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Evidence for novel polaronic Fermi-liquid in doped oxides

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Abstract

The modern multi-polaron theory predicts a *polaronic* Fermi-liquid state, but such a state has not been unambigously confirmed by experiment so far. Here we report theoretical and experimental studies of the isotope effects on the *low temperature kinetic* properties of the doped ferromagnetic manganites. Our results provide clear evidence for a novel polaronic Fermi-liquid state in doped oxides.

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Over the last decades, the very interesting phenomena such as high-temperature superconductivity and colossal magnetoresistance have been discovered in several doped oxides (e.g., cuprates [1], bismuthates [2], and manganites [3,4]). These doped oxides are characterized by significant carrier densities ($\geq 10^{21}$ cm^{-3}) and low mobility of the order or even less than the Mott-Ioffe-Regel limit ($ea^2/\hbar \sim 1$ cm ^{2}/Vs, where a is the lattice constant) at room temperature. The very nature of the low-temperature 'metallic' state of these materials can be hardly understood within the framework of the canonical theory of metals. However, it has been known for many decades starting from the pioneering work by Landau [5] that the local lattice deformation can transform electrons into 'self-trapped' polarons in doped ionic insulators. Further extensive theoretical studies (for review see [6,7] have shown that polarons behave like heavy particles, and can be mobile with metallic conduction at sufficiently low temperatures. Under certain conditions [8] they form a polaronic Fermi liquid with some properties being very different from ordinary metals. One of them is the effective mass of polarons. It is well known that electrons can change their mass in solids due to the interactions with ions, spins, and themselves. The renormalized (effective) mass of electrons is independent of the ion mass M in ordinary metals where the Migdal adiabatic approximation is believed to be valid. However, if the interactions between electrons and nuclear ions are strong and/or the adiabatic approximation is not applied electrons form polarons (quasiparticles dressed by lattice distortions), so their effective mass m^* will depend on M. This is because the polaron mass $m^* = m\exp(A/\omega)$ [9], where m is the band mass in the absence of the electron-phonon interaction, A is a constant, and ω is a characteristic phonon frequency which depends on the masses of ions. Hence, there is a large isotope effect on the carrier mass in polaronic metals, in contrast to the zero isotope effect in ordinary metals.

The total exponent of the isotope effect on m^* is defined as $\alpha_{m^*} = \sum -d\ln m^*/d\ln M_i$ (M_i is the mass of the ith atom in a unit cell). From this definition and the expression for the polaron mass m^* mentioned above, one readily finds

$$\alpha_{m^*} = -\frac{1}{2}\ln(m^*/m). \tag{1}$$

Interestingly, the same isotope exponent is predicted for the inter-site bound pairs of polarons (i.e., bipolarons) [9,10]. A large isotope effect on the effective mass of supercarriers has been observed in the high-temperature cuprate superconductors [11–13], which suggests that polarons and/or bipolarons should be involved in high-temperature superconductivity. Furthermore, in doped manganites, the polaronic nature of the carriers in the high-temperature paramagnetic insulating state has been shown by several independent experimental results such as the sign anomaly of the Hall effect [14], the Arrhenius behavior of the drift and Hall mobilities [14], the giant oxygen-isotope shift of the Curie temperature T_C [15], and the electron paramagnetic resonance linewidth which is proportional to the conductivity [16].

On the other hand, the very nature of the charge carriers in the low-temperature metallic state of doped manganites has not been understood. In a theory of colossal magnetoresistance for ferromagnetic manganites [17] polarons are considered as the carriers even in the low-temperature metallic state, while others [18,19] believe that polaronic effects are not important at low temperatures. In particular, it was shown that, in low T_C polycrystalline manganites (T_C <120K), the resistivity and T_C are extremely sensitive to the external pressure [19] and chemical pressure [20] while the thermopower is small and nearly pressure independent [19]. This unusual behavior in the low T_C materials was taken as evidence for a vibronic electronic state [19], or may be explained in terms of a percolative conduction mechanism [20]. In contrast, from isotope-effect studies [21], it was concluded that polaronic effects may be involved in the ferromagnetic metallic state of the low T_C compounds. Moreover, in high T_C materials, it is even less clear whether polaronic effects are important in the low temperature metallic state.

Theoretically it has been demonstrated that the coherent motion of small polarons can lead to metallic conduction at low temperatures [6,22,8]. However, there have been no convincing and clear experiments to confirm this theoretical prediction. In order to provide decisive evidence for the 'metallic' polaronic liquid in doped ferromagnetic manganites, it is essential to study the isotope effects on the low-temperature kinetic and thermodynamic properties. These properties are dominated by the impurity and electron-electron scatter-

ings, so the only source of the isotope effect might be the polaron mass dependence on the ion mass. For example, replacing ^{16}O with ^{18}O will increase the polaron mass, leading to the changes in the residual resistivity, and in other kinetic and thermodynamic properties. Here we provide clear evidence for the polaronic liquid in doped ferromagnetic manganites at low temperatures from theoretical and experimental studies of the isotope effects on kinetic and thermodynamic properties.

We first define and calculate three exponents: $\beta_C = d\ln C_{el}/d\ln m^*$, $\beta_\rho = d\ln \rho_o/d\ln m^*$, and $\beta_S = d\ln S/d\ln m^*$ in the polaronic liquid. Here C_{el} is the electronic specific-heat at low temperatures, ρ_o is the residual resistivity, and S is the low-temperature thermopower. The above definition is equivalent to $C_{el} \propto (m^*)^{\beta_C}$, $\rho_o \propto (m^*)^{\beta_\rho}$, and $S \propto (m^*)^{\beta_S}$. We assume that the Fermi level (chemical potential) lies above the mobility edge, so polarons propagate in the Bloch states as heavy fermions [22,8] scattered by the short-range impurity potential. Since doped holes in manganites mainly reside on the oxygen orbitals [23,24], their bands are almost one-dimensional (1D) due to a large anisotropy in the hopping integrals (i.e., $t_{pp\pi} = -(1/4)t_{pp\sigma}$ [6]). The polaronic mass enhancent factor could be larger along the directions with a much smaller hopping integral [25]. This will increase the anisotropy further so that the polaron bands become more 1D-like. Then the polaronic propagator, $G(k,E) = [E - k^2/2m^* - \Sigma(k,E)]^{-1}$ is found analytically with the self-energy, which is momentum (k)-independent in the non-crossing approximation,

$$\sigma(\epsilon) = \frac{\epsilon}{3} - \left(\frac{1-i3^{1/2}}{2}\right)\left[\frac{1}{16} + \frac{\epsilon^3}{27} + \left(\frac{1}{256} + \frac{\epsilon^3}{216}\right)^{1/2}\right]^{1/3} - \left(\frac{1+i3^{1/2}}{2}\right)\left[\frac{1}{16} + \frac{\epsilon^3}{27} - \left(\frac{1}{256} + \frac{\epsilon^3}{216}\right)^{1/2}\right]^{1/3}. \quad (2)$$

Here we introduce a dimensionless energy, $\epsilon = E/\epsilon_0$, and the self-energy $\sigma(\epsilon) = \Sigma(E)/\epsilon_0$ using the scattering rate $\epsilon_0 = (D^2 m^*/\hbar^2)^{1/3}$ as the energy unit. The constant D is the second moment of the Gaussian white-noise potential, $D = 2(n_{im}v^2/a^2)$, where n_{im} is the impurity density, and v is the coefficient of the δ-function impurity potential. This self-energy is almost exact above the mobility edge as shown with the exact solution for a

one-dimensional particle in a random white-noise potential [26].

The specific heat is proportional to the density of states (DOS) at the (dimensionless) Fermi level, $\mu = E_F/\epsilon_0$. Assuming three degenerate spin-polarized oxygen bands, we find

$$\frac{C_{el}}{T} = \frac{2\pi k_B^2}{\hbar a^2}\left(\frac{m^*}{\epsilon_0}\right)^{1/2} g(\mu) \tag{3}$$

with $g(\epsilon) = \Im\sigma(\epsilon)$. The residual conductivity ρ_o^{-1} and the low-temperature thermopower S are found with the Kubo formalism [27] as

$$\rho_o^{-1} = \frac{e^2}{2\pi^2 (m^*)^2}\int_{-\infty}^{\infty} dk k^2 \left[\Im G(k,\mu)\right]^2, \tag{4}$$

and

$$S = -\frac{\pi^2 k_B^2 T}{3e\epsilon_0}\frac{d\ln\rho_o}{d\mu}. \tag{5}$$

If there coexist electron-like and hole-like polarons, Eq. 5 must be multiplied by a factor f_S with $|f_S| \leq 1$. We would like to mention that Eq. 4 should be underestimated due to the fact that the bands are not strictly one-dimensional and that the transport relaxation rate is generally smaller than ϵ_0. Calculating the integral in Eq. 4 leads to

$$\rho_o^{-1} = \frac{e^2}{2a^2\pi(m^*\epsilon_0)^{1/2}} F(\mu), \tag{6}$$

where

$$F(\mu) = \frac{\tilde{\mu}[\tilde{\mu} + (\tilde{\mu}^2 + g^2)^{1/2}] + g^2/2}{g[(\tilde{\mu}^2 + g^2)(\tilde{\mu} + (\tilde{\mu}^2 + g^2)^{1/2})]^{1/2}}, \tag{7}$$

$\tilde{\mu} = \mu - \Re\sigma(\mu)$ and $g = g(\mu)$.

These expressions allow us to calculate the exponents: β_C, β_ρ, and β_S. The number of carriers does not depend on the isotope substitution [28], so that the chemical potential itself depends on the polaron mass with the exponent

$$\frac{d\mu}{d\ln m^*} = -\frac{2N}{3g}, \tag{8}$$

where $N = \int_{-\infty}^{\mu} g(\epsilon)d\epsilon$ is proportional to the total number of states under the Fermi level. Keeping that in mind we finally obtain

$$\beta_C = \frac{1}{3}\left(1 - \frac{2Ng'}{g^2}\right), \tag{9}$$

$$\beta_\rho = \frac{2}{3}\left(1 + \frac{NF'}{gF}\right), \tag{10}$$

and

$$\beta_S = \frac{1}{3}\left(\frac{2N(F'/F - F''/F')}{g} - 1\right). \tag{11}$$

The prime and the double primes mean the first and the second derivative with respect to μ. The numerical results for the exponents are shown in Fig. 1 as a function of the Fermi level E_F/ϵ_0. To compare with the experimentally measured oxygen-isotope isotope exponents one should multiply $\beta_C, \beta_\rho, \beta_S$ by the oxygen-isotope exponent $\alpha^O_{m^*}$. Because of the logarithmic dependence on the mass ratio (see Eq. 1), the magnitude of the exponent α_{m^*} is expected to be of the order of unity. At large filling, i.e., $\mu >> 1$, the theory predicts sizable isotope effects on all thermodynamic and kinetic properties with the exponents:

$$\beta_C = 1; \beta_\rho = 2; \beta_S = 1. \tag{12}$$

This is precisely what one expects from the simple Born approximation for the impurity scattering. The ratio remains the same for any dimension of the polaron spectrum in this doping regime. Interestingly, if the doping is not so high, $\mu \leq 1$, the exponent for the thermopower is very small (see Fig. 1), while the exponent for the residual resistivity remains larger than unity. For $\mu = 0.25$ and $\alpha^O_{m^*} \simeq \alpha_{m^*} \simeq 1.1$,

$$\beta_C \simeq 0.45; \beta_\rho \simeq 1.45; \beta_S \simeq -0.03, \tag{13}$$

and

$$\alpha^O_C \simeq -0.5; \alpha^O_\rho \simeq -1.6; \alpha^O_S \simeq 0.0, \tag{14}$$

i.e., negligible isotope effect on S, and more than 3 times lower isotope effect on C_{el} than on ρ_o. Applying the exact density of states in the random potential [26], we find even a

somewhat lower exponent β_C for $\mu \leq 0$ than that in Fig. 1. All these findings are in sharp contrast with those in ordinary metals where $\alpha_{m^*} = 0$, and there are no isotope effects on C_{el}, ρ_o and S.

The choice of $\mu = 0.25$ and $\alpha^O_{m^*} = -1.1$ is consistent with the experimental results for $Nd_{0.67}Sr_{0.33}MnO_3$. Using the measured bare plasma frequency $\hbar\Omega_p \simeq 3.3$ eV [29] and the total carrier density per cell of 0.33, we obtain the bare hopping integral $t = t_{pp\sigma} = \hbar^2/2a^2m$ = 0.9 eV, which is very close to the value (0.87 eV) calculated for La_2CuO_4 [30]. From Eq. 1 and $\alpha_{m^*} \simeq \alpha^O_{m^*} \simeq -1.1$, we get the effective hopping integral $t^* = \hbar^2/2a^2m^* \simeq 100$ meV. By setting $\mu = 0.25$, $t^* = 100$ meV, $\epsilon_0 = 27$ meV, we calculates $C_{el}/T = 26$ mJ/moleK^2 and ρ_o = 185 $\mu\Omega$ cm by using Eq. 3 and Eq. 6, respectively. These values are in remarkably good agreement with the measured values: $C_{el}/T = 25(3)$ mJ/moleK^2 [31], and $\rho_o \simeq 170$ $\mu\Omega$ cm for the best single crystals [32,33]. Furthermore, the value of ϵ_0 is in good agreement with the measured transport relaxation rate ($\sim$24 meV) for $La_{0.7}Ca_{0.3}MnO_3$ [34]. This justifies the choice of $\mu = 0.25$ and $\alpha^O_{m^*} = -1.1$ for $Nd_{0.67}Sr_{0.33}MnO_3$. For $La_{0.75}Ca_{0.25}MnO_3$, we calculate $C_{el}/T = 7.5$ mJ/moleK^2 and $\rho_o = 150$ $\mu\Omega$ cm by setting $\mu = 0.70$ and $\alpha^O_{m^*} = -0.7$. The measured ρ_o for $La_{0.75}Ca_{0.25}MnO_3$ is 120-150 $\mu\Omega$ cm (see Fig. 2a and discussion in Ref. [28]). From our preliminary specific-heat data for $La_{0.75}Ca_{0.25}MnO_3$, we estimate C_{el}/T $\simeq$ 7 mJ/moleK^2, which is slightly lower than the one ($\sim$8 mJ/moleK^2) for $La_{0.8}Ca_{0.2}MnO_3$ [35]. With $\mu = 0.70$ and $\alpha^O_{m^*} = -0.7$, we have

$$\alpha^O_C \simeq -0.5; \alpha^O_\rho \simeq -1.2; \alpha^O_S \simeq -0.17, \tag{15}$$

Experimentally, the oxygen-isotope effect on the intrinsic low-temperature resistivity has been studied [28] in high-quality thin films of $La_{0.75}Ca_{0.25}MnO_3$ and $Nd_{0.7}Sr_{0.3}MnO_3$. Our thin films were thick (2000 $\mathring{A}$), and were annealed at 940^{o}C for 10 hours, so no strains could remain in the films. We did back-exchange, and we knew precisely the oxygen-isotope enrichment (more details can be found in Ref. [28,37]). In Fig. 2 we plot the low-temperature resistivity of the oxygen-isotope exchanged films of (a) $La_{0.75}Ca_{0.25}MnO_3$; (b) $Nd_{0.7}Sr_{0.3}MnO_3$. In both cases, the residual resistivity ρ_o for the ^{18}O samples is larger than for the ^{16}O sam-

ples by about 15%. The observed large oxygen-isotope effect on ρ_o is consistent with the above theoretical prediction for polaronic metals. From the definition of the oxygen-isotope exponent: $\alpha_\rho^O = -d\ln\rho_o/d\ln M_O$ (M_O is the oxygen mass), we obtain $\alpha_\rho^O = -1.4(2)$ for $Nd_{0.7}Sr_{0.3}MnO_3$, and $\alpha_\rho^O = -1.2(2)$ for $La_{0.75}Ca_{0.25}MnO_3$. These values are in excellent agreement with Eq. 14 and Eq. 15, respectively.

The quantitative agreement between the resistivity data and the theory indicates that the low-temperature metallic state in doped manganites is a polaronic Fermi-liquid. To further confirm this, one should experimentally demonstrate that α_S^O is negligible and that α_C^O is rather small in these materials. Our preliminary results of the oxygen-isotope effect on the low-temperature specific heat in $La_{1-x}Ca_xMnO_3$ are consistent with Eq. 15 within the experimental uncertainty. The results will be published elsewhere. Here we show the data of the thermopower S at low temperatures for the isotope-exchanged $La_{1-x}Ca_xMnO_3$. The detailed measurement technique was described in Ref. [36]. Fig. 3a shows the thermopower S below 60 K for the ^{16}O and ^{18}O samples of $La_{1-x}Ca_xMnO_3$ with $x = 0.20$ and 0.25. The isotope-exchanged samples are the same as those reported in [37]. It is apparent that the oxygen-isotope effect on S is negligible below 20 K for both compositions ($\alpha_S^O \simeq 0.0$), as seen more clearly from Fig. 3b. This is in sharp contrast to the large oxygen-isotope shift of the Curie temperature T_C, which is 21 K for $x = 0.20$ and 14.5 K for $x = 0.25$ [37]. The fact that the slope of the linear part in S (see Fig. 3) is very sensitive to the doping or the carrier concentration and that the ^{16}O and ^{18}O samples have the same slope implies that the carrier concentrations for two isotope samples of a fixed x should be nearly the same, in agreement with the previous results [28,37]. The negligible α_S^O and the substantial $\alpha_\rho^O \sim$ -1.3 observed in these samples are in quantitative agreement with the theoretical prediction (Eq. 14 and Eq. 15). We are not aware of any other theoretical models which can quantitatively explain the experimental values of several physical quantities, such as electronic specific heat, residual resistivity, scattering rate, and three different oxygen-isotope exponents. Therefore, the present theoretical and experimental studies of the low-temperature kinetic and thermodynamic properties provide strong evidence for the existence of novel polaronic Fermi-liquid in

doped metallic oxides (e.g., magnetoresistive manganites and high-temperature superconductive cuprates).

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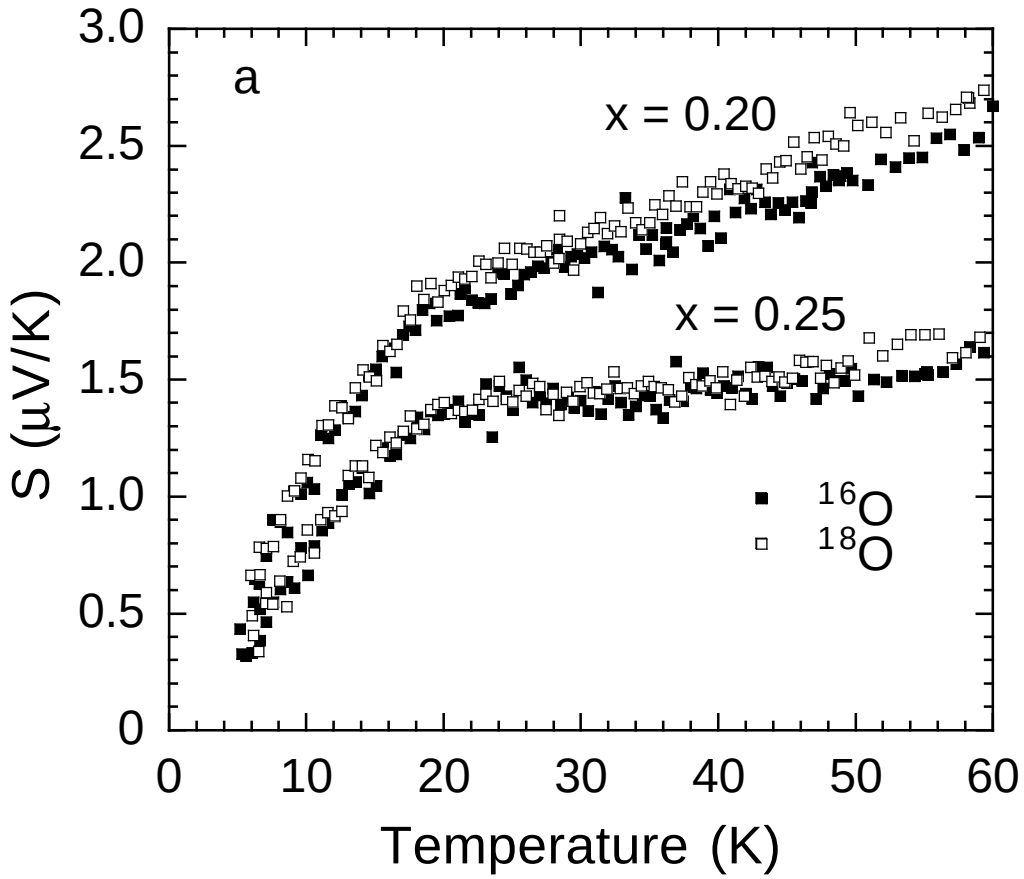
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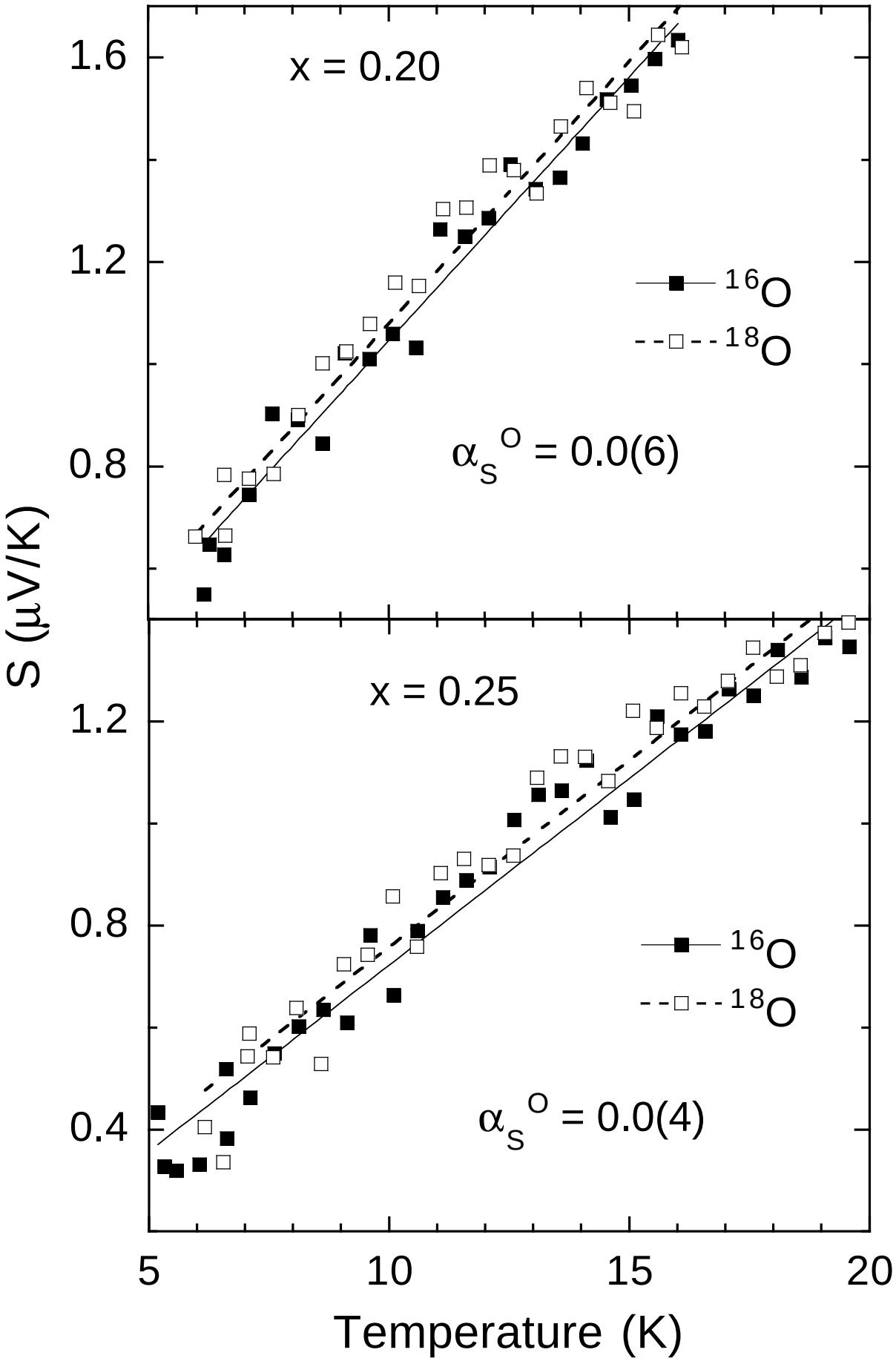
Fig.1. The numerical results for three exponents: β_ρ, β_C, and β_S as a function of the Fermi level $\mu \equiv E_F/\epsilon_0$. See text for the definition of the exponents.

Fig.2. The low-temperature resistivity of the oxygen-isotope exchanged films of (a) $La_{0.75}Ca_{0.25}MnO_3$; (b) $Nd_{0.7}Sr_{0.3}MnO_3$. In both cases, the residual resistivity ρ_o for the ^{18}O samples is larger than for the ^{16}O samples by 15% [28]. The oxygen-isotope exponent α_ρ^O was calculated from the definition: $\alpha_\rho^O = -d\ln\rho_o/d\ln M_O$ (M_O is the oxygen mass).

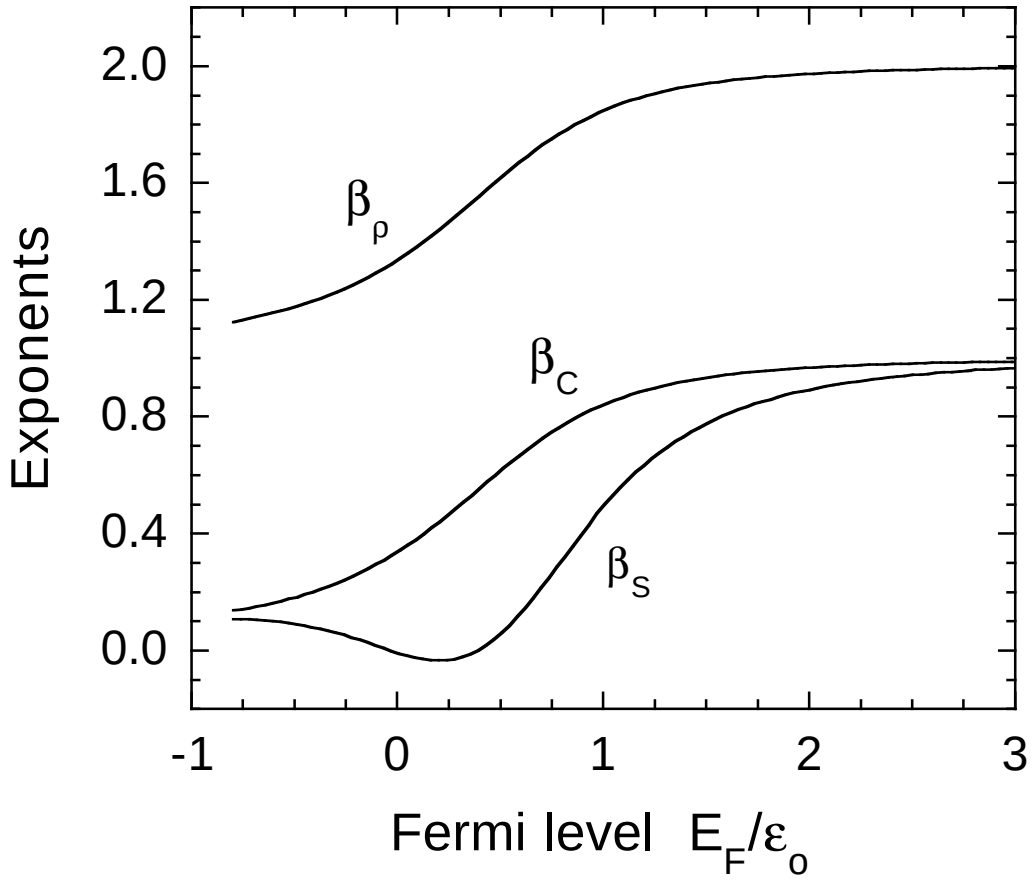
Fig.3. The temperature dependence of the thermopower S of the ^{16}O and ^{18}O samples of $La_{1-x}Ca_xMnO_3$ below 60 K (Fig. 3a) and below 20 K (Fig. 3b). The solid and dash lines in Fig. 3b are the fitted curves by $S = S_o + BT$ with a constraint that the absolute value of S_o should be less than the systematic measurement error 0.1 μV/K (Ref. [36]). The magnitudes of the exponent α_S^O were estimated from the values of the fitting parameter B.

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x = 0.20
^{16}O
^{18}O
α_S^O = 0.0(6)
x = 0.25
^{16}O
^{18}O
α_S^O = 0.0(4)
S (μV/K)
Temperature (K)
1.6
1.2
0.8
1.2
0.8
0.4
5
10
15
20

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