Thermoelectric and Hall transport in Correlated Matter: A new approach

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I outline a new approach to the difficult problem of computing transport constants in correlated matter, using a combination of analytical and numerical methods. Our technique exploits the simplifications gained by going to frequencies larger than some characteristic scale, where the Kubo formulas simplify somewhat.

A careful computation of the Kubo type formulas reveal important corrections to known formulas in literature for the thermpower, and thermal conductivity, including new sum rules. Finally these are put together into a numerical computation for small clusters where these ideas are bench marked and several new predictions are made that should be of interest to materials community.

References:

 Strong Correlations Produce the Curie-Weiss Phase of NaxCoO2
 J. O. Haerter, M. R. Peterson, and B. S. Shastry Phys. Rev. Lett.
 97, 226402 (2006)
 Finite-temperature properties of the triangular lattice t-J model and applications to NaxCoO2, J. O. Haerter, M. R. Peterson, and B. S. Shastry, Phys. Rev. B 74, 245118 (2006)

3) Sum rule for thermal conductivity and dynamical thermal transport coefficients in condensed matter B. S. Shastry, Phys. Rev. B 73, 085117 (2006)

Mike Peterson Jan Haerter The Boltzmann theory approach to transport:

A very false approach for correlated matter, unfortunately very Strongly influential and pervasive.

Need for alternate view point.



wikipedia

Vulcan death grip – Derived from a Star Trek classic episode where a non- existar "Vulcan death grip" was used to fool Romulans that Spock had killed Kirk.





First serious effort to understand Hall constant in correlated matter: S S, Boris Shraiman and Rajeev Singh, Phys Rev Letts (1993)

Introduced object

 $R_{H}^{*} = \lim_{B \to 0} \lim_{\omega \to \infty} \frac{\rho_{xy}(\omega)}{B}$

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

$$\sigma^{\alpha,\beta}(\omega_c) = \frac{i}{\hbar N_s v \omega_c} \left[\langle \tau^{\alpha,\beta} \rangle - \hbar \sum_{n,m} \frac{p_n - p_m}{\epsilon_n - \epsilon_m + \hbar \omega_c} \langle n | \hat{J}_{\alpha} | m \rangle \langle m | \hat{J}_{\beta} | n \rangle \right]$$

 $\mathbf{R}_{H}^{*} = \frac{-iN_{s}v}{B\hbar} \frac{\langle [J^{x}, J^{y}] \rangle}{\langle \tau^{xx} \rangle^{2}}$

•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

Faraday Rotation and the Hall Constant in Strongly Correlated Fermi Systems

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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.



Anomalous high-temperature Hall effect on the triangular lattice in $Na_x CoO_2$

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The Hall coefficient R_H of Na_xCoO₂ (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 for strong correlation. We discuss the effect of hopping on a triangular lattice and compare R_H with a recent prediction by Kumar and Shastry.







Phenomenological interpretations of the ac Hall effect in the normal state of $YBa_2Cu_3O_7$

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Thermoelectric phenomena

$$\Theta^{xx} = -\lim_{k_x
ightarrow 0} rac{d}{dk_x} [\hat{J}^Q_x(k_x), \hat{K}(-k_x)]$$

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

$$Re \kappa(\omega) = \frac{\pi}{\hbar T} \delta(\omega) \bar{D}_Q + Re \kappa_{reg}(\omega) \text{ with}$$

$$Re \kappa_{reg}(\omega) = \frac{\pi}{T\mathcal{L}} \left(\frac{1 - e^{-\beta\omega}}{\omega} \right) \sum_{\epsilon_n \neq \epsilon_m} p_n |\langle n| \hat{J}_x^Q |m\rangle|^2 \delta(\epsilon_m - \epsilon_n - \hbar\omega), \quad (1)$$

$$\bar{D}_Q = \frac{1}{\mathcal{L}} \left[\langle \Theta^{xx} \rangle - \hbar \sum_{\epsilon_n \neq \epsilon_m} \frac{p_n - p_m}{\epsilon_m - \epsilon_n} |\langle n| \hat{J}_x^Q |m\rangle|^2 \right]. \quad (2)$$

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

$$\int_{0}^{\infty} Re \ \kappa(\omega) d\omega = \frac{\pi}{2\hbar T \mathcal{L}} \langle \Theta^{xx} \rangle. \tag{1}$$

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

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

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

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

Thermo-power follows similar logic:

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

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

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

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

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

High frequency limits that are feasible and sensible similar to $\ \mathsf{R}^{\star}$

$$\mathbf{L}^{*} = \frac{\langle \Theta^{xx} \rangle}{T^{2} \langle \tau^{xx} \rangle}$$
(1)
$$\mathbf{Z}^{*}T = \frac{\langle \Phi^{xx} \rangle^{2}}{\langle \Theta^{xx} \rangle \langle \tau^{xx} \rangle}.$$
(2)
$$S^{*} = \frac{\langle \Phi^{xx} \rangle}{T \langle \tau^{xx} \rangle}.$$
(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 paper: 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...

Anharmonic Lattice example

$$\begin{split} H &= \sum_{j} H_{j} \\ H_{j} &= \left[\frac{\vec{p}_{j}^{2}}{2m_{j}} + U_{j} \right]; \qquad U_{j} = \frac{1}{2} \sum_{i \neq j} V_{j,i}, \qquad (1) \\ F_{i,j} &= -\frac{\vec{\partial}}{\vec{\partial}\vec{u}_{i}} V_{i,j} \\ J_{x}^{E}(\vec{k}) &= \frac{1}{4} \sum_{i,j} \frac{(X_{i} - X_{j})}{m_{i}} e^{ik_{x}X_{i}} \left\{ p_{x,i}, F_{i,j}^{x} \right\} \\ \Theta^{xx} &= (\hbar \omega_{0}^{2} a_{0}^{2}) \left[\frac{1}{m} \sum_{i} p_{i} p_{i+1} + \frac{k_{s}}{4} \sum_{i} (u_{i-1} - u_{i+1})^{2} \right]. \\ \langle \Theta^{xx} \rangle &= \mathcal{L}(\hbar^{2} \omega_{0}^{3} a_{0}^{2}) \int_{0}^{\pi} \frac{dk}{\pi} \left[(\frac{1}{2} + \frac{1}{e^{\beta \omega_{k}} - 1}) \left\{ \frac{\omega_{k}}{\omega_{0}} \cos(k) + \frac{\omega_{0}}{\omega_{k}} \sin^{2}(k) \right\} \right], \end{split}$$

Thermo power operator for Hubbard model

 au^{xx}

$$\Phi^{xx} = -\frac{q_e}{2} \sum_{\vec{\eta},\vec{\eta'},\vec{r}} (\eta_x + \eta'_x)^2 t(\vec{\eta}) t(\vec{\eta'}) c^{\dagger}_{\vec{r}+\vec{\eta}+\vec{\eta'},\sigma} c_{\vec{r},\sigma} - q_e \mu \sum_{\vec{\eta}} \eta_x^2 t(\vec{\eta}) c^{\dagger}_{\vec{r}+\vec{\eta},\sigma} c_{\vec{r},\sigma} + \frac{q_e U}{4} \sum_{\vec{r},\vec{\eta}} t(\vec{\eta}) (\eta_x)^2 (n_{\vec{r},\vec{\sigma}} + n_{\vec{r}+\vec{\eta},\vec{\sigma}}) (c^{\dagger}_{\vec{r}+\vec{\eta},\sigma} c_{\vec{r},\sigma} + c^{\dagger}_{\vec{r},\sigma} c_{\vec{r}+\vec{\eta},\sigma}).$$
(1)

This object can be expressed completely in Fourier space as

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

$$+ \frac{q_e U}{2\mathcal{L}} \sum_{\vec{l},\vec{p},\vec{q},\sigma,\sigma'} \frac{\partial^2}{\partial l_x^2} \left\{ \varepsilon_{\vec{l}} + \varepsilon_{\vec{l}+\vec{q}} \right\} c_{\vec{l}+\vec{q},\sigma}^{\dagger} c_{\vec{l},\sigma} c_{\vec{p}-\vec{q},\vec{\sigma}'}^{\dagger} c_{\vec{p},\vec{\sigma}'}.$$

$$(3)$$

$$^x = \frac{q_e^2}{\hbar} \sum_{\vec{k}} \eta_x^2 t(\vec{\eta}) c_{\vec{r}+\vec{\eta},\sigma}^{\dagger} c_{\vec{r},\sigma} \text{ or }$$

$$(1)$$

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

$$(2)$$

Free Electron Limit and Comparison with the Boltzmann Theory

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

$$\langle \tau^{xx} \rangle = \frac{2q_e^2}{\mathcal{L}} \sum_p n_{\vec{p}} \frac{d}{dp_x} \left[v_{\vec{p}}^x \right]$$

$$\langle \Theta^{xx} \rangle = \frac{2}{\mathcal{L}} \sum_p n_{\vec{p}} \frac{d}{dp_x} \left[v_{\vec{p}}^x (\varepsilon_{\vec{p}} - \mu)^2 \right]$$

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

$$(1)$$

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

$$\langle \tau^{xx} \rangle = 2q_e^2 \rho_0(\mu) \langle (v_{\vec{p}}^x)^2 \rangle_{\mu}$$

$$\langle \Theta^{xx} \rangle = T^2 \frac{2\pi^2 k_B^2}{3} \rho_0(\mu) \langle (v_{\vec{p}}^x)^2 \rangle_{\mu}$$

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

$$(1)$$

We may form the high frequency ratios

$$S^{*} = T \frac{\pi^{2} k_{B}^{2}}{3q_{e}} \frac{d}{d\mu} \log \left[\rho_{0}(\mu) \langle (v_{\vec{p}}^{x})^{2} \rangle_{\mu} \right]$$

$$L^{*} = \frac{\pi^{2} k_{B}^{2}}{3q_{e}^{2}}.$$
(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 $i/\omega \rightarrow \tau$.

Some new results for strong correlations and triangular lattice: Thermopower formula to replace the Heikes-Mott-Zener formula PRL 97, 226402 (2006)

PHYSICAL REVIEW LETTERS

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Strong Correlations Produce the Curie-Weiss Phase of Na_xCoO₂

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Magnetic field dep of S(B) vs data

PHYSICAL REVIEW B 74, 245118 (2006)

Finite-temperature properties of the triangular lattice t-J model and applications to Na_xCoO₂

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







$R_H/_{Transport} = R_H^* + \int_0^\infty \Im m[R_H(\omega)]/\omega \ d\omega/\pi$



T-t'-J model, typical frequency dependence is very small . This is very encouraging for the program of x_eff





Remarkably, a small t' of either sign shifts the zero crossing of Hall constant significantly! **Conclusions:**

•New and rather useful starting point for understanding transport phenomena in correlated matter

•Kubo type formulas are non trivial at finite frequencies, and have much structure

•We have made several successful predictions for NCO already

•Can we design new materials using insights gained from this kind of work?

Useful link for this kind of work:

http://physics.ucsc.edu/~sriram/sriram.html