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Effects of electronic correlations on the thermoelectric power of the cuprates

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We show that important anomalous features of the normal-state thermoelectric power S of high-T_c materials can be understood as being caused by doping-dependent short-range antiferromagnetic correlations. The theory is based on the fluctuation-exchange approximation applied to the Hubbard model in the framework of the Kubo formalism. First, the characteristic maximum of S as a function of temperature can be explained by the anomalous momentum dependence of the single-particle scattering rate. Second, we discuss the role of the actual Fermi-surface shape for the occurrence of a sign change of S as a function of temperature and doping. [S0163-1829(97)51232-X]

The thermoelectric power (TEP), S, of the high-T_c materials in the normal state exhibits anomalous features[1–5] that are not well understood at present. For example, its temperature dependence shows a characteristic maximum that can be found in all optimally and underdoped cuprates, which is in contrast to the conventional linear-T behavior of a weakly correlated Fermi liquid. Furthermore, a better understanding of the doping dependence, which has been shown to be universal for a large class of high-T_c materials,[2] is needed.

Experimentally, the TEP is positive for the *underdoped* cuprates. It increases rapidly as a function of temperature, reaches a maximum $S^\star$ at a temperature $T^\star$ and falls off almost linearly with temperature. The size of the maximum $S^\star$ and the value of $T^\star$ decrease rapidly with increasing doping, while the slope for the fall off at $T>T^\star$ is almost independent of the doping concentration. The TEP of nearly *optimally doped* materials shows a similar behavior below room temperature, but the overall size of the TEP is reduced. Furthermore, the TEP for most of the optimally doped samples changes sign at approximately room temperature.[2] For *overdoped* samples, the behavior of the TEP is very different: It is negative and decreases linearly with increasing temperature. At present there are only two classes of high-T_c materials that do not follow this generic trend, namely $La_{2-x}Sr_xCuO_4$ (LSCO) and $YBa_2Cu_3O_{7-\delta}$ (YBCO). For LSCO the TEP remains positive in the overdoped regime, where it exhibits a maximum of decreasing height for increasing hole concentration,[5] whereas YBCO shows a negative TEP in the overdoped regime but with a positive slope at temperatures $T>T^\star$.[2,3] Recently, Bernhard and Tallon[3] presented strong experimental evidence that the chain contribution is responsible for this nongeneric TEP of YBCO while the contribution of the CuO_2 planes follows the generic behavior.

Theoretically, important open problems concerning the characteristic behavior of the TEP remain, although there have been several attempts[1] to explain this behavior, including the van Hove scenario[6,7] and phonon drag effects.[8] The most important open questions are the physical origin of the characteristic temperature scale $T^\star$ and of the doping dependence of the sign of S.

In this paper, we demonstrate that short-range antiferromagnetic (AF) correlations are responsible for salient features of the temperature and doping dependence of the TEP. In particular, we show that for underdoped systems the occurrence of a pseudogap in the single-particle excitation spectrum, and for optimally doped systems the pronounced increase of the scattering rate near the ''hot-spot'' regions [near $(\pi,0)$], are essential. As was proposed by Stojković *et al.*,[9] the latter phenomenon is also of importance for the description of the temperature dependence of the Hall coefficient. Furthermore, we discuss the influence of the shape of the Fermi surface of LSCO and YBCO and of the bilayer coupling.

We note that AF spin correlations have recently also been argued to be intimately related to the normal-state pseudogap in underdoped cuprates and to a sizable deformation of the quasiparticle band structure and Fermi surface as a function of doping.[10,11] These ideas have been corroborated with evidence from quantum-Monte-Carlo (QMC) simulations in a recent work by Preuss *et al.*[12]

The thermoelectric power S is determined by the relation between the electrical current $\mathbf{j}$ and the temperature gradient ∇T: $\mathbf{j}=\sigma_{\rm dc}(\mathbf{E}-S\nabla T)$, where $\sigma_{\rm dc}$ is the dc conductivity and $\mathbf{E}$ an applied electrical field. In order to account for the strong electronic correlations and the pronounced short-range AF correlations of the cuprates, we use the two-dimensional single-band Hubbard model with $\epsilon(\mathbf{k})=-2t(\cos k_x+\cos k_y)-4t'\cos k_x\cos k_y-2t''[\cos(2k_x)+\cos(2k_y)]$ for the microscopic calculation of S. We set $t=0.25$ eV and $U=4t$. The values of the longer-range transfer integrals will be given below. The transport coefficients $\sigma_{\rm dc}$ and S are calculated within the Kubo-formalism as

$$\sigma_{\rm dc}=e^2\int d\omega[-\partial f(\omega)/\partial\omega]\sigma(\omega), \tag{1}$$

$$S=-\frac{e}{\sigma_{\rm dc}T}\int d\omega[-\partial f(\omega)/\partial\omega]\omega\sigma(\omega), \tag{2}$$

with the differential conductivity

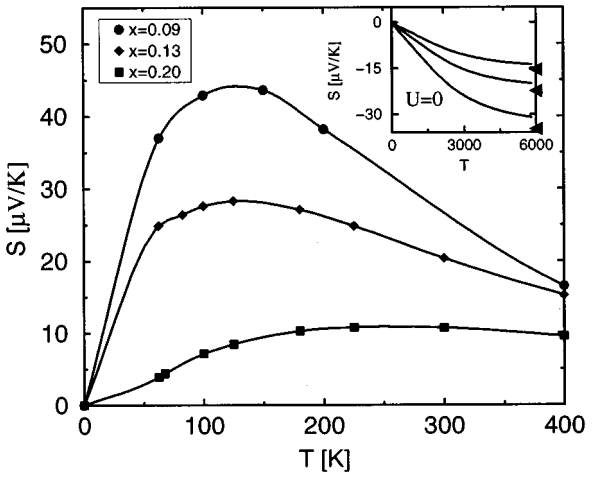

FIG. 1. Thermopower S for the LSCO system as a function of temperature for different doping concentrations. The inset shows S of the tight-binding model for the same doping values. The triangles indicate the high-temperature limit given by the generalized Heike formula.

$$\sigma(\omega)=\frac{2\pi}{N}\sum_{\mathbf{k}} v_{\mathbf{k},x}^2 A^2(\mathbf{k},\omega)\,. \tag{3}$$

Equations (1) and (2) are obtained following, for example, the derivation of Moreno and Coleman[13] and approximating the full vertex function by the velocity $v_{\mathbf{k},x}=\nabla_x\epsilon(\mathbf{k})$. Here e is the electronic charge, N the number of sites, $f(\omega)$ the Fermi function, and $A(\mathbf{k},\omega)=-(1/\pi)\mathrm{Im}G(\mathbf{k},\omega)$ the spectral function. The single-particle propagator $G(\mathbf{k},\omega)$ is calculated within the fluctuation-exchange approximation (FLEX).[14] This approximation includes the interaction of the electrons with charge and spin fluctuations. It includes feedback effects for low-energy excitations resulting from the AF correlations built up by these low-energy states. This makes FLEX a useful approach for the determination of (intrinsically low-energy) transport properties, although its application for high-energy features (occurrence of Hubbard bands) is limited.[15] We employ a real-frequency approach to this approximation, which directly yields $A(\mathbf{k},\omega)$.[16] The electron energy ω is counted from the chemical potential.

In Fig. 1, we show the TEP for the single-layer LSCO system as function of temperature for different electron occupation numbers $n=1-x$. Here LSCO is described by $t'=t''=0$ to ensure that the Fermi surface is closed around (0,0).[17] For all fillings, our theory yields the interesting result of a positive TEP which decreases with increasing hole concentration and shows pronounced maxima at about 150 K for the two lowest doping levels. For $x=0.20$, the maximum is very broad and shifts to higher temperatures. The behavior of the TEP is very similar to the experimental observation in LSCO.[5,18] Note that also the absolute values of the calculated TEP are in good agreement with experimental data.[5] For the dc resistivity we find similar results as in Ref. 19.

To understand the role of electronic correlations for the T dependence and sign of S and to relate them to previous theories, it is of interest to contrast them with the TEP of the $U=0$ system. Here S is obtained from a tight-binding model calculated in the relaxation-time approximation [the $U=0$ calculations along the lines of Eqs. (1) to (3) yield a vanishing TEP] with a momentum and frequency-independent relaxation time; see the inset of Fig. 1. In this approach, S is negative and proportional to T in the relevant temperature range ($T<600$ K). In the high-temperature limit, the TEP deviates from this linear behavior for $T\sim t$ and finally approaches the values given by a generalized high-temperature Heike's formula.[20] Such a $U=0$ theory, in which no details of the relaxation or scattering rate are included, still fails to describe experiments even when a more sophisticated phenomenological band structure with a pronounced flattening of the dispersion near the Fermi level[21] as found in photoemission experiments[22,23] is included. Thus, the strong qualitative disagreement between the above $U=0$ and our FLEX results for the interacting system implies that for an understanding of the experimental data it is necessary to take into account the momentum and frequency dependence of the scattering rate. In order to demonstrate in more detail the role of electronic correlation effects for $S(T)$ in our approach, we now show (i) that our results for the sign of S are caused by the anomalous $\mathbf{k}$ dependence of the scattering rate $1/\tau(\mathbf{k})=-\mathrm{Im}\Sigma(\mathbf{k},\omega=0)$ due to AF correlations, and (ii) that the temperature dependence of S is determined by the characteristic temperature scales of these correlations. It is instructive to consider the differential conductivity $\sigma(\omega)$ of Eq. (3), since the TEP, from Eq. (2), is a measure of the asymmetry of $\sigma(\omega)$ with respect to $\omega=0$ for energies of the order of k_BT. Thus the TEP is very sensitive to excitations of states which are close to but not directly at the Fermi surface.

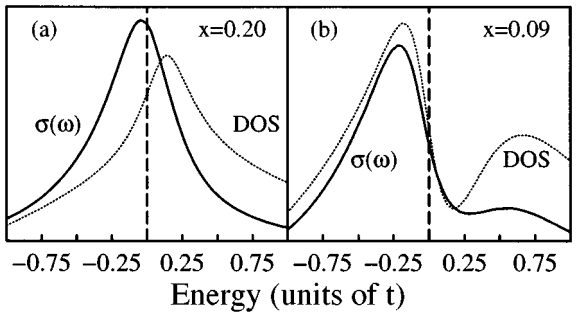

FIG. 2. Differential conductivity (solid lines) and density of states (dotted lines), both in arbitrary units, for two doping levels and $T=200$ K. All for $U=4t$.

In Fig. 2(a), we show $\sigma(\omega)$ and the density of states $\varrho(\omega)=\Sigma_{\mathbf{k}}A(\mathbf{k},\omega)/N$ for $x=0.20$. Although $\varrho(\omega)$ is peaked for $\omega>0$ (as in the uncorrelated case), the differential conductivity $\sigma(\omega)$ exhibits its maximum for $\omega<0$, yielding the positive value of S. This behavior can be understood if one takes into account that in Eq. (3) momentum states with small velocity $\mathbf{v}_{\mathbf{k}}$ and large line width (large scattering rates) are suppressed. Antiferromagnetic spin fluctuations lead to large scattering rates for momenta near $\mathbf{k}_F+\mathbf{Q}$ [$\mathbf{k}_F$ Fermi momentum, $\mathbf{Q}=(\pi,\pi)$] which, for LSCO, are everywhere outside of the Fermi surface [see Fig. 3(a)].

Thus the scattering rate of unoccupied states (holes) near the Fermi surface is larger than the scattering rate of the corresponding occupied states (electrons). This suppresses the contribution of the holes (at $\omega>0$) to $\sigma(\omega)$, which as a result has a maximum at $\omega<0$. This effect is illustrated in detail in Fig. 3(c), where we show the spectral function $A(\mathbf{k},\omega)$ in the vicinity of $\omega=0$. It can be clearly seen that

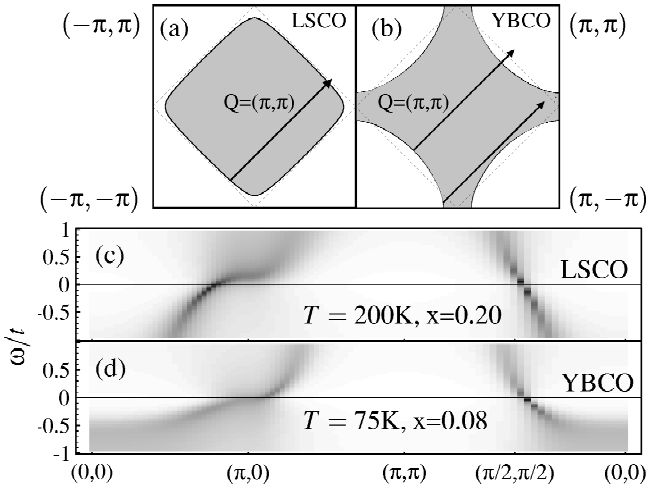

FIG. 3. Fermi surface, (a,b), and spectral function, (c,d), for the LSCO and YBCO parameters, respectively. In (c) and (d) dark (white) areas correspond to large (small) spectral weight.

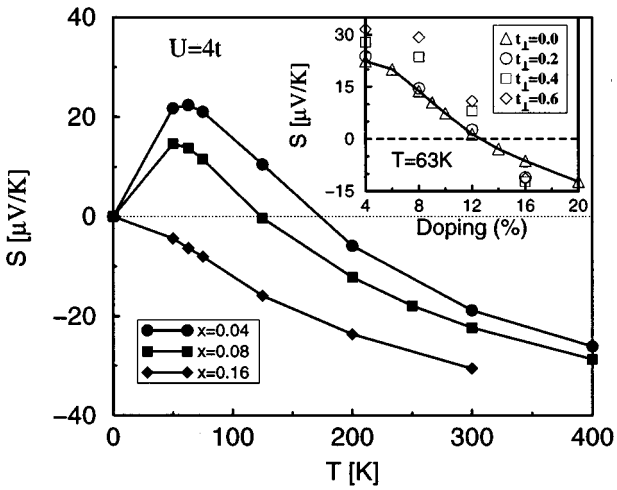

FIG. 4. TEP for the YBCO parameters as a function of temperature for different hole dopings (inset: doping dependence).

the spectral weight is more sharply peaked for occupied than for unoccupied states around $(\pi,0)$. The suppression of states near $(\pi,0)$ due to the low velocities near the van Hove singularity also contributes to the absolute magnitude of S but does not determine its sign and order of magnitude. This demonstrates that the thermoelectric power is determined by the combined momentum and energy dependence of the scattering rate, making transport for holes less coherent than for electrons, yielding $S>0$. The transition to conventional behavior occurs at temperatures for which the AF correlation length becomes smaller than the range of the hopping matrix (determined by t, t', and t''), such that no pronounced momentum dependencies in the scattering rate occur.[9,12] For decreasing doping concentration, the strength of the AF correlations increases, leading to the appearance of a pseudogap in the density of states (DOS) as a precursor of the AF phase transition.[11,12] The FLEX calculation for $U=4t$ (Ref. 11) yields results for the pseudogap qualitatively similar to those obtained in QMC simulations.[12] The density of states showing the pseudogap is presented in Fig. 2(b). The asymmetry in $\varrho(\omega)$ caused by the AF fluctuations is even more pronounced in $\sigma(\omega)$ such that these correlations cause in addition to the positive sign a pronounced increase of the magnitude of S. For temperatures smaller than the characteristic energy set by AF correlations, the TEP is linear in T but with a sign and magnitude as discussed above. When T becomes larger than this energy scale, new thermally activated states behave rather conventional, leading to a decrease of $S(T)$. The maximum $S^{\star}$ thus arises from the competition of these two effects and is consequently a thermal measure of the above-mentioned characteristic energy scale. Note, however, the weak doping dependence of $T^{\star}$ in our theory. We believe that this shortcoming is related to the difficulties in the theoretical description of the single-particle excitation spectrum of underdoped systems rather than to the approximate treatment of the transport problem. We believe that a better description of the detailed momentum dependence of the pseudogap, in agreement with recent angular resolved photoemission experiments[22] and QMC results[12] is essential here.

The inset in Fig. 4 displays our results for the doping dependence of the planar contribution to the TEP of a generic system, labelled YBCO. To generate a Fermi surface that is closed around (π,π) as observed for various high-T_c materials[22] we set $t'=-0.38t$ and $t''=-0.06t$ [see Fig. 3(b)]. In contrast to the LSCO system and in agreement with the general trends in the experiments, the thermopower changes its sign as a function of the doping concentration. In order to understand the origin of this behavior, we plot in Fig. 4 the TEP as a function of temperature for different doping values. As in LSCO, a maximum of the TEP occurs for small doping and small temperatures. However, for increasing T, the sign of S becomes negative. For larger doping, S decreases until it becomes negative at all temperatures. In agreement with the experimental observation,[3] we find that the negative slope of the TEP for large T is almost doping independent. These phenomena can also be understood in terms of the pronounced momentum dependence of the scattering rates caused by AF spin fluctuations. We find that for low doping, the scattering rates are maximal for momentum states near $\mathbf{k}_F+\mathbf{Q}$. However, as can be seen in Fig. 3(b), these states are solely outside the Fermi surface for $\mathbf{k}_F+\mathbf{Q}$ near $(\pi/2,\pi/2)$, while they are within the Fermi surface for $\mathbf{k}_F+\mathbf{Q}$ near $(\pi,0)$, in contrast to the LSCO system. If these two regions in momentum space were of equal importance for the suppression of near-Fermi surface states in $\sigma(\omega)$, they would cancel each other, leading to a negative S, as found for $U=0$. However, the transport properties are dominated by the quasiparticles near the diagonal of the Brillouin zone.[24] This can also be observed in the spectral function plot for YBCO in Fig. 3(d), in which the states near $(\pi/2,\pi/2)$ are much more coherent than the ones near $(\pi,0)$. For $\mathbf{k}_F$ near $(\pi/2,\pi/2)$, the holes near the Fermi surface are closer to $\mathbf{k}_F+\mathbf{Q}$ than the corresponding electron states and it follows that $S>0$ for sufficiently strong AF correlations. Finally, due to the more subtle competition of different regions in momentum space for the YBCO parameters, the weakening of AF correlations for increasing doping or temperature leads to the more rapid (as compared to LSCO) transition to negative values of the TEP, shown in Fig. 4. For a quantitative comparison of our results in Fig. 4 with experiment, it should be noted that the magnitude of S as well as the doping

level at which S changes its sign for low T are smaller than in experiment. One reason for this quantitative difference might be the neglect of the bilayer coupling in YBCO which was shown to enhance the in-plane AF correlations.[25,26] By taking bilayer coupling into account via an interlayer hopping $t_\perp$, we solved the FLEX equations and determined the TEP. Note, that $t_\perp = 0.4t$ is a reasonable value for the interlayer hopping in YBCO.[26] The results are also shown in the inset of Fig. 4 for different values of $t_\perp$ and demonstrate that indeed a considerable increase of the absolute magnitude of S occurs due to interlayer spin correlations.

Concerning the influence of the band-structure parametrization used for YBCO, it might be argued that the velocity $v_{\mathbf{k},\alpha}$ on the Fermi surface depends more strongly on the direction of $\mathbf{k}$ than for other parametrizations [e.g., $t' = -0.45t$ and $t'' = 0$ (Ref. 9)]. However this is not essential for our results, since the agreement with experiments would be even better if we were to neglect this $\mathbf{k}$ dependence, which suppresses S. We have used this parametrization since it yields better agreement of the calculated spin susceptibility with the spin-excitation spectrum deduced from NMR experiments.[27]

In summary, we have shown that the anomalous features of the in-plane thermopower can be understood as consequences of short-range AF correlations. The opening of a pseudogap and the anomalous momentum dependence of the single-particle scattering rate provide an explanation of the general trends of both temperature (maximum and sign change) and doping dependence of the in-plane thermopower. This demonstrates that the thermoelectric power is a sensitive probe of the anomalous nature of the low-energy excitations, caused by the antiferromagnetic correlations of the cuprates.

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