A cluster Mott Insulator on the pyrochlore lattice

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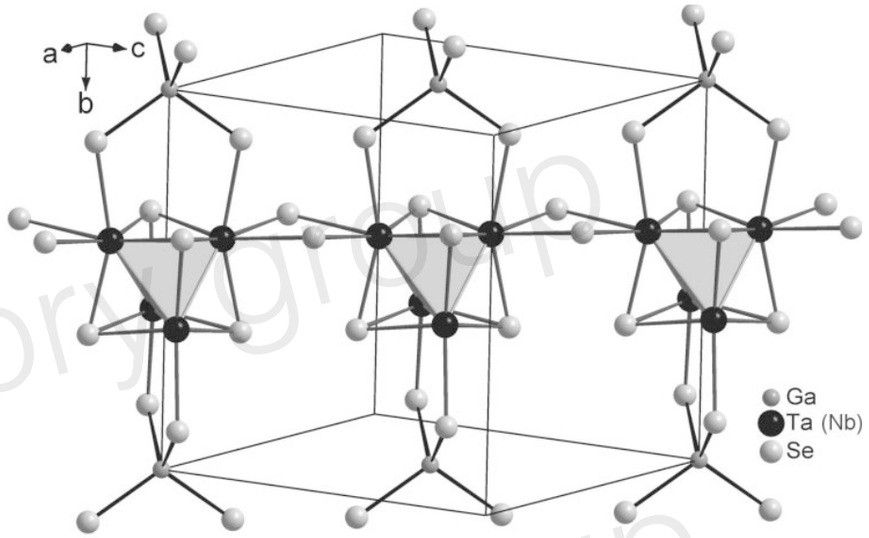
Outline

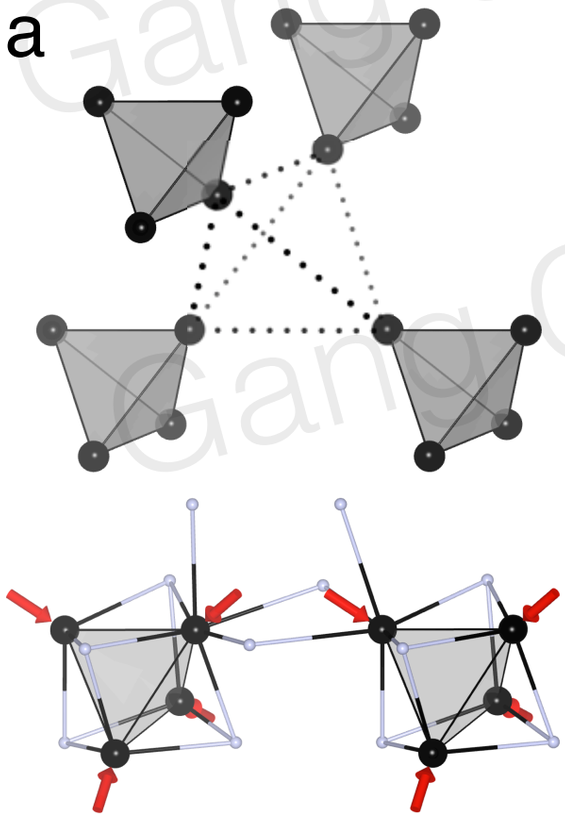
- Some experiments on AM_4X_8
- Some Models
- Mean field theory
- Beyond mean field theory

Lacunar spinel compound: AM4X8

A= Ga,Ge;
M=V, Mo, Ta, Nb, Cr
X (Q) = Se,Te,S

M: fractional valence charge
Pyrochlore lattice system





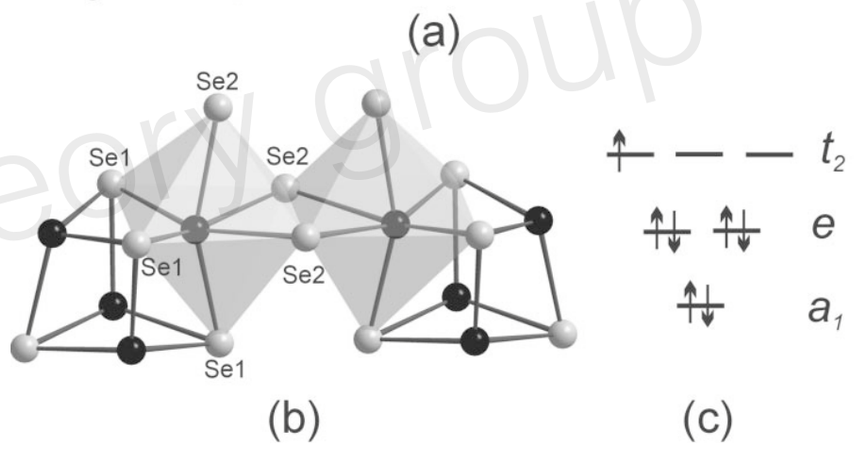


FIG. 1. (a) Linkage of the Ta_4Se_4 cluster units via bridging Se2 atoms and their connection with the $GaSe_4$ tetrahedra in the fcc $GaMo_4S_8$ structure. (b) (Ta,Nb) atoms shifted off the centers of distorted edge-sharing Se_6 octahedra ($d_{\text{Ta-Se1}} = 2.508$ Å; $d_{\text{Ta-Se2}} = 2.643$ Å). (c) Molecular orbital (MO) scheme for the *M-M* bonding orbitals of a M_4 cluster with ideal T_d symmetry for seven electrons per cluster.

Local molecules/clusters

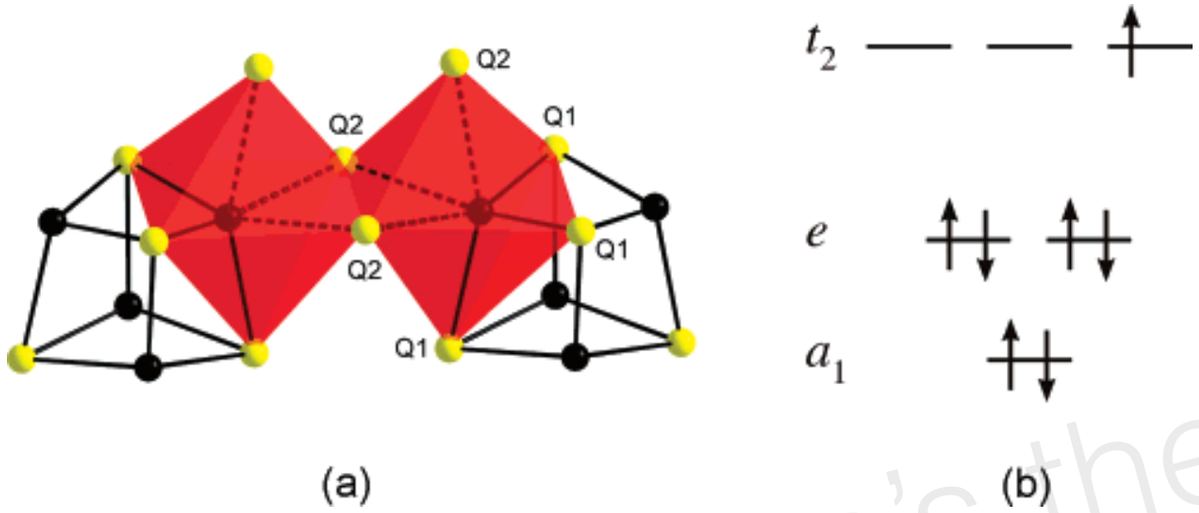


Figure 2. (a) Connection of two M_4Q_4 cubes in GaM_4Q_8. The M atoms (black spheres) are located in distorted, edge-sharing MQ_6 octahedra (red) with three shorter $M-Q1$ (solid) and three longer $M-Q2$ bonds (dashed). (b) MO scheme of the tetrahedral M_4-cluster with M = V, Nb, Ta, occupied by seven electrons according to the formula $GaM_4^{3.25+}Q_8^{2-}$.

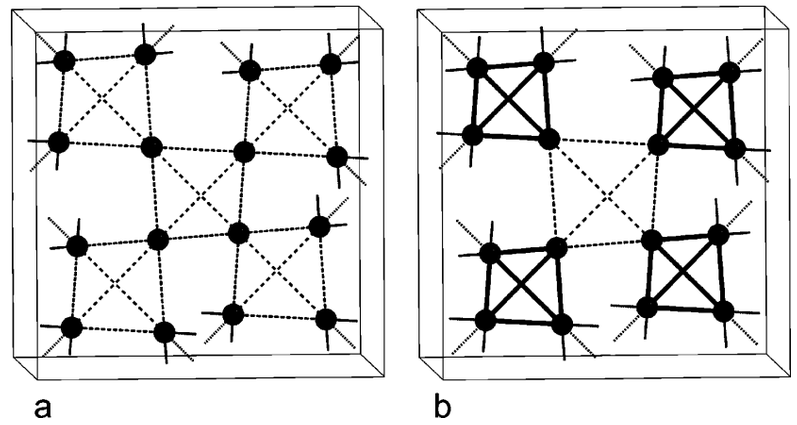


Figure 2. Substructures of the metal atoms in the spinel-type (a) and in the $GaMo_4S_8$ structure (b).

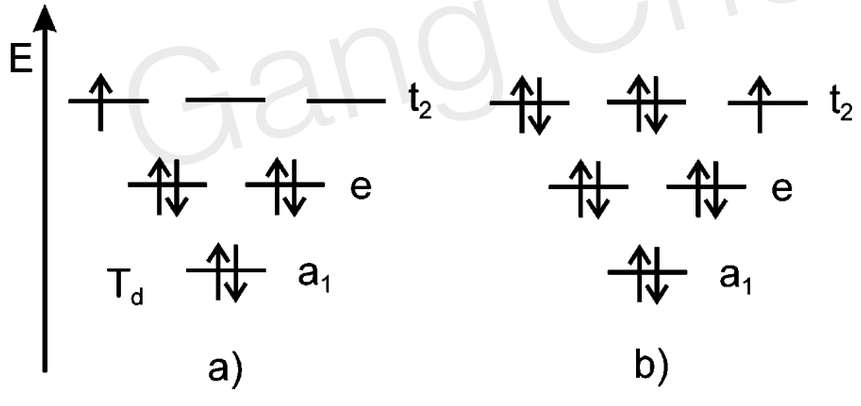


Figure 3. Molecular orbital schemes for the tetrahedral M_4 clusters: (a) GaV_4S_8 and (b) $GaMo_4S_8$.

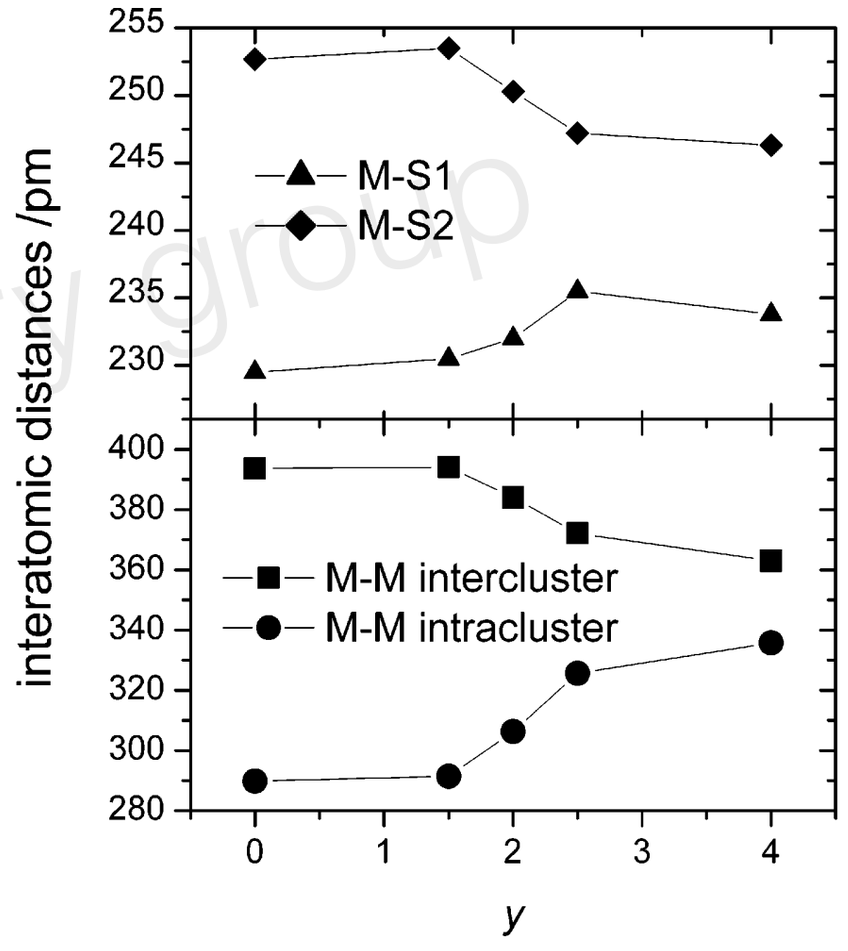


Figure 5. $M-S$ and $M-M$ (M = V/Cr) distances in the series $Ga_xV_{4-y}C$

Metal-insulator transition

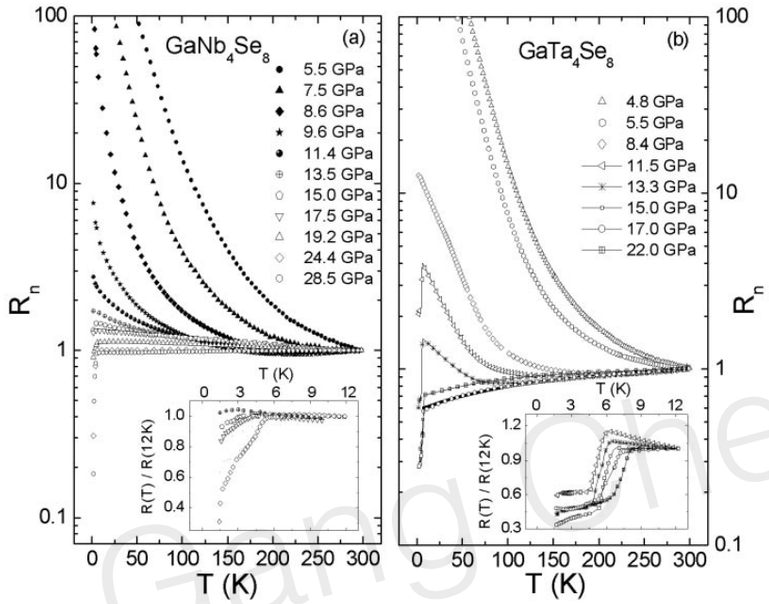


FIG. 2. Temperature dependence of the normalized electrical resistance $R_n = [R(T)/R(297\ \mathrm{K})]$ of $GaNb_4Se_8$ (a) and $GaTa_4Se_8$ (b) at different pressures up to 28.5 GPa. The insets show the drop of R_n at high pressures and low temperatures.

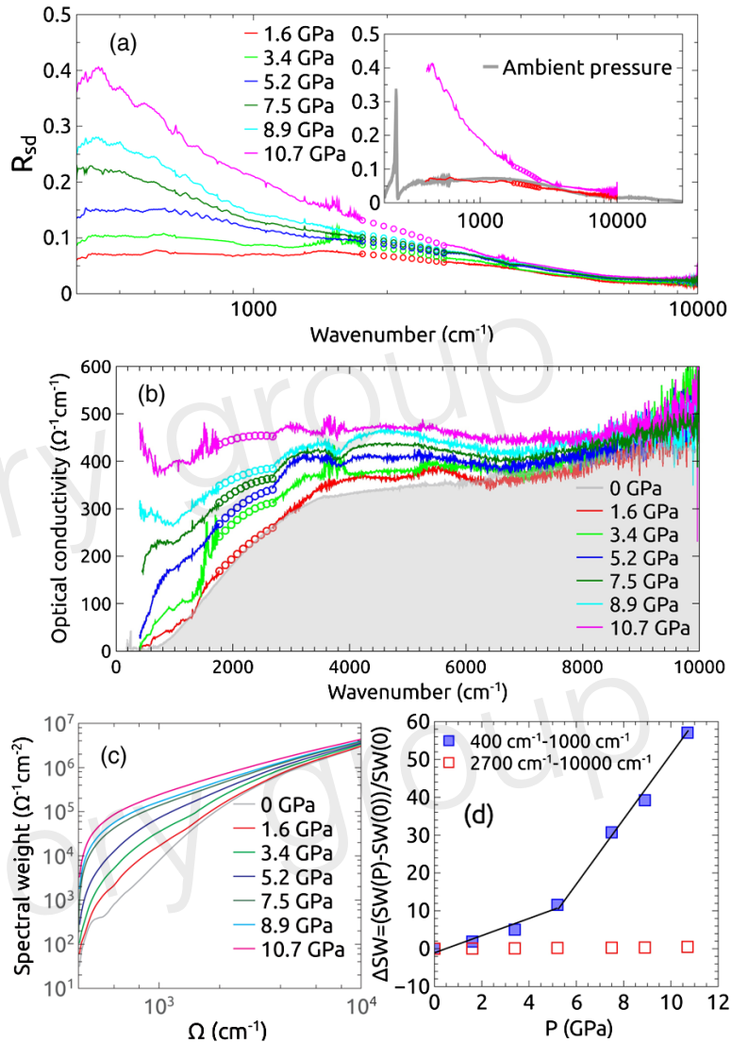


FIG. 1 (color online). (a) Renormalized $R_{\mathrm{sd}}(\omega)$ reflectivity at the sample-diamond interface from 1.6 GPa (lowest) to 10.7 GPa (highest curve), compared to $R_{\mathrm{sd}}^{\mathrm{cal}}(\omega)$ (see text). (b) Pressure-dependent optical conductivity $\sigma(\omega)$ from 1.6 GPa (lowest) to 10.7 GPa (highest curve). The dashed area corresponds to ambient pressure optical conductivity. (c) Spectral weight $\mathrm{SW}_{\Omega_0}^{\Omega}(P) = \int_{\Omega_0}^{\Omega} \sigma(\omega, P)d\omega$, integrated between $\Omega_0 = 400\ \mathrm{cm}^{-1}$ and Ω for various pressures from 0 GPa (lowest) to 10.7 GPa (highest curve). (d) Relative spectral weight $\frac{\mathrm{SW}_{\Omega_1}^{\Omega_2}(P) - \mathrm{SW}_{\Omega_1}^{\Omega_2}(0)}{\mathrm{SW}_{\Omega_1}^{\Omega_2}(0)}$ as a function of pressure, integrated at low energy ($\Omega_1, \Omega_2 = 400, 1000\ \mathrm{cm}^{-1}$) and high energy ($\Omega_1, \Omega_2 = 2700, 10\,000\ \mathrm{cm}^{-1}$). Solid lines are a guide for the eye.

GaNbS	10GPa	Mott to superconductor	Tc = 2.1K	noncentrosymmetric superconductor
GaTaSe	11.5GPa	Mott to superconductor	Tc=5.8K	noncentrosymmetric superconductor
GaNbSe	13GPa	Mott to superconductor	Tc= 2.9K	noncentrosymmetric superconductor

Some magnetism

GaV4S8 7e in the cluster	M=1.7	FM at Tc=10K	
GeV4S8 8e in the cluster	M=2.8	AFM at Tc=13K, q=[1/2,1/2,0]	might be some charge order?
GaNb4S8 s=1/2 cluster	-298K Curie-Weiss	spin singlet state with spin gap 200K	structural and magnetic xtion at 21K
GaMo4S8	structural xtion at 46.5K,	weakly FM below 13K	
GaNb4Se8	M=1.73		weak soc
GaTa4Se8	M=0.7		strong sox
GaMo4Se8		FM below 26.7K	

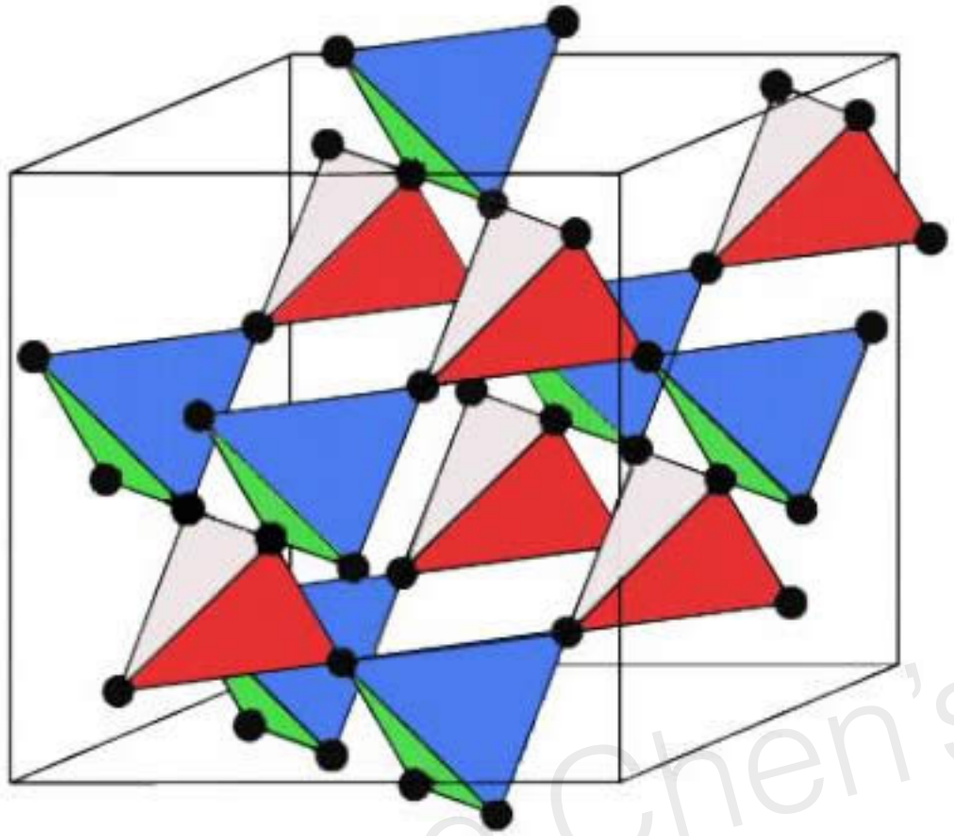
Basic features of these materials:

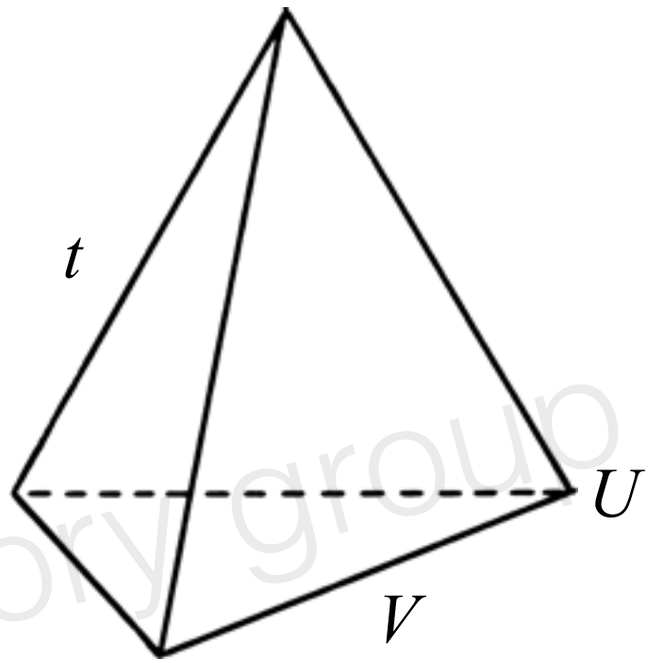
- Structure: 3D pyrochlore lattice, break inversion, chemical doping can do something;

- charge transport: mostly Mott insulator, can become metal or superconductor under high pressure, electrons are localized to the cluster not to lattice sites;

- magnetism: not very clear. Very limited experiments suggests some ordering in some material. Current “theory” assumes molecular/cluster orbitals and spins, spin-orbit coupling seems to be important in some compounds

Pyrochlore lattice Hubbard model





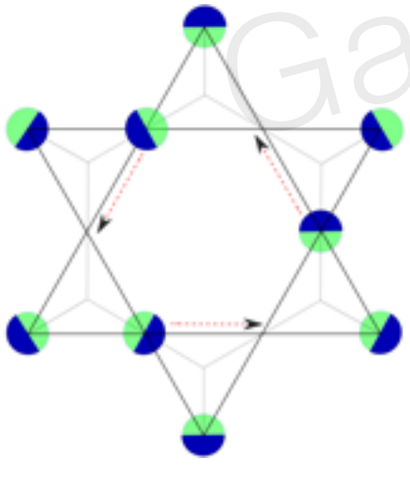
$$H = -\sum_{\langle ij \rangle} t(c^{\dagger}_{i\sigma} c_{j\sigma} + h.c.) + V \sum_{\langle ij \rangle} n_i n_j + \frac{U}{2} \sum_i (n_i - \frac{1}{2})^2 - \mu \sum_i n_i$$

- 1/2 filling: 1 electron per site
- 1/4 filling: 2 electron per tetrahedron
- 1/8 filling: 1 electron per tetrahedron

site mott insulator vs cluster mott insulator

For 1/2 filling, electron becomes localized on the lattice site in the strong Mott insulator. At weak Mott regime, maybe interpreted as “metallic in short distance and insulator in long distance”.

For 1/4 filling, electron becomes localized on the tetrahedral cluster in the strong Mott insulator. There are two electrons per cluster. Since there are empty sites, electron can do some collective motion.



Similar for 1/8 filling, there is only 1 electron in each cluster

Metal-insulator transition

$c^\dagger_{i\sigma} = e^{i\theta_i} f^\dagger_{i\sigma}$ rotor takes care of the bosonic charge

fermion takes care of the spin

with a constraint $L^z_i = (\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma}) - \frac{1}{2},$

One may justify/understand this from many body wavefuction

$$\Psi_c(r_1, r_2, \cdots) = \Psi_\theta(r_1, r_2, \cdots) \times \Psi_f(r_1, r_2, \cdots)$$

For 1/2 filling,

metal: superfluid wavefct for rotor

insulator: boson mott insulator wavefct for rotor

in terms of operator,

$$\langle \theta \rangle \neq 0 \quad \text{or} \quad \langle \theta \rangle = 0$$

For 1/4 filling, one needs a different boson wavefct

$$H = -\sum_{\langle ij\rangle} t(c^\dagger_{i\sigma}c_{j\sigma} + h.c.) + V\sum_{\langle ij\rangle} n_i n_j + \frac{U}{2}\sum_i (n_i - \frac{1}{2})^2 - \mu\sum_i n_i$$

With slave rotor decoupling

$$c^\dagger_{i\sigma}c_{j\sigma} \sim \langle e^{i(\theta_i-\theta_j)}\rangle f^\dagger_{i\sigma}f_{j\sigma} + e^{i(\theta_i-\theta_j)}\langle f^\dagger_{i\sigma}f_{j\sigma}\rangle$$

then we have two Hamiltonian for spin and charge, respectively,

$$H_{\rm S} = -\sum_{\langle ij\rangle,\sigma} t^{\rm eff}_{ij}(f^\dagger_{i\sigma}f_{j\sigma} + h.c.) - \sum_{i,\sigma}(\mu + h_i) f^\dagger_{i\sigma}f_{i\sigma}$$

$$H_{\rm C} = -\sum_{\langle ij\rangle} J^{\rm eff}_{ij}(e^{i\theta_i - i\theta_j} + h.c.) + V\sum_{\langle ij\rangle} L^z_i L^z_j$$

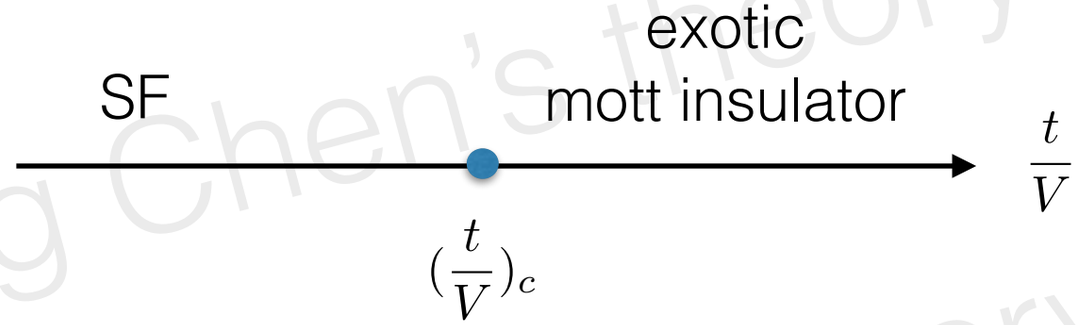
$$+3V\sum_i L^z_i + \sum_i h_i(L^z_i + \frac{1}{2}) + \frac{U}{2}\sum_i (L^z_i)^2.$$

Here, $t^{\rm eff}_{ij} = t\langle e^{i\theta_i - i\theta_j}\rangle$, $J^{\rm eff}_{ij} = t\sum_\sigma \langle f^\dagger_{i\sigma}f_{j\sigma}\rangle$

Charge sector Hamiltonian is nothing but a XXZ model

identify $e^{\pm i\theta} = L^{\pm}$

we know there are only two phases of this model

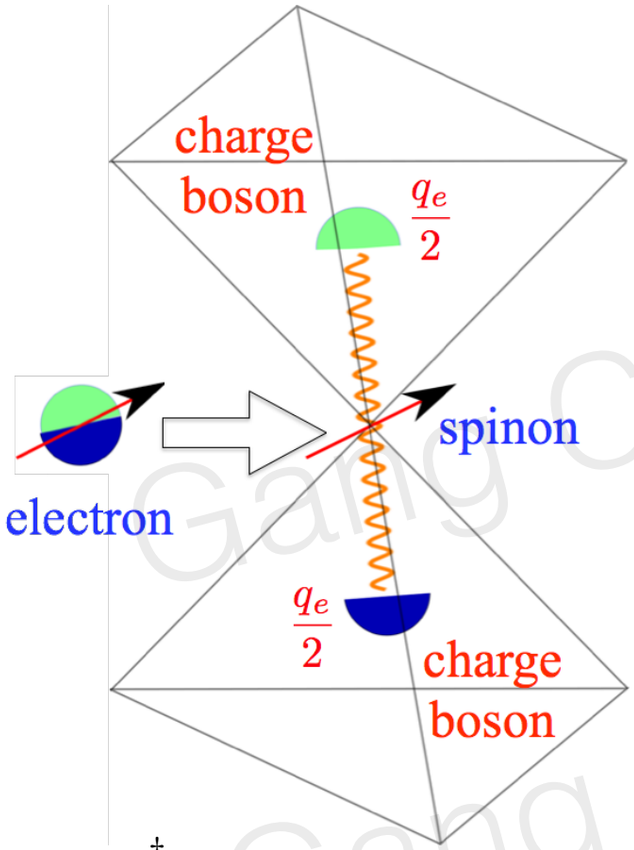


This exotic mott insulator is just quantum spin ice.
rotor fractionalizes into two bosons, each carries half the rotor charge quantum number.

Mott xtion occurs when the boson charge condenses

measure change with respect to the parent state

In terms of calculation (if one is interested), one needs to combine the slave-rotor mean field theory with the gauge mean field theory for the quantum ice together.



$$c^\dagger_{i\sigma} = \Phi^\dagger_{\mathbf{r}} \Phi_{\mathbf{r}'} l^+_{\mathbf{r}\mathbf{r}'} f^\dagger_{i\sigma}.$$

we need 2 internal U(1) gauge fields,
one for spin-charge separation
one for charge fractionalization

Operator	$Q^{\rm em}$	$Q^{\rm sc}$	$Q^{\rm c}$
$c^\dagger_{i\sigma}$	q_e	0	0
$f^\dagger_{i\sigma}$	0	1	0
$e^{i\theta_i}$	q_e	-1	0
$\Phi^\dagger_{\mathbf{r}},\ \mathbf{r}\in \mathrm{I}$	$q_e/2$	$-1/2$	1
$\Phi^\dagger_{\mathbf{r}},\ \mathbf{r}\in \mathrm{II}$	$-q_e/2$	$1/2$	1

$c^\dagger_1$

$c^\dagger_1 c_2 c^\dagger_3$

$c^\dagger_1 c_2 c^\dagger_3 c_4 c^\dagger_5$

$c^\dagger_1 c_2 c^\dagger_3 c_4 c^\dagger_5 c_6 c^\dagger_7$

$c^\dagger_1 c_2 c^\dagger_3 c_4 c^\dagger_5 c_6 c^\dagger_7 c_8 c^\dagger_9$

.....

introduce one charge to the system, but shared by two tetrahedra on its ends

Mott transition

in terms of wavefct, $\Psi_c = \Psi_{\Phi,\mathrm{I}}\Psi_{\Phi,\mathrm{II}}\Psi_f$

metal: superfluid wavefct for two bosons
mott insulator: bosonic mott insulator for two bosons

here, we assume inversion symmetry, so two bosons behave the “same”.

in terms of operator,

metal: $\langle\Phi\rangle \neq 0$

insulator: $\langle\Phi\rangle = 0$

Electron Green's function

$$G_{e,\alpha\beta}(i,j;\tau) = -\langle T c_{i\alpha}(\tau) c^{\dagger}_{j\beta}(0)\rangle = \Delta^2 G_{\mathrm{I}}(\mathbf{r}_i - \mathbf{r}_j, \tau) G_{\mathrm{II}}(\mathbf{r}'_j - \mathbf{r}'_i, -\tau) G_{f,\alpha\beta}(i,j;\tau)$$

Electron spectral function

$$\begin{aligned} A_e(\mathbf{k},\omega) = \sum_{\mathbf{k}_1,\mathbf{k}_2} \int_{E_1,E_2} & f(E_1)[n(E_1+E_2-\omega) - n(E_2)] \\ & \times \Delta^2 A_{\mathrm{I}}(\mathbf{k}_2, E_2) A_{\mathrm{II}}(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}, E_1+E_2-\omega) \\ & \times A_f(\mathbf{k}_1, E_1), \end{aligned} \tag{5}$$

1. activated behaviour in the mott phase: gap = 2* boson gap

2. pseudogap-like at mott transition point, A(w) ~ w^4

Beyond mean field theory, include gauge fluctuations

$$\mathcal{L} = \mathcal{L}_\Phi + \mathcal{L}_f + \mathcal{L}_\mathcal{A} + \mathcal{L}_a + \mathcal{L}_{bf} \tag{6}$$

$$\mathcal{L}_\Phi = \left|[\partial_\mu - i(\mathcal{A}_\mu - \frac{a_\mu}{2})]\Phi_{\rm I}\right|^2 + \left|[\partial_\mu - i(\mathcal{A}_\mu + \frac{a_\mu}{2})]\Phi_{\rm II}\right|^2$$
$$+ m^2[|\Phi_{\rm I}|^2 + |\Phi_{\rm II}|^2] + u[|\Phi_{\rm I}|^4 + |\Phi_{\rm II}|^4] + v|\Phi_{\rm I}|^2|\Phi_{\rm II}|^2$$

$$\mathcal{L}_f = \psi^\dagger_\sigma(\partial_\tau - ia_0 - \mu_f)\psi_\sigma + \frac{1}{2m_f}|(\nabla - i\mathbf{a})\psi_\sigma|^2$$

$$\mathcal{L}_\mathcal{A} = \frac{1}{4g_A^2}(\partial_\mu \mathcal{A}_\nu - \partial_\nu \mathcal{A}_\mu)^2, \;\; \mathcal{L}_a = \frac{1}{4g_a^2}(\partial_\mu a_\nu - \partial_\nu a_\mu)^2$$

$$\mathcal{L}_{f\Phi} = \lambda|\psi_\sigma|^2(|\Phi_{\rm I}|^2 + |\Phi_{\rm II}|^2).$$

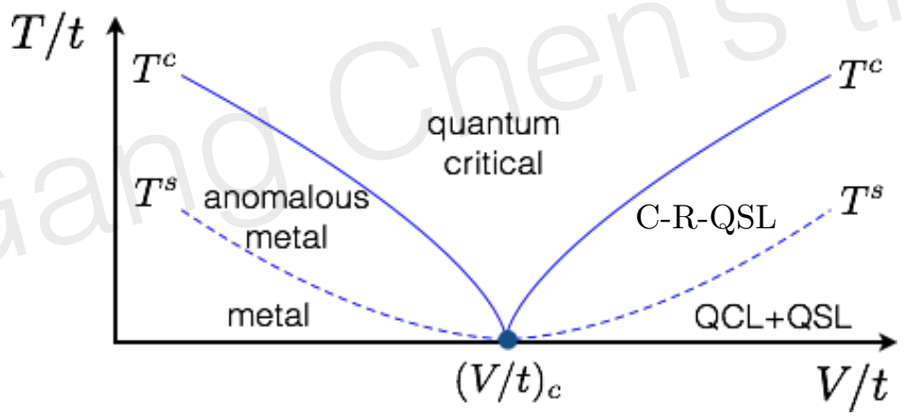


FIG. 2. The finite temperature crossover in the vincinty of the weakly first-order Mott transition.

Crossover in heat capacity and electric conductivity

heat capacity crossover signals the zs=3 dynamical exponent spin sector dominates the thermodynamics

$$C \approx \begin{cases} T \ln\ln 1/T & T > |V - V_c|^{3/2} \\ \gamma_1 T \ln 1/T & T < (V - V_c)^{3/2} \\ \gamma_2 T & T < (V_c - V)^{3/2} \end{cases}$$

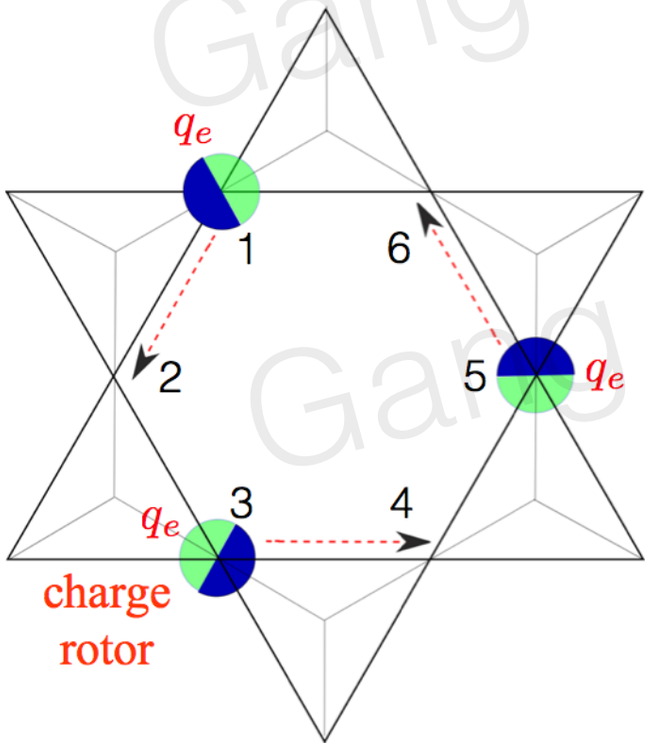
electric resistivity signals the zc=1 dynamical exponent and

$$\rho_c = \rho_f + (\rho_I^{-1} + \rho_{II}^{-1})^{-1}$$

note: the resistivity gap in the mott regime is single boson gap.

How about 1/8 filling?

With slave rotor, we obtain a xxz model for charge sector. The mott state (which is incompressible) corresponds to the 1/2 magnetization plateau of the xxz model. There is only one electron in each cluster, which turns into a Gauss law for the low energy physics.



Like quantum spin ice state, the low energy theory is also the U(1) compact QED.

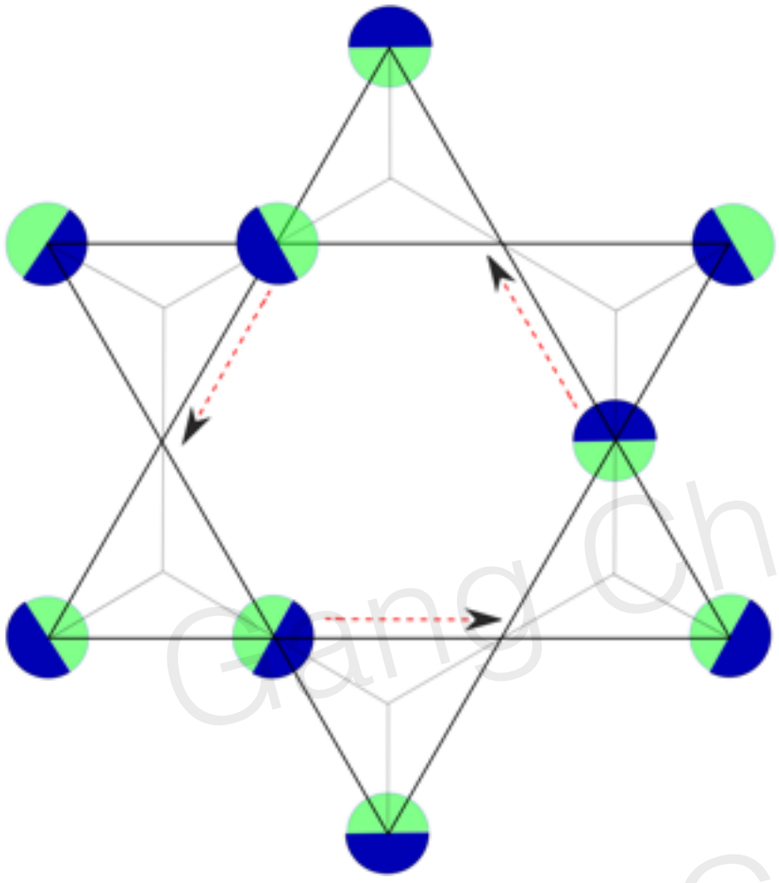
bose hubbard model at n=1,2,3,4,5, fixed density, there is emergent particle-hole symmetry and xy transition as one tune U/t.

here, we stay on the 1/2 magnetization plateau or fix the density/filling of the boson and tune the interaction.

we measure the charge of “quasiparticle” respect to the original state.

Strong Mott regime: spin becomes important

For 1/4 filling, the spin interaction appears in 2nd order



$$J_{ex} \sim \frac{t^2}{U+V}$$

we also the ring hopping

$$H_{\text{eff}} = -\frac{3t^3}{V^2} \sum_{\text{hexagon } \alpha\beta\gamma} \sum (c^\dagger_{1\alpha} c_{2\alpha} c^\dagger_{3\beta} c_{4\beta} c^\dagger_{5\gamma} c_{6\gamma}$$
$$+ c^\dagger_{1\alpha} c_{6\alpha} c^\dagger_{5\beta} c_{4\beta} c^\dagger_{3\gamma} c_{2\gamma} + h.c.).$$

not sure which one is larger. If exchange is dominant, one may favor some electron configuration that optimize the spin state first, then one may not have the U(1) quantum charge ice state. In opposite limit, one have quantum charge ice, spin is likely to form a quantum spin liquid.

Because in that limit, we have..

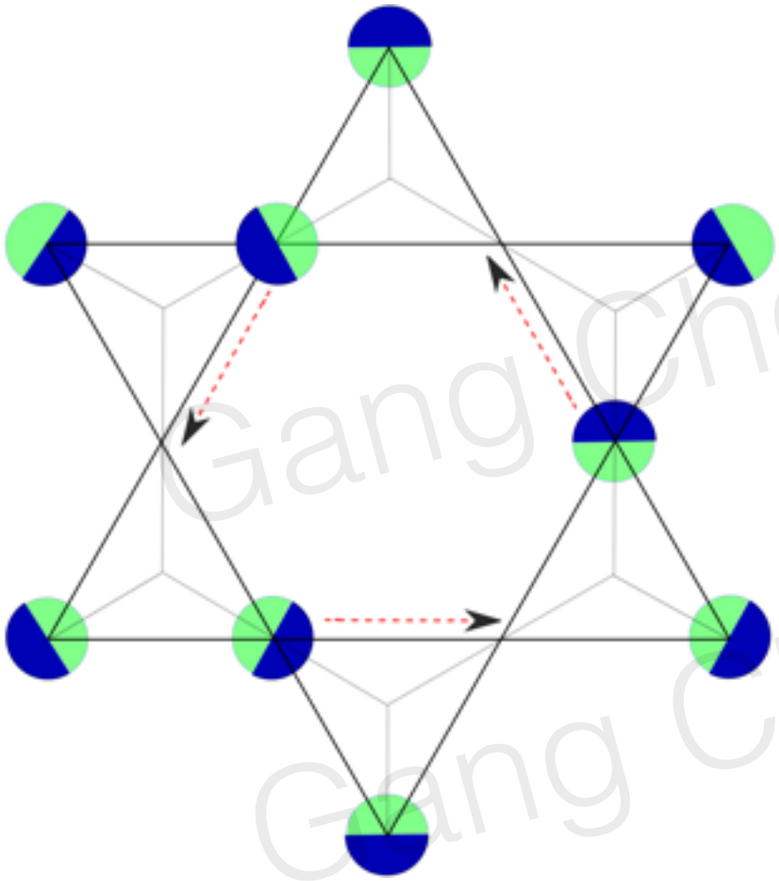
Remark:

1. the leading order spin interaction is not usual spin exchange nor ring exchange, it cannot be written into the standard spin operator. This is an important feature brought by the cluster Mott insulator.

2. for this kind of spin interaction, it is hard to define a mean mean field that couples to the spin, so it is more likely for the spins to form a quantum spin liquid than the usual magnets.

How about 1/8 filling

$V \gg t$ the leading interaction is the following ring hopping of electrons



$$H_{\text{eff}} = -\frac{3t^3}{V^2} \sum_{\text{hexagon}} \sum_{\alpha\beta\gamma} (c^\dagger_{1\alpha} c_{2\alpha} c^\dagger_{3\beta} c_{4\beta} c^\dagger_{5\gamma} c_{6\gamma}$$
$$+ c^\dagger_{1\alpha} c_{6\alpha} c^\dagger_{5\beta} c_{4\beta} c^\dagger_{3\gamma} c_{2\gamma} + h.c.).$$

spin-charge separation:
c = b f

Boson is hardcored because of the large onsite U

Decouple the electron ring hopping

$$c^\dagger_{1\alpha} c_{2\alpha} c^\dagger_{3\beta} c_{4\beta} c^\dagger_{5\gamma} c_{6\gamma} \Rightarrow \langle b^\dagger_1 b_2 b^\dagger_3 b_4 b^\dagger_5 b_6 \rangle f^\dagger_{1\alpha} f_{2\alpha} f^\dagger_{3\beta} f_{4\beta} f^\dagger_{5\gamma} f_{6\gamma}$$

$$\langle f^\dagger_{1\alpha} f_{2\alpha} f^\dagger_{3\beta} f_{4\beta} f^\dagger_{5\gamma} f_{6\gamma} \rangle b^\dagger_1 b_2 b^\dagger_3 b_4 b^\dagger_5 b_6$$

The charge sector Hamiltonian

$$H_c \sim - \sum_{hexagon} b^\dagger_1 b_2 b^\dagger_3 b_4 b^\dagger_5 b_6 + h.c$$

identical to the effective model to quantum spin ice! so we expect a quantum charge ice ground state for the charge sector.

What do we do for the spin?

$$f^\dagger_{1\alpha} f_{2\alpha} f^\dagger_{3\beta} f_{4\beta} f^\dagger_{5\gamma} f_{6\gamma} \to \langle f^\dagger_{3\beta} f_{4\beta}\rangle \langle f^\dagger_{5\gamma} f_{6\gamma}\rangle f^\dagger_{1\alpha} f_{2\alpha}$$

or directly from the original Hamiltonian

$$c^\dagger_{1\alpha} c_{2\alpha} \sim \langle b^\dagger_1 b_2\rangle f^\dagger_{1\alpha} f_{2\alpha}$$

This will lead to a quantum spin liquid with spinon Fermi surface

(the choice of hopping for 1/4 filling leads to some flat band at Fermi level. the flatness can be broken by extra hopping)

At finite temperature ?

Crossover instead of transition.

more general model for the actual quantum spin ice may allow 1st transition. Here we probably do not expect phase transition. One reason is that in the numerics, one does not see finite transition transition.

Crossover temperature $T \sim \frac{t^3}{V^2}$

quantum charge ice turns to a classical charge ice

What did we find?

We find a new phase whose spin and charge are both exotic. It seems to be a natural case to think for the cluster Mott insulator.

Experimental consequence:

1. Thermodynamics: Cv ~ T ln1/T + T^3
   spin susceptibility: chi ~ const (but complicated by SOC)

2. Transport: charge is fractionalized. difference between tunnelling conductance and electric conductance.

3. In (charge) gap physics: note the external magnetic flux can couple to internal ones, this may lead to the in gap optica conductivity/absorption (not looked at right now). (These are for the experts in the audience!)

Metal-insulator transition

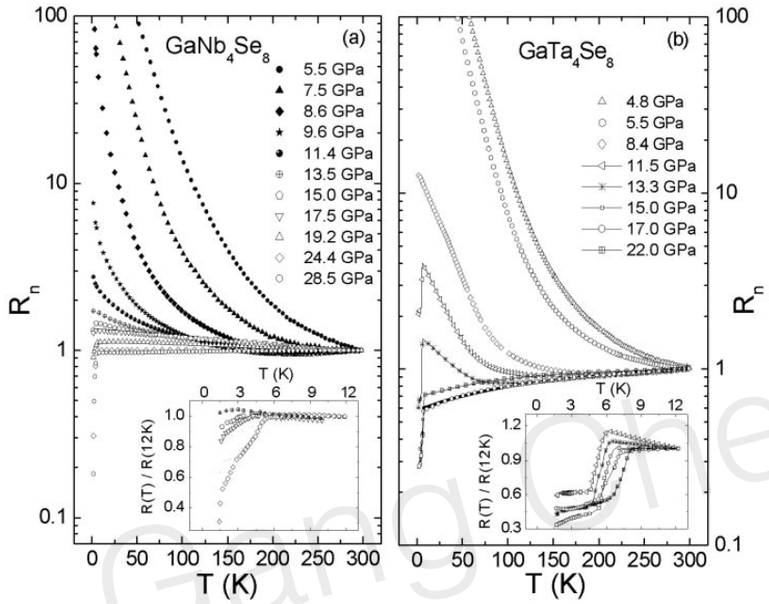


FIG. 2. Temperature dependence of the normalized electrical resistance $R_n = [R(T)/R(297\ \mathrm{K})]$ of $GaNb_4Se_8$ (a) and $GaTa_4Se_8$ (b) at different pressures up to 28.5 GPa. The insets show the drop of R_n at high pressures and low temperatures.

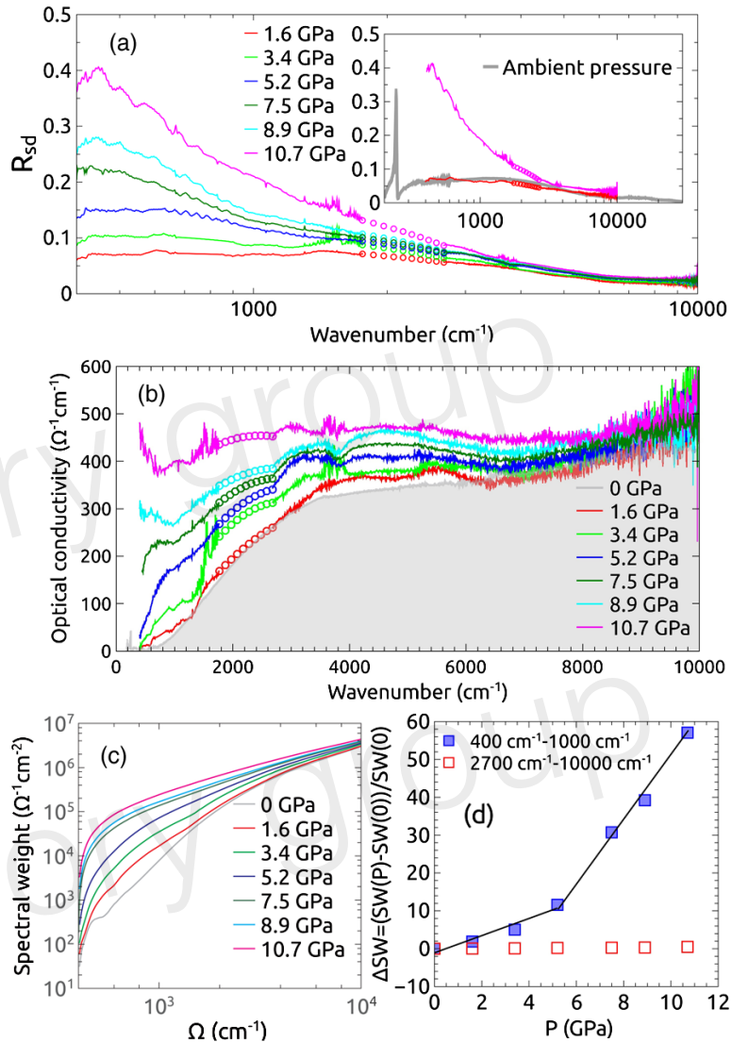


FIG. 1 (color online). (a) Renormalized $R_{\mathrm{sd}}(\omega)$ reflectivity at the sample-diamond interface from 1.6 GPa (lowest) to 10.7 GPa (highest curve), compared to $R_{\mathrm{sd}}^{\mathrm{cal}}(\omega)$ (see text). (b) Pressure-dependent optical conductivity $\sigma(\omega)$ from 1.6 GPa (lowest) to 10.7 GPa (highest curve). The dashed area corresponds to ambient pressure optical conductivity. (c) Spectral weight $\mathrm{SW}_{\Omega_0}^{\Omega}(P) = \int_{\Omega_0}^{\Omega} \sigma(\omega, P)d\omega$, integrated between $\Omega_0 = 400\ \mathrm{cm}^{-1}$ and Ω for various pressures from 0 GPa (lowest) to 10.7 GPa (highest curve). (d) Relative spectral weight $\frac{\mathrm{SW}_{\Omega_1}^{\Omega_2}(P) - \mathrm{SW}_{\Omega_1}^{\Omega_2}(0)}{\mathrm{SW}_{\Omega_1}^{\Omega_2}(0)}$ as a function of pressure, integrated at low energy ($\Omega_1, \Omega_2 = 400, 1000\ \mathrm{cm}^{-1}$) and high energy ($\Omega_1, \Omega_2 = 2700, 10\,000\ \mathrm{cm}^{-1}$). Solid lines are a guide for the eye.

Kagome Hubbard model at 1/6 filling