A Sum Rule for Thermal Conductivity and Dynamical Thermal Transport Coefficients in Condensed Matter

Sriram Shastry
UCSC, Santa Cruz, CA

Work supported by
DMR 0408247

Motivation:

Experiments:

$Na_x Co O_2$:Happy symbiosis of basic theory and technology.

- Large Thermo power
- Huge B dependence of Thermopower
- Strange Hall constant

Skutterudites:

- Cages and Rattlers; or how to manipulate lattice thermal conductivity

Heavy Fermi Systems

Theory:

- Updating Boltzmann theory: long lived almost free quasi particles are not a good starting point for most of these materials.
- Effect of strong correlations on transport.
- Understanding Mott Hubbard physics: what are holes?

A rhetorical question: What is a hole?

K-space point of view A. H. Wlison (1935)

Real space point of view: Mott Hubbard holes

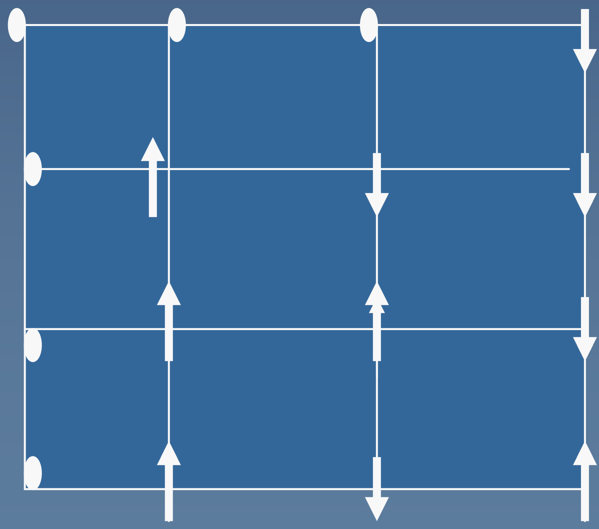
Imagine a band with strong correlations and n= number of electrons per site. As n~1 we approach the Mott insulating state, so should we think of it as a system with x=(1-n) holes?

HALL CONSTANT is a good example to think about.

Hall Effect in Strongly Correlated Matter

Standard expression says that Hall constant is a measure of carrier concentration:

$$R_H = 1/nec$$

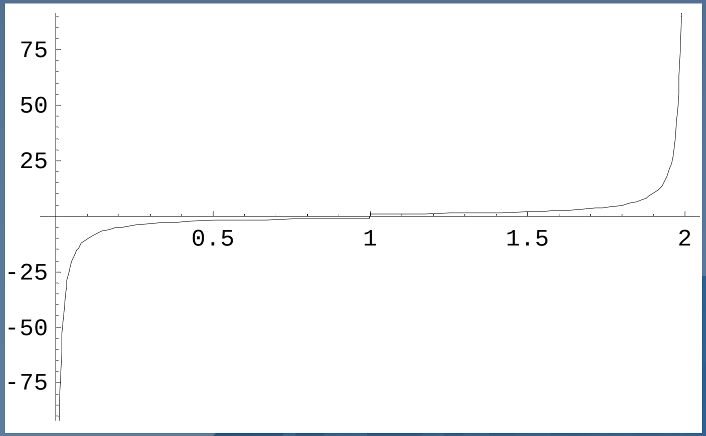


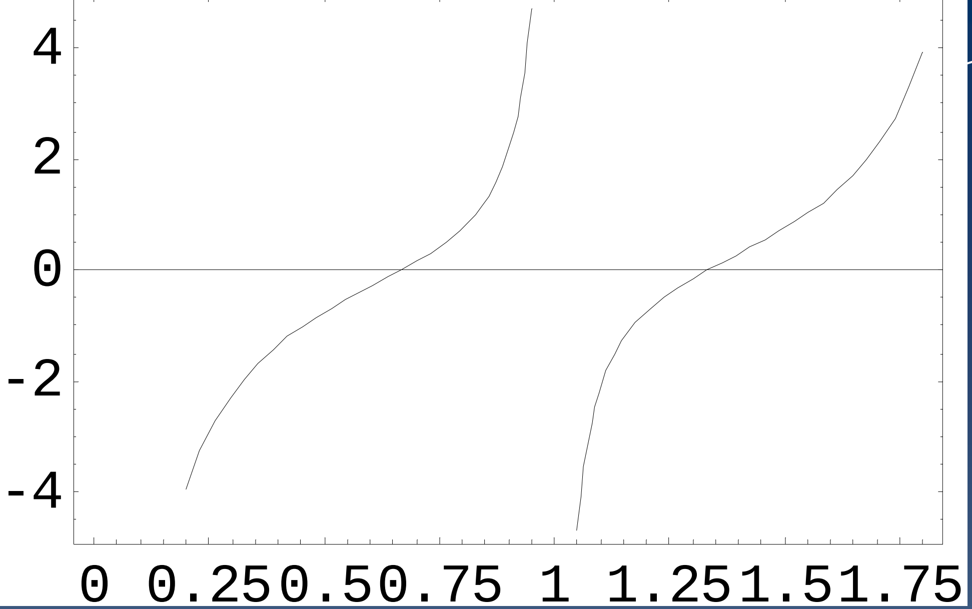
Question: What is "n" for a Mott Hubbard system? Electron number of hole number (measured from half filling)?

Real space versus k space!!

$$-1/|e|n$$

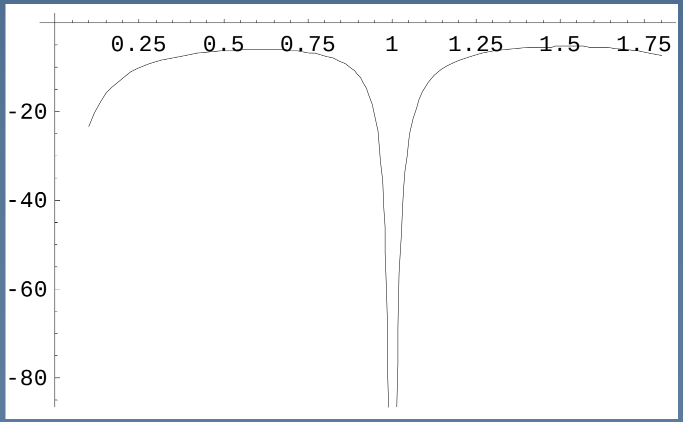
Naïve expectation from Band picture

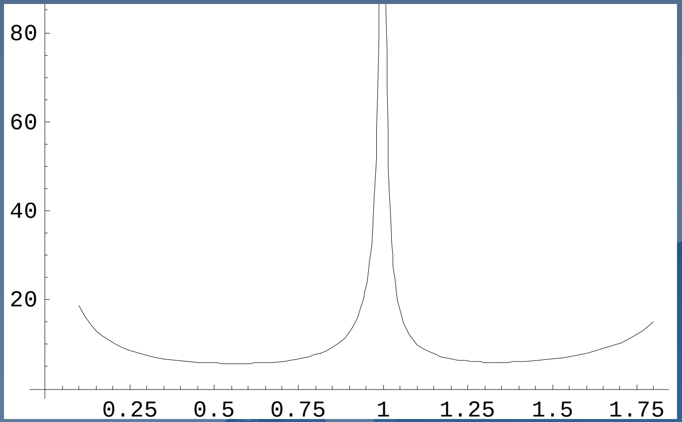




Behaviour for square lattice Mott Hubbard system. Also expected for triangular lattice at low T (work in progress). Notice there are THREE zero crossings

t<0 Triangular lattice at T> |t| is always hole like. No zero crossings in either case.





Question: What about the Fermi surface curvature and all that!!

$$\sigma_{xy} = 2(e^3/\hbar)B \sum_{\mathbf{k}} \left[\frac{-\partial f_{\mathbf{k}}}{\partial \varepsilon}\right](v_y \tau_{\mathbf{k}}) \left[v_y \left[\frac{\partial}{\partial k_x}\right] - v_x \left[\frac{\partial}{\partial k_y}\right]\right](v_x \tau_{\mathbf{k}})$$

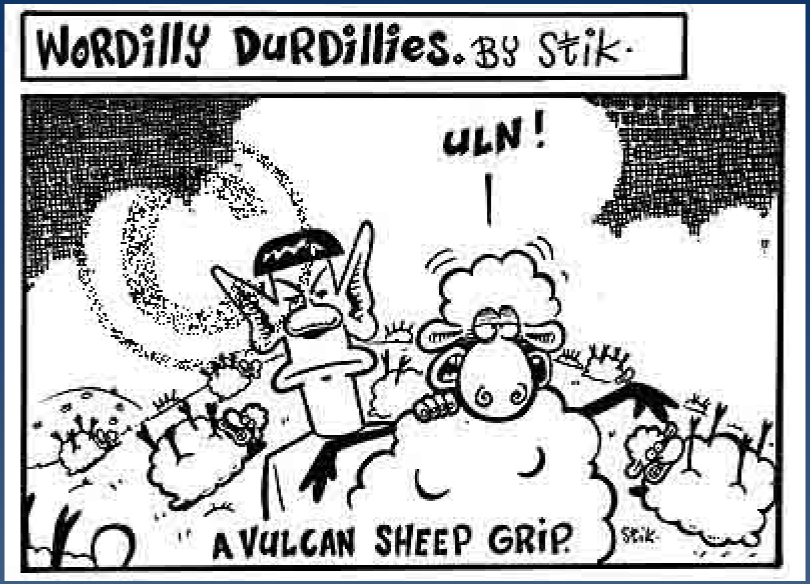
Vulcan Death Grip of the Boltzmann approach view:

Need to break out.

Hall constant is a reactive response and the fact that we can express it as an integral over the Fermi surface curvature for free electron theory is EXCEEDINGLY misleading. It is true only for free electrons and there is no reason why a reactive response is to be expressed in terms of delta functions conserving energy over the FS. Same fact also realized in QHE literature.

Valid Theoretical Possibility that:

Fermi surface curvature remains electron like whereas Hall constant changes sign.



wikipedia

Vulcan death grip –
A variant of Vulcan nerve pinch derived from a Star Trek classic episode where a **non- existant** "Vulcan death grip" was used to fool Romulans that Spock had killed Kirk.

How do we calculate any of these things reliably? T-J models are the correct framework but notoriously hard

- Auxiliary fields: slave bosons/fermions/... nice but hardly reliable in the phyisical cases
- Numerical techniques QMC, diagonalization, High Temperature expansions,,promising but defeated by Kubo formulas involving sums over states and a tricky DC limit in most cases.

Shastry, Shraiman Singh (1993) broke new ground for Hall constant. Basic idea: :

Think high frequency

First serious effort to understand Hall constant in correlated matter:

S S, Boris Shraiman and Rajiv Singh, Phys Rev Letts (1993)

Introduced object

$$R_H^* = \lim_{B \to 0} \lim_{\omega \to \infty} \rho_{xy}(\omega) / B$$

- **Easier to calculate than transport Hall constant**
- **Captures Mott Hubbard physics to large extent**

Motivation: **Drude theory** has

$$\sigma_{xy}(\omega) = \sigma_{xy}(0)/(1+i\omega\tau)^2$$

$$\sigma_{xx}(\omega) = \sigma_{xx}(0)/(1+i\omega\tau)$$

Hence **relaxation time cancels out in the Hall resistivity**

$$\rho_{xy}(\omega) = \frac{\sigma_{xy}}{(\sigma_{xx})^2}$$

Why not compute at high frequencies from Kubo's formulas directly:

$$\sigma_{xx}(\omega) = \frac{i}{N\mathrm{v}\omega}\left[<\tau_{xx}> - \tfrac{1}{Z}\sum \frac{e^{-\beta\varepsilon_n} - e^{-\beta\varepsilon_m}}{\varepsilon_m - \varepsilon_n - \omega} |< n \,|\, J^x \,|\, m >|^2 \right]$$

$$= \frac{i<\tau_{xx}>}{N\mathrm{v}\omega} + o(1/\omega^3)$$

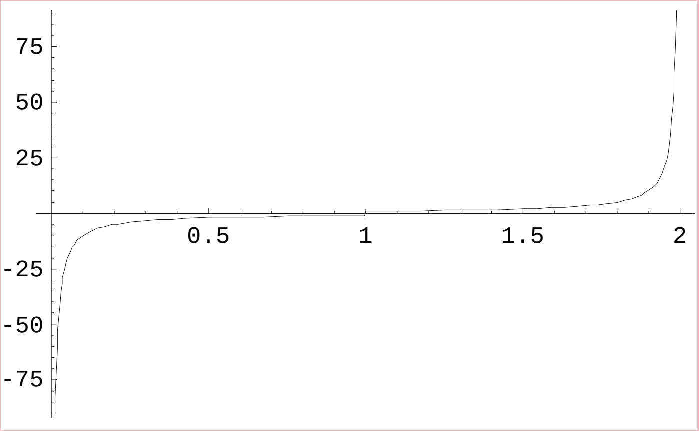
Here τ is the stress tensor (k.e.) and v the cell volume

$$\sigma_{xy}(\omega) = \frac{-i}{N\mathrm{v}\omega Z}\sum \frac{e^{-\beta\varepsilon_n} - e^{-\beta\varepsilon_m}}{\varepsilon_m - \varepsilon_n - \omega} < n \,|\, J^x \,|\, m >< m \,|\, J^y \,|\, n >$$

$$= \frac{i}{Nv\omega^2} < [J^x, J^y] > + o(1/\omega^3)$$

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

- 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, not too useful for Hubbard model.
  - This asymptotic formula usually requires ω to be larger than J



Naïve expectation from Band theory for Hall constant with one zero crossing (at half filling).

To leading order in T/J we find

$0 < n < 1$

$$R = -\frac{v}{|e|c}\left[\frac{kT}{4t}\frac{(2-n)}{n(1-n)} + \frac{(1-n/2)(1-3n/2)}{n(1-n)}\right]$$

Tr and square lattice

$1<n<2$

Tr lattice only

$$R = -\frac{v}{|e|c}\left[\frac{kT}{4t}\frac{(n)}{(2-n)(n-1)} + \frac{n(4-3n)}{4(2-n)(n-1)}\right]$$

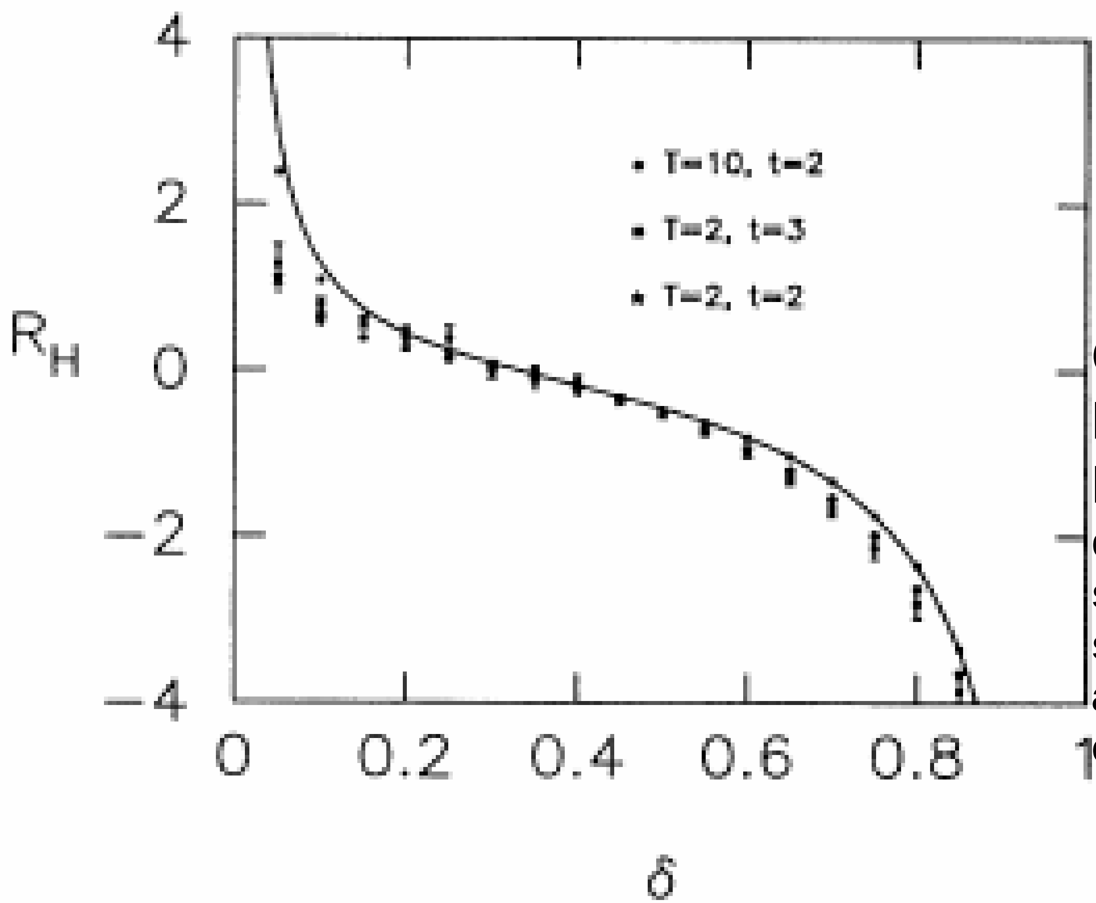
VOLUME 70, NUMBER 13 PHYSICAL REVIEW LETTERS 29 MARCH 1993

Faraday Rotation and the Hall Constant in Strongly Correlated Fermi Systems

B. Sriram Shastry and Boris I. Shraiman
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

Rajiv R. P. Singh
University of California, Davis, California 95616

(Received 30 December 1992)



Comparison with Hidei Takagi and Bertram Batlogg
data for LSCO showing change of sign of Hall constant at delta=.33 for squar e lattice

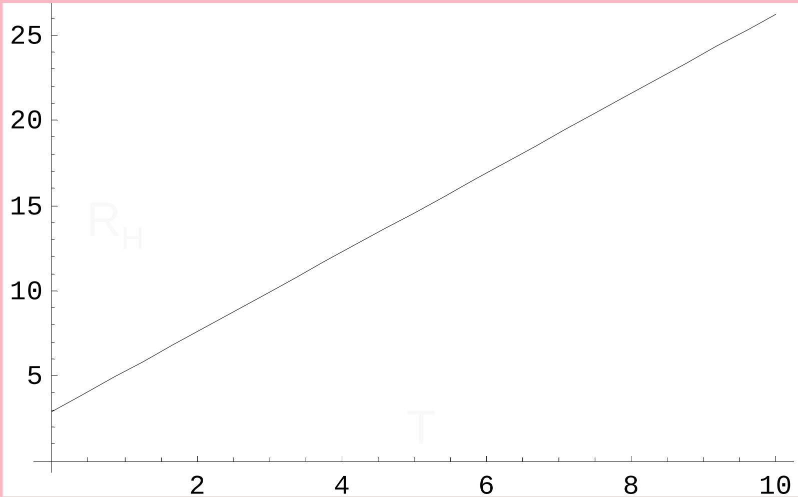
[10] The lattice structure and the statistics of the particles plays a crucial role in the behavior of R_H^*, and through it on $R_H(\omega)$. This is brought out in a calculation of the leading high-temperature behavior of R_H^* in the triangular lattice t-J model. We find $R_H^* \sim (\beta t)^{-1}\frac{1+\delta}{\delta(1-\delta)}$, which in contrast to the square lattice result, Eq. (11), does not change sign with δ, but rather with t. Furthermore $R_H^* \sim T$ so that, in a sense, the semiclassical limit $1/ne$ does not exist at all. This highly nontrivial behavior is a consequence of a "fermionic frustration" on the triangular lattice, the same calculation for hard core bosons gives $R_H^* \sim -(\beta t)^{-1}\frac{1-3\delta}{\delta(1-\delta)}$, which indeed changes sign at $\delta = \frac{1}{3}$.

$$R_H^* = -\frac{v}{4|e|}\frac{k_B T}{t}\frac{1+\delta}{\delta(1-\delta)}$$

Here $\delta = \rho - 1$.

Since Fermi temperature seems low, the large T limit may work, **so we predict:** R_H will not saturate with T.

Predict linear T dependence and known slope.

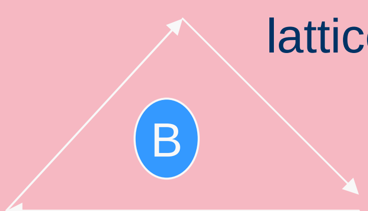


As a function of T, Hall constant is LINEAR for triangular lattice!!

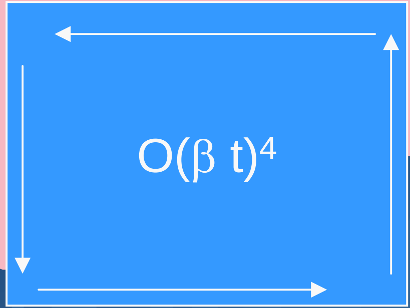
We suggest that transport Hall = high frequency Hall constant!!

- **Origin of T linear behaviour in triangular lattice has to do with frustration. Loop representation of Hall constant gives a unique contribution for triangular lattice with sign of hopping playing a non trivial role.**

Triangular lattice



$O(\beta t)^3$

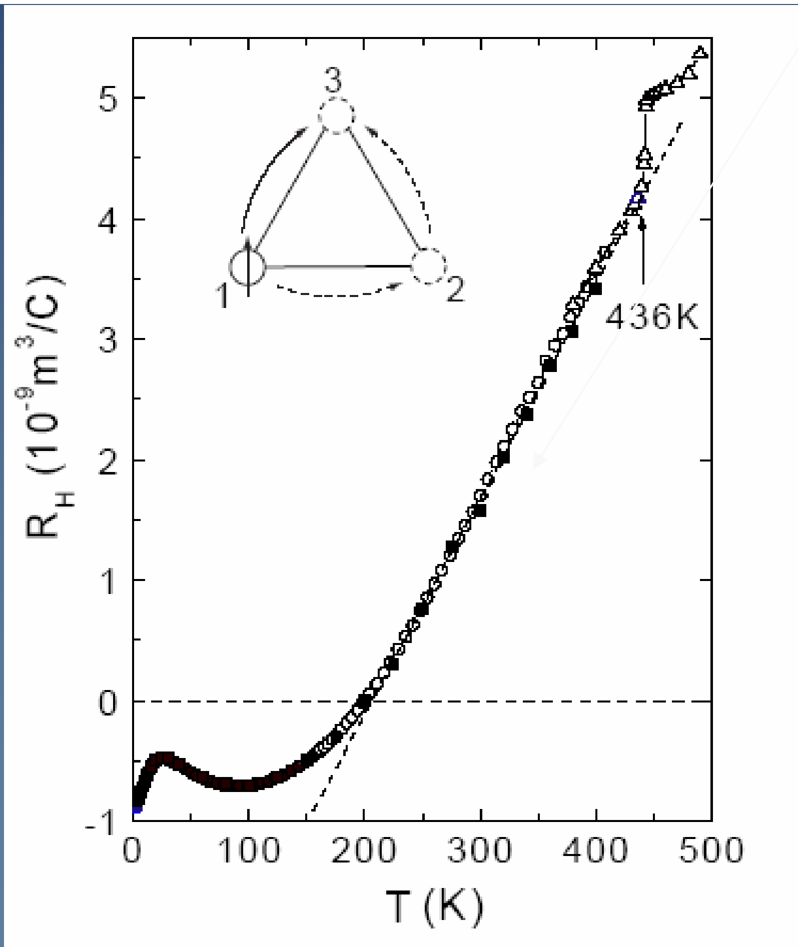


square lattice

Anomalous high-temperature Hall effect on the triangular lattice in Na_xCoO_2

Yayu Wang[1], Nyrissa S. Rogado[2], R. J. Cava[2,3], and N. P. Ong[1,3]

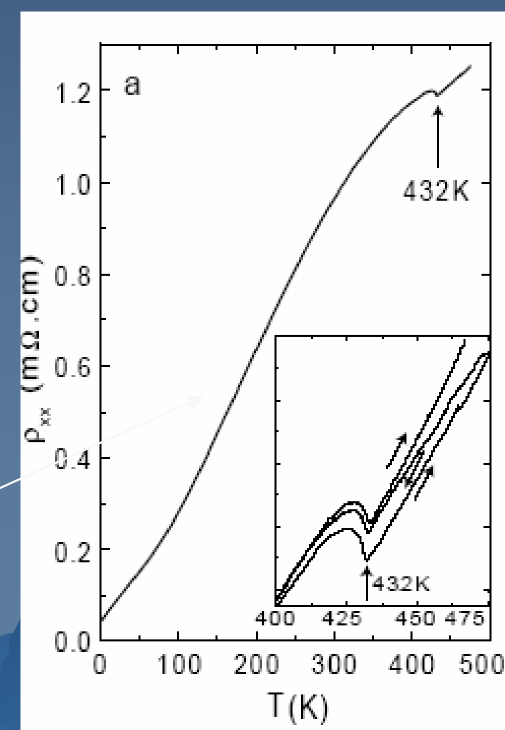
The Hall coefficient R_H of Na_xCoO_2 ($x = 0.68$) behaves anomalously at high temperatures (T) From 200 to 500 K, R_H increases linearly with T to 8 times the expected Drude value, with no sign of saturation. Together with the thermopower Q, the behavior of R_H provides firm evidence fo strong correlation. We discuss the effect of hopping on a triangular lattice and compare R_H with a recent prediction by Kumar and Shastry.



Hall constant as a function of T for x=.68 (CW metal). T linear over large range 200⁰ to 436⁰ (predicted by theory of triangular lattice transport KS)

STRONG CORRELATIONS & Narrow Bands

T Linear resistivity



Spin entropy as the likely source of enhanced thermopower in $Na_xCo_2O_4$

Yayu Wang*, Nyrissa S. Rogado†, R. J. Cava†‡ & N. P. Ong*‡

NATURE | VOL 423 | 22 MAY 2003 |

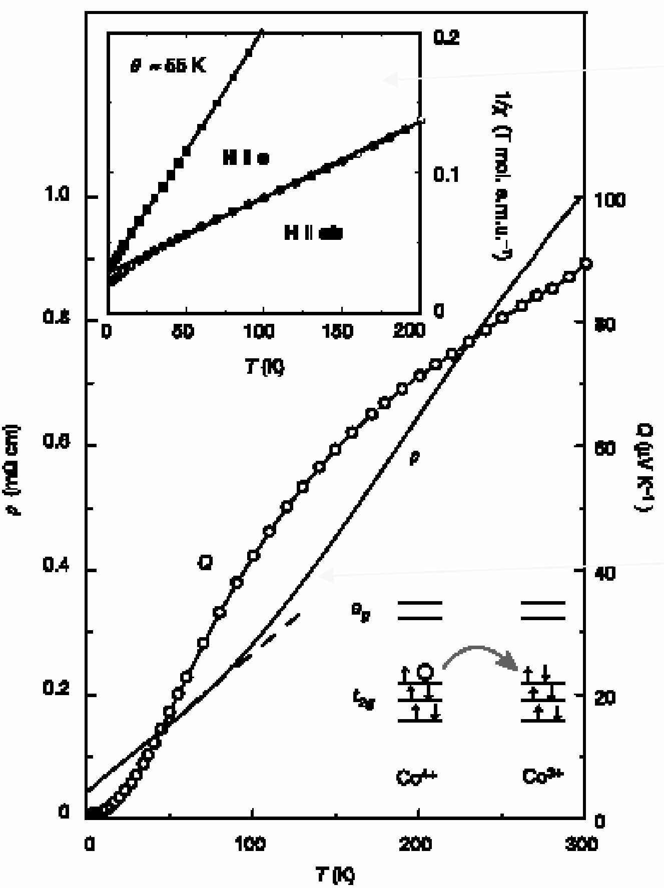


Figure 1 The temperature (*T*) dependence of magnetic and transport properties of single-crystal $Na_xCo_2O_4$ and electronic states in the Co ions. The in-plane thermopower *Q*

Static χ inverse looks linear already at 50^0, this is remarkable for a good metal (ρ~ .1 mΩ cm)!!

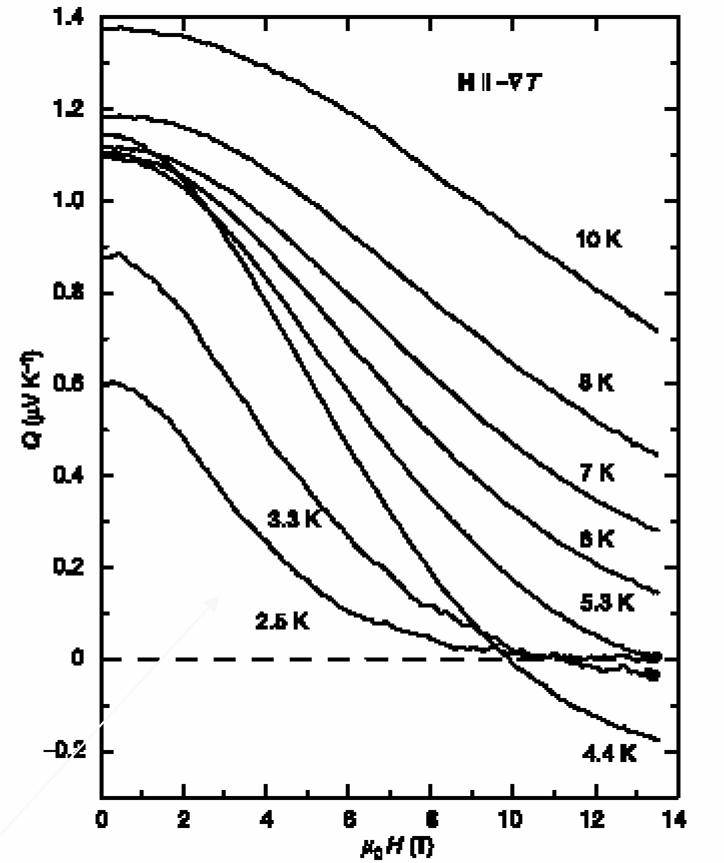
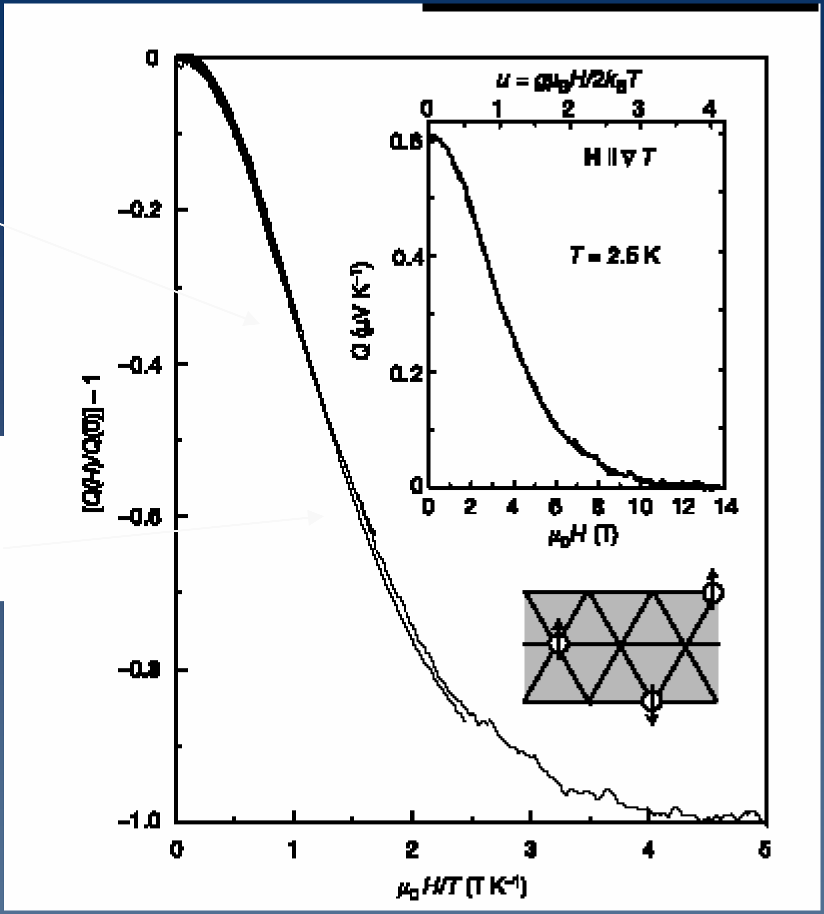


Figure 2 The in-plane thermopower *Q* versus an in-plane **H**||(−∇*T*) at selected *T*. At

Strongly magnetic field dependent thermopower, already anomolous due to large magnitude

All data for different T collapses to single curve



Entropy of free spin ½ particles!!

$$\sigma(H,T)/\sigma(0,T) = \{\ln[2\cosh(u)] - u\tanh(u)\}/\ln(2)$$

$$u = g\mu_B H/2k_B T.$$

Remarkably successful fit possible to Heikes Zener Mott formula for thermopower, interpreted as spin contribution of entropy per particle (i.e. neglecting transport issues, a great simplification of Kubo's exact formula)

G Beni (1974), P Chaikin and Beni for Hubbard model in limit of t->0:

Success is quite surprising, and hints at localized character of electron spins despite being a good conductor.

Triangular lattice: High Temperature expansion result is a **prediction**, namely non saturation of R_H and that the slope is a measure of bandwidth.

Many things to do further: currently under study

ω T dependence.

Experimentally: Wish list: Can someone help with determining the R(ω) curve from optics to dc?

Lesson learnt from Hall study: Find appropriate combinations of Kubo response functions that may be insensitive to frequency.

YES

$$\rho_{xy}(\omega)$$

NO

$$\sigma_{xy}(\omega)$$

$$\sigma_{xx}(\omega)$$

A Sum Rule for Thermal Conductivity and Dynamical Thermal Transport Coefficients in Condensed Matter -I

Cond-mat/0508711

S S Summer 05, August 29, 05

New formalism with new results:->

- Thermal conductivity sum rule, analogous to plasma sum rule
- Thermo-power formula that is better than the Heikes- Mott - Zener formula. Transport contribution is evaluated and is correct at ALL temperatures for the free electron case, and presumably close to DC answer at all frequencies
- Thermoelectric figure of merit
- Lorentz ratio
- Nernst effect

Explicit results for NCO- a useful prediction regarding design of higher Thermopower materials.

WORK in progress-

$$R_H^* = \lim_{B\to 0;\omega\to\infty} \frac{1}{B}\rho_{xy}(\omega)$$

Formalism gives results that are typically the analogs of the Hall constant R* and are equal time correlators of non trivial operators. Hard to evaluate, but not impossible, unlike the DC counterparts.

Need frequency dependence of various objects

$\kappa(\omega)$ $\rho(\omega)$ $S(\omega)$ $L(\omega)$ $Z(\omega)$

$$K = K_0 + K_1 e^{-i\omega_c t},$$

with adiabatic switching from the infinitely remote past $t = -\infty$ as usual, and $K_0 = \sum_r K(r) = \sum_r (H(r) - \mu\ n(r))$. Here $H(r)$ is the energy density, and since we are mainly dealing with lattice models, we sum over r. The operator

$$K_1 = \sum_r \psi(r) K(r),$$

where $\psi(r)$ is a small (pseudo) gravitational field with some spatial variation such that its average is zero.

Physical idea of Luttinger is that $\psi(r)$ is a pseudo temperature, it is a mechanical analog of temperature. The action $\beta(r)K(r)$ tries to be locally constant, and so if $\psi(r)$ increases locally then T increases locally too. Finally use Einstein relation to relate the response to $\nabla\psi$ to that to ∇T, but basically think of

$$\nabla\psi(r) = \frac{\nabla T(r)}{T}$$

Kubo Formulas at finite frequencies: (in fact infinite)

Somewhat strangely, it is better to do the case of general non dissipative systems and then to specialize to dissipative systems

$$\sigma(\omega_c) = \frac{i}{\hbar\omega_c} D_c + \frac{i\hbar}{\mathcal{L}} \sum_{n,m} \frac{p_n - p_m}{\epsilon_m - \epsilon_n} \frac{|\langle n|\hat{J}_x|m\rangle|^2}{\epsilon_n - \epsilon_m + \hbar\omega_c}.$$

where the charge or Meissner stiffness is given by

$$D_c = \frac{1}{\mathcal{L}} \left[\langle \tau^{xx} \rangle - \hbar \sum_{n,m} \frac{p_n - p_m}{\epsilon_m - \epsilon_n} |\langle n|\hat{J}_x|m\rangle|^2 \right].$$

The more familiar superfluid density $\rho_s(T)$ is defined in terms of D_c by

$$\frac{q_e^2 \rho_s(T)}{m} = \frac{D_c(T)}{\hbar}.$$

We may rewrite this in a more compact form as

$$\sigma(\omega_c) = \frac{i}{\hbar\omega_c} D_c + \frac{1}{\mathcal{L}} \int_0^\infty dt\, e^{i\omega_c t} \int_0^\beta d\tau \langle \hat{J}_x(-t - i\tau)\hat{J}_x(0)\rangle, .$$

Usual Kubo formula does not have first term

The sum rule for the real part of the conductivity (an even function of ω) follows as

$$\int_0^\infty Re\,\sigma(\omega)d\omega = \frac{\pi}{2\hbar\mathcal{L}}\langle\tau^{xx}\rangle.$$

For normal systems, D_c is zero, but whether it is zero or not the sum rule is valid. In cases where Dc=0,

$$\frac{1}{\mathcal{L}}\int_0^\beta d\tau\langle\hat{J}_x(-i\tau)\hat{J}_x(0)\rangle = \frac{1}{\mathcal{L}}\langle\tau^{xx}\rangle$$

Therefore one can get the result of the sum rule more easily from RHS than from LHS. We next play the same game with thermal conductivity. Here the fundamental paper is due to Luttinger (1963)

Grand canonical ensemble perturbation theory with a pseudo graviational field:

$$\Theta^{xx} = - \lim_{k_x \to 0} \frac{d}{dk_x} [\hat{J}_x^Q(k_x), \hat{K}(-k_x)]$$

Here we commute the Heat current with the energy density to get the thermal operator

$$\begin{aligned} Re\ \kappa(\omega) &= \frac{\pi}{\hbar T} \delta(\omega) \bar{D}_Q + Re\ \kappa_{reg}(\omega) \ \text{ with} \\ Re\ \kappa_{reg}(\omega) &= \frac{\pi}{T \mathcal{L}} \left(\frac{1 - e^{-\beta\omega}}{\omega} \right) \sum_{\epsilon_n \neq \epsilon_m} p_n |\langle n | \hat{J}_x^Q | m \rangle|^2 \delta(\epsilon_m - \epsilon_n - \hbar\omega), \\ \bar{D}_Q &= \frac{1}{\mathcal{L}} \left[\langle \Theta^{xx} \rangle - \hbar \sum_{\epsilon_n \neq \epsilon_m} \frac{p_n - p_m}{\epsilon_m - \epsilon_n} |\langle n | \hat{J}_x^Q | m \rangle|^2 \right]. \end{aligned}$$

Printing error

The sum rule for the real part of the thermal conductivity (an even functi
of ω) follows

$$\int_0^\infty Re\ \kappa(\omega) d\omega = \frac{\pi}{2 \hbar T \mathcal{L}} \langle \Theta^{xx} \rangle.$$

Comment: New sum rule. Not noted before in literature.

$$\kappa(\omega_c) = \frac{i}{T\hbar\omega_c} D_Q + \frac{1}{T\mathcal{L}} \int_0^\infty dt e^{i\omega_c t} \int_0^\beta d\tau \langle \hat{J}_x^Q(-t - i\tau) \hat{J}_x^Q(0) \rangle.$$

Where

$$D_Q = \frac{1}{\mathcal{L}} \left[\langle \Theta^{xx} \rangle - \hbar \sum_{n,m} \frac{p_n - p_m}{\epsilon_m - \epsilon_n} |\langle n | \hat{J}_x^Q | m \rangle|^2 \right].$$

In normal dissipative systems, the correction to Kubo's formula is zero, but it is a useful way of rewriting zero, it helps us to find the frequency integral of second term, hitherto unknown!!

So, what does Θ look like and what is its value? Answer is model dependent, and in brief, Θ is the specific heat times a velocity

$$\frac{\Theta^{xx}}{\hbar T} = \frac{1}{d} C_\mu v_{eff}^2$$

Thermo-power follows similar logic:

$$< \hat{J}_x >= \sigma(\omega)E_x + \gamma(\omega)(-\nabla T)$$

then the thermopower is

$$S(\omega) = \frac{\gamma(\omega)}{\sigma(\omega)}.$$

$$\Phi^{xx} = -\lim_{k\to 0}\frac{d}{dk_x}[\hat{J}_x(k_x), K(-k_x)].$$

This is the thermo electric operator

$$\gamma(\omega_c) = \frac{i}{\hbar\omega_c T\mathcal{L}}\left[< \Phi^{xx} > -\hbar\sum_{n,m}\frac{p_n - p_m}{\epsilon_n - \epsilon_m + \hbar\omega_c}\langle n|\hat{J}_x|m\rangle\langle n|\hat{J}_x^Q|m\rangle\right].$$

$$D_\gamma = \frac{1}{\mathcal{L}}\left[< \Phi^{xx} > -\hbar\sum_{n,m}\frac{p_n - p_m}{\epsilon_m - \epsilon_n}\langle n|\hat{J}_x|m\rangle\langle n|\hat{J}_x^Q|m\rangle\right].$$

$$\gamma(\omega_c) = \frac{i}{\hbar\omega_c T}D_\gamma + \frac{1}{T\mathcal{L}}\int_0^\infty dt e^{i\omega_c t}\int_0^\beta d\tau\langle\hat{J}_x(-t - i\tau)\hat{J}_x^Q(0)\rangle,$$

High frequency limits that are feasible and sensible similar to R*

$$\mathbf{L}^* = \frac{\langle \Theta^{xx} \rangle}{T^2 \langle \tau^{xx} \rangle} \tag{1}$$

$$\mathbf{Z}^* T = \frac{\langle \Phi^{xx} \rangle^2 T^2}{\langle \Theta^{xx} \rangle \langle \tau^{xx} \rangle}. \tag{2}$$

$$S^* = \frac{\langle \Phi^{xx} \rangle}{T \langle \tau^{xx} \rangle}. \tag{3}$$

Hence for any model system, armed with these three operators, we can compute the Lorentz ratio, the thermopower and the thermoelectric figure of merit!

So we naturally ask

- what do these operators look like
- how can we compute them
- how good an approximation is this?

In the preprint: several models worked out in detail

- Lattice dynamics with non linear disordered lattice
- Hubbard model
- Inhomogenous electron gas
- Disordered electron systems
- Infinite U Hubbard bands

- Lots of detailed formulas: we will see a small sample for Hubbard model and see some tests...

Thermo power operator for Hubbard model

$$\Phi^{xx} = -\frac{q_e}{2}\sum_{\vec{\eta},\vec{\eta'},\vec{r}} (\eta_x+\eta_x')^2 t(\vec{\eta})t(\vec{\eta'})c^\dagger_{\vec{r}+\vec{\eta}+\vec{\eta'},\sigma}c_{\vec{r},\sigma} - q_e\mu\sum_{\vec{\eta}}\eta_x^2 t(\vec{\eta})c^\dagger_{\vec{r}+\vec{\eta},\sigma}c_{\vec{r},\sigma} + \frac{q_eU}{4}\sum_{\vec{r},\vec{\eta}} t(\vec{\eta})(\eta_x)^2(n_{\vec{r},\bar{\sigma}}+n_{\vec{r}+\vec{\eta},\bar{\sigma}})(c^\dagger_{\vec{r}+\vec{\eta},\sigma}c_{\vec{r},\sigma}+c^\dagger_{\vec{r},\sigma}c_{\vec{r}+\vec{\eta},\sigma}). \tag{1}$$

This object can be expressed completely in Fourier space as

$$\Phi^{xx} = q_e\sum_{\vec{p}}\frac{\partial}{\partial p_x}\left\{v_p^x(\varepsilon_{\vec{p}}-\mu)\right\}c^\dagger_{\vec{p},\sigma}c_{\vec{p},\sigma} \tag{2}$$

$$+\frac{q_eU}{2\mathcal{L}}\sum_{\vec{l},\vec{p},\vec{q},\sigma,\sigma'}\frac{\partial^2}{\partial l_x^2}\left\{\varepsilon_{\vec{l}}+\varepsilon_{\vec{l}+\vec{q}}\right\}c^\dagger_{\vec{l}+\vec{q},\sigma}c_{\vec{l},\sigma}c^\dagger_{\vec{p}-\vec{q},\bar{\sigma}'}c_{\vec{p},\bar{\sigma}'}. \tag{3}$$

$$\tau^{xx} = \frac{q_e^2}{\hbar}\sum\eta_x^2\, t(\vec{\eta})\, c^\dagger_{\vec{r}+\vec{\eta},\sigma}c_{\vec{r},\sigma} \quad \text{or} \tag{1}$$

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

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

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

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

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

Interesting by product of these formulas: at T=0, $<\Phi>$ must vanish being entropy current, and hence the chemical potential can be expressed as a ratio of two operators. This is pretty surprising, and can be verified in some cases: half filled Hubbard model in any dimension for bipartite lattices $\mu = U/2$

Free Electron Limit and Comparison with the Boltzmann Theory

It is easy to evaluate the various operators in the limit of $U \to 0$, and this exercise enables us to get a feel for the meaning of these various somewhat formal objects. We note that

$$\begin{aligned} \langle \tau^{xx} \rangle &= \frac{2q_e^2}{\mathcal{L}} \sum_p n_{\vec{p}} \frac{d}{dp_x} \left[v_{\vec{p}}^x \right] \\ \langle \Theta^{xx} \rangle &= \frac{2}{\mathcal{L}} \sum_p n_{\vec{p}} \frac{d}{dp_x} \left[v_{\vec{p}}^x (\varepsilon_{\vec{p}} - \mu)^2 \right] \\ \langle \Phi^{xx} \rangle &= \frac{2q_e}{\mathcal{L}} \sum_p n_{\vec{p}} \frac{d}{dp_x} \left[v_{\vec{p}}^x (\varepsilon_{\vec{p}} - \mu) \right]. \end{aligned} \quad (1)$$

At low temperatures, we use the Sommerfield formula after integrating by parts, and thus obtain the leading low T behaviour:

$$\begin{aligned} \langle \tau^{xx} \rangle &= 2q_e^2 \rho_0(\mu) \langle (v_{\vec{p}}^x)^2 \rangle_\mu \\ \langle \Theta^{xx} \rangle &= T^2 \frac{2\pi^2 k_B^2}{3} \rho_0(\mu) \langle (v_{\vec{p}}^x)^2 \rangle_\mu && (1) \\ \langle \Phi^{xx} \rangle &= T^2 \frac{2q_e \pi^2 k_B^2}{3} \left[\rho_0'(\mu) \langle (v_{\vec{p}}^x)^2 \rangle_\mu + \rho_0(\mu) \frac{d}{d\mu} \langle (v_{\vec{p}}^x)^2 \rangle_\mu \right], && (2) \end{aligned}$$

We may form the high frequency ratios

$$S^* = T \frac{\pi^2 k_B^2}{3q_e} \frac{d}{d\mu} \log \left[\rho_0(\mu)\langle (v_{\vec{p}}^x)^2 \rangle_\mu\right]$$
$$L^* = \frac{\pi^2 k_B^2}{3q_e^2}. \quad (1)$$

It is therefore clear that the high frequency result gives *the same* Lorentz number as well as the thermopower that the Boltzmann theory gives in its simplest form.

The thermal conductivity cannot be found from this approach, but basically the formula is the same as the Drude theory with $i/\omega \to \tau$.

Some new results for strong correlations and triangular lattice:

Thermopower formula to replace the Heikes-Mott-Zener formula

Leading High temperature term for the Triangular lattice and application to Sodium Cobalt Oxide

$$S^* = -\frac{\mu}{q_e T} + \frac{q_e \Delta}{T \langle \tau^{xx} \rangle}$$

where

$$\Delta = -\frac{1}{2} \sum_{\vec{\eta}, \vec{\eta'}, \vec{r}} (\eta_x + \eta'_x)^2 \; t(\vec{\eta}) \; t(\vec{\eta'}) \; Y_{\sigma', \sigma}(\vec{r} + \vec{\eta}) \; \langle c^\dagger_{\vec{r}+\vec{\eta}+\vec{\eta'}, \sigma'} c_{\vec{r}, \sigma} \rangle$$

This is a very useful alternate formula to the Heikes-Mott-Zener formula where the second term in Eq above is thrown out. It interpolates very usefully between the standard formulas for low temperature as well as at high temperature. The second term represents the "transport" contribution to the thermopower, whereas the first term is the thermodynamic or entropic part, which dominates at high temperaturefor S^* we can actually make a systematic expansion in powers of βt, unlike the dc counterpart.

Leading high temp expansion:

$$\langle \tau^{xx} \rangle = 6\mathcal{L}q_e^2 t \langle \tilde{c}_1^\dagger \tilde{c}_0 \rangle = 3\mathcal{L}q_e^2 \beta t^2 n(1-n).$$

$$\begin{aligned} \left\{ \tilde{c}_{\vec{r},\sigma}, \tilde{c}^\dagger_{\vec{r'},\sigma'} \right\} &= \delta_{\vec{r},\vec{r'}} \left\{ \delta_{\sigma,\sigma'}(1-n_{\vec{r},\bar{\sigma}}) + (1-\delta_{\bar{\sigma},\sigma'})\tilde{c}^\dagger_{\vec{r},\sigma}\tilde{c}_{\vec{r},\bar{\sigma}} \right\} \\ &\equiv Y_{\sigma,\sigma'}\, \delta_{\vec{r},\vec{r'}} \end{aligned} \qquad (1)$$

$$\Delta \sim -\frac{3}{2}\mathcal{L}t^2 \sum_{\sigma,\sigma'} \langle Y_{\sigma',\sigma}(\vec{\eta})\tilde{c}^\dagger_{\vec{\eta}+\vec{\eta'},\sigma'}\tilde{c}_{\vec{0},\sigma} \rangle.$$

The spins must be the same to the leading order in βt where we generate a hopping term $\tilde{c}^\dagger_{\vec{0},\sigma}\tilde{c}_{\vec{\eta}+\vec{\eta'},\sigma}$ from an expansion of $\exp(-\beta K)$, and hence a simple estimation yields

$$\Delta = -\frac{3}{4}\mathcal{L}t^3\beta n(1-n)(2-n) + O(\beta^3).$$

This together with $\mu/k_B T = \log(n/2(1-n)) + O(\beta^2 t^2)$ gives us the result for $0 \le n \le 1$

$$S^* = \frac{k_B}{q_e}\left\{ \log[2(1-n)/n] - \beta t \frac{2-n}{4} + O(\beta^2 t^2) \right\},$$

$$S^* = \frac{k_B}{q_e}\left\{\log[2(n-1)/(2-n)] + \beta t\frac{n}{4} + O(\beta^2 t^2)\right\}$$

for $1 \leq n \leq 2$ using particle hole symmetry.

From the temperature dependence of the data of Terasaki et aland assuming $S \sim S^*$one finds that $t = -110^0 K$, and with this, $S^* \sim 120\mu V/K$, fairly close to the observed value.

Note that in these high Temp expansions:

1. Correction is O(β) for triangular lattice but O(β^2) for square lattice. Hence larger transport correction for triangular lattice.
2. Prediction. If sign of t is +ve then (unfrustrated case) S will reach its asymptotic Heikes-Mott-Zener value FROM ABOVE, hence a peak in S must exist. Such a case must be the largest S for any metallic system.
3. Go FOR IT GUYS!! Let us find a realization of this.

Conclusions:

- New and promising formalism.
- Lots of computations are being carried out or planned at Santa Cruz.
- Thanks to my colleagues at SC: Jan Haerter and Mike Peterson
- Many thanks to NSF for funding and to ICAM and Croation organizers for inviting.