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ELECTRONIC PROPERTIES OF SOLID

Thermoelectric Power and Hall Effect in Correlated Metals and Doped Mott–Hubbard Insulators: DMFT Approximation

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Abstract—We present comparative theoretical investigation of thermoelectric power and Hall effect in the Hubbard model for correlated metal and Mott insulator (considered as prototype cuprate superconductor) for different concentrations of current carriers. Analysis is performed within standard DMFT approximation. For Mott insulator we consider the typical case of partial filling of the lower Hubbard band (hole doping). We calculate the dependence of thermopower on doping level and determine the critical concentration of carriers corresponding to sign change of thermopower. An anomalous dependence of thermopower on temperature is obtained significantly different from linear temperature dependence typical for the usual metals. The role of disorder scattering is analyzed on qualitative level. The comparison with similar studies of the Hall effect shows, that breaking of electron-hole symmetry leads to the appearance of the relatively large interval of band-fillings (close to the half-filling) where thermopower and Hall effects have different signs. We propose a certain scheme allowing to determine the number of carriers from ARPES data and perform semi-quantitative estimate of both thermopower and Hall coefficient using the usual DFT calculations of electronic spectrum.

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1. INTRODUCTION

The problem of doping dependence of thermopower and Hall effect in strongly correlated systems is known for a long time. It remains actual with respect to experimental studies of these effects in copper-oxide (cuprate) high-temperature superconductors at different doping levels. One of the basic concepts in cuprate physics is that these systems are strongly correlated and their metallic (and superconducting) state is realized via doping of the prototype phase of the Mott insulator, which in the simplest case can be described in the framework of the Hubbard model.

Most developed method of theoretical description of the Hubbard model for the wide range of parameters of this model remains the dynamical mean-field approach (DMFT) [1–3]. Systematic studies of concentration and temperature dependencies of the Hall effect was performed in our recent papers [4, 5]. Similar papers on concentration and temperature dependencies of the empower are practically absent. This paper is devoted to the study of this problem in comparison with previous results on the Hall effect.

2. THERMOPOWER AND HALL COEFFICIENT. GENERAL RELATIONS

As noted above the general method to study the Hubbard model is the dynamical mean-field theory (DMFT) [1–3], which gives an exact description of the system in the limit of infinite dimensions (lattice with an infinite number of nearest neighbors). Going beyond this approximation [6, 7] is usually much more complicated. The aim of the present work is the systematic study of concentration and temperature dependence of thermopower at different dopings of the lower Hubbard band within the standard DMFT approximation. We also compare in detail the results for thermopower with similar results obtained in our previous papers using the similar approach [4, 5].

Anticipating the possible comparison with experimental data for cuprates in the following we consider the two-dimensional model of electronic spectrum in tight-binding approximation:

$$\varepsilon(\mathbf{p}) = -2t(\cos(p_x a) + \cos(p_y a)) - 4t'\cos(p_x a)\cos(p_y a), \quad (1)$$

where a is (square) lattice parameter.

For this two-dimensional model in the following we shall consider a number of typical cases:

(1) spectrum with hopping only between nearest neighbors ($t' = 0$) and complete electron-hole symmetry,

(2) spectrum with $t'/t = -0.25$ qualitatively corresponding to electron in systems like LSCO,

(3) spectrum with $t'/t = -0.4$, qualitatively corresponding to situation observed in YBCO.

Thermopower is determined by the following expression [1, 8, 9]:

$$S = -\frac{k_B}{e}\frac{1}{T}\frac{\int_{-\infty}^{\infty} d\varepsilon \varepsilon \tau(\varepsilon)\left(-\frac{\partial f(\varepsilon)}{\partial \varepsilon}\right)}{\int_{-\infty}^{\infty} d\varepsilon \tau(\varepsilon)\left(-\frac{\partial f(\varepsilon)}{\partial \varepsilon}\right)}, \quad (2)$$

where k_B is Boltzmann's constant (in the following we write temperature in the units of energy assuming $k_B = 1$), e is electronic charge, $f(\varepsilon) = (e^{\frac{\varepsilon}{T}} + 1)^{-1}$— Fermi distribution. Below the values of thermopower are shown in units of $\frac{k_B}{e} \approx 86\left[\frac{\mu V}{K}\right]$.

Relaxation parameter introduced in Eq. (2) is given by:

$$\tau(\varepsilon) = \sum_{\mathbf{p}\sigma}\left(\frac{\partial \varepsilon(\mathbf{p})}{\partial p_x}\right)^2 A^2(\mathbf{p}\varepsilon) \quad (3)$$

and is completely determined by Green's function $G(\mathbf{p}\varepsilon)$ spectral density:

$$A(\mathbf{p}\varepsilon) = -\frac{1}{\pi}\mathrm{Im}\, G^R(\mathbf{p}\varepsilon). \quad (4)$$

In standard [1–3] self-energy of Green's function is local, i.e. independent of momentum. Due to this locality both the usual and Hall conductivities are also completely determined by the spectral density $A(\mathbf{p}\varepsilon)$. The usual (diagonal) static conductivity is given by [1]:

$$\sigma_{xx} = \frac{\pi e^2}{2\hbar}\int_{-\infty}^{\infty} d\varepsilon\left(-\frac{\partial f}{\partial \varepsilon}\right)\tau(\varepsilon), \quad (5)$$

while Hall (non-diagonal) conductivity determined by:

$$\sigma_{xy}^H = -\frac{2\pi^2 e^3 a^2 H}{3\hbar^2}\int_{-\infty}^{\infty} d\varepsilon\left(-\frac{\partial f}{\partial \varepsilon}\right)\tau_H(\varepsilon), \quad (6)$$

where H is the magnetic field along z-axis. Here we also introduced Hall relaxation parameter:

$$\tau_H = \sum_{\mathbf{p}\sigma}\left(\frac{\partial \varepsilon(\mathbf{p})}{\partial p_x}\right)^2 \frac{\partial^2 \varepsilon(\mathbf{p})}{\partial p_y^2} A^3(\mathbf{p}\varepsilon). \quad (7)$$

Thus, the Hall coefficient is:

$$R_H = \frac{\sigma_{xy}^H}{H\sigma_{xx}^2} \quad (8)$$

and it is also completely determined by spectral density $A(\mathbf{p}\varepsilon)$, which we shall calculate within DMFT. Effective single Anderson impurity model of DMFT in this work, as in [4, 5], will be solved using the numerical renormalization group approach (NRG) (NRG) [11].

Consider the case of low temperatures ($T \ll E_F$). For any function $\Phi(\varepsilon)$ (if integral is converges) we have:

$$\int_{-\infty}^{\infty} d\varepsilon \Phi(\varepsilon)\left(-\frac{\partial f}{\partial \varepsilon}\right) = \Phi(0) + \frac{\pi^2}{6}T^2\left(\frac{\partial^2 \Phi}{\partial \varepsilon^2}\right)_{\varepsilon=0} + \ldots \quad (9)$$

Then for diagonal conductivity from Eq. (5) we get:

$$\sigma_{xx} = \frac{e^2}{\hbar}\frac{\pi}{2}\tau(0), \quad (10)$$

and thermopower at low temperatures is given by:

$$S = -\frac{k_B}{e}\frac{\pi^2}{3}T\left.\frac{\frac{d\tau(\varepsilon)}{d\varepsilon}}{\tau(\varepsilon)}\right|_{\varepsilon=0} = -\frac{k_B}{e}\frac{\pi^2}{3}T\left.\frac{d\ln\tau(\varepsilon)}{d\varepsilon}\right|_{\varepsilon=0}, \quad (11)$$

so that from Eqs. (11) and (10) we immediately obtain Seebeck's expression [8, 10]:

$$S = -\frac{k_B}{e}\frac{\pi^2}{3}T\frac{d\ln\sigma_{xx}(\mu)}{d\mu}, \quad (12)$$

where μ is the chemical potential (from which we always count the energy ε).

Thus at low T the absolute value of thermopower linearly increases with the growth of T, while the sign of S is completely determined by the sign of $\left.\frac{d\tau(\varepsilon)}{d\varepsilon}\right|_{\varepsilon=0}$ or $\frac{d\sigma_{xx}(\mu)}{d\mu}$. In the following we shall be interested in dependence on band-filling $n = n_\uparrow = n_\downarrow = \int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)N(\varepsilon)$ (we shall consider only the paramagnetic state), where $N(\varepsilon)$ is the density of states per single spin projection. If we explicitly take into account the chemical potential Eq. (11) can be rewritten as:

$$S = -\frac{k_B}{e}\frac{\pi^2}{3}T\frac{\frac{d\tau(\mu)}{d\mu}}{\tau(\mu)} = -\frac{k_B}{e}\frac{\pi^2}{3}T\frac{dn}{d\mu}\frac{\frac{d\tau(n)}{dn}}{\tau(n)}. \quad (13)$$

Here $\frac{dn}{d\mu} > 0$ as at low temperatures $\frac{dn}{d\mu} = \frac{d}{d\mu}\int_{-\infty}^{\mu} d\varepsilon N(\varepsilon) = N(\mu) > 0$. Thus, the sign of S is completely determined by the sign of $\frac{d\tau(n)}{dn}$ or $\frac{d\sigma_{xx}(n)}{dn}$.

Hall conductivity (6) at low temperatures with the account of Eq. (9) takes the form:

$$\sigma_{xy}^{H} = -\frac{2\pi^2 e^3 a^2 H}{3\hbar^2}\tau_{\mathrm{H}}(0) \tag{14}$$

and the Hall coefficient

$$R_{\mathrm{H}} = -\frac{a^2}{e}\frac{8}{3}\frac{\tau_{\mathrm{H}}(0)}{\tau^2(0)}. \tag{15}$$

The sign of the Hall coefficient is completely determined by the sign of $\tau_{\mathrm{H}}(0)$ or by the sign of $\tau_{\mathrm{H}}(n)$. We see that the signs of thermopower and Hall coefficient are determined, strictly speaking, by completely different expressions, so that the band-fillings corresponding to their sign change, in general, can be quite different.

3. THERMOPOWER AND HALL COEFFICIENT IN THE ABSENCE OF CORRELATIONS ($U = 0$)

To study this situation in more details we shall consider first the case when electron correlations are absent (Hubbard interaction $U = 0$). In the absence correlations and other scattering processes of electrons (ideal conductor) both numerator and denominator in Eq. (2) for thermopower and in Eq. (8) for the Hall coefficient diverge, thus to regularize our calculations we have to introduce some weak electron scattering, taking the single-electron Green's as:

$$G^{R}(\mathbf{p}\varepsilon) = \frac{1}{\varepsilon - \varepsilon(\mathbf{p}) + i\gamma}, \tag{16}$$

where $\gamma \ll t$ is scattering frequency (e.g. by impurities), so that the spectral density takes the form:

$$A(\mathbf{p}\varepsilon) = \frac{1}{\pi}\frac{\gamma}{(\varepsilon - \varepsilon(\mathbf{p}))^2 + \gamma^2}. \tag{17}$$

Dependencies of thermopower and Hall coefficient obtained directly from Eqs. (2) and (8) using the spectral density from Eq. (17) with $\gamma/8t = 0.005$, are shown in Fig. 1, both for the case of complete electron-hole symmetry ($t' = 0$) and for $t'/t = -0.4$, typical e.g., for cuprates like YBCO. We can see that in the case of complete electron-hole symmetry thermopower (Fig. 1a) is linear in temperature in accordance with Eqs. (11) and (12), up to high temperatures $T/8t \approx 0.06$, while R_{H} (Fig. 1c) is practically independent of T. Both thermopower and Hall coefficient change sign at band half-filling. In case of pretty strong breaking of such symmetry ($t'/t = -0.4$) we observe a noticeable deviation from linear dependence of thermopower on temperature (cf. Fig. 1b), at high $T/8t \approx 0.06$, and Hall coefficient (Fig. 1d) at high T also acquires a noticeable dependence on temperature. The sign change of S is observed at filling $n \approx 0.65$, which is significantly larger than half-filling, while the Hall coefficient changes its sign at band-filling noticeably lower than half-filling, and with the growth of temperature this deviation from half-filling further grows and for $T/8t \approx 0.06$ R_{H} changes sign at $n \approx 0.22$. Thus for $t'/t = -0.4$ we observe rather wide region of band-fillings n, where the Hall coefficient R_{H} already has positive sign (hole-like), while the thermopower S is still negative (electron-like). As was shown above the sign of thermopower is completely determined by the sigh of $\frac{d\tau(n)}{dn}$, while the sigh of Hall coefficient is determined by the sign of $\tau_{\mathrm{H}}(n)$. Thus, to clarify situation it may be useful to analyze the dependencies of $\tau(n)$ and $\tau_{\mathrm{H}}(n)$. But both relaxation parameter τ and Hall relaxation parameter τ_{H} diverge as $\tau \sim \frac{1}{\gamma}$ and $\tau_{\mathrm{H}} \sim \frac{1}{\gamma^2}$ for $\gamma \to 0$, so that it is useful to introduce some reduced relaxation parameters, which are independent of γ, but are characterized by some characteristics of spectrum at the Fermi surface. We shall see that such parameters will be useful also for the analysis of systems with strong electronic correlations.

For $\gamma \ll t$ spectral density has a narrow peak at $\varepsilon \sim \varepsilon(\mathbf{p})$, then:

$$\begin{aligned}\tau(\varepsilon) &= \sum_{\mathbf{p}\sigma}\left(\frac{\partial\varepsilon(\mathbf{p})}{\partial p_x}\right)^2\left(\frac{1}{\pi}\frac{\gamma}{(\varepsilon - \varepsilon(\mathbf{p}))^2 + \gamma^2}\right)^2 \\ &= \int_{-\infty}^{\infty} d\xi\left[\sum_{\mathbf{p}\sigma}\left(\frac{\partial\varepsilon(\mathbf{p})}{\partial p_x}\right)^2\delta(\xi - \varepsilon(\mathbf{p}))\right] \\ &\quad\times\left(\frac{1}{\pi}\frac{\gamma}{(\varepsilon - \xi)^2 + \gamma^2}\right)^2\end{aligned} \tag{18}$$

so that we can write:

$$\tau(\varepsilon) \approx \tau_0(\varepsilon)\int_{-\infty}^{\infty} d\xi\left(\frac{1}{\pi}\frac{\gamma}{(\varepsilon - \xi)^2 + \gamma^2}\right)^2 = \frac{1}{2\pi\gamma}\tau_0(\varepsilon), \tag{19}$$

where we have introduced

$$\tau_0(\varepsilon) = \sum_{\mathbf{p}\sigma}\left(\frac{\partial\varepsilon(\mathbf{p})}{\partial p_x}\right)^2\delta(\varepsilon - \varepsilon(\mathbf{p})). \tag{20}$$

Similarly

$$\begin{aligned}\tau_{\mathrm{H}}(\varepsilon) &= \sum_{\mathbf{p}\sigma}\left(\frac{\partial\varepsilon(\mathbf{p})}{\partial p_x}\right)^2\frac{\partial^2\varepsilon(\mathbf{p})}{\partial p_y^2}A^3(\varepsilon,\mathbf{p}) \\ &\approx \tau_{0\mathrm{H}}(\varepsilon)\int_{-\infty}^{\infty} d\xi\left(\frac{1}{\pi}\frac{\pi}{\xi^2 + \gamma^2}\right)^3 = \frac{3}{8}\frac{1}{\pi^2\gamma^2}\tau_{0\mathrm{H}}(\varepsilon),\end{aligned} \tag{21}$$

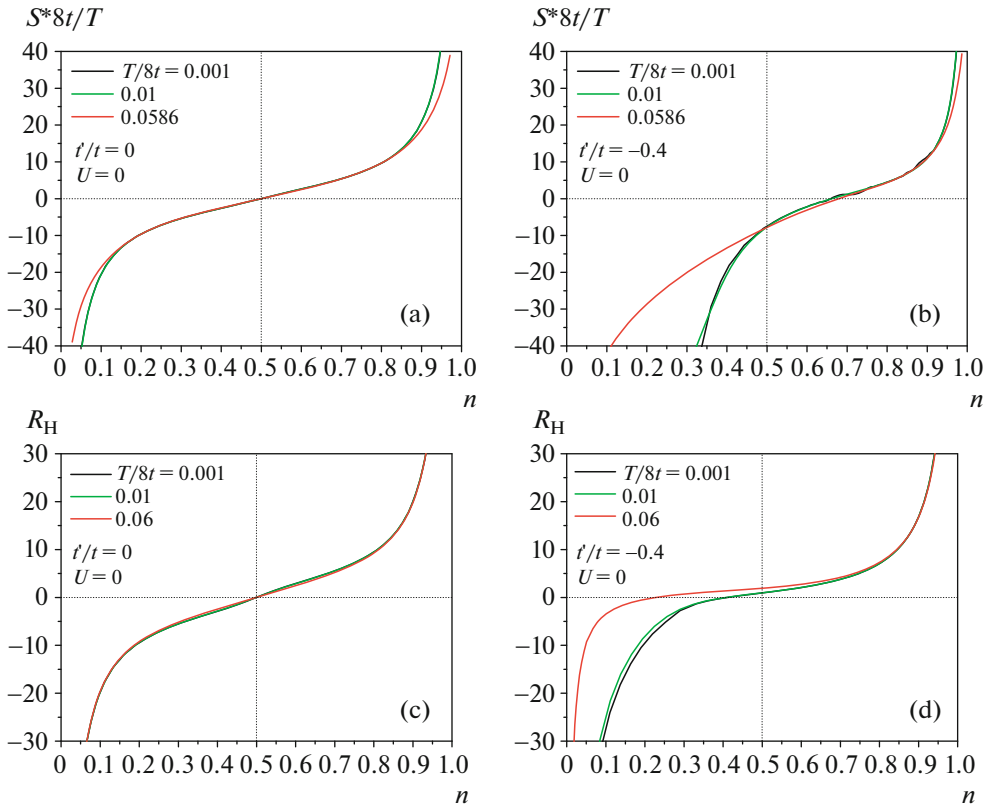

Fig. 1. Dependence of thermopower (a, b) and Hall coefficient (c, d) on band-filling for $t' = 0$—left column (a,c) and $t'/t = -0.4$—right column (b, d).

where we have introduced

$$\tau_{0H}(\varepsilon) = \sum_{\mathbf{p}\sigma}\left(\frac{\partial\varepsilon(\mathbf{p})}{\partial p_x}\right)^2 \frac{\partial^2\varepsilon(\mathbf{p})}{\partial p_y^2}\delta(\varepsilon - \varepsilon(\mathbf{p})). \qquad (22)$$

Then at low temperature ($T \ll E_F$) instead of Eq. (13) we obtain for the thermopower:

$$S = -\frac{k_B}{e}\frac{\pi^2}{3}T\frac{\frac{d\tau_0(\mu)}{d\mu}}{\tau_0(\mu)} = -\frac{k_B}{e}\frac{\pi^2}{3}T\frac{dn}{d\mu}\frac{\frac{d\tau_0(n)}{dn}}{\tau_0(n)} \qquad (23)$$

and the sign of S is opposite to the sign of $\frac{d\tau_0(n)}{dn}$.

For Hall coefficient instead of Eq. (15) we obtain

$$R_H = -\frac{a^2}{e}\frac{4\tau_{0H}(n)}{\tau_0^2(n)} \qquad (24)$$

and the sign of R_H is opposite to the sign of $\tau_{0H}(n)$. Note that $\frac{\partial^2\varepsilon(\mathbf{p})}{\partial p_y^2}$ is the only sign-changing entity in Eq. (22).

In Fig. 2 we show dispersions $\varepsilon(\mathbf{p})$ along symmetry directions in Brillouin zone (Figs. 2a, 2b) and Fermi surfaces, corresponding to different band-fillings (Figs. 2c, 2d) for two choices of t'/t. In the case of complete electron-hole symmetry ($t' = 0$) for half-filled band we observe the change of the type of Fermi surface (Fig. 2c) from electron pocket around Γ—point of the Brillouin zone, characteristic for $n < 0.5$, to hole pocket around M-point of the Brillouin zone, characteristic for $n > 0.5$. Due to electron-hole symmetry the regions of quadratic electron spectrum (close to M-point) and similar hole spectrum (close to M-point) are just the same (Fig. 2a). Thus τ_{0H} and correspondingly R_H change sign at half-filling (Fig. 2f). Because of electron-hole symmetry $\tau_0(n) = \tau_0(0.5 + n)$ ($n \le 0.5$) and derivative $\frac{d\tau_0(n)}{dn}$ (this means also S) also can change the sign only at half-filling (cf. Fig. 2e).

If electron-hole symmetry is broken ($t'/t = 0.4$) the region quadratic electron dispersion close to Γ-point of the Brillouin zone becomes very narrow, while the region of quadratic hole dispersion close to M-point becomes much wider (cf. Fig. 2b). Correspondingly the region where the Fermi surface is electron-like pocket around M-point is limited to small fillings $n < 0.3$, while in a wide region of fillings $n \ge 0.3$ the Fermi surface is hole-like pocket around M-point of the Brillouin zone (Fig. 2d). Thus the sign change of τ_{0H}

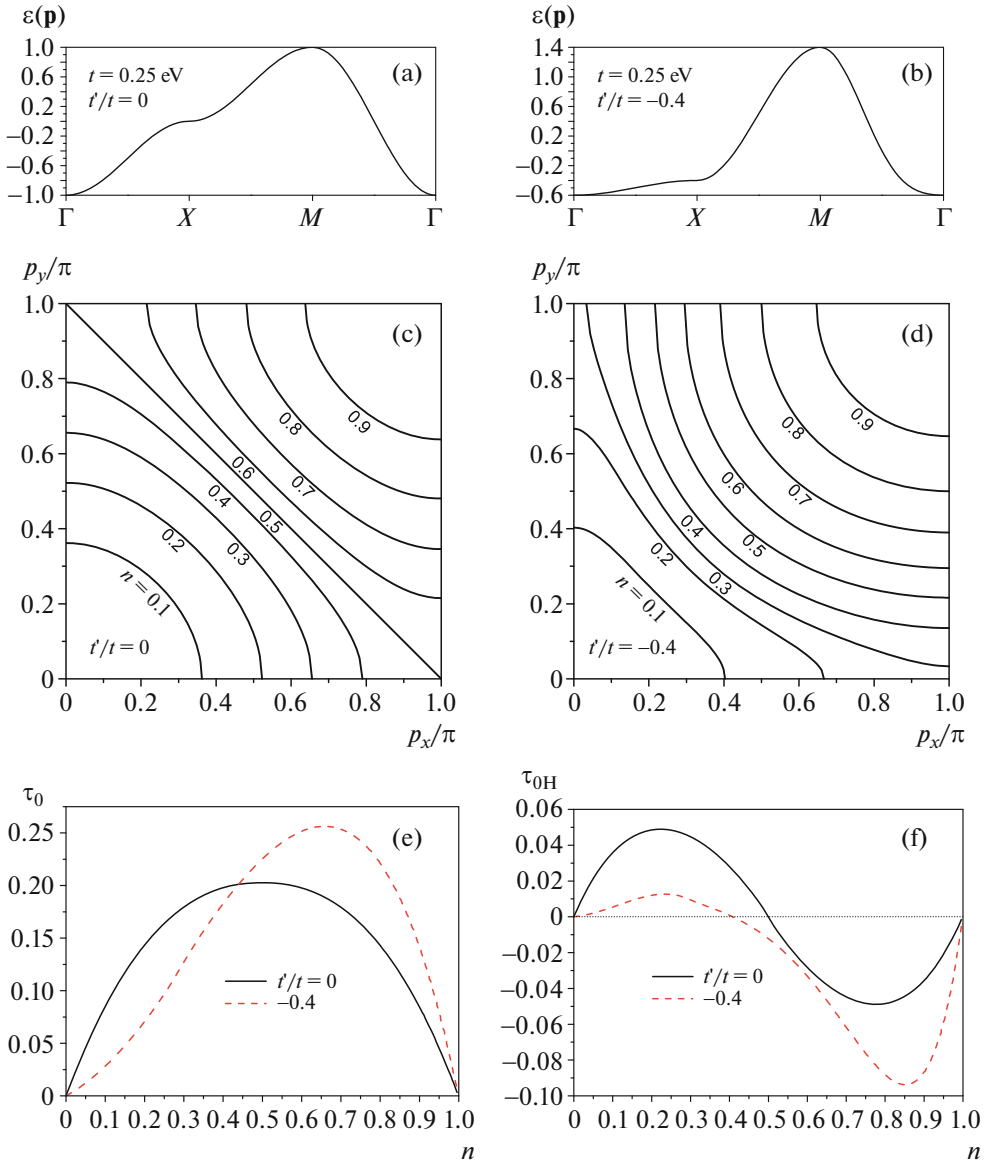

Fig. 2. Electron spectrum along symmetry directions in the Brillouin zone (a, b), Fermi surfaces for different band-fillings for (c, d) and dependences of reduced relaxation parameters τ_0 and τ_{0H} on band-filling (e, f) for $t' = 0$, $t'/t = -0.4$.

and correspondingly of R_H is observed for fillings $n \approx 0.4$ below half-filling (Fig. 2e). More so, the positive values of $\tau_{0H}(n)$ for $n < 0.4$ is significantly smaller than the absolute value of $\tau_{0H}(n)$ for $n > 0.4$, so that the "smearing" due to the derivative of distribution function $\left(-\frac{\partial f}{\partial \varepsilon}\right)$ in Eq. (6) at high temperatures leads to R_H changing its sign at much lower fillings (Fig. 1d). In particular, for $T/8t = 0.06$ R_H changes sign at $n \approx 0.2$.

On the contrary, $\tau_0(n)$ for $t'/t = -0.4$ (Fig. 2e) has a maximum at n larger than half-filling, so that $\frac{d\tau_0(n)}{dn}$ (and correspondingly S) changes its sign for $n \approx 0.66$. Naturally the behavior of $\tau_0(n)$ is more or less symmetric around its maximum, and "smearing" due to the derivative of distribution function in Eq. (2) close to this maximum leads only to small changes of S, so that the filling $n \approx 0.66$, where thermopower changes its sign is only weakly dependent on the temperature growth (cf. Fig. 1b).

Thus with the growth of $|t'|$ the band-filling, corresponding to the sign change of R_H, moves to the region of n below half-filling of the band, while the band-filling, corresponding to sign change of S, more farther into the region of $n > 0.5$. As a result, the growth of $|t'|$ close to half-filling leads to formation of a wider region of band-fillings, where Hall coefficient and

thermopower have different signs, and the growth of the temperature makes this region even wider. Below we shall see that this tendency is observed also in systems with systems with strong electronic correlations.

4. THERMOPOWER IN STRONGLY CORRELATED METAL AND DOPED MOTT INSULATOR

Before going to the results of our DMFT calculations we shall present an elementary qualitative analysis along the lines of [4, 5]. It is more or less obvious that deep in the Mott insulator state with well-defined upper and lower Hubbard bands under the hole doping both thermopower and Hall coefficient are in fact determined by filling of the lower Hubbard band (the upper band is much upper in energy and is practically empty). Under this situation in the model with electron-hole symmetry (in two-dimensional case this corresponds to a spectrum with $t' = 0$) we can estimate the band-filling, corresponding to sign change of thermopower, in very simple way. We shall consider only the paramagnetic phase with $n_\uparrow = n_\downarrow = n$, so that in the following n denotes electron density per single spin projection and total electron density is $2n$. In the following we consider only hole-doping of Mott insulator and the number of these "holes" is given by $p = 1 - 2n$.

In the case of electron-hole symmetry it is natural to suppose that both thermopower and Hall coefficient change sign close to half-filling of the lower Hubbard band $n_0 \approx 1/2$. Consider the states with "upper" spin projection, then the full number of available states in the lower Hubbard band id $1 - n_\downarrow, = 1 - n$. Then the band-filling is determined by $n = n_\uparrow = n_0(1 - n) \approx 1/2(1 - n)$. Thus for the filling at which both thermopower and Hall effect we obtain the estimate $n_c \approx 1/3$, with corresponding "hole" concentration $p_c = 1 - 2n_c \approx 1/3$ [4, 5].

In general case situation is obviously more complicated. In strongly correlated systems both thermopower and Hall effect (as well as other electronic properties) are significantly dependent on temperature. At low temperatures in these systems considered in DMFT approximation along with lower and upper Hubbard bands an additional narrow quaiparticle band forms close to the Fermi level leading to the so-called quasiparticle peak in the density of states [1–3]. In doped Mott insulator this peak is placed near the upper edge of the lower Hubbard band. Thus, at low temperatures both thermopower and Hall coefficient are mainly determined by filling of this quasiparticle band. At high enough temperature (of the order or larger than the width of quasiparticle peak) this peak is smeared and thermopower is completely determined by the filling of the lower Hubbard band. Correspondingly, during the analysis of thermopower, as well as Hall effect [4, 5], it is necessary to consider two different temperature regimes.

In Fig. 3 we show the dependence of thermopower on band-filling, both for strongly correlated metal ($U/8t = 1$) and doped Mott insulator ($U/8t = 4, 10$). We see that in high-temperature regime (curves with filled symbols) thermopower, even in the model with complete electron-hole symmetry with $t' = 0$ (Fig. 3a), in doped Mott insulator changes sign at filling much closer to half-filling ($n = 0.42$), than follows from our qualitative estimate. Note that Hall coefficient [4, 5] in this case changes its sign at $n_H \sim 0.36$ (cf. Fig. 4a), which is quite close to our qualitative estimate. For comparison in Fig. 3a blue dashed line shows an exact result for thermopower [14]:

$$S = -\frac{k_B}{e}\ln\frac{2p}{1-p} = -\frac{k_B}{e}\ln\frac{1-2n}{n} \qquad (25)$$

for the limit of $U \gg t$. It can be seen that in this atomic limit thermopower changes sign at $n = 1/3$ in complete accordance with our estimate. It is possible that this significant difference is due to our use of DMFT approximation. Cluster DMFT [9] and Monte-Carlo calculations [12] give thermopower values for $U \gg t$ much closer to an exact result of Eq. (25). However the main deficiency of these approaches is the possibility to perform calculations only in the region of high enough temperatures $T \sim t$.

For noticeable breaking of electron-hole symmetry our qualitative estimate becomes invalid both for the Hall coefficient [4, 5] and thermopower (cf. Fig. 3b). Note that in accordance with our analysis of the case of $U = 0$ given above the deviation from electron-hole symmetry leads to decreasing values of the filling, corresponding to change of the sign of Hall coefficient [4, 5], and increasing values of the filling, corresponding the sign change of thermopower (cf. Fig. 4b). Thus, in strongly correlated systems breaking of electron-hole systems leads to the appearance of rather wide region of band-fillings, where Hall coefficient and thermopower have different signs. To compare high-temperature behavior of thermopower and Hall coefficient [4, 5] in hole doped Mott dielectric we show corresponding band-filling dependencies in Fig. 4 for $t'/t = 0$; -0.4.

In the low-temperature limit (curves with unfilled symbols in Fig. 3) the presence of quasiparticle peak leads to the absence of sign change of thermopower in doped Mott insulator, which remains negative (electron-like) in the whole region of hole dopings. Note that Hall coefficient at low temperatures becomes positive (hole-like) only in a narrow region close to half-filling, i.e. at very small hole doping [4, 5]. In low-temperature regime the width and the amplitude of quasiparticle peak depend both on band-filling and temperature. Significant dependence of quasiparticle peak on band-filling in low-temperature regime leads

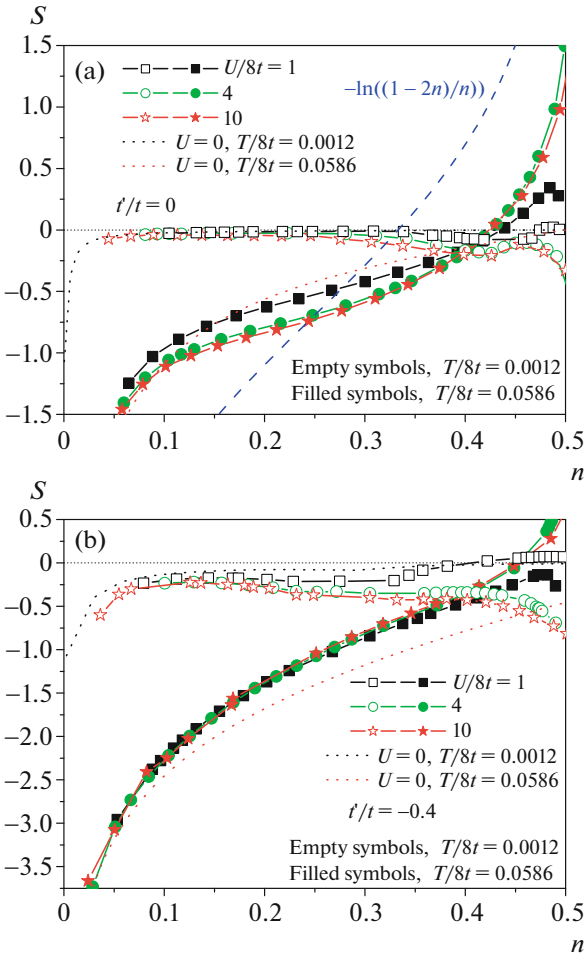

Fig. 3. Dependence of thermopower for the case of $t' = 0$ (a) and $t'/t = -0.4$ (b) on band-filling for different strength of electron correlations U in the low-temperature regime (empty symbols) and in high-temperature regime (filled symbols).

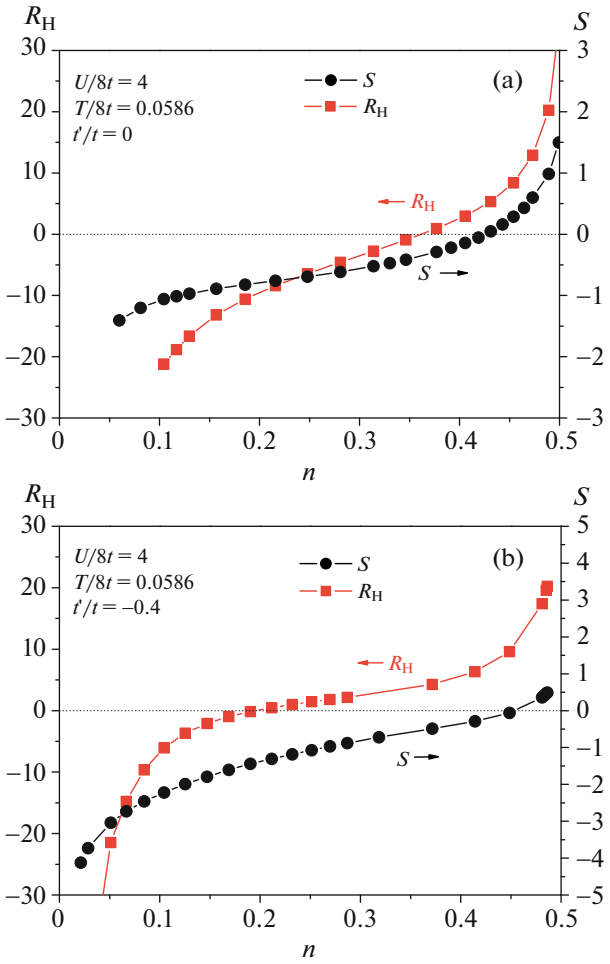

Fig. 4. Comparison of high-temperature dependencies of thermopower and Hall coefficient [4, 5] on band-filling for $t' = 0$ (a) and $t'/t = -0.4$ (b).

to the appearance of rather wide region of band-fillings, where thermopower decreases with the growth of n (cf. Fig. 3). Such anomalies on filling, related to quasiparticle peak, are stronger for thermopower than for Hall coefficient [4, 5].

Smearing and disappearance of quasiparticle peak may be not only due to the growth of temperature, but also due to disordering [6, 15], as well as due to pseudogap fluctuations, which are totally neglected in the local DMFT approach [6, 13]. Thus, the region of applicability of simple estimates and qualitative behavior of thermopower, given above in the framework of DMFT for high-temperature regime, in reality may be much wider.

In general case calculation of disorder scattering effects (more so pseudogap fluctuations) upon thermopower is rather complicated problem. As a simple estimate below we present the results of calculations using Eqs. (2) and (3), where we use the values of spectral density $A(\mathbf{p}\varepsilon)$ for disordered Hubbard model, obtained within DMFT + Σ approach [6, 13]. Disorder parameter Δ denotes here an effective scattering rate of electrons by the random field of impurities (in self -consistent Born approximation). It is clear that such approach, taking into account only the influence of disorder in the spectral density, is oversimplified, but it seems reasonable for qualitative analysis.

In Fig. 5 we compare dependencies of thermopower on band-filling in the absence of disorder (empty symbols) and for the case impurity scattering with $\Delta/8t = 0.25$ in Mott insulator with $U/8t = 4$. While in high-temperature regime disorder only weakly affects the Hall coefficient [4, 5] for different values of t'/t, for the thermopower disorder influence, even in this regime, is rather noticeable. Disorder leads to the decrease of thermopower for fillings close to half-filling (low hole doping), leading to the decrease of hole doping, corresponding to sign change of S. At high enough hole doping < when thermopower is negative, the growth of disorder leads to

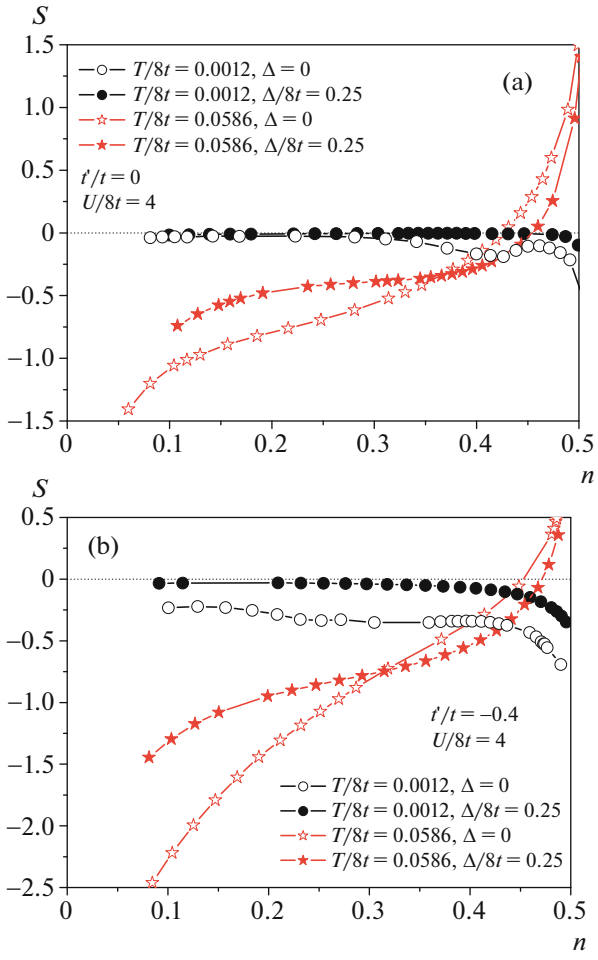

Fig. 5. Dependence of thermopower on bandfilling in the presence of impurity scattering ($\Delta/8t = 0.25$, filled symbols) and in its absence ($\Delta = 0$, empty symbols) for $t' = 0$ (a) and $t'/t = -0.4$ (b).

rather significant decrease of the absolute value of S. In low-temperature regime, where thermopower is negative at any hole dopings, increasing disorder leads to a strong decrease of the absolute value of thermopower.

In Fig. 6 we show the dependencies of thermopower on band-filling and temperature in the case of Mott insulator with $U/8t = 4$ for different models of electron spectrum, for the case of complete electron-hole symmetry $t' = 0$ and for $t'/t = -0.25$ and $t'/t = -0.4$, typical correspondingly for cuprates like LSCO and YBCO. On the filling dependence of S we observe smooth evolution from low-temperature to high-temperature regime. While at low temperatures for values of t'/t, considered here, thermopower is negative at all fillings, increasing of the temperature close to half-filling leads to the appearance of the region of positive values of thermopower. Hole doping at which S changes its sign increases with temperature and decreases with the growth of $|t'/t|$. It is necessary to mote, that for Hall coefficient [4, 5] the growth of $|t'/t|$ leads to the decrease of the region of hole dopings with positive values of Hall coefficient and to the increase of doping value, corresponding to the sign change of R_H. Situation is here is in many respects similar to the case of absence of correlations ($U = 0$)—the growth of $|t'/t|$ leads to the increase of band-filling, corresponding to sign change of thermopower and the decrease of the filling, corresponding to sign change of R_H.

In Figs. 6b, 6d, 6f we show the temperature dependencies of thermopower for different band-fillings. In all cases we observe the significant dependence of S on temperature. For low temperatures thermopower is negative and its absolute value increases with the growth of the temperature. At high temperatures close to half-filling (low hole doping) thermopower is increasing with rising temperature changing sign of S, leading to positive values of S at high T. At small fillings (high hole doping) the absolute value of S at high temperatures continues to increase with the growth of T, though slower than at low temperatures.

Recently quantum Monte-Carlo calculations of thermopower were done for the Hubbard model [12] at reasonable temperature $T = t/4$. The results of these calculations nicely reproduce the experimental results on thermopower for a number of hole-doped cuprates (cf. Fig. 1 in [12]). In particular they reproduce the sign change of thermopower at hole doping $p \approx 0.15$. In Fig. 7 we compare the results of our DMFT calculations with Monte-Carlo results of [12]. We can see that the results of our DMFT calculations of thermopower at $U/8t = 4$ are close to Monte-Carlo results at $U/8t \approx 1$, and correspondingly to the room temperature experimental data for thermopower in cuprates. Note that in the framework of DMFT at $U/8t = 1$ the system still remains (even at half-filling) the strongly correlated metal, while at $U/8t = 4$ it becomes the doped Mott insulator, which is usually considered as typical for cuprates.

5. ESTIMATING CARRIER CONCENTRATION, THERMOPOWER AND HALL COEFFICIENT FROM ARPES DATA

Measuring thermopower and Hall coefficient are among the major experimental methods to determine the type and concentration of current carriers. However, as was demonstrated above, especially under the conditions of broken electron-hole symmetry ($t' \neq 0$) a wide region of band-fillings appear, where thermopower and Hall effect produce evidence for different type of charge carriers (p–n anomaly). As was shown in [4, 5] in the interval of doping, where Hall effect changes its sign, Hall number (the number of carriers formally determined from this effect) sharply increases and can not be used to determine the real number of carriers.

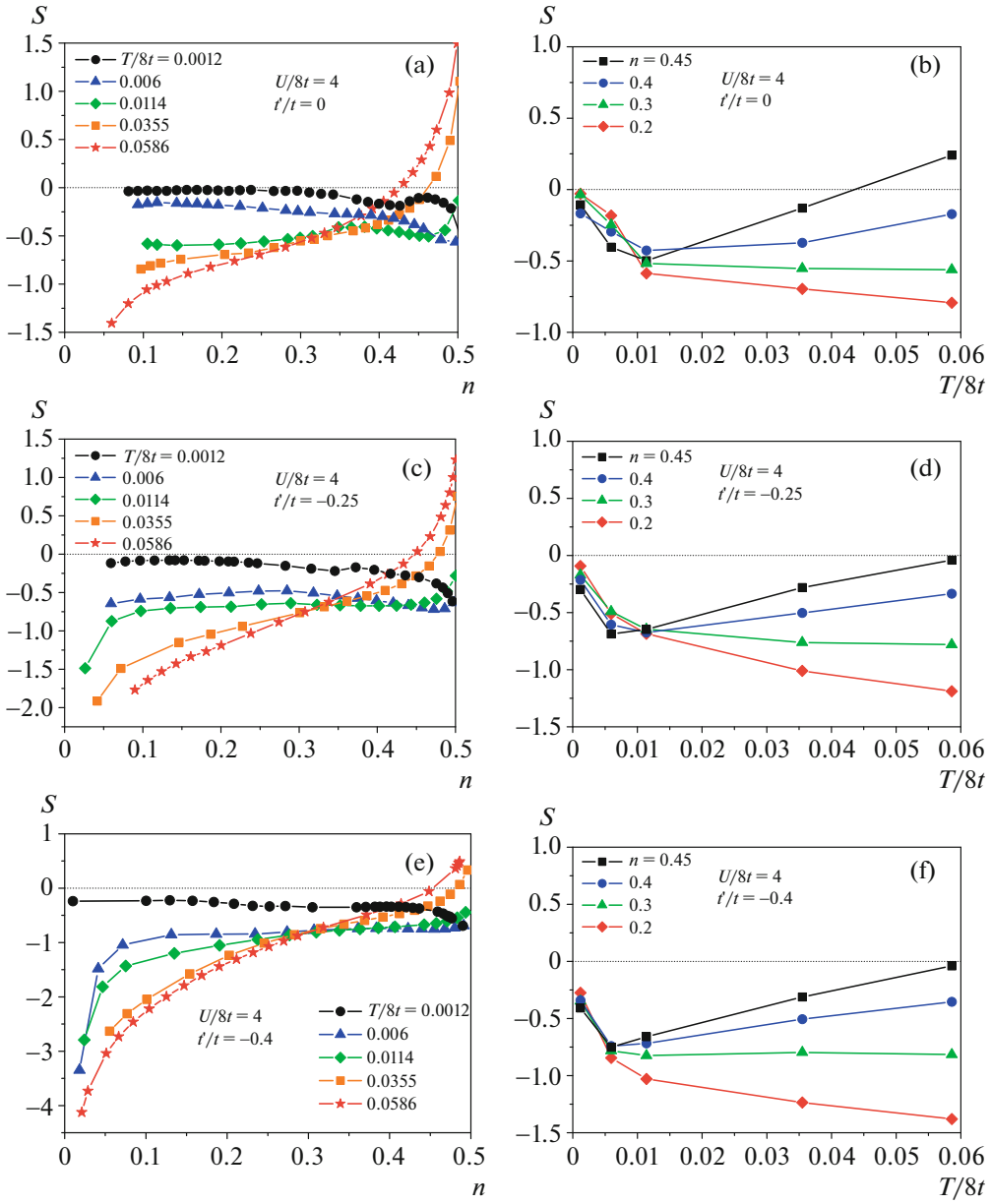

Fig. 6. Dependence of thermopower on band-filling for different temperatures (a, c, e) and temperature dependence of S for different band-fillings (b, d, f).

Below we shall show, that a semiquantitative estimate of the number of charge carriers, as well as of thermopower and Hall coefficient in doped Mott insulator can be done using ARPES data and electron spectrum obtained within the standard DFT calculations.

In systems with strong electronic correlations, doped Mott insulator including, Fermi-liquid description still holds, Fermi surface is well defined, as well as quasiparticles near it [16]. In Fig. 8 we show the spectral density $A(\mathbf{p}, 0)$ at the Fermi level, obtained within DMFT in high-temperature regime, when the temperature is noticeably larger than the width of quasi-particle peak, but still much lower than the Fermi energy. Band-filling is taken to be equal to $n = 0.19$, at which, in the case of $t'/t = -0.4$, Hall coefficient changes its sign [4, 5]. We see that spectral density is much "smeared," but it still has a maximum at the Fermi surface, shown in Fig. 8 by black curve, obtained from equation:

$$\mu - \varepsilon(\mathbf{p}) - \mathrm{Re}\,\Sigma(0) = 0, \qquad (26)$$

where μ is the chemical potential, determined from band-filling within DMFT, while $\Sigma(0)$ is local (DMFT) self-energy at the Fermi level.

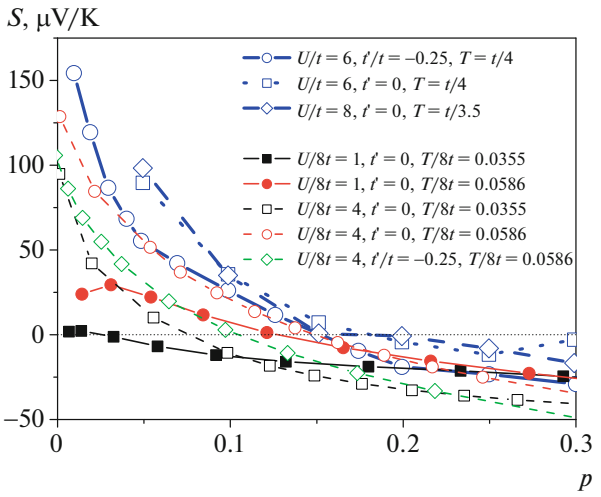

Fig. 7. Comparison of DMFT results and Monte-Carlo calculations [12] (thick blue lines) for the dependence of thermopower on hole doping p.

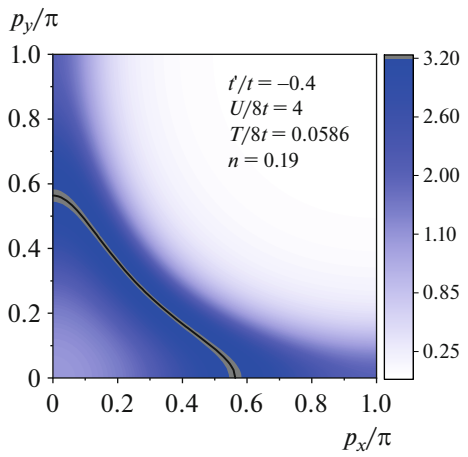

Fig. 8. Spectral density in the quarter of the Brillouin zone. Curve represents the Fermi surface obtained from Eq. (26).

Let us introduce now $\mu_{\text{eff}} = \mu - \text{Re}\Sigma(0)$, then instead of Eq. (26) we obtain an equation:

$$\mu_{\text{eff}} - \varepsilon(\mathbf{p}) = 0, \tag{27}$$

determining the Fermi surface like in the absence of electronic correlations ($U = 0$), but with chemical potential μ_{eff}. Obviously in general case spectral density is much "smeared" around the Fermi surface, but its maximum is still on it (Fig. 8). Thus for qualitative (but much simpler) estimate of Hall coefficient and thermopower we can use expressions given by, Eqs. (23) and (24), corresponding to $U = 0$, but with $\mu \to \mu_{\text{eff}}$. Naturally, the filling n_0, corresponding to chemical potential μ_{eff} in the system with no correlations ($U = 0$), does not coincide with real band-filling n in correlated system. It seems reasonable to assume that this n_0 corresponds to the filling of the lower Hubbard band. Then from n_0 we can easily get the total filling of the whole band n. The total number of states in the lower Hubbard band is $1 - n$, so that the total filling is $n = n_0(1 - n)$ and we get:

$$n = \frac{n_0}{1 + n_0}. \tag{28}$$

In Fig. 9 we show relations between n_0 and n, as well as simple estimates for Hall coefficient and thermopower, compared with exact DMFT results. In high-temperature regime ($T/8t = 0.0586$), when quasiparticle peak in the density of states vanishes, we can see that Eq. (28) is more or less confirmed. Estimates for thermopower S_0 and especially for Hall coefficient obtained in this approach are also sufficiently close to exact results.

In low-temperature regime the presence of quasiparticle peak in the density of states of DMFT makes Eq. (28) invalid and n_0 in fact just coincides with n. The estimate for Hall coefficient R_{H0} is still quite close to exact DMFT results (except anomalies in R_H, related to quasiparticle peak for $t' = 0$ [4, 5] (Fig. 9h). At the same time the estimate of thermopower at low temperatures is unsatisfactory. It is possibly related to more important role of "smearing" of spectral density around the Fermi surface in calculations of thermopower and anomalies related to filling of quasiparticle peak.

It is necessary to note once more, that anomalies observed in low-temperature regime are determined by the presence of quasiparticle peak in DMFT approximation, while it vanishes with increase of temperature or due to disorder, as well as due to account of nonlocal correlations outside DMFT. Thus, it is quite possible that qualitatively the results obtained in high-temperature regime are more or less general.

Using ARPES data for doped Mott insulator we can determine n_0 (in fact it is just the area of the Brillouin zone below the Fermi surface) and then, using Eq. (28), define the total filling n and hole doping level as $p = 1 - 2n$. Standard DFT calculations of electron spectrum, with the account of filling of uncorrelated band n_0, allow using Eqs. (23) and (24) to estimate both R_H and S. In fact these results are in total agreement with the picture of "hidden" Fermi-liquid introduced in [16].

6. CONCLUSIONS

We have studied the behavior of thermoelectric power in metallic phase originating by doping the Mott insulator. We mainly concentrated on the case of

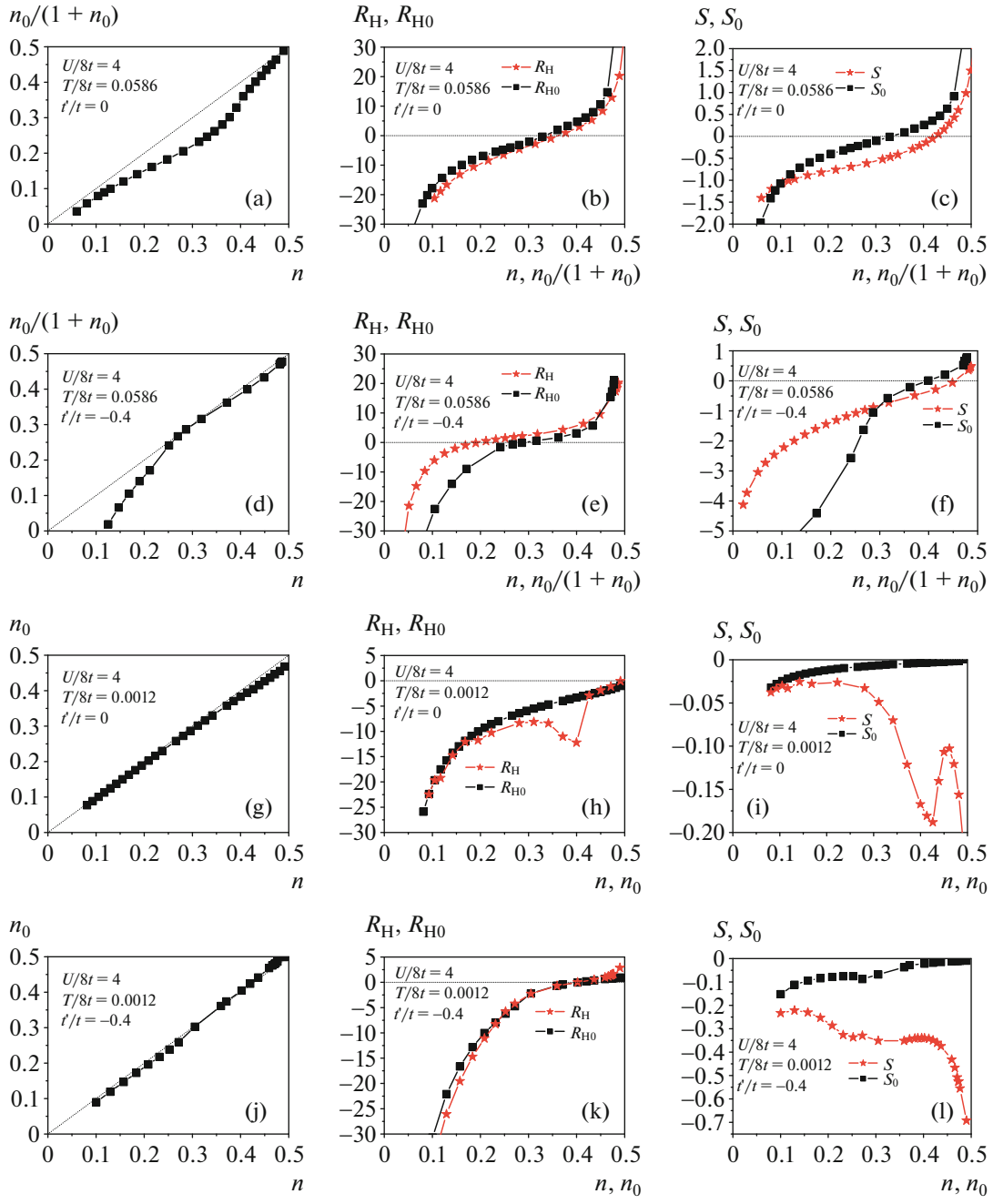

Fig. 9. Relation between the effective band-filling of the band without correlations n_0 and total band-filling of correlated band n (a, d, g, j). Comparison of estimates for Hall coefficient R_{H0} (b, e, h, k) and thermopower S_0 (c, f, i, l) with DMFT results (R_H, S).

hole doping, characteristic for the major part of cuprates. We have considered a number of two-dimensional tight-binding models of electron spectrum, fitting the electronic structure of cuprates. In all of these models in doped Mott insulator we observe anomalous temperature dependence of thermopower S which is significantly different from linear dependence typical to usual metals. In low-temperature limit S is mainly determined by filling of quasiparticle peak, leading to negative values of thermopower at any hole doping and anomalous dependence of S on filling, when the negative (electron-like) thermopower increases its absolute value with addition of electrons. For thermopower these low-temperature anomalies, connected with the filling of quasiparticle peak, are much stronger than similar anomalies for Hall coefficient.

In high-temperature limit, when the quasiparticle peak is essentially damped, S is mainly determined by the filling of lower (for hole doping) or upper (for electron doping) Hub-bard band. In this limit the qualitative estimate shows that the sign change of both S and Hall effect in the simplest (symmetric, $t' = 0$) case takes place at band-filling $n = 1/3$ per single spin projection, corresponding to hole doping $p = 1 - 2n = 1/3$.

DMFT calculations show that S changes its sign at $n = 0.42$, while Hall effect sign change takes place at $n = 0.36$. Thus, even in the case of electron-hole symmetry there is narrow region of dopings, where R_H is already positive, while S remains negative. The breaking of electron-hole symmetry leads to the increase of filling n, corresponding to sign change of S, and to the decrease of n, corresponding to sign change of R_H, leading to rather wide region of fillings, where thermopower and Hall effect have different signs. We have also proposed a scheme, allowing to obtain the number of charge carriers in doped Mott insulator form ARPES data using DFT calculations of electronic spectrum and to perform a semiquantitative estimate of both Hall coefficient and thermopower.

In principle, on the qualitative level, the possibility of different signs of thermopower and Hall effect is known for rather long time and was observed in a number of experiments in disordered systems [17, 18]. Systematic studies of thermopower in cuprates for different doping levels was performed in [19, 20]. It was demonstrated that thermopower of a number of cuprates changes its sign close to $p = 1 - 2n \approx 0.1$–0.2, in the vicinity of optimal doping level $p \approx 0.16$, corresponding to the maximum of superconducting temperature. In [21] this behavior of thermopower was interpreted as connected to the presence of a nearby quantum critical point, related with separation of the upper Hubbard band. Unfortunately, no comparison was made in these works with the available data on Hall effect. In [22–25] the measurements of Hall effect were done for several cuprates at low temperatures in extremely strong magnetic fields (in the normal phase), which also demonstrated the anomalous behavior (growth of the Hall number of carriers) at doping levels $p \approx 0.2$–0.25. I these works this anomalous behavior of Hall effect was also related to the nearby quantum critical point, corresponding to the closure of the pseudogap. In [4, 5] this behavior of Hall effect was interpreted as due to the approach to the point, where Hall effect changes its sign (which actually was not observed in the samples studied in [22–25]), with no relation to any quantum critical point. The DMFT results obtained above are in qualitative agreement with experimental data. In particular, the hole doping level of Mott insulator, corresponding to the change of sign of the thermopower is always lower, than the doping level where the Hall coefficient changes its sign. This stresses the importance of systematic comparative studies of Hall effect and thermopower in cuprates.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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