Thermoelectric Effects in Correlated Matter

Work supported by
NSF DMR
0408247

Sriram Shastry
UCSC
Santa Cruz

Work supported by
DOE, BES DE-FG02-
06ER46319

Aspen Center for Physics:

14 August, 2008

Intro

- High Thermoelectric power is very desirable for applications.
- Usually the domain of semiconductor industry, e.g. Bi_2Te_3. However, recently correlated matter has found its way into this domain.
- Heavy Fermi systems (low T), Mott Insulator Junction sandwiches (Harold Hwang 2004)
- Sodium Cobaltate Na_xCoO_2 at $x \sim .7$ Terasaki, Ong, …..

What is the Seebeck Coefficient S?

$$\begin{aligned}\frac{1}{\Omega}\langle \hat{J}_x \rangle &= L_{11}E_x + L_{12}(-\nabla_x T/T)\\ \frac{1}{\Omega}\langle \hat{J}_x^Q \rangle &= L_{21}E_x + L_{22}(-\nabla_x T/T),\end{aligned}$$

where $(-\nabla_x T/T)$ is regarded as the *external driving thermal force*, and $\hat{J}_x^Q$ is the heat current operator.

$$\begin{aligned}\text{Thermopower} \quad S(\omega) &= \frac{L_{12}(\omega)}{TL_{11}(\omega)}\\ \text{Lorentz Number} \quad \mathbf{L}(\omega) &= \frac{\kappa_{zc}(\omega)}{T\sigma(\omega)}\\ \text{Figure of Merit} \quad \mathbf{Z}(\omega)T &= \frac{S^2(\omega)}{\mathbf{L}(\omega)}. \qquad (1)\end{aligned}$$

$$S_{Kubo} = \left[\frac{\int_0^\infty \; dt \; \int_0^\beta \; d\tau \; \langle \hat{J}_x^E(t - i\tau) \hat{J}_x(0) \rangle}{\int_0^\infty \; dt \; \int_0^\beta \; d\tau \; \langle \hat{J}_x(t - i\tau) \hat{J}_x(0) \rangle} \; - \frac{\mu(0)}{q_e} \right] + \frac{\mu(0) - \mu(T)}{q_e}.$$

S= Transport part + Thermodynamic part Write

$$S_{Kubo} = S_{Tr} + S_{Heikes-Mott},$$

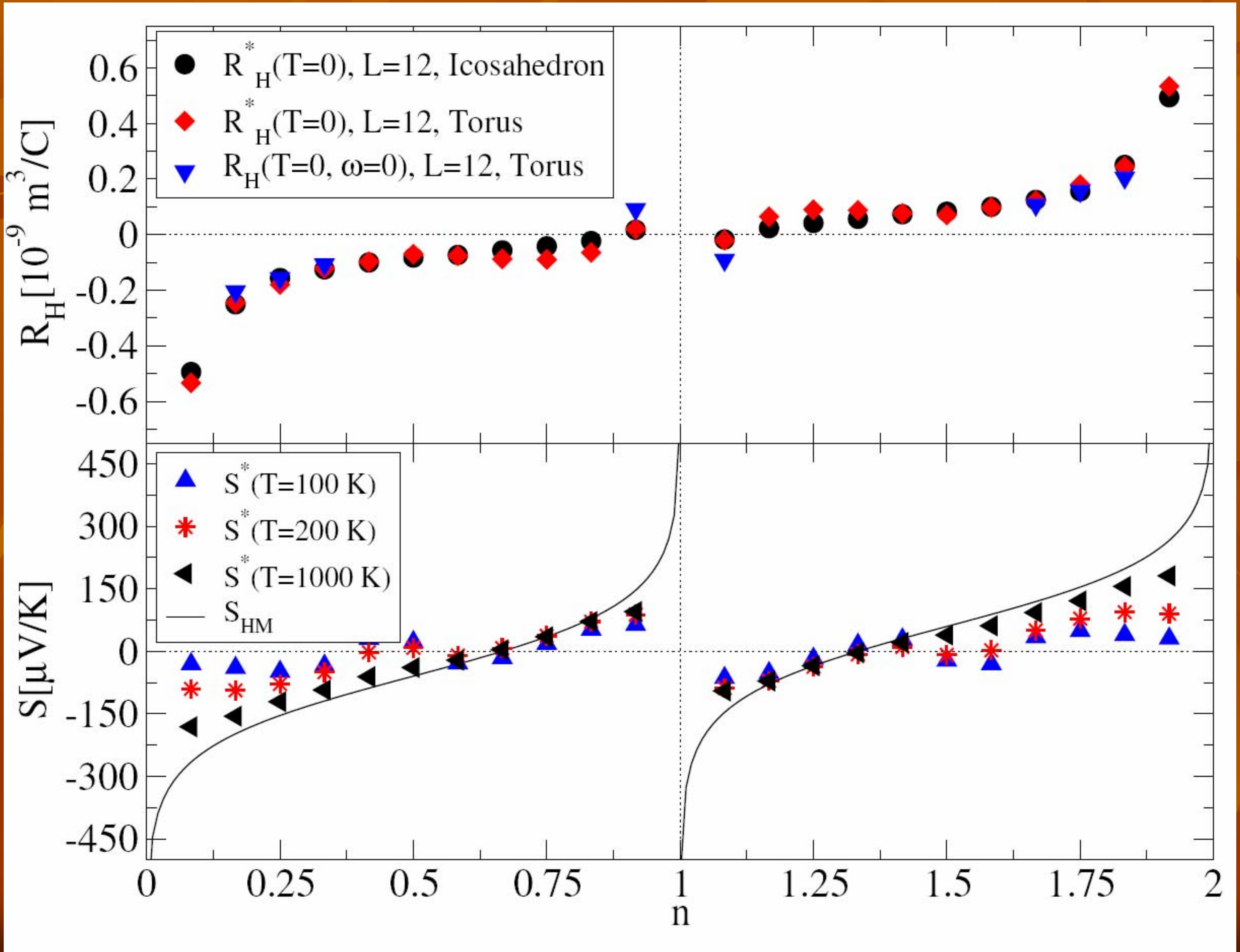
Where the first term is the difficult Transport part of S.

Similarly thermal conductivity and resitivity are defined with appropriate current operators. The computation of these transport quantities is brutally difficult for correlated systems.

Hence seek an escape route……….That is the rest of the story!

Triangular lattice Hall and Seebeck coeffs: (High frequency objects)

Notice that these variables change sign thrice as a band fills from 0->2. Sign of Mott Hubbard correlations.



Considerable similarity between Hall constant and Seebeck coefficients.

Both gives signs of carriers---(Do they actually ???)

Zero crossings tell a tale. These objects are sensitive to half filling and hence measure Mott Hubbard hole densities.

Brief story of Hall constant to motivate the rest.

The Hall constant at finite frequencies: S Shraiman Singh- 1993

High T_c and triangular lattices---

Consider a novel dispersion relation

(Shastry ArXiv.org 0806.4629)

$$\Re e R_H(0) = R_H^*(\Omega) + \frac{2}{\pi}\int_0^{\Omega} \frac{\Im m R_H(\nu)}{\nu}\, d\nu \; .$$

- Here Ω is a cutoff frequency that determines the RH*. LHS is measurable, and the second term on RHS is beginning to be measured (recent data exists).
- The smaller the Ω, closer is our RH* to the transport value.
- We can calculate RH* much more easily than the transport value.
- For the tJ model, it would be much closer to the DC than for Hubbard type models. This is obvious since cut off is max{t,J} rather than U!!

$$\begin{aligned} \hbar\omega &\gg \{|t|, U\}_{max} \\ \hbar\omega &\gg \{|t|, J\}_{max}. \end{aligned}$$

ANALOGY between Hall Constant and Seebeck Coefficients

New Formalism SS (2006) is based on a finite frequency calculation of thermoelectric coefficients. Motivation comes from Hall constant computation (Shastry Shraiman Singh 1993- Kumar Shastry 2003)

$$\rho_{xy}(\omega) = \frac{\sigma_{xy}(\omega)}{\sigma_{xx}(\omega)^2} \to BR_H^* \text{ for } \omega \to \infty$$

$$R_H^* = R_H(0) \text{ in Drude theory}$$

Perhaps ω dependence of R_H is weak compared to that of Hall conductivity.

$$R_H^{\ *} = \frac{-i2\pi}{hB} N\mathrm{v} < [J^x, J^y] > / < \tau_{xx} >^2$$

- Very useful formula since
  - Captures Lower Hubbard Band physics. This is achieved by using the Gutzwiller projected fermi operators in defining J's
  - Exact in the limit of simple dynamics (e.g few frequencies involved), as in the Boltzmann eqn approach.
  - Can compute in various ways for all temperatures (exact diagonalization, high T expansion etc…..)
  - We have successfully removed the dissipational aspect of Hall constant from this object, and retained the correlations aspect.
  - Very good description of t-J model.
  - This asymptotic formula usually requires ω to be larger than J

Need similar high frequency formulas for S and thermal conductivity.

Requirement::: $L_{ij}(\omega)$

Did not exist, so had lots of fun with Luttinger's formalism of a gravitational field, now made time dependent.

$$K_{tot} = \sum K(r)(1 + \psi(r,t))$$

$$\nabla(\psi(r,T)) \sim \nabla T(r,t)/T$$

$$
\begin{array}{cccc}
 & \text{i=1} & \text{i=2} & \\
 & \text{Charge} & \text{Energy} & \\
\mathcal{I}_i & \hat{J}_x(q_x) & \hat{J}^Q_x(q_x) & \\
\mathcal{U}_i & \rho(-q_x) & K(-q_x) & \\
\mathcal{Y}_i & E^x_q = iq_x\phi_q & iq_x\psi_q. & \qquad (1)
\end{array}
$$

$$
L_{ij}(\omega) = \frac{i}{\Omega\omega_c}\left[\langle\mathcal{T}_{ij}\rangle - \sum_{n,m}\frac{p_m - p_n}{\varepsilon_n - \varepsilon_m + \omega_c}(\mathcal{I}_i)_{nm}(\mathcal{I}_j)_{mn}\right], \qquad (1)
$$

$$
\langle\mathcal{T}_{ij}\rangle = -\lim_{q_x\to 0}\langle[\mathcal{I}_i, \mathcal{U}_j]\rangle\frac{1}{q_x}. \qquad (2)
$$

Stress tensor	Thermal operator	Thermoelectric operator
$\mathcal{T}_{11}$	$\mathcal{T}_{22}$	$\mathcal{T}_{12} = \mathcal{T}_{21}$
τ^{xx}	Θ^{xx}	Φ^{xx}
$-\frac{d}{dq_x}\left[\hat{J}_x(q_x), \rho(-q_x)\right]_{q_x\to 0}$	$-\frac{d}{dq_x}\left[\hat{J}^Q_x(q_x), K(-q_x)\right]_{q_x\to 0}$	$-\frac{d}{dq_x}\left[\hat{J}_x(q_x), K(-q_x)\right]_{q_x\to 0}$
		(1)

We thus see that a knowledge of the three operators gives us a interesting starting point for correlated matter:

$$\begin{aligned}
\text{High Freq Thermopower} \quad & S^* = \frac{\langle \Phi^{xx} \rangle}{T\langle \tau^{xx} \rangle} \\
\text{High Freq Lorentz Number} \quad & \mathbf{L}^* = \frac{\langle \Theta^{xx} \rangle}{T^2\langle \tau^{xx} \rangle} - (S^*)^2 \\
\text{High Freq Figure of Merit} \quad & \mathbf{Z}^* T = \frac{\langle \Phi^{xx} \rangle^2}{\langle \Theta^{xx} \rangle \langle \tau^{xx} \rangle - \langle \Phi^{xx} \rangle^2}.
\end{aligned} \tag{1}$$

$$\kappa_{zc}(\omega) = \frac{1}{T}\left[L_{22}(\omega) - \frac{L_{12}(\omega)^2}{L_{11}(\omega)}\right],$$

This leads to interesting sum rules a là the f-sum rule for conductivity.

$$\int_{-\infty}^{\infty} \frac{d\nu}{\pi} \Re e\ \kappa_{zc}(\nu) = \frac{1}{T\Omega}\left[\langle \Theta^{xx} \rangle - \frac{\langle \Phi^{xx} \rangle^2}{\langle \tau^{xx} \rangle}\right].$$

Thermo power operator for Hubbard model

$$\Phi^{xx} = -\frac{q_e}{2}\sum_{\vec\eta,\vec\eta',\vec r}(\eta_x+\eta'_x)^2 t(\vec\eta)t(\vec\eta')c^\dagger_{\vec r+\vec\eta+\vec\eta',\sigma}c_{\vec r,\sigma} - q_e\mu\sum_{\vec\eta}\eta_x^2 t(\vec\eta)c^\dagger_{\vec r+\vec\eta,\sigma}c_{\vec r,\sigma} + \frac{q_eU}{4}\sum_{\vec r,\vec\eta}t(\vec\eta)(\eta_x)^2(n_{\vec r,\bar\sigma}+n_{\vec r+\vec\eta,\bar\sigma})(c^\dagger_{\vec r+\vec\eta,\sigma}c_{\vec r,\sigma}+c^\dagger_{\vec r,\sigma}c_{\vec r+\vec\eta,\sigma}).$$

This object can be expressed completely in Fourier space as

$$\Phi^{xx} = q_e\sum_{\vec p}\frac{\partial}{\partial p_x}\left\{v_p^x(\varepsilon_{\vec p}-\mu)\right\}c^\dagger_{\vec p,\sigma}c_{\vec p,\sigma} + \frac{q_eU}{2\mathcal{L}}\sum_{\vec l,\vec p,\vec q,\sigma,\sigma'}\frac{\partial^2}{\partial l_x^2}\left\{\varepsilon_{\vec l}+\varepsilon_{\vec l+\vec q}\right\}c^\dagger_{\vec l+\vec q,\sigma}c_{\vec l,\sigma}c^\dagger_{\vec p-\vec q,\bar\sigma'}c_{\vec p,\bar\sigma'}.$$

$$\tau^{xx} = \frac{q_e^2}{\hbar}\sum\eta_x^2\,t(\vec\eta)\,c^\dagger_{\vec r+\vec\eta,\sigma}c_{\vec r,\sigma} \quad \text{or}$$

$$= \frac{q_e^2}{\hbar}\sum_{\vec k}\frac{d^2\varepsilon_{\vec k}}{dk_x^2}c^\dagger_{\vec k,\sigma}c_{\vec k,\sigma}$$

$$\Theta^{xx} = \sum_{p,\sigma} \frac{\partial}{\partial p_x} \left\{ v^x_{\vec{p}} (\varepsilon_{\vec{p}} - \mu)^2 \right\} \; c^\dagger_{\vec{p},\sigma} c_{\vec{p},\sigma} + \frac{U^2}{4} \sum_{\eta,\sigma} t(\vec{\eta}) \eta_x^2 (n_{\vec{r},\bar{\sigma}} + n_{\vec{r}+\vec{\eta},\bar{\sigma}})^2 c^\dagger_{\vec{r}+\vec{\eta},\sigma} c_{\vec{r},\sigma}$$

$$-\mu U \sum_{\vec{\eta},\sigma} t(\vec{\eta}) \eta_x^2 (n_{\vec{r},\bar{\sigma}} + n_{\vec{r}+\vec{\eta},\bar{\sigma}}) c^\dagger_{\vec{r}+\vec{\eta},\sigma} c_{\vec{r},\sigma}$$

$$-\frac{U}{8} \sum_{\vec{\eta},\vec{\eta}',\sigma} t(\vec{\eta}) t(\vec{\eta}') (\eta_x + \eta'_x)^2 \left\{ 3 n_{\vec{r},\bar{\sigma}} + n_{\vec{r}+\vec{\eta},\bar{\sigma}} + n_{\vec{r}+\vec{\eta}',\bar{\sigma}} + 3 n_{\vec{r}+\vec{\eta}+\vec{\eta}',\bar{\sigma}} \right\} c^\dagger_{\vec{r}+\vec{\eta}+\vec{\eta}',\sigma} c_{\vec{r},\sigma}$$

$$+\frac{U}{4} \sum_{\vec{\eta},\vec{\eta}',\sigma} t(\vec{\eta}) t(\vec{\eta}') (\eta_x + \eta'_x) \eta'_x c^\dagger_{\vec{r}+\vec{\eta},\sigma} c_{\vec{r},\sigma} \left\{ c^\dagger_{\vec{r}+\vec{\eta},\bar{\sigma}} c_{\vec{r}+\vec{\eta}+\vec{\eta}',\bar{\sigma}} + c^\dagger_{\vec{r}-\vec{\eta}',\bar{\sigma}} c_{\vec{r},\bar{\sigma}} - h.c. \right\} . \quad (1)$$

Unpublished- For Hubbard model using “Ward type identity” can show a simpler result for \Phi.

$$\langle \Phi^{xx} \rangle = \frac{q_e}{c} k_B T \; \sum_{m,\sigma,\vec{k}} G_\sigma(k, i\omega_m) \left[\frac{d}{dk_x} \; (v^x_k (\varepsilon_k - \mu)) \; + \frac{d^2 \varepsilon_k}{dk_x^2} \; \Sigma_\sigma(k, i\omega_m) \right]$$

Hydrodynamics of energy and charge transport in a band model:

This involves the fundamental operators in a crucial way:

$$\left\{\frac{\partial}{\partial t}+\frac{1}{\tau_c}\right\}\delta J(r)=\frac{1}{\Omega}\langle\tau^{xx}\rangle\left[\frac{1}{q_e^2}\frac{\partial\mu}{\partial n}(-\nabla\rho)-\nabla\phi(r)\right]+\frac{1}{\Omega}\langle\Phi^{xx}\rangle\left[\frac{1}{C(T)}(-\nabla K(r))-\nabla\Psi\right]$$

$$\left\{\frac{\partial}{\partial t}+\frac{1}{\tau_E}\right\}\delta J^Q(r)=\frac{1}{\Omega}\langle\Phi^{xx}\rangle\left[\frac{1}{q_e^2}\frac{\partial\mu}{\partial n}(-\nabla\rho)-\nabla\phi(r)\right]+\frac{1}{\Omega}\langle\Theta^{xx}\rangle\left[\frac{1}{C(T)}(-\nabla K(r))-\nabla\Psi\right]$$

Einstein diffusion term of charge

Energy diffusion term

$$\frac{\partial\rho}{\partial t}+\nabla J(r)=0$$

$$\frac{\partial K(r)}{\partial t}+\nabla J^Q(r)=p_{ext}(r)$$

Continuity

Input power density

These eqns contain energy and charge diffusion, as well as thermoelectric effects. Potentially correct starting point for many new nano heating expts with lasers.

And now for some results:

Triangular lattice t-J exact diagonalization (full spectrum)

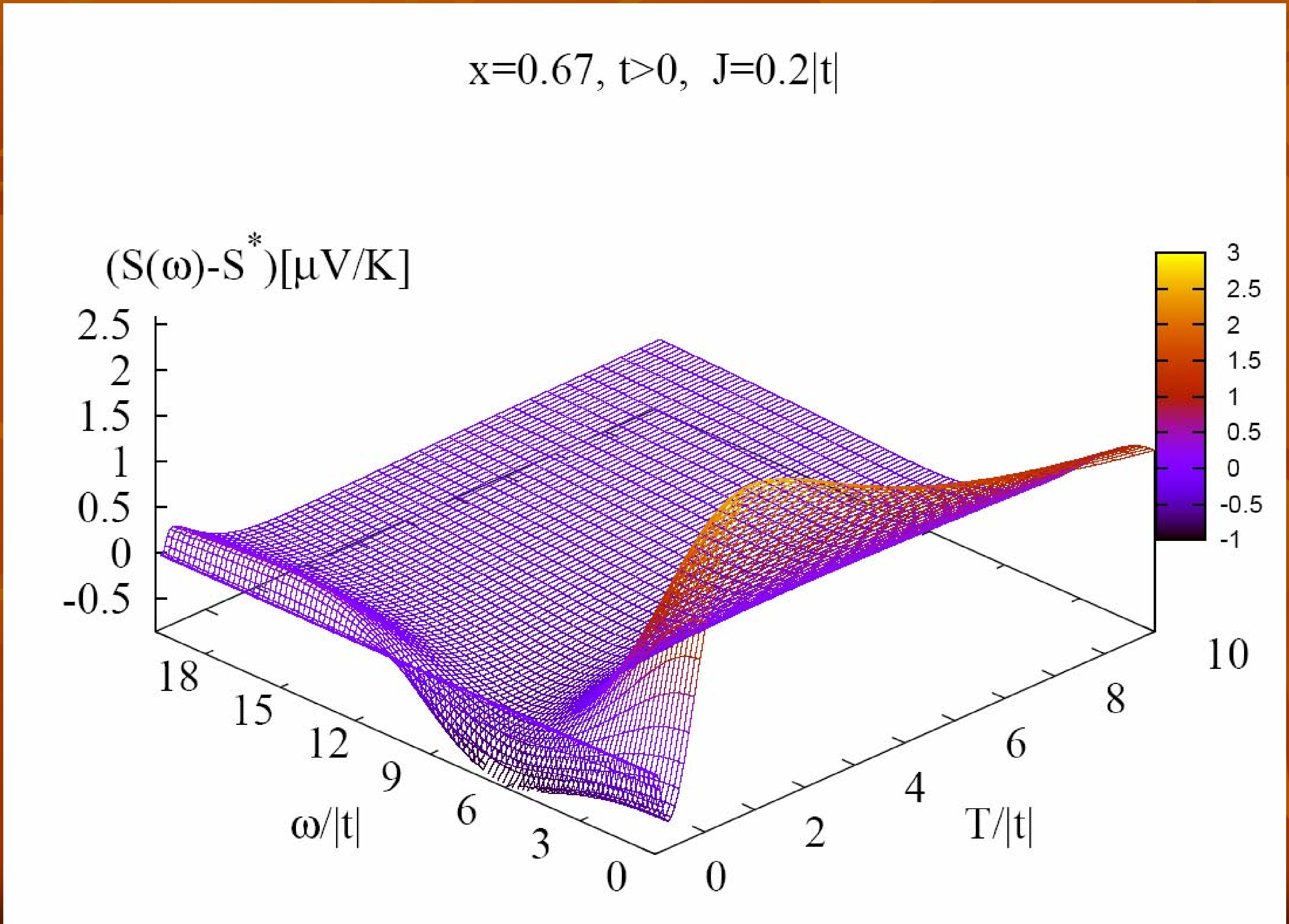
Collaboration and hard work by:-

J Haerter, M. Peterson, S. Mukerjee (UC Berkeley)

How good is the S* formula compared to exact Kubo formula?

A numerical benchmark: Max deviation 3% anywhere !!

As good as exact!



Results from this formalism:

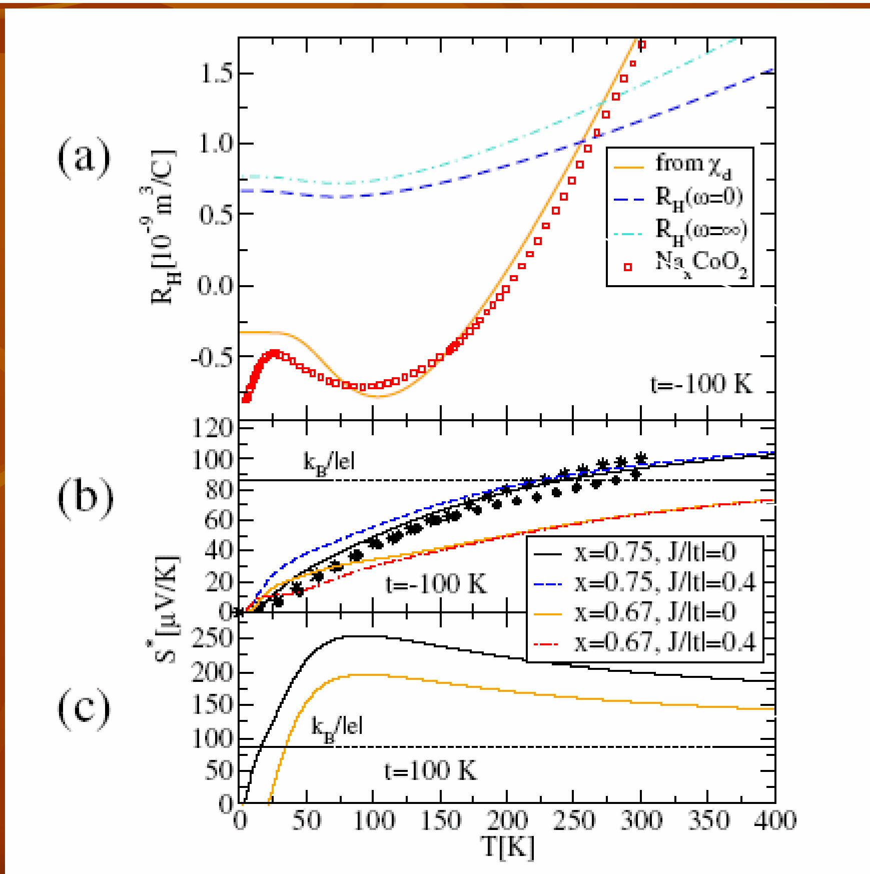
PRL 97, 226402 (2006) PHYSICAL REVIEW LETTERS week ending 1 DECEMBER 2006

Strong Correlations Produce the Curie-Weiss Phase of Na_xCoO_2

Jan O. Haerter, Michael R. Peterson, and B. Sriram Shastry

Physics Department, University of California, Santa Cruz, California 95064, USA

(Received 21 July 2006; published 28 November 2006)



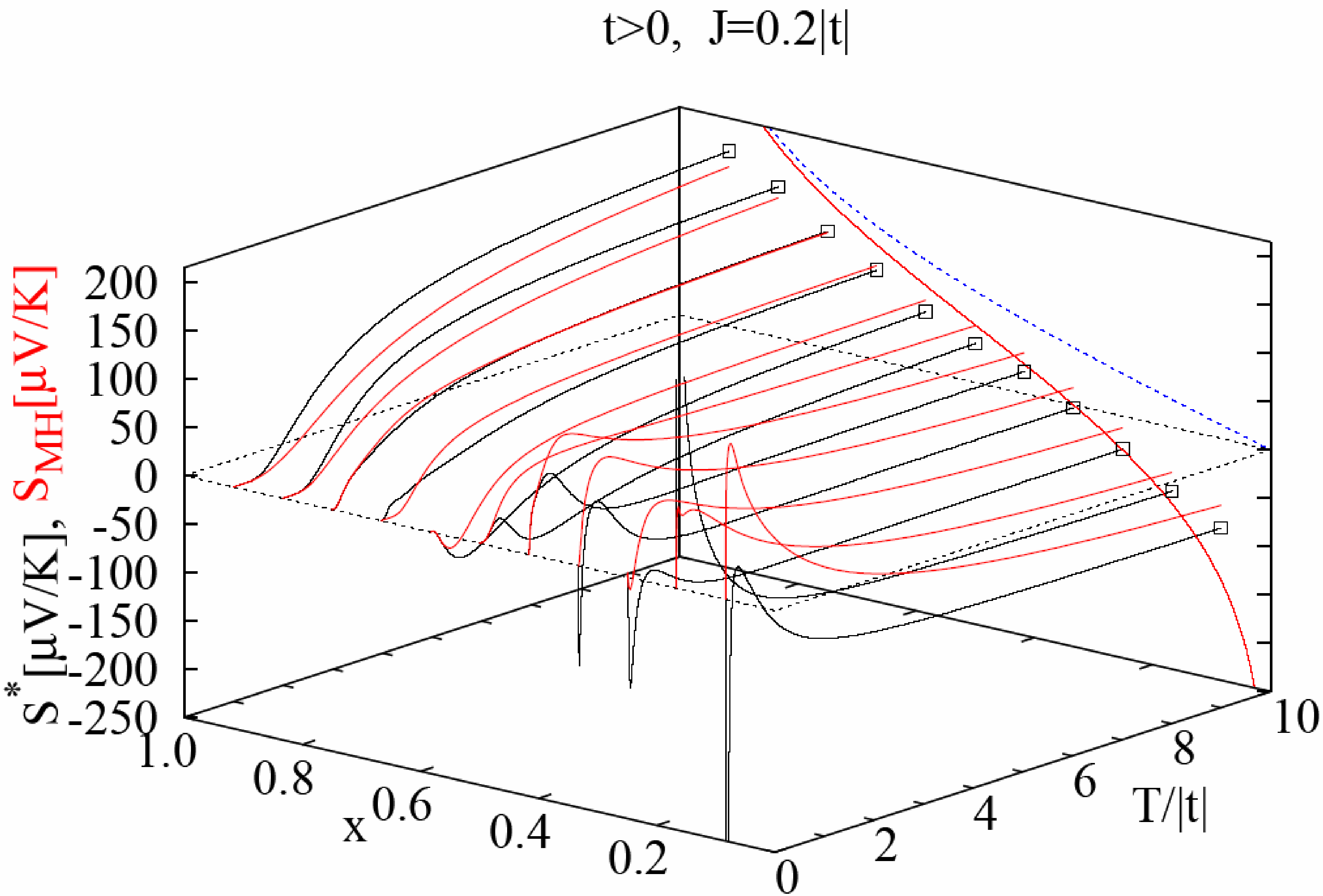
T linear Hall constant for triangular lattice predicted in 1993 by Shastry Shraiman Singh! Quantitative agreement hard to get with scale of "t"

Comparision with data on absolute scale!

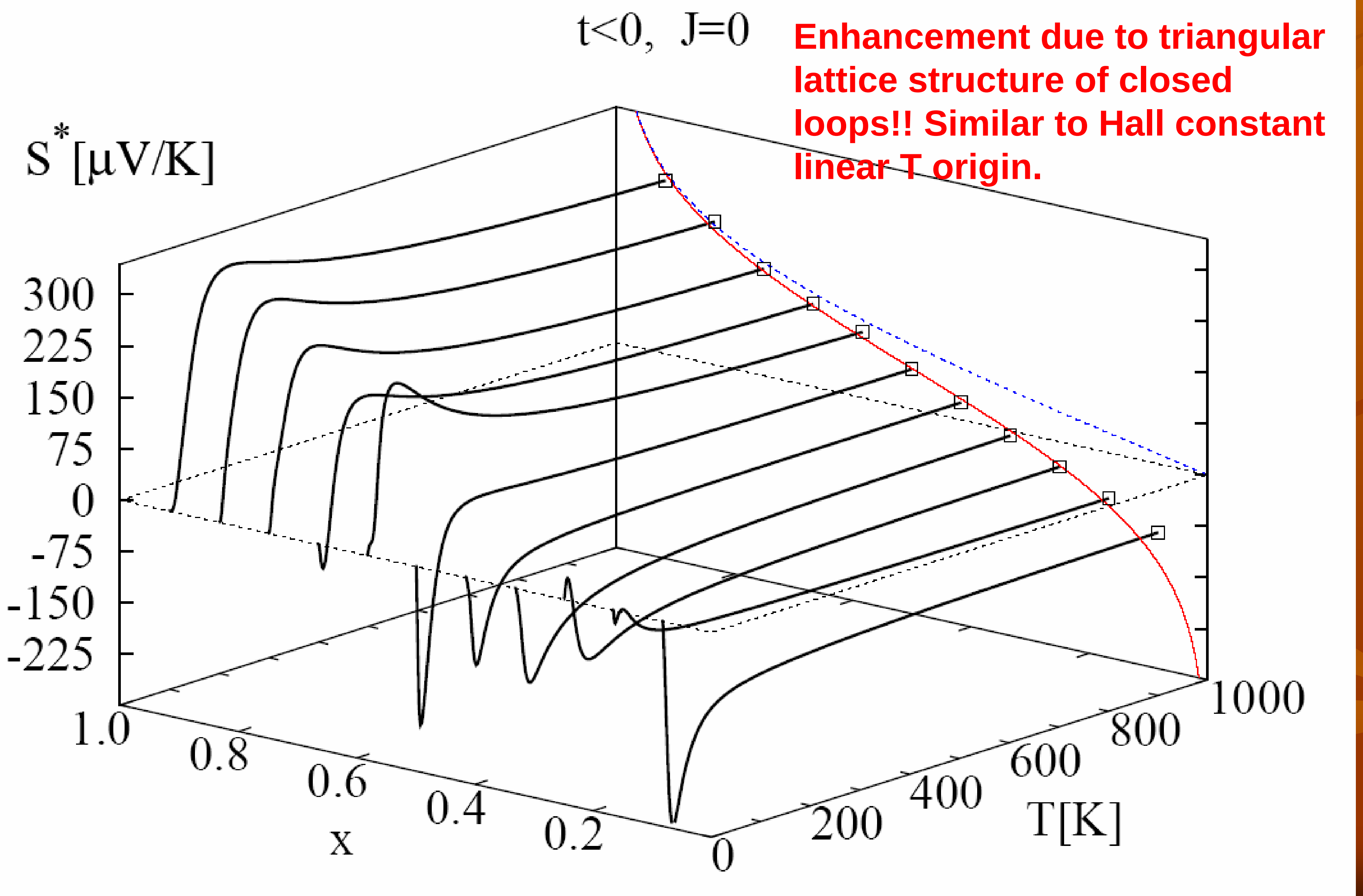
Prediction for t>0 material

S* and the Heikes Mott formula (red) for Na_xCo O2.

Close to each other for t>o i.e. electron doped cases

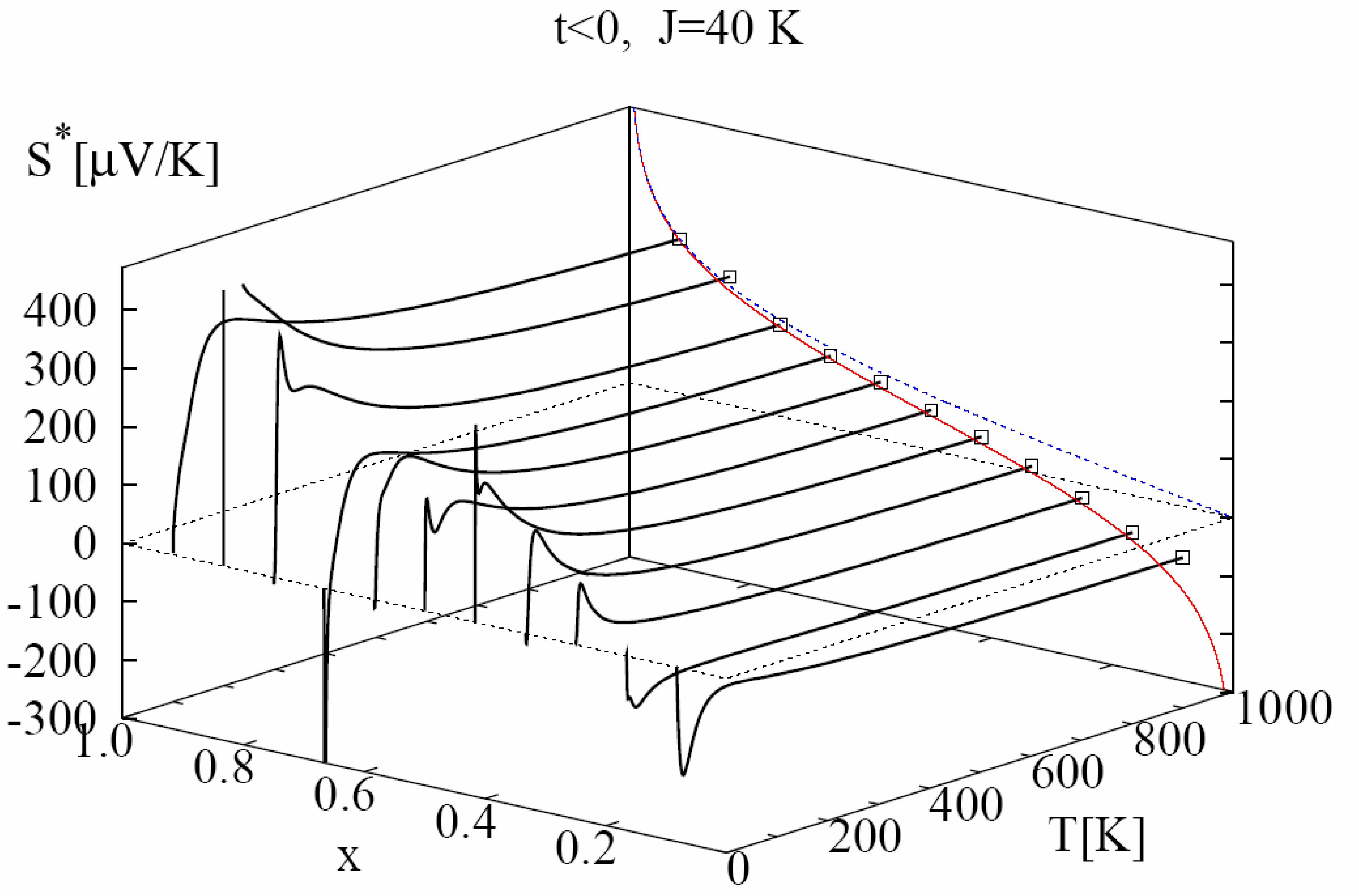


Predicted result for t<0 i.e. fiducary hole doped CoO_2 planes. Notice much larger scale of S* arising from transport part (not Mott Heikes part!!).



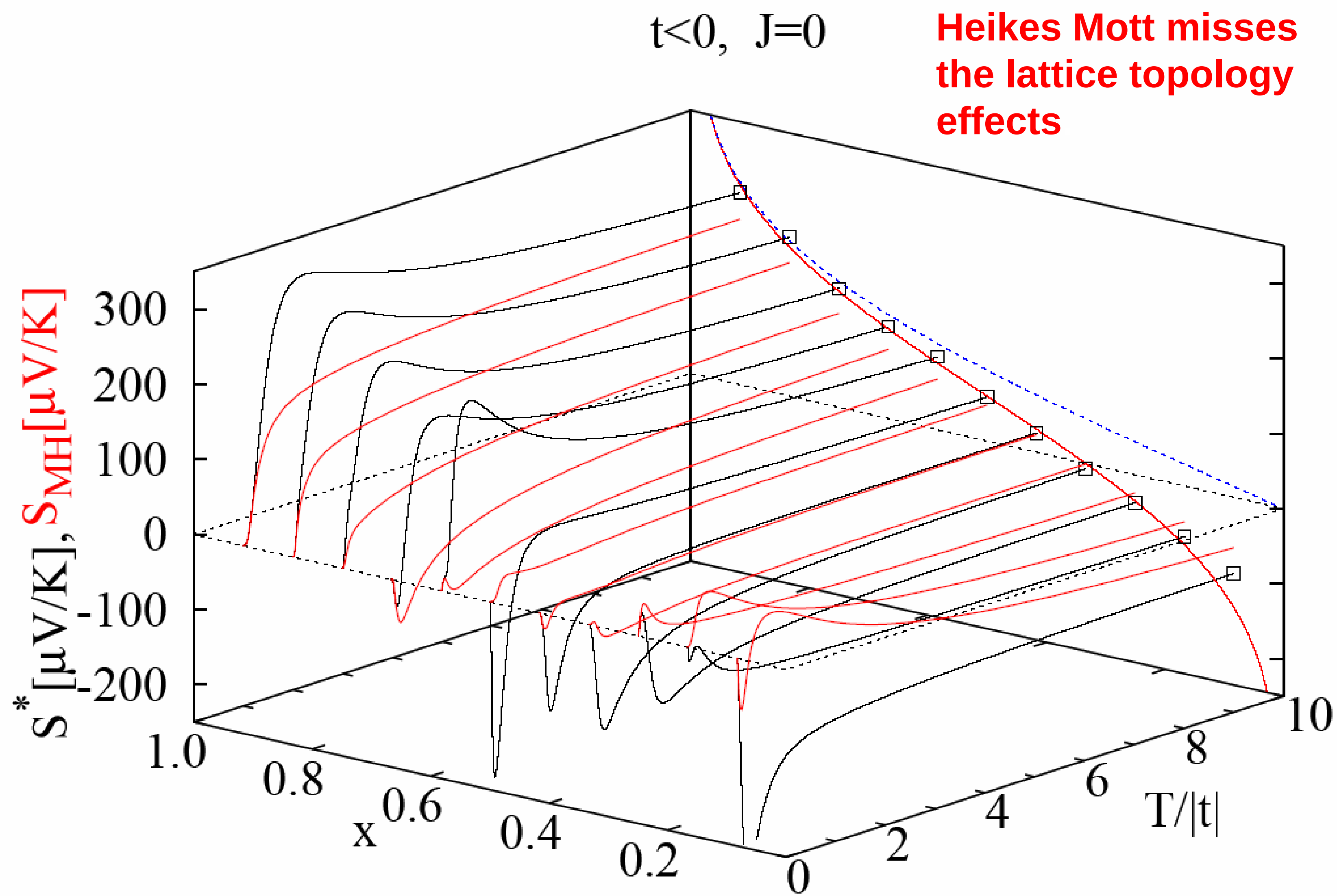
Predicted result for t<0 i.e. fiducary hole doped CoO_2 planes.

Different J higher S.



Predictions of S* and the Heikes Mott formula (red) for fiducary hole doped CoO2.

Notice that S* predicts an important enhancement unlike Heikes Mott formula



Z*T computed from S* and Lorentz number. Electronic contribution only, no phonons. Clearly large x is better!!

Quite encouraging.