta\eta)^2, \tag{10}$$

around the mean value of the order parameter η_0, which is defined as a stable solution of equation $\partial\mathcal{F}/\partial\eta=0$ we can explicitly calculate the Gaussian integrals. Due to the fact that η_0 is given by Eq. (4) below T^* and vanishes at $T\geqslant T^*$, we obtain finally

$$S_{fl}^{-}(T,H)=\frac{Ak_BT^*}{4\alpha(H)(T^*-T)+4\zeta(H)}, \quad T\leqslant T^* \tag{11}$$

and

$$S_{fl}^{+}(T,H)=\frac{Ak_BT^*}{2\alpha(H)(T-T^*)-2\zeta(H)}, \quad T\geqslant T^*. \tag{12}$$

As we shall see below, for the experimental range of parameters under discussion, $\zeta(H)/\alpha(H)\gg|T^*-T|$. Hence with a good accuracy we can linearize Eqs. (11) and (12) and obtain for the fluctuation contribution to the magneto-TEP:

$$\Delta S_{fl}^{\pm}(T,H)\simeq S_{p,fl}^{\pm}(H)\pm B_{fl}^{\pm}(H)(T^*-T), \tag{13}$$

where

$$S_{p,fl}^{-}(H)=-\frac{Ak_BT^*\Delta\zeta(H)}{4\zeta^2(0)}, \quad S_{p,fl}^{+}(H)=-2S_{p,fl}^{-}(H), \tag{14}$$

and

$$B_{fl}^{-}(H)=-\frac{Ak_BT^*\Delta\alpha(H)}{4\zeta^2(0)}\left(1-\frac{2}{z}\right),\quad B_{fl}^{+}(H)=-2B_{fl}^{-}(H). \tag{15}$$

Furthermore, it is quite reasonable to assume that $S_p^-=S_p^+\equiv S_p$, where the magneto-TEP peak (dip) values are defined as follows, $S_p^-=S_{p,av}+S_{p,fl}^-$ and $S_p^+=S_{p,fl}^+$. The above equations allow us to fix the arbitrary parameter A yielding $A=-4\zeta^2(0)\alpha(0)(1+z)/3ek_BT^*\beta n_e$. This in turn leads to the following expressions for the fluctuation contribution to peaks and slopes through their average counterparts [see Eqs. (7) and (8)]: $S_{p,fl}^+(H)=(2/3)S_{p,av}(H)$, $S_{p,fl}^-(H)=-(1/3)S_{p,av}(H)$, $B_{fl}^-(H)=-(1/2)B_{av}(H)$, and $B_{fl}^+(H)=B_{av}(H)$. Finally, the total contribution to the observable magneto-TEP reads [cf. Eq. (1)]

$$\Delta S(T,H)=S_p(H)\pm B^{\pm}(H)(T^*-T), \tag{16}$$

where

$$S_p(H)=-\frac{(1+z)E_k^0}{3eT^*}\left(\frac{H}{H_0}\right), \tag{17}$$

$$B^+(H)\equiv B_{fl}^+(H)=\left(\frac{n_e}{n_i}\right)\frac{(z-2)E_k^0}{JST^*}S_p(H), \tag{18}$$

and

$$B^-(H)\equiv B_{av}(H)+B_{fl}^-(H)=\left[\frac{3z}{(z+1)(z-2)}-\frac{1}{2}\right]B^+(H). \tag{19}$$

Here $E_k^0=\hbar^2/2m\xi_0^2(0)$, and $z=n_iJS/M_sH_0$. Notice that within our model the asymmetry of slopes ratio $B^-(H)/B^+(H)$ originates from the balance of the exchange n_iJS and localization induced magnetic M_sH_0 energies.

D. Magnetization and the critical temperatures

Before turning to the comparison of our theoretical findings with the experimental data, let us discuss the critical temperatures which control the magnetic (T_C) and carrier localization metal-insulator (T_{MI}) phase transitions. According to the adopted model, these two temperatures are defined through the spontaneous magnetization $M=M_{av}+M_{fl}^-$ as follows: $M(T_C)=0$ and $M(T_{MI})=M_0$. Here $M_0\propto H_0$ is the critical magnetization at which the zero-temperature localization length $\xi_0(H)=\xi_0(0)(1-H/H_0)^{-1}\propto(1-M/M_0)^{-1}\to\infty$ marking the M-I phase transition. According to Sec. III, the average magnetization reads $M_{av}(T)\equiv\mathcal{M}(\eta_0)=M_s\eta_0^2(T)$, where $M_s=n_i\mu_B$ is the saturated magnetization, and the equilibrium order parameter $\eta_0(T)$ is defined by Eq. (4). Now, for the self-consistency of our approach, we need to find the fluctuation contributions to the magnetization as well. Following the lines of the previous section, we obtain

$$M_{fl}^-(T,H)=\frac{Ck_BT^*}{4\alpha(H)(T^*-T)+4\zeta(H)},\quad T\leqslant T^* \tag{20}$$

and

$$M_{fl}^+(T,H)=\frac{Ck_BT^*}{2\alpha(H)(T-T^*)-2\zeta(H)},\quad T\geqslant T^*. \tag{21}$$

As usual, to fix the constant C, we assume that $M(T^*)=M^+(T^*)$, where $M^+=M_{fl}^+$ is the magnetization above T^*. As a result, we obtain $C=-4M_s\zeta^2/3k_B\beta T^*$ which leads to the following expression for the total magnetization below T^*:

$$M=M_{av}+M_{fl}^-=M_s\left(\eta_0^2-\frac{\zeta^2}{3\beta^2\eta_0^2}\right), \tag{22}$$

with ζ, β, and η_0 defined earlier. Given the above definitions, the two critical temperatures are related to each other and to the magneto-TEP peak temperature T^* within our model as follows:

$$T_{MI}=\left(1-\frac{2M_0H_0}{n_eE_k^0-n_iJS}\right)T_C, \tag{23}$$

with

$$T_C=\left(1+\frac{yn_iJS}{n_eE_k^0}\right)T^*,\quad y=1-\frac{1}{\sqrt{3}}. \tag{24}$$

Let us compare now the obtained theoretical expressions with our magneto-TEP experimental data on $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ (see Fig. 3). First, by comparing the ratios $(B^-(H)/B^+(H))_{exp}$ and $(B^-(H)/B^+(H))_{theor}$, we obtain $z\simeq 3$ for the slopes asymmetry parameter leading to $JS=3\mu_BH_0$. Then, using Eq. (18), B_{exp}^+, $T^*=170$ K, and just obtained z, we get $E_k^0/JS=2.5(n_i/n_e)$ which in turn brings about $T_C=195$ K for the Curie temperature (this value falls into the reported range of the FM transition temperatures for this class of manganites[5–8]). Using this temperature and assuming $S=2$ for an effective Mn spin, we can estimate the value of the exchange energy J (via the mean-field expression for the critical field $H_0=3k_BT_C/2S\mu_B$). The result is: $JS=40$ meV, which agrees with other reported estimates of this parameter.[11] Next, using[16] $n_e/n_i=0.7/3$ for an estimate of the free-to-localized carrier number density ratio in $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$ (which leads to the saturated magnetization $M_s=n_i\mu_B\simeq 4n_e\mu_B$), Eq. (17) yields $\xi_0\simeq 5$ Å for the estimate of the localization length[5,13,19,20] (using a free-electron mass m_e for m). Besides, from Eq. (23) we immediately get a simple relationship between the two critical temperatures, $T_{MI}/T_C=1-4M_0/9M_s$, which allows us to estimate the critical magnetization M_0. Using $T_{MI}=160$ K (deduced from the GMR data on the same sample as a peak temperature, see Fig. 1), we obtain $M_0=0.4M_s$, in a good agreement with the localization theory prediction.[13] It is also worth noting that the found localization energy E_k^0 is of the order of the Fermi energy E_F, as expected for manganites.[11] To conclude with the estimates, we note that $\zeta(H)T^*/\alpha(H)\simeq 1$ which *a posteriori* justifies the use of the linearized Eq. (13) for the fluctuation region $|1-T/T^*|\ll 1$. As is seen in Fig. 3, this criterion is well met in our case.

Finally, to put our model predictions [in particular, the field-dependent behavior of the TEP given by Eqs. (16) and (17)] to test, in Fig. 4 we present the experimental data ob-

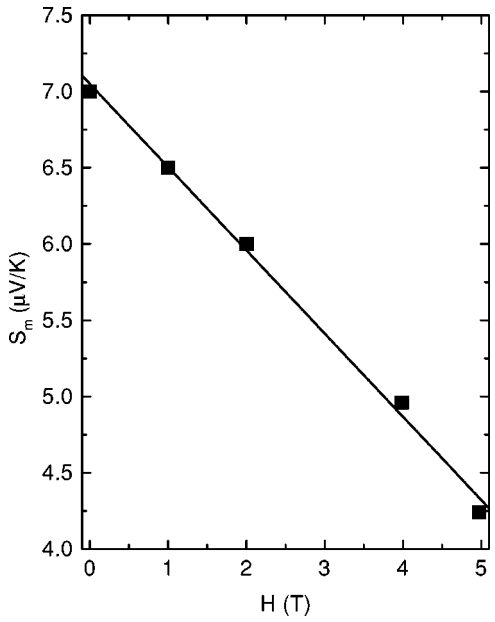

FIG. 4. Field dependence of the TEP maximum S_m calculated according to Eq. (25). The experimental points are deduced from the TEP data (Ref. 21) on $La_{0.67}Ca_{0.33}MnO_3$.

tained in Ref. 21 for a similar material. As is seen, with a good accuracy the data points follow a linear field dependence (straight line), in agreement with the theory predicting [see Eqs. (16) and (17)]

$$S_m(H) = S_m(0)\left(1 - \frac{H}{H_m}\right) \tag{25}$$

for the maximum of the TEP $S_m(H) \equiv S(T^*,H)$. Here, $H_m = [3eT^* S_m(0)/(1+z)E_k^0]H_0$ and we neglected a small field dependence of the peak temperature T^*.[21] The linear fit according to Eq. (25) yields $H_m = 14$ T for a characteristic field. In turn, using the above-obtained estimates for H_0, z, and E_k^0 along with T^* and $S_m(0)$, we get $H_m \simeq 0.07 H_0$ which leads to the expected value of the Curie field $H_0 = 3k_B T_C/2S\mu_B \simeq 200T$.

IV. CONCLUSION

To account for the observed Λ-like shape of the temperature dependence of the magneto-TEP $\Delta S(T,H)$ in $La_{0.6}Y_{0.1}Ca_{0.3}MnO_3$, exhibiting a field-dependent peak at some temperature T^* (lying in between the charge carrier localization temperature T_{MI} where the observed negative magnetoresistivity has a minimum, and magnetic transition temperature T_C which marks the occurence of the spontaneous magnetization), we adopted the ideas of the localization model and introduced a free-energy functional of Ginzburg-Landau (GL)-type describing the phase transition from paramagnetic (insulator) to ferromagnetic (metal) state near T^*. Calculating both average and fluctuation contributions to the total magnetization and magneto-TEP within the GL theory, we were able to successfully fit the data and estimate some important model parameters (including the metal-insulator T_{MI} and magnetic T_C transition temperatures, localization length ξ_0, electron-spin exchange coupling constant J, and the saturated magnetization M_s, all in a reasonable agreement with existing microscopic theories. The Gaussian fluctuations both above and below T^* are found to substantially contribute to the peak value $S_p(H) \equiv \Delta S(T^*,H)$ of the observed magneto-TEP, amounting to 67 and 33 %, respectively.

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