Two novel Frustrated Mott Hubbard systems: Can RVB win this time

Feb 9, 2004

Sriram Shastry

UC Santa Cruz

Recently discovered systems Sr Cu_2 $(BO_3)_2$ and Na_x Co O_2 have in common that they are Mott Hubbard Systems with frustrated exchange and also display electronic frustration, i.e. a non trivial dependence on the sign of the electronic hopping. Several experiments on these display novel behaviour, such as magnetization plateaus, and in the case of Na_x Co O_2 , also superconductivity.

We discuss these systems within the framework of the Resonating Valence Bond theory, and suggest that they are a better testing ground for RVB as compared to High Tc systems for a variety of reasons.

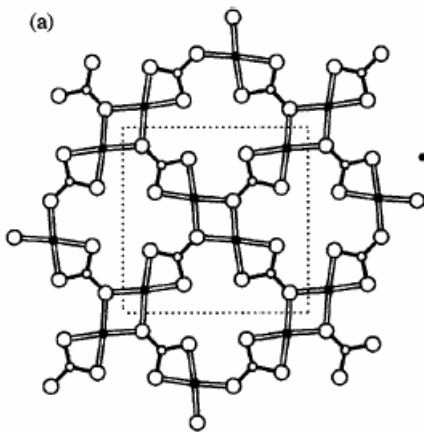
$SrCu_2(BO_3)_2$: A Mott Hubbard Semimetal

New system with interesting connection with past theoretical speculations:

VOLUME 82, NUMBER 15 PHYSICAL REVIEW LETTERS 12 APRIL 1999

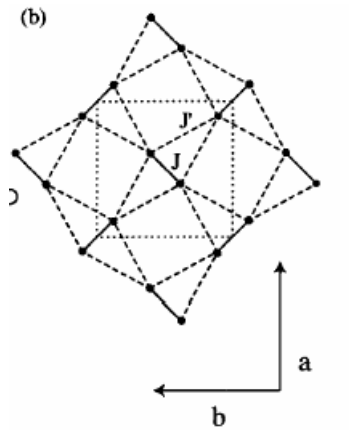
Exact Dimer Ground State and Quantized Magnetization Plateaus in the Two-Dimensional Spin System $SrCu_2(BO_3)_2$

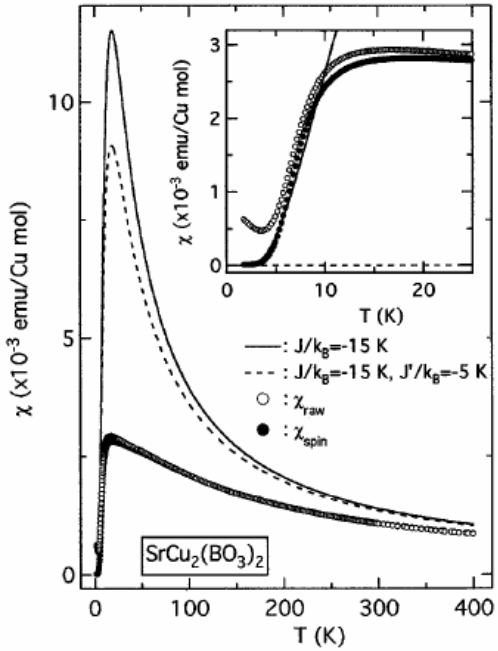
H. Kageyama,[1,2,*] K. Yoshimura,[1,3,†] R. Stern,[3] N. V. Mushnikov,[2] K. Onizuka,[2] M. Kato,[1] K. Kosuge,[1] C. P. Slichter,[3] T. Goto,[2] and Y. Ueda[2]

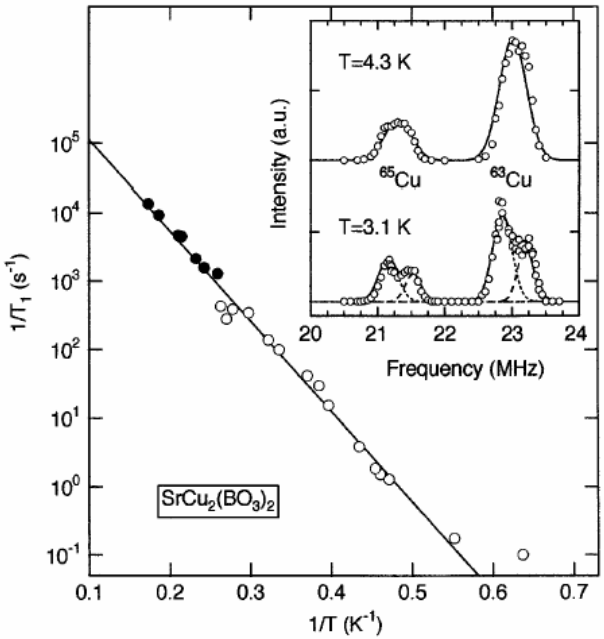


2-D Layered system with spin gap

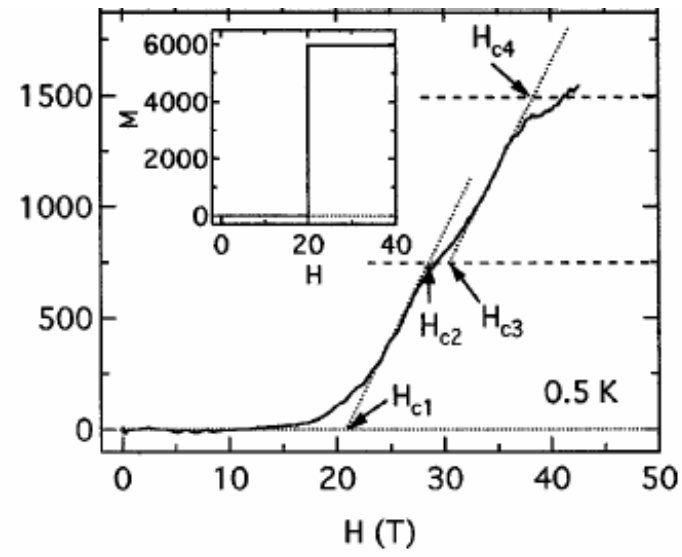
Copper lattice ->





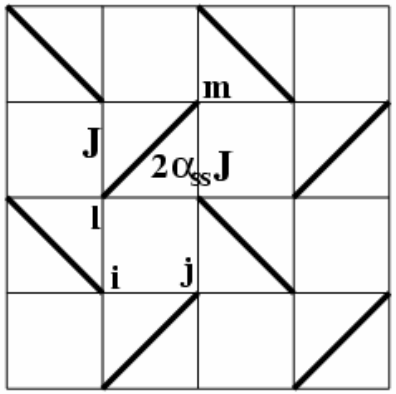


Spin gap ~30^0 K



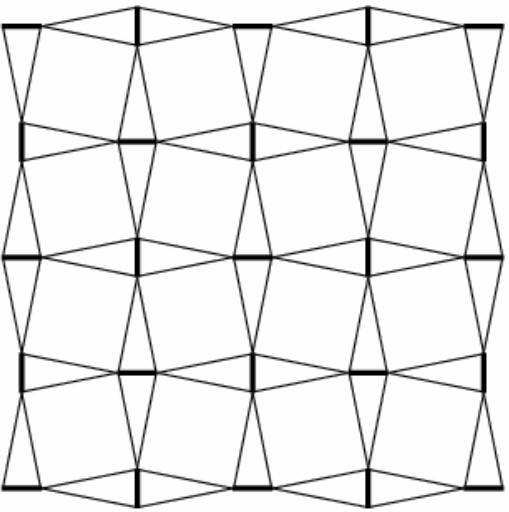
Magenitzation plateaus at M/M_0= ¼, 1/8

←



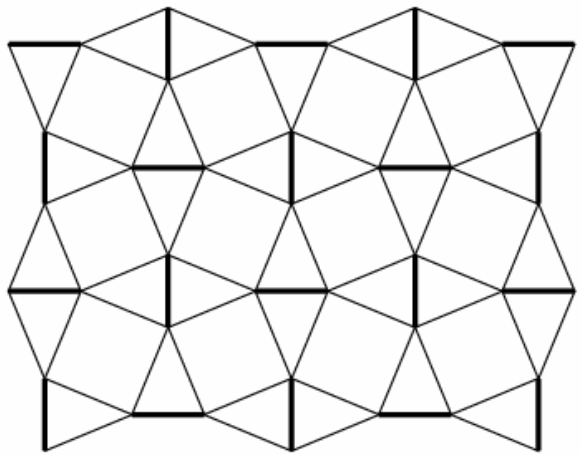
(a)

$\theta = \frac{\pi}{2}$



(c)

$\theta = \frac{\pi}{8}$

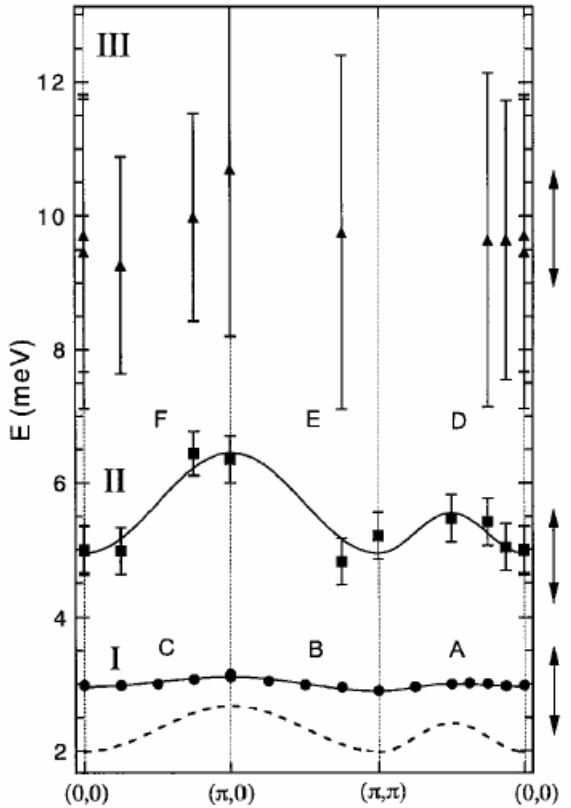


(b)

$\theta = \frac{\pi}{4}$

Realized soon (K Ueda and S Miyahara (1999) PRL that this is topologically equivalent to a model solved earlier by Shastry & Sutherland (1981). This is a remarkable theoretical model that we found years ago, an exactly solvable 2 dimensional spin ½ Heisenberg model. Key to solvability; frustration.

Condition for dimer ground state is easily met J'/J>2. Critical point and no trivial phase transitions as J'/J is varied.



Q dependence of the excitation energies of bands

Flat triplet band at 3mev and dispersive pair of triplets at ~ 6mev.

←

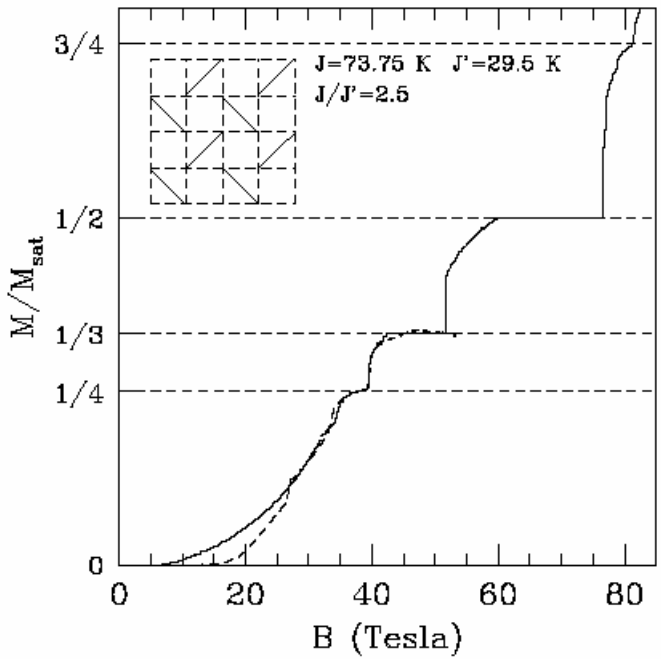
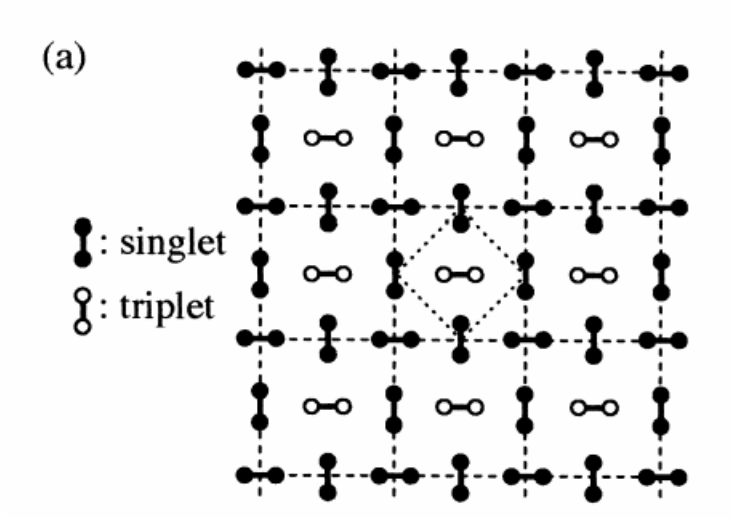


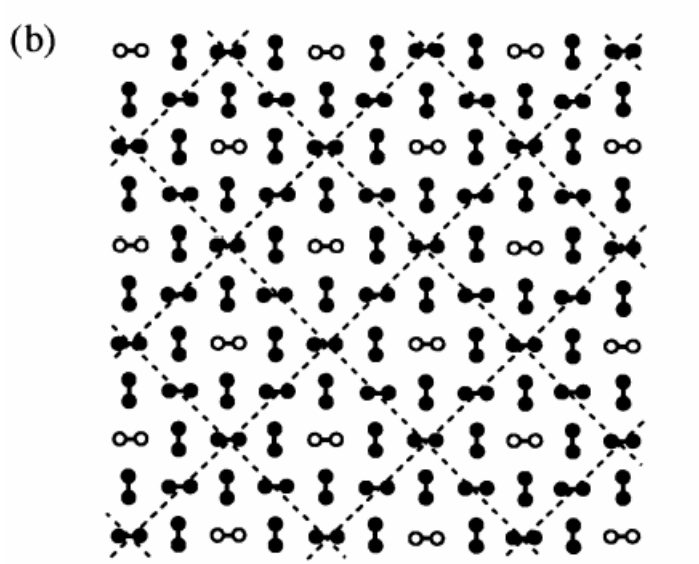
FIG. 1. Comparison between the magnetization curve of $SrCu_2(BO_3)_2$ measured by Onizuka *et al.* (dashed line) and the mean-field result (solid line). Inset: Shastry-Sutherland lattice. The exchange interaction is J on black links and J' on the dotted ones.

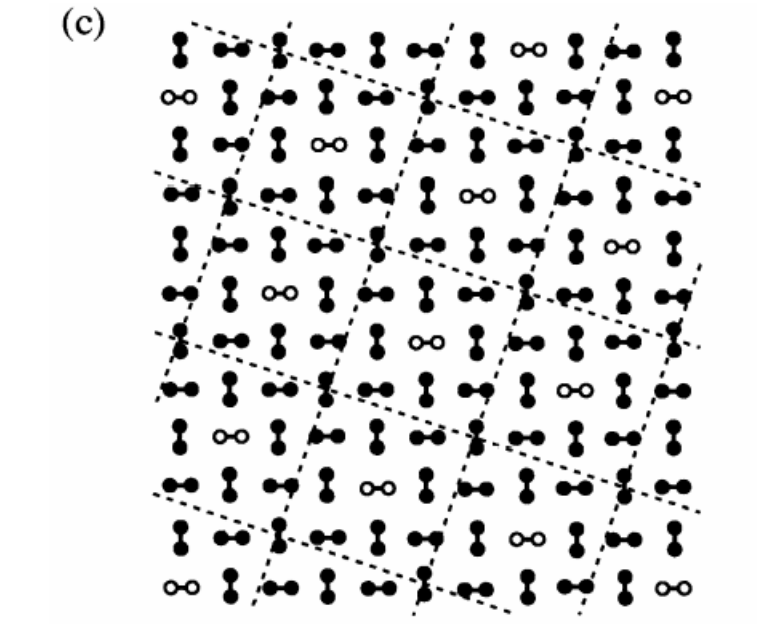
Magnetization Plateaus of $SrCu_2(BO_3)_2$ from a Chern-Simons Theory

G. Misguich,[1] Th. Jolicoeur,[2] and S. M. Girvin[3,4]

PHYSICAL REVIEW LETTERS 27 AUGUST 2001







Patterns of localized bosons (each boson is a triplet) corresponding to m/ms ¼,1/8, 1/10.

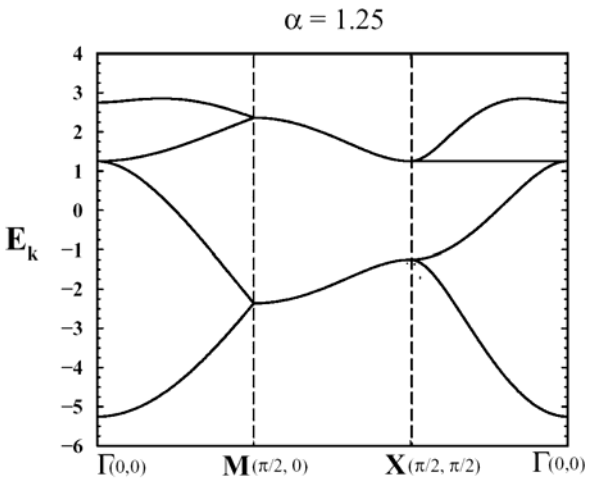
Heavy strongly interacting bosons crystallize in substrate potential (.

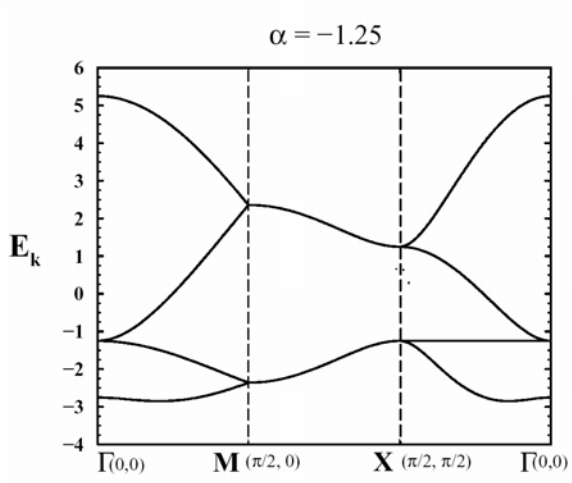
Plateaus at 1/3 and ½ not given by this argument:

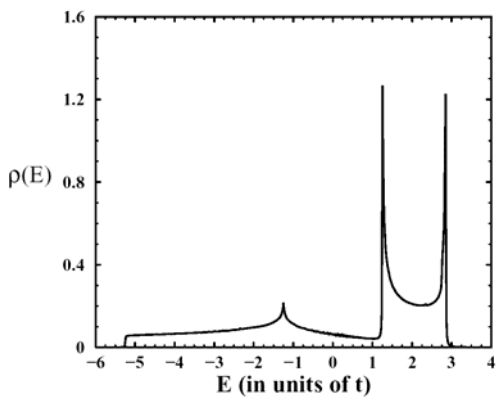
What about doping? Is SCBO a band insulator or a Mott Insulator?

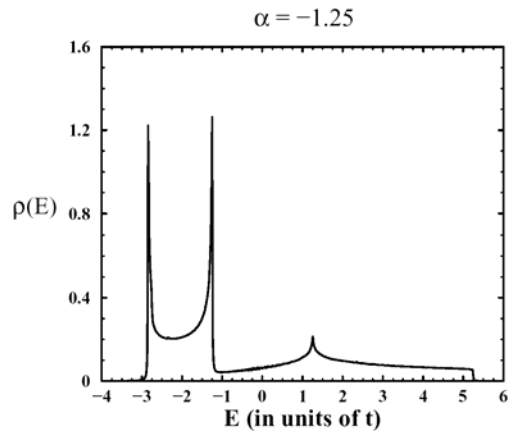
Shastry/Kumar Yukawa conference: Prg Theo Phys Suppl 145, 1 (2002). And Cond Mat 2002

$$H = -t \sum_{<ij>,\sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c.) - \alpha t \sum_{<l,m>,\sigma} (c^{\dagger}_{l,\sigma} c_{m,\sigma} + h.c.) + U \sum_{r} n_{r,\uparrow} n_{r,\downarrow}$$





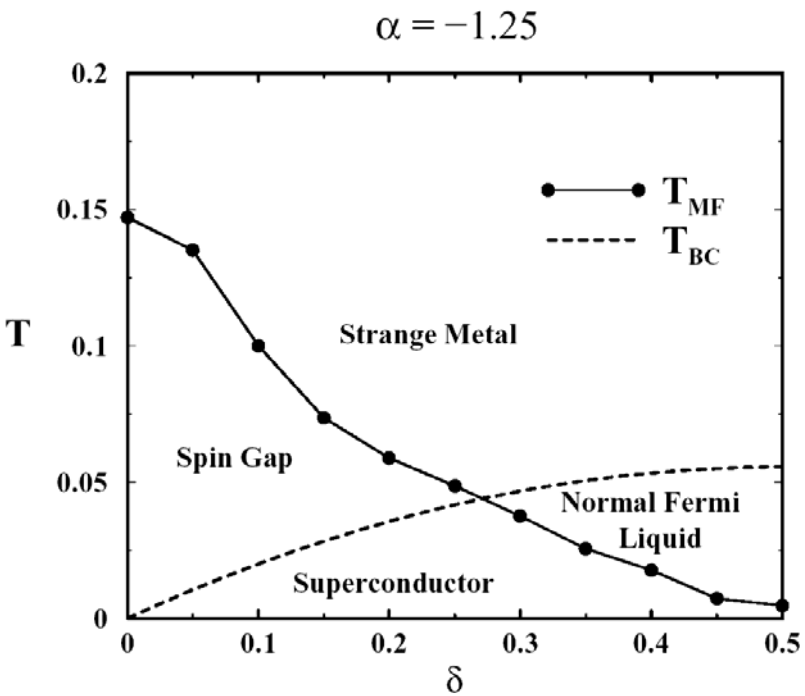


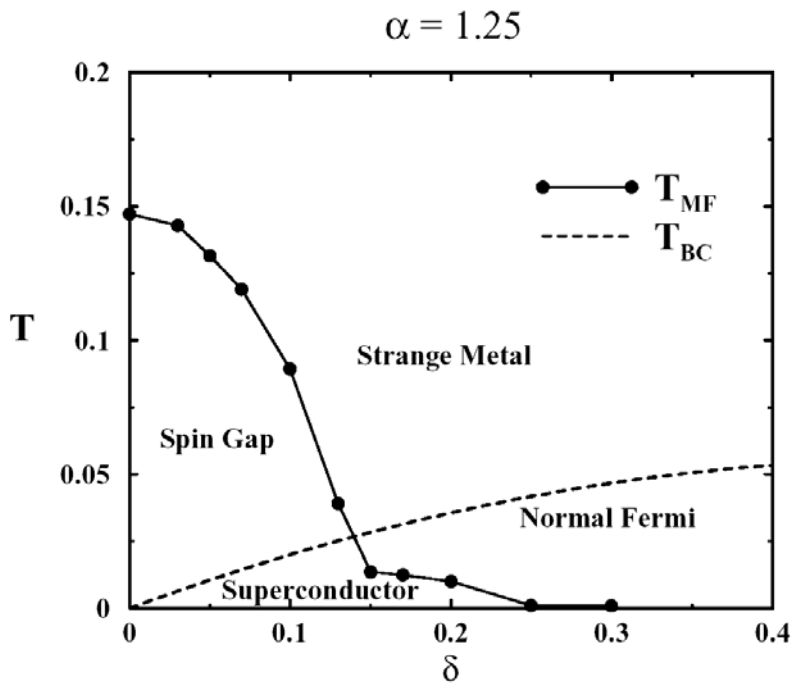


A semi metal: somewhat like graphite but with a quadratic touching of bands as opposed to linear, so the DOS is finite, like in a metal (linear specific heat etc). Much more metallic than Graphite.

RVB is a possible description. (what is RVB? Minimally tJ MFT)

IN fact **RVB is exact at half filling in this case**, it **does** reproduce the singlet ground state (not so Resonating Valence Bond state). How about doping? SC with complex order parameter results: Hole doping phase diagram: Max Tc approximately 10^0K. **ORDER PARAMETER IS INTRINSICALLY COMPLEX**





Another frustrated Mott Hubbard system Na_x Co O_2

Experiments :

- Early papers:
  - Terasaki (1997) Discovered high thermopower in a reasonable metal
  - Thermopower calculation within Band Structure: D J Singh (2000).
- Recent surge of activity:
  - Takada et al (2003 March 6, Nature) Superconductivity in hydrated system
  - Ong and Cava (2003 May Nature) Magnetic Field dependence of S.
  - Lots more since
- Theory:
  - RVB Scenario, Kumar & SS (April 9) RVB type calculation+ Hall constant prediction, Baskaran (March 31) cond-mat Wang, Lee and Lee (April 16) cond-mat RVB
  - Some unresolved issues.

Large thermoelectric power in $NaCo_2O_4$ single crystals

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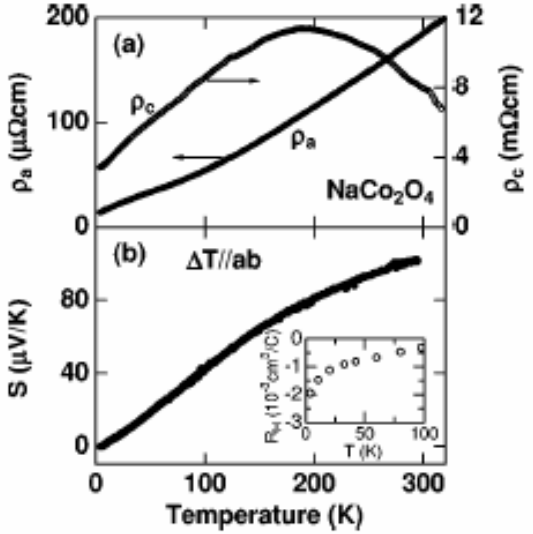
(Received 18 July 1997)

We measured and analyzed the transport properties of single-crystal $NaCo_2O_4$, which is a metallic transition-metal oxide consisting of a two-dimensional triangle lattice of Co. Reflecting the crystal structure, the resistivity is highly anisotropic between in- and out-of-plane directions, and the in-plane resistivity is as low as 200 $\mu\Omega$ cm at 300 K. Most strikingly, the in-plane thermoelectric power of $NaCo_2O_4$ is about 100 μV/K at 300 K, which is nearly ten times larger than that of typical metals. The large thermoelectric power and the low resistivity suggest that $NaCo_2O_4$ is a potential thermoelectric material. [S0163-1829(97)52144-8]



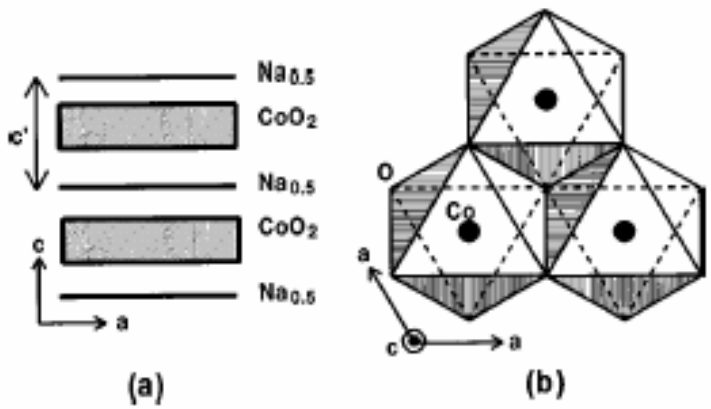


FIG. 1. A schematic picture of the crystal structure of $NaCo_2O_4$. (a) The layered structure; (b) the CoO_2 layer.

TABLE I. Various physical parameters for $NaCo_2O_4$ and Bi_2Te_3 (Ref. 6) at 300 K. ρ, S, and μ are resistivity, thermoelectric power, and mobility, respectively. Note that ρ and S of $NaCo_2O_4$ are the in-plane data.

Parameters	Unit	$NaCo_2O_4$	Bi_2Te_3
ρ	mΩ cm	0.2	1
$\lvert S\rvert$	μV/K	100	200
S^2/ρ	μW/K^2 cm	50	40
μ	cm^2/V s	13	150

Fuita, Mochida, Nakamura (2001): More recent studies:

The dimensionless variable Z T exceeds unity at 800K !

Table I. Various parameters for $Na_xCoO_{2-\delta}$ single crystal, $Na_xCoO_{2-\delta}$ polycrystal and $Si_{0.95}Ge_{0.05}$ [22] are compared at 300 K and 800 K, ρ, S, $S^2 \cdot \rho^{-1}$, κ and Z are the resistivity, thermoelectric power, power factor, thermal conductivity and figure-of-merit, respectively.

		$Na_xCoO_{2-\delta}$ single crystal		$Na_xCoO_{2-\delta}$ polycrystal		$Si_{0.95}Ge_{0.05}$ typical p-type	
		300 K	800 K	300 K	800 K	300 K	800 K
ρ	$m\Omega \cdot cm$	0.29	0.52	2	3.6	0.77	1.7
S	$\mu V K^{-1}$	83	200	100	170	200	338
$S^2 \cdot \rho^{-1}$	$mW m^{-1} K^2$	2.38	7.69	0.50	0.81	5.2	6.7
κ	$W m^{-1} K^{-1}$	19.0	5.1	2.0	2.1	10.0	9.5
Z	mK^{-1}	0.12	1.5	0.25	0.39	0.52	0.71
ZT	—	0.03	1.2	0.08	0.31	0.16	0.57

Electronic structure of $NaCo_2O_4$

D. J. Singh
Code 6391, Naval Research Laboratory, Washington, DC 20375
(Received 10 January 2000)

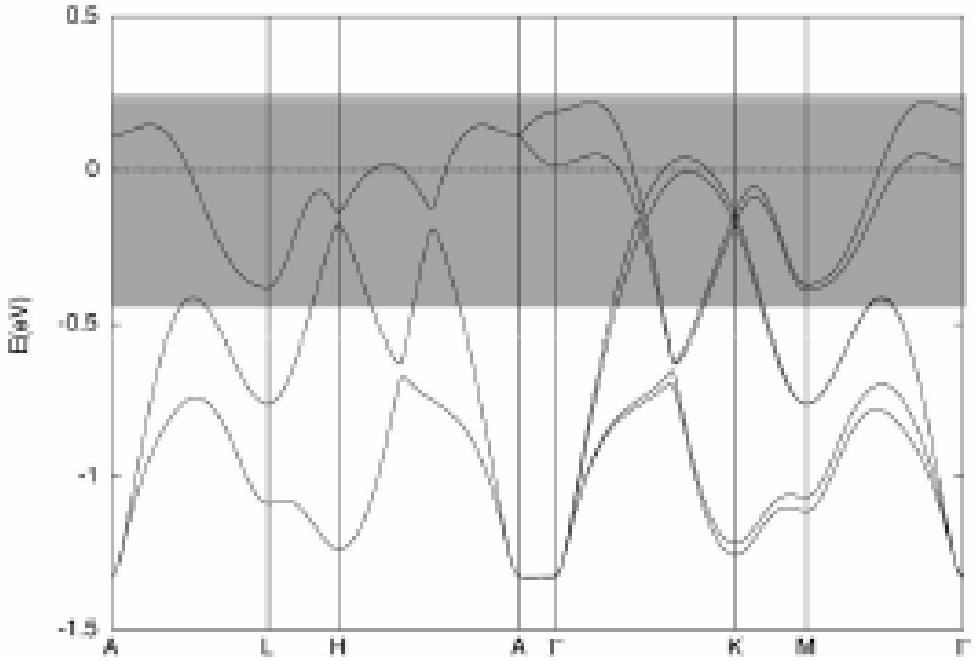


FIG. 1. LDA paramagnetic band structure of $NaCo_2O_4$. The

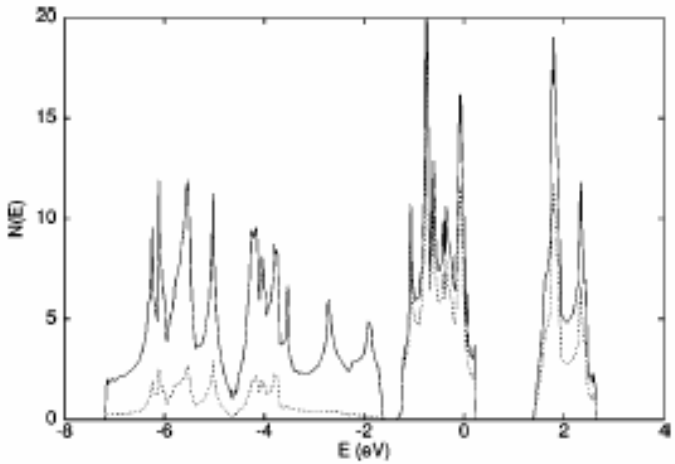


FIG. 2. LDA paramagnetic DOS, $N(E)$, of $NaCo_2O_4$. The DOS

Superconductivity in two-dimensional CoO_2 layers

NATURE | VOL 422 | 6 MARCH 2003

Kazunori Takada*‡, Hiroya Sakurai†, Eiji Takayama-Muromachi†, Fujio Izumi*, Ruben A. Dilanian* & Takayoshi Sasaki*‡

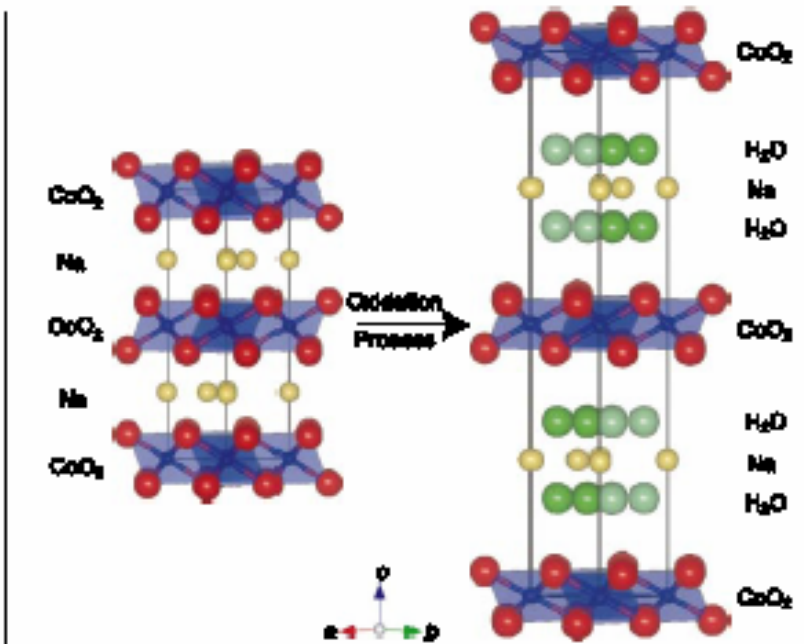


Figure 1 Structural views of $Na_{0.7}CoO_2$ (left) and $Na_xCoO_2 \cdot yH_2O$ (right), where Na and

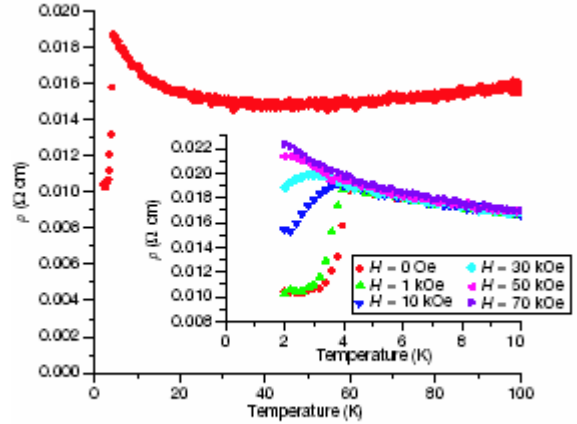
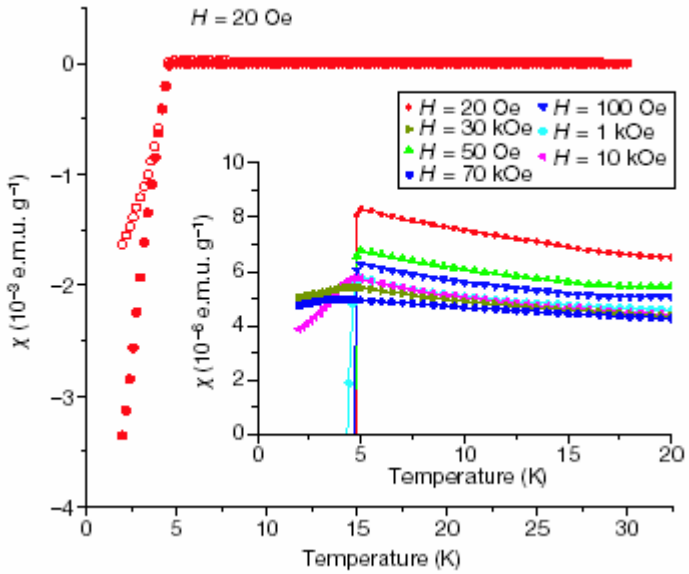


Figure 4 Resistivity (ρ) of $Na_xCoO_2 \cdot yH_2O$ under zero magnetic field. The inset figure



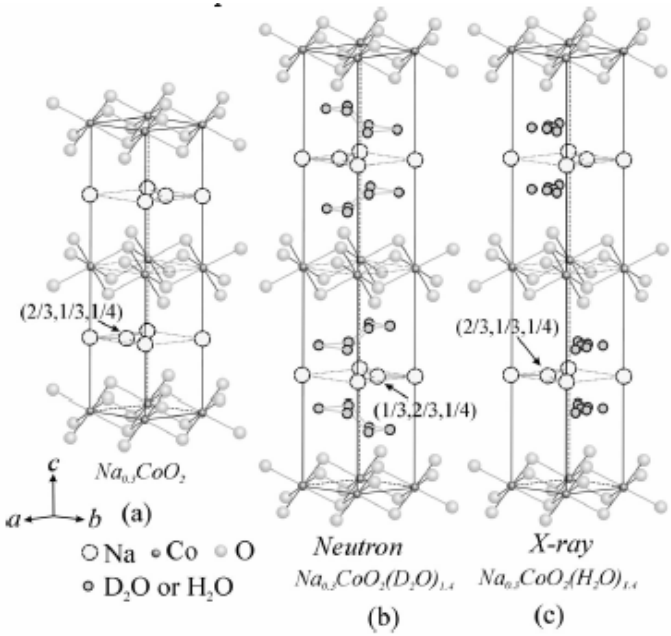


Fig 5. Structures of (a) $Na_{0.3}CoO_2$, (b) the superconducting phase model used in the present work and (c) the model reported in ref. [4]. Note that the positions for the Na and water centers are different in (b) and (c).

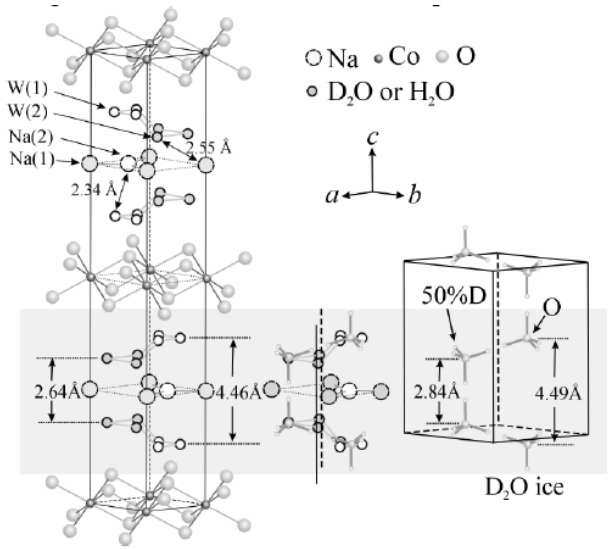


Fig. 4. Structure model for the Na_xCoO_2 1.4(D_2/H_2O) superconductor (left). The figure on the right shows the D_2O ice structure.[24] For comparison, the figure in the middle shows the ice molecule superposed on the water block in the structure of the superconducting phase, demonstrating that the dimensions of the water block are close to those found in free ice.

Neutron data:
Cava/Lynn

NATURE | VOL 423 | 22 MAY 2003 |

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*‡

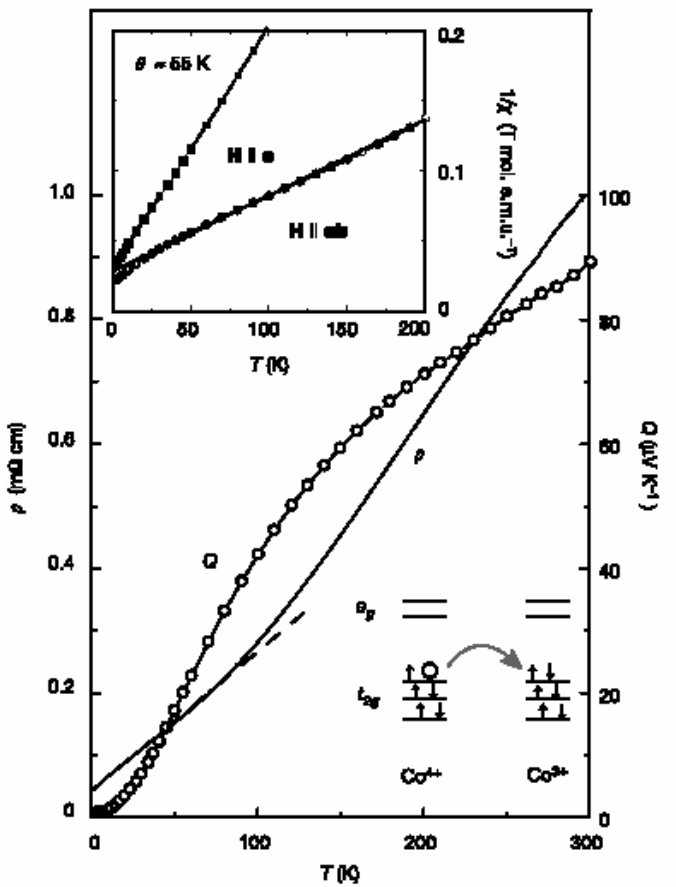


Figure 1 The temperature (T) dependence of magnetic and transport properties of single-crystal $Na_xCo_2O_4$ and electronic states in the Co ions. The in-plane thermopower Q

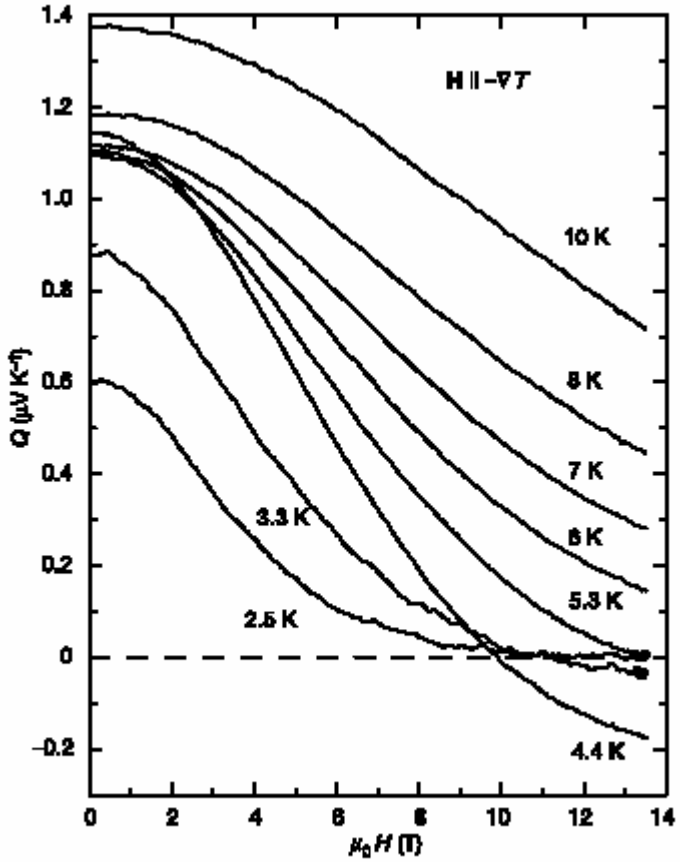


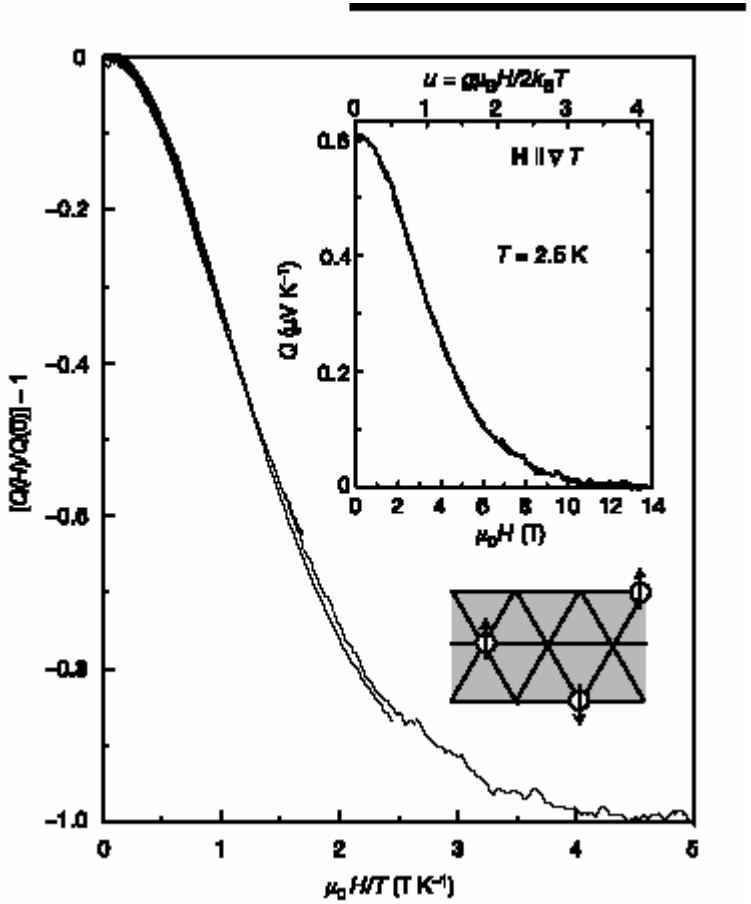
Figure 2 The in-plane thermopower Q versus an in-plane $\mathbf{H}\|(-\nabla T)$ at selected T. At

Hubbard model in small hopping limit (Heikes Formula): G Beni (1974):Q=-1/T($S_2/S_1+\mu/e$) Q~- entropy/e, entropy~k_B Log(g_{spin} g_{config}):

All data for different T collapses to single curve →

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

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



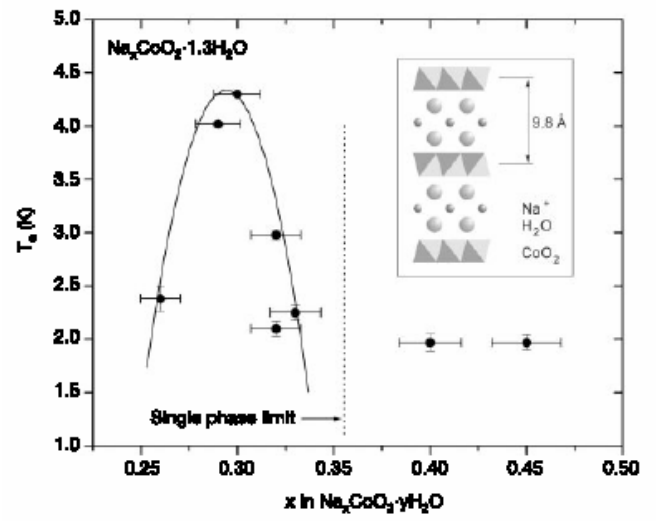


Figure 4 The superconducting phase diagram for $Na_xCoO_2 \cdot 1.3H_2O$. Main panel, T_c as a

← Cava, Nature May Tc versus x

ARPES/FS two preprints at x~.6,.7

ARPES on $Na_{0.6}CoO_2$: Fermi surface, extended flat dispersion, and unusual band splitting

H.-B. Yang,[1] S.-C. Wang,[1] A.K.P. Sekharan,[1] H. Matsui,[2] S. Souma,[2] T. Sato,[2] T. Takahashi,[2] T. Takeuchi,[3] J.C. Campuzano,[4] R. Jin,[5] B.C. Sales,[5] D. Mandrus,[5] Z. Wang,[1] H. Ding[1]

Fermi surface and quasiparticle dynamics of $Na_{0.7}CoO_2$ investigated by Angle-Resolved Photoemission Spectroscopy

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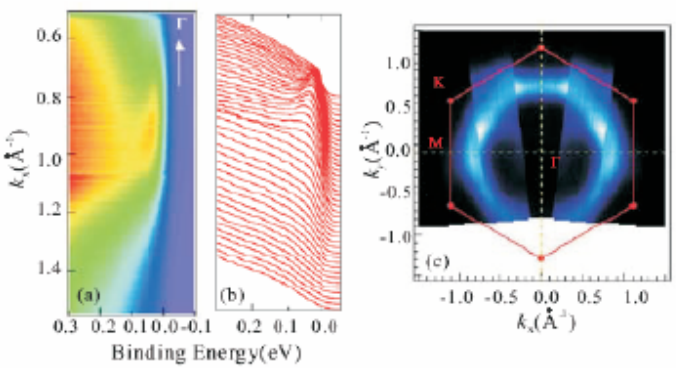


FIG. 2: Fermi Surface : (a)Γ →M Fermi crossing. Color red reflects the highest intensity - yellow to green to blue is in the order of decreasing intensity. (b) EDCs corresponding to the image plot in (a). (c) n(k) plot generated by integrating within 75 meV of Fermi level. A large hole-pocket is centered around the Γ-point. The Fermi surface, exhibiting some hexagonal anisotropy, is the inner edge of pocket as shown over the complete Brillouin zone.

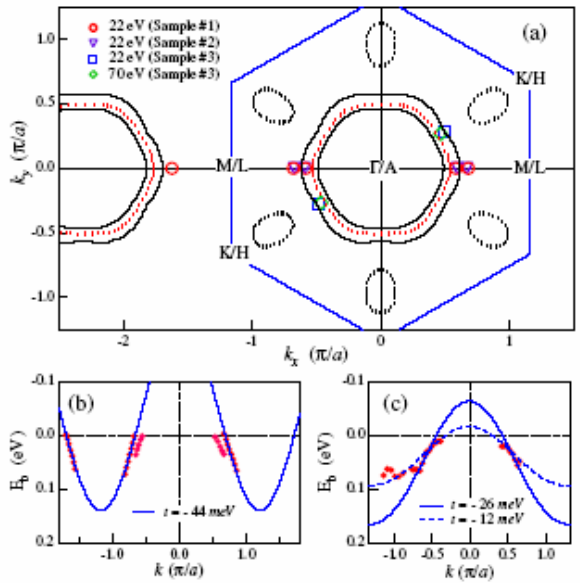


FIG. 4: (a) Measured FS crossings (symbols) comparing to the calculated FS in $k_z = 0$ (black solid lines) and $k_z = 0.5$ (red dashed lines) planes. The blue hexagon is the 2D Brillouin zone. (b) Extracted band positions along Γ-M (red dots) and a tight binding fit with $t = -44\ meV$ (solid line) (c) Extracted band positions along Γ-K (red dots) and two tight binding fits with $t = -12\ meV$ (solid line) and $t = -26\ meV$ (dashed line).

- Theoretical ideas:
  - Triangular lattice, hence frustrated in electronic sense as well as usual spin language
  - Possibility of non phononic SC, perhaps RVB, “the original lattice”, since low Tc
  - Temperature dependent susceptibility suggest low energy scale, hence possibly strongly correlated
  - Ferromagnetism / Antiferromagnetism? SDW state?
  - Charge Ordering?

Kumar and SS

Baskaran, 2003

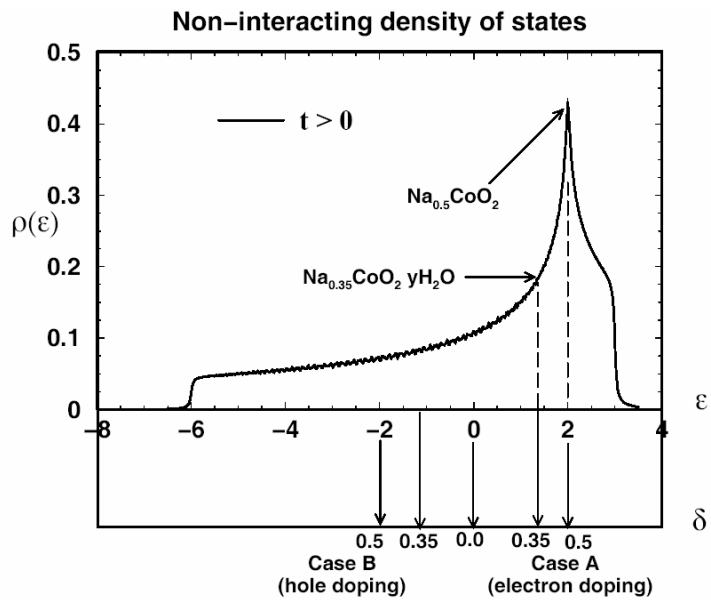
Q H Wang, Dung Hai Lee, Patrick Lee 2003

- t-J Model

$$\mathrm{H} = -t \sum_{<i,j>,\sigma} \mathcal{P} c^{\dagger}_{i\sigma} c_{j\sigma} \mathcal{P} + \mathrm{J} \sum_{<i,j>} (\vec{S}_i \cdot \vec{S}_j - \frac{n_i n_j}{4})$$

The fermi surface for triangular lattice tight binding band structure, $\epsilon(\mathrm{k}) = -2t(\cos(k_x) + 2\cos(k_x/2)\cos(\sqrt{3}k_y/2)$, gives a density of states

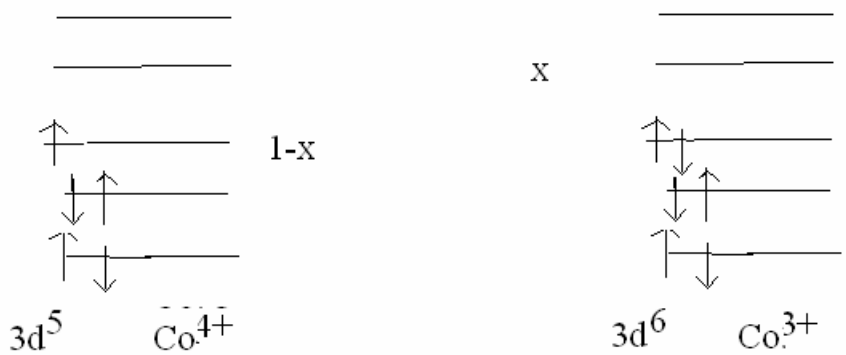
- Case A: Here we have either
  - (i) $t > 0$ and electron doping, or
  - (ii) $t < 0$ with hole doping, and
- Case B: where we have either
  - (i) $t > 0$ and hole doping, or
  - (ii) $t < 0$ and electron doping.

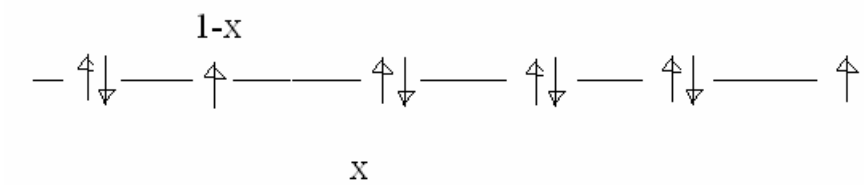


Co: $3d^7\ 4s^2$
Co^{4+} ---> $3d^5$
Co^{3+} ----> $3d^6$

$Na_x\ Co\ O_2$ gives x as fraction of Co^{3+}

Hence number of "singlons" i.e. projected electrons is "x"





Interpret x as number of electrons in a Gutzwiller projected model. NMR is consistent with this, experiments show x as the relative fraction of 3+ and 4+ states of Cobalt. This is called a low spin CF splitting and familiar in Co chemistry.

X= 0 gives 1 electron per “site” and should be a Mott Insulator. Prediction

Hall Constant in a strongly interacting fermi system:

•Highly mysterious. Boltzmann theory is not a good starting point, since z is either zero or very very small, so quasiparticle dominance fails.

•Big anomalies in High Tc: Boebinger etal (Nature 2003) show failure of Boltzmann theory

•R~1/(nec) ~ 1/(1+x)n' ec is far from seen, actually one sees R~1/x e c divergent as x->0.

•Theory is in poor shape, a new direction was charted by us 1993 using high frequency Hall constant. Started with the amusing observation that in the Drude model, while $\sigma(\omega)$ is frequency dependent, $\rho(\omega)$ is **independent** of ω due to complete cancellation. While cannot calculate anything at low ω, large ω is still managable:

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Faraday Rotation and the Hall Constant in Strongly Correlated Fermi Systems

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$$\sigma_{\alpha,\beta}(\omega) = \frac{ie^2}{\omega\Omega}\left[\langle\tau_{\alpha,\beta}\rangle - \frac{1}{\mathcal{Z}}\sum \frac{\exp-(\beta\epsilon_\nu) - \exp-(\beta\epsilon_\mu)}{\epsilon_\mu - \epsilon_\nu - \omega - i\eta}\langle\nu|J_\alpha|\mu\rangle\langle\mu|J_\beta|\nu\rangle\right].$$

The Hall resistivity, at a frequency ω, may be obtained from

$$R_H(\omega) = a_0^3\frac{\partial}{\partial B}\left(\frac{\sigma_{x,y}(\omega)}{\sigma_{x,x}(\omega)\sigma_{y,y}(\omega) - \sigma_{x,y}(\omega)\sigma_{y,x}(\omega)}\right)_{B=0}$$

in the weak field regime. The relevant part of $\sigma_{x,y}$ is antisymmetric in x, y, and h

$$\sigma_{[x,y]}(\omega) = -\frac{ie^2}{\mathcal{Z}\Omega}\sum \frac{\exp-(\beta\epsilon_\nu) - \exp-(\beta\epsilon_\mu)}{(\epsilon_\mu - \epsilon_\nu)^2 - \omega^2}\langle\nu|J_x|\mu\rangle\langle\mu|J_y|\nu\rangle.$$

For large ω ($>> \epsilon_\mu - \epsilon_\nu$), we have

$$\sigma_{[x,y]}(\omega) = \frac{ie^2}{\Omega\omega^2}\left[\langle[J_x, J_y]\rangle + \frac{1}{\omega^2}\langle[[[J_x, H], H], J_y]\rangle + O(1/\omega^4)\right],$$

while

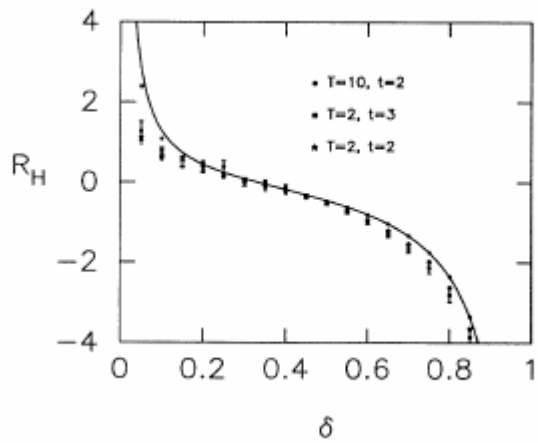
$$\sigma_{x,x}(\omega) = \frac{ie^2}{\Omega\omega}\left[\langle\tau_{x,x}\rangle + \frac{1}{\omega^2}\langle[[J_x, H], J_x]\rangle + O(1/\omega^4)\right].$$

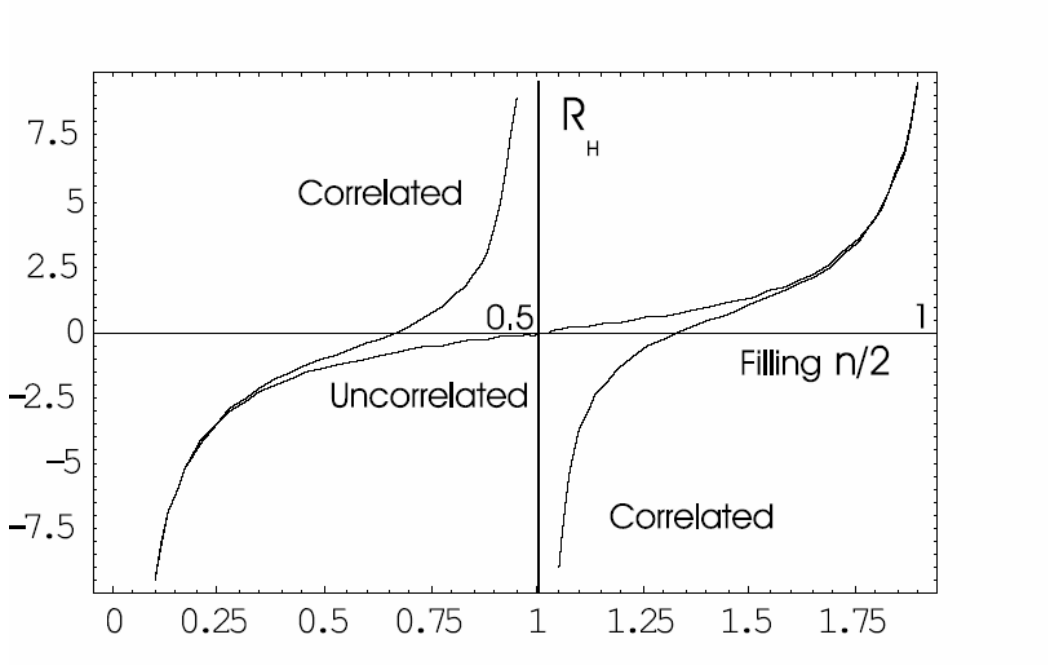
$$R_H^* = \frac{\tau_0}{4}\left[\frac{1 - \frac{3\beta J}{2}}{\delta} - \frac{4}{1-\delta} + 3 + \frac{3\beta J}{2}\right] + O(\beta^2).$$

$$R_H(\omega) = \frac{R_H^*}{1 - \Sigma_H(\omega)},$$

with

$$R_H^* = \lim_{B\to 0}\left(-\frac{ia_0^3 N}{Be^2}\frac{\langle[J_x, J_y]\rangle}{\langle\tau_{x,x}\rangle^2}\right).$$





Correlated e's n square lattice: Hall constant is shown to have **THREE** zero crossings, in contrast to single crossing for free e's

Hall constant versus filling for triangular lattice at fixed T versus filling is predicted to have NO crossings!!

[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.

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

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(Dated: May 8, 2003)

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 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.

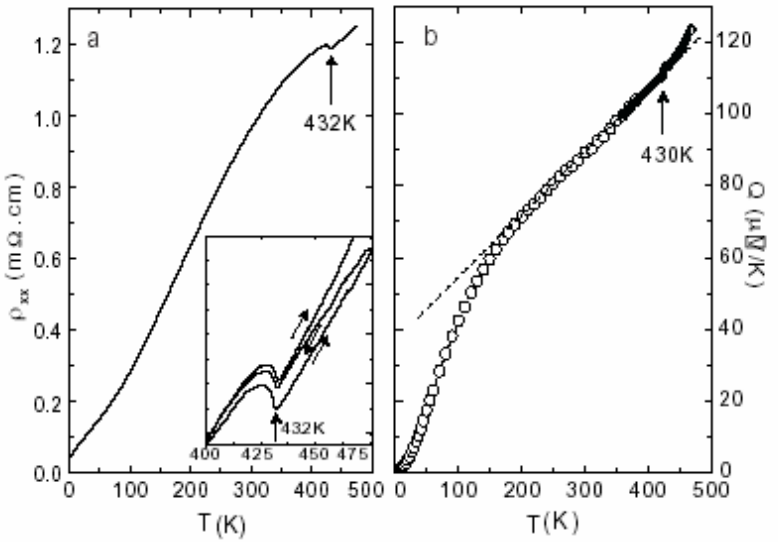


FIG. 1: (a) The in-plane resistivity ρ of Na_xCoO_2 ($x = 0.68$). ρ is linear in T from 2 to 80 K, but has a steeper slope above 100 K. The inset shows slight hysteresis in ρ in the vicinity of the transition at $T_D = 430$ K. (b) The in-plane thermopower Q of Na_xCoO2. The anomaly at T_D is just resolved in Q.

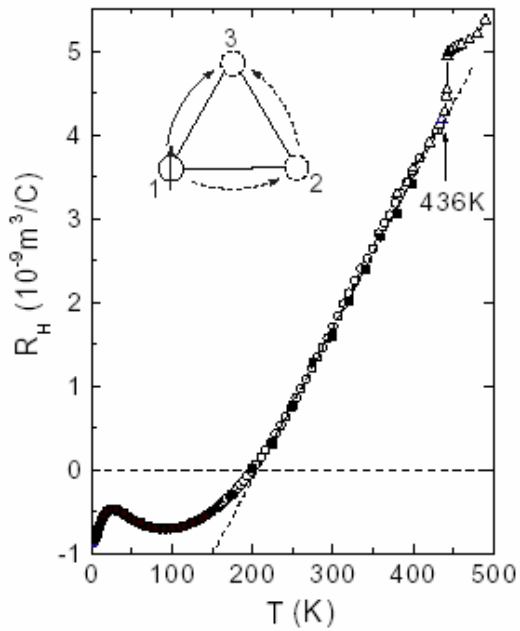


FIG. 2: The T dependence of the Hall coefficient R_H in Na_xCoO_2 showing anomalous T-linear increase between 200 and 500 K. The open circles are measurements using Method

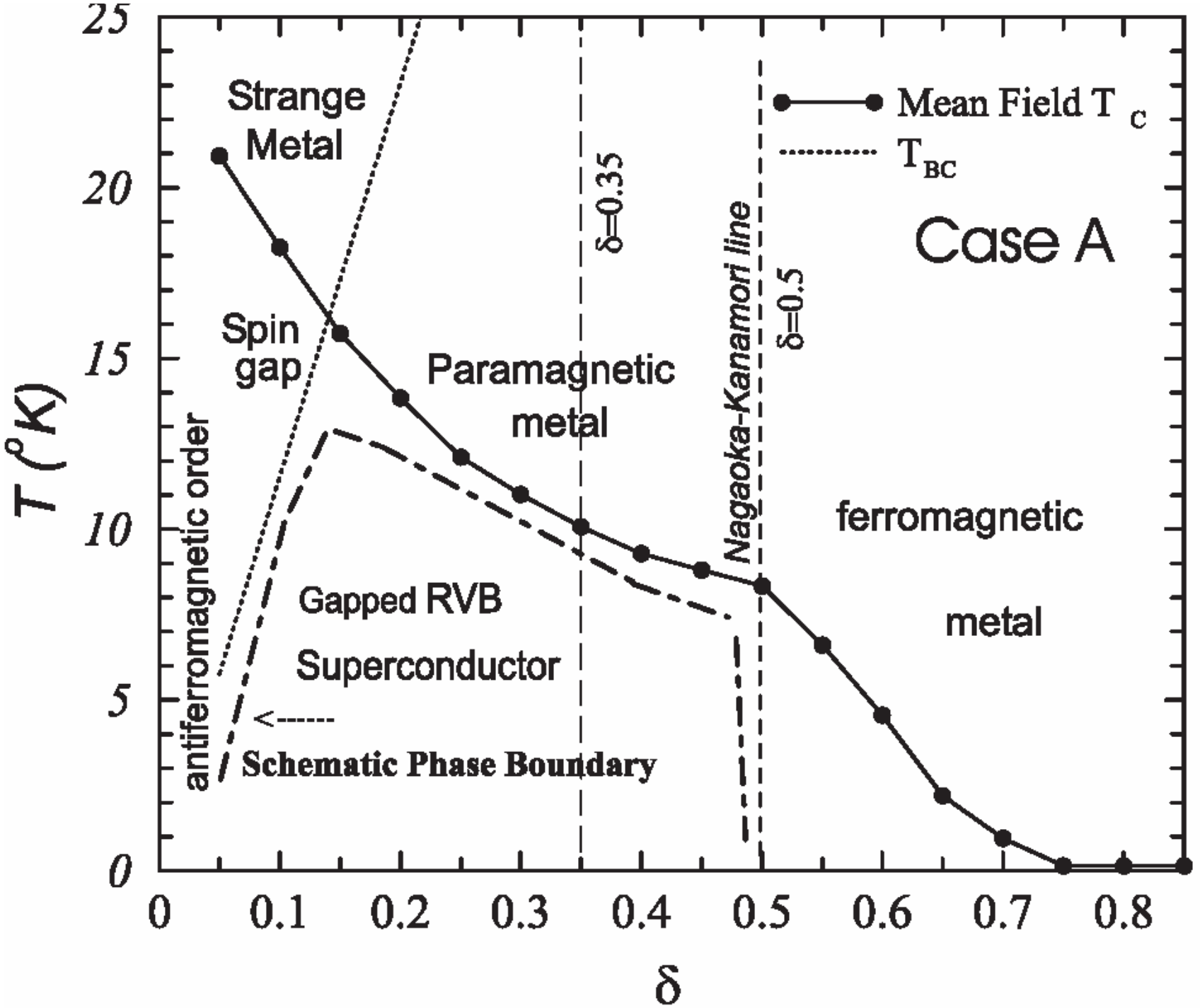
Prediction for $\omega >> \{J,t\}_{min}$ is (with v = volume of unit cell and x = δ)

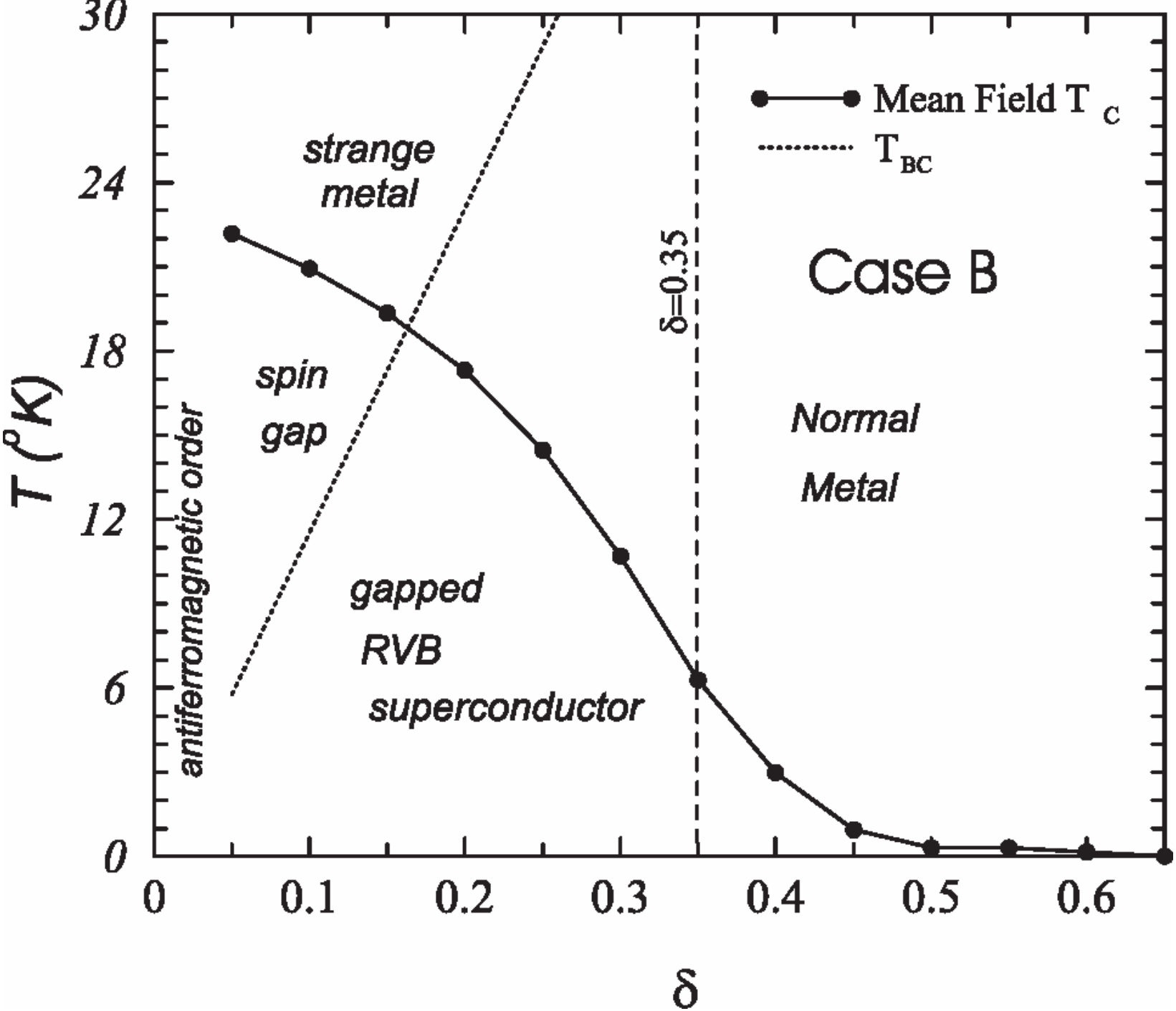
$$R_H^* = -\frac{v}{4|e|}\frac{k_B T}{t}\frac{1+\delta}{\delta(1-\delta)}$$

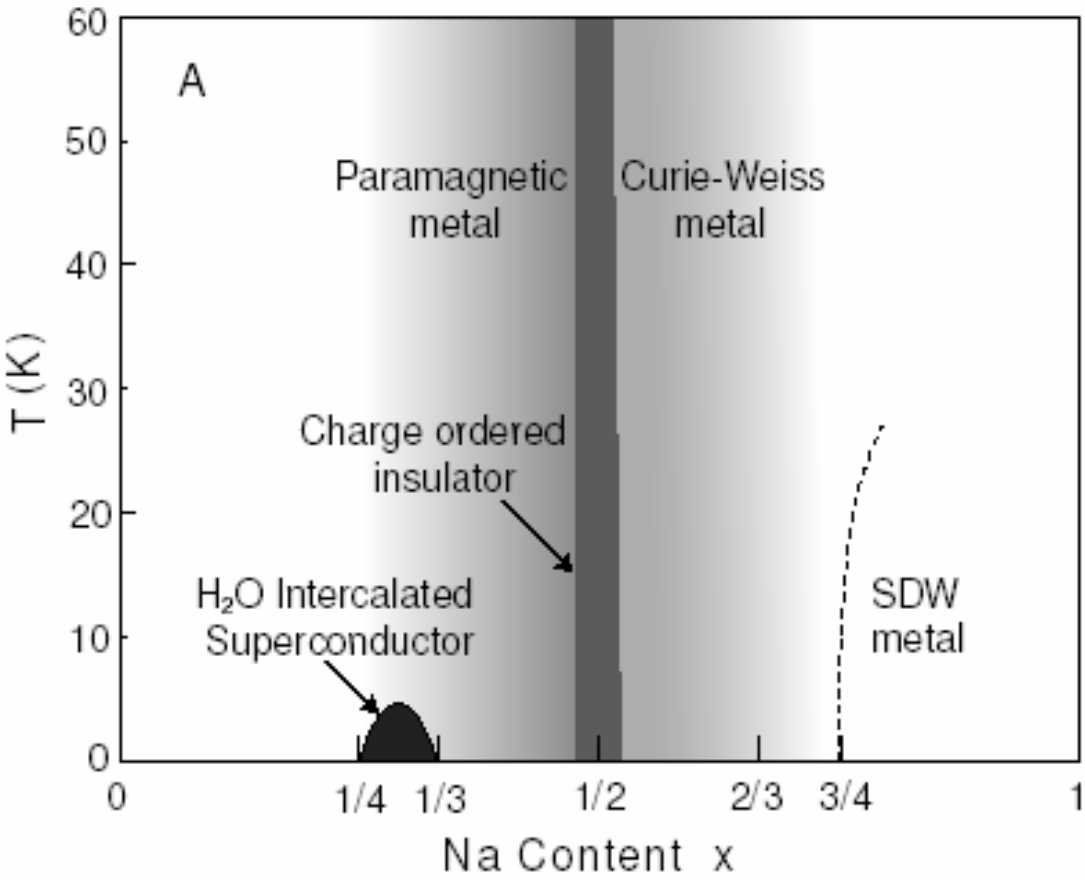
From Cava Ong (transport) Hall measurement we find on comparing with our prediction (for large frequencies):

- **Indeed Hall constant is linear in T over large range (200 to 400 K)**
- Slope can be used to deduce hopping: t <0 for x~.7 and |t| ~ 60^0K hence bandwidth ~550^0K.
- Fermi surface for this sign has unoccupied region around Γ point, ie hole like as seen in two recent experiments.
- Particularly interesting would be x~0 where Mott Hubbard physics is dominant.
- **Business with frequency**: **wanted $\rho_{xy}(w)$ for correlated systems from transport to large w.**

ρ preferable to σ


Strange
Metal
Spin
gap
Paramagnetic
metal
antiferromagnetic order
Gapped RVB
Superconductor
Schematic Phase Boundary
δ=0.35
Nagaoka-Kanamori line
δ=0.5
ferromagnetic
metal
Mean Field T c
T BC
Case A
T (°K)
δ
0
5
10
15
20
25
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8

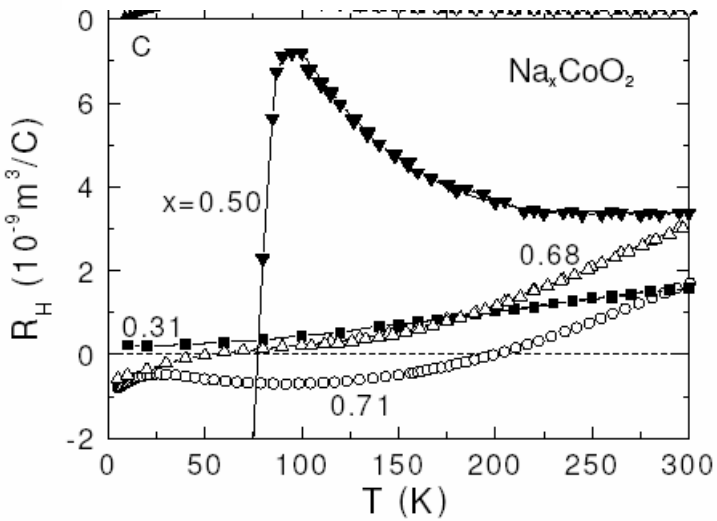

30
24
18
12
6
0
T (°K)
0
0.1
0.2
0.3
0.4
0.5
0.6
δ
Mean Field T c
T BC
strange
metal
spin
gap
antiferromagnetic order
gapped
RVB
superconductor
δ=0.35
Case B
Normal
Metal



Most recent experiments by Cava/Ong show a phase diagram that has new components: charge ordering at x=.5

(condmat Dec 18, 2003)

Is this due to degenerate bands and a "chemical transition"?



Open Questions:

Fermi surface, renormalization effects, sign of t?

Optimium Tc?

Prediction of time reversal violating superconducting state?

Magnetism? Ferro : Nagaoka/Kanamori for t>0 and other orderings for t<0.

Possibly a fermi surface switching transition as a function of x? t<0 to t>0 due to competing bands, a chemical transition.

Battery connections? Li_x Co O_2

Big Question:

Why is there a low energy scale in this problem? T_f should be 3-4,000 degrees but seems to be in 100's.

Conclusions:

- **Frustrated and low energy scale Mott Hubbard systems are GOOD.**
- **Testing grounds for theories, better than High Tc systems with concomitant high energy scales.**
- **Theoretically good since classical correlated systems, while being highly non trivial, are yet much easier to deal with than degenerate strongly correlated systems. If T_f is low, we can do reliable theory.**