Mott transition and anomalous resistive state in the pyrochlore molybdates

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received 13 May 2017; accepted in final form 14 August 2017; published online 8 September 2017

PACS 71.27.+a – Strongly correlated electron systems; heavy fermions
PACS 71.30.+h – Metal-insulator transitions and other electronic transitions
PACS 75.10.-b – General theory and models of magnetic ordering

Abstract – The rare-earth based pyrochlore molybdates involve orbitally degenerate electrons of Hund’s coupled to local moments. The large Hund’s coupling promotes ferromagnetism, the superexchange between the local moments prefers antiferromagnetism, and Hubbard repulsion tries to open a Mott gap. The phase competition is tuned by the rare-earth ionic radius, decreasing which leads to change from a ferromagnetic metal to a spin disordered highly resistive ground state, and ultimately an “Anderson-Mott” insulator. We attempt a quantitative theory of the molybdates by studying their minimal model on a pyrochlore geometry, using a static auxiliary field based Monte Carlo. We establish a thermal phase diagram that closely corresponds to the experiments, predict the hitherto unexplored orbital correlations, quantify and explain the origin of the anomalous resistivity, and present dynamical properties across the metal-insulator transition.

Introduction. – Traditional Mott materials involve a strong on-site Coulomb interaction that, beyond a critical value, and at integer filling, inhibits electron motion [1]. This, in a clean material, leads to an abrupt change in the zero-temperature state from perfectly conducting to non-conducting. The nonconducting state typically has strong antiferromagnetic (AF) correlations, if not long-range order, since that lowers the kinetic energy.

The Mott transition on a frustrated structure brings in a novelty since the AF ordered state in the Mott phase cannot be realized and one may have the signatures of a “spin liquid” [2,3]. Such phases are realised in some triangular lattice organics [4–6]. The pyrochlores [7] are also highly frustrated structures, much studied for possible spin liquid phases, but the rare-earth molybdates, $R_2Mo_2O_7$, add additional twists to the Mott problem: i) the Mott transition in these materials occur in the background of overall ferromagnetic correlation [7–9], and ii) the zero temperature resistivity seems to grow continuously with the control parameter [10] (see next) rather than having an abrupt zero to infinity transition. These features owe their origin to the additional degrees of freedom, and couplings, involved in these materials.

The $R_2Mo_2O_7$ family exhibit ground states that vary from a ferromagnetic metal (FM) to a spin glass metal (SG-M) and then a spin glass insulator (SG-I) as the rare-earth radius $r_R$ is reduced [11]. Materials with $R = Nd$ and $Sm$ are metallic, $R = Tb$, Dy, Ho, Er, and $Y$ are insulating, and $R = Gd$ is on the verge of the insulator-metal transition (IMT) [9,10,12]. The highest observed ferromagnetic transition temperature $T_c$ is $\sim 100$ K, in Nd, while the spin glass transition temperature, $T_{SG}$ is typically $\sim 20$ K [13–15].

The unusual features in transport include very large residual resistivity, $\sim 10 \text{ m}\Omega \text{cm}$ close to the metal-insulator transition [10], prominent anomalous Hall effect in metallic samples, e.g., $Nd_2Mo_2O_7$ [16–20], and magnetic-field-driven metallisation in the weakly insulating samples, e.g., $Gd_2Mo_2O_7$ [21].

The qualitative physics behind these effects is not a mystery, but several major questions remain unanswered, e.g., on i) the simultaneity of the metal-insulator and magnetic transition in the ground state, ii) the thermal scales for the magnetic transitions, iii) the orbital state, whose fate is entangled with the spin state, and iv) the transport near the Mott transition.

We employ a real space approach, using a static auxiliary orbital field to handle the Hubbard interaction, and solve the resulting “electron-local moment-orbital moment” problem via a Monte Carlo scheme on the pyrochlore lattice. After making parameter choices...
suggested by \textit{ab initio} estimates [22,23] our main results are as follows.

i) We obtain a phase diagram with ferromagnetic metal and spin disordered metal and insulator phases. However, our disordered state is a “spin liquid” in contrast to the experimental spin glass. ii) The FM four disordered state is a “spin liquid” in contrast to the exchange spin disordered metal and insulator phases. However, our transport results bear almost quantitative correspondence with experiments [10,12], explain the high residual resistivity in terms of spin and orbital disorder scattering, and predict a highly nonmonotonic temperature dependence for samples of the form Gd$_2$-$_x$Sm$_x$Mo$_2$O$_7$. iv) The temperature and correlation dependence of our intermediate frequency optical spectral weight is very similar to the experiments [10,12] but changing the cutoff frequency reveals peculiarities, also reflected in the single-particle weight.

\textbf{Model and method.} – The R$_2$Mo$_2$O$_7$ structure consists of two interpenetrating pyrochlore lattices, one formed by Mo cations and the other by R. The Mo atom has octahedral oxygen coordination. The resulting crystal field splits the fivefold degenerate Mo 4d states into doubly degenerate $e_g$ and triply degenerate $t_{2g}$ manifolds, and a trigonal distortion splits the $t_{2g}$ further into a nondegenerate $a_{1g}$ and a doubly degenerate $e'_g$ [22]. The Mo cation is nominally tetravalent and has two electrons on average. The deeper $a_{1g}$ electron behaves like a local moment, and the single electron in the two $e'_g$ orbitals is the “itinerant” degree of freedom [22]. The $e_g$ states remain unoccupied. The accepted model [24] for these degrees of freedom takes the following form:

$$H = \sum_{ij} t_{ij} \alpha^\beta \epsilon_{i\alpha}^\dagger \epsilon_{j\beta}^\dagger + J_{\text{AF}} \sum_{ij} S_i \cdot S_j + U \sum_i n_{i\alpha} n_{i\beta},$$

where $H_{\text{AF}} = J_{\text{AF}} \sum_{ij} S_i \cdot S_j$. The first term is the kinetic energy, involving nearest-neighbour intra- and inter-orbital $e'_g$ hopping. The second term is the Hund’s coupling between the $a_{1g}$ local moment $S_i$ and the $e'_g$ electrons, $J_{\text{AF}}$ is the AF superexchange coupling between local moments at neighbouring sites on the pyrochlore lattice, and $U$ represents onsite $e'_g$ Coulomb matrix elements.

To simplify the computational problem we treat the localized spins $S_i$ as classical unit vectors, absorbing the size $S$ in the magnetic couplings. Also, to reduce the size of the Hilbert space we assume that $J_{\text{AF}}/t \gg 1$, where $t$ is the typical hopping scale, so that only the locally “spin aligned” fermion state is retained. In this local basis the hopping matrix elements are dictated by the orientation of the $S_i$ on neighbouring sites. These lead to a simpler model:

$$H = \sum_{ij} \tilde{t}_{ij}^\alpha \bar{\epsilon}_{i\alpha}^\dagger \bar{\epsilon}_{j\beta}^\dagger + J_{\text{AF}} \sum_{ij} S_i \cdot S_j + U \sum_i n_{i\alpha} n_{i\beta},$$

where the fermions are now “spinless”. $U > 0$ is the inter-orbital Hubbard repulsion. The effective hopping is determined by the orientation of the localized spins. If $S_i = (\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i)$ and $S_j = (\sin \theta_j \cos \phi_j, \sin \theta_j \sin \phi_j, \cos \theta_j)$ then $t_{ij}^\alpha = \cos \theta_i \cos \theta_j + \sin \theta_i \sin \theta_j e^{-i(\phi_i-\phi_j)}$, with $t_{11}^1 = t_{12}^2 = t$ and $t_{12}^1 = t_{12}^2 = t'$. We set $t' = 1.5t$ as suggested by the density functional theory [23] and keep only nearest-neighbour hopping.

The first two terms represent fermions in a classical spin background and the resulting magnetic phase competition has been studied on a pyrochlore lattice [25]. While these results are interesting they miss out on the large correlation scale, $U$, that drives the Mott transition. One option is to treat the model within the dynamical mean-field theory (DMFT) [26], but then the spatial character crucial to the pyrochlore lattice is lost.

We opt to handle the problem in real space as follows: i) We use a Hubbard-Stratonovich (HS) [27–29] transformation that decouples $Un_{i\alpha} n_{i\beta}$ in terms of an auxiliary orbital moment $\Gamma_i(\tau)$, coupling to $O_i = \sum_{\mu\nu} \xi_{i\mu}^\dagger \xi_{i\nu} c_{i\mu\nu}$, and a scalar field $\Phi_i(\tau)$ coupling to $n_i$ at each site [30]. ii) An exact treatment of the resulting functional integral requires quantum Monte Carlo. Here we retain only the zero Matsubara frequency modes of $\Gamma_i$ and $\Phi_i$, i.e., approximate them as classical fields. iii) The spatial thermal fluctuations of $\Gamma_i$ are completely retained, while $\Phi_i$ is treated at the saddle point level, setting $\Phi_i \to \langle \Phi_i \rangle = (U/2)\langle n_i \rangle = U/2$ at half-filling (since charge fluctuations are expensive at large $U$). This method overall is known as a “static path approximation” (SPA) to the functional integral for the many-body partition function and has been used earlier in several problems [31–33]. This leads to a more tractable problem,

$$H_{\text{eff}}(S_i, \Gamma_i) = -\frac{1}{\beta} \log \text{Tr} e^{-\beta H_{\text{eff}}} + H_{\text{AF}} + \frac{U}{4} \sum_i \Gamma_i^2,$$

$$H_{\text{eff}}(S_i, \Gamma_i) = \sum_{ij} \tilde{t}_{ij}^\alpha \bar{\epsilon}_{i\alpha}^\dagger \bar{\epsilon}_{j\beta}^\dagger - \tilde{\mu} \sum_i n_i - \frac{U}{2} \sum_i \Gamma_i \cdot O_i,$$

with $\tilde{\mu} = \mu - U/2$, $\mu$ being the chemical potential. The localized spin and orbital moment configurations follow the distribution

$$P(S_i, \Gamma_i) \propto \text{Tr} e^{-\beta H_{\text{eff}}}.$$
single-particle eigenvalues and eigenstates of $H$ calculated by using the Kubo formula [36] as follows: 

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\text{grounds. The optical conductivity for the molybdates is}
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\text{rare-earth molybdates.}
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gap, \Delta(\mathbf{q}) \text{, the solution of the model above — generating the equilibrium configurations of } \{\mathbf{S}_i, \mathbf{\Gamma}_i\} \text{ through iterative diagonalisation of } H_{\text{eff}}(\mathbf{S}_i, \mathbf{\Gamma}_i). \text{ To access large sizes within reasonable time we use a cluster algorithm for estimating the update cost [34,35]. Results in this paper are for a } 6 \times 6 \times 6 \text{ pyrochlore lattice (384 sites), using a cluster of } 3 \times 3 \times 3 \text{ pyrochlore unit cells (108 sites).}
\]

From the equilibrium configurations we calculate the thermally averaged magnetic structure factor $S_{\text{mag}}(\mathbf{q}) = \frac{1}{N} \sum_i \mathbf{S}_i \cdot \mathbf{S}_j e^{i \mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}$ and orbital structure factor $S_{\text{orb}}(\mathbf{q}) = \frac{1}{N} \sum_i \mathbf{\Gamma}_i \cdot \mathbf{\Gamma}_j e^{i \mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}$ at each temperature.

Electronic properties are calculated by diagonalising $H_{\text{eff}}(\mathbf{S}_i, \mathbf{\Gamma}_i)$ on the $6 \times 6 \times 6$ lattice on equilibrium backgrounds. The optical conductivity for the molybdates is calculated by using the Kubo formula [36] as follows:

\[
\sigma^{xx}(\omega) = \frac{\sigma_0}{N} \left( \sum_{n,m} \frac{f(\epsilon_n) - f(\epsilon_m)}{\epsilon_m - \epsilon_n} J_{nm}^m \right) \delta(\omega - (\epsilon_m - \epsilon_n)),
\]

where $J_{nm}^m = \langle n|J_x|m\rangle$ and the current operator $J_x$ is given by

\[
J_x = -i \sum_{\mathbf{r},\mathbf{r}'} \left[ (\epsilon_{\mathbf{r}'} - \epsilon_{\mathbf{r}}) \tilde{c}_{\mathbf{r}'} \tilde{c}_{\mathbf{r}} - \text{h.c.} \right].
\]

$f(\epsilon)$ is the Fermi function, $\epsilon_n$ and $|n\rangle$ are the single-particle eigenvalues and eigenstates of $H_{\text{el}}(\mathbf{S}_i, \mathbf{\Gamma}_i)$, respectively. The conductivity is in units of $\sigma_0 = e^2/(\hbar a_0)$, where $a_0$ is the lattice constant. $N$ is the total number of lattice sites. The dc conductivity is obtained as a low-frequency average of the optical conductivity over a window $\sim 0.05t$.

We study the Mott transition in the molybdate family with changing ionic radius of the rare-earth cation ($r_R$) by tuning the $U/t$ in our model.

**Results.** We will discuss the physics of the model for a wide range of $J_{AF}-U$ in a separate paper and focus here on parameters appropriate to the molybdates. Following *ab initio* estimates [22,23] we use $t = 0.1\text{ eV}$ and $J_{AF} = 0.02\text{ eV}$. The calibration of $U/t$ in terms of $r_R$ is based on the optical gap. We discuss this here briefly.

**Parameter calibration.** In our calculation, the pyrochlore lattice constant $a_0 = a/4$, where $a$ is the FCC lattice constant. For molybdates we have $a \sim 1\text{.0 nm}$ [22] and thus $\sigma_0 \sim 10^3 (\Omega\text{cm})^{-1}$. For $T \sim 0$, the optical gap $\Delta$ is determined by linearly extrapolating the decreasing edge of the optical conductivity spectra in the low-energy regime. Figure 1(a) shows the comparison of experimental gap [12] $\Delta_{\text{expt}} = \Delta(r_R)$ and theoretical gap $\Delta_{\text{th}} = \Delta(U/t)$ (see fig. 1(b)) at low temperature. We “calibrate” the $U/t$ of our model in terms of $r_R$ using $\Delta(r_R) \sim \Delta(U/t)$ in the insulating regime (finite optical gap). We try a linear fitting of this data and extrapolate it to lower $U/t$ values, to have an estimate of the $U/t$ in terms of $r_R$ in the metallic regime (zero optical gap). Our calibration (see fig. 1(c)) suggests that for the rare-earth molybdates $U/t$ seems to vary from $\sim 5$–9 as $R$ varies from Nd to Ho.

We now discuss results for the chosen $t$, $J_{AF}$, and $U/t$, using absolute scales, and compare with available experimental data [10,37].

**Phase diagram.** Figure 2(a) shows the experimental phase diagram. At large $r_R$, where the $U/t$ ratio is relatively small, the ground state is a ferromagnetic metal with a moment $\sim 1.4\mu_B$ per Mo [17,37]. The magnetisation seems to diminish slowly as $r_R$ reduces (fig. 2(c)), and then rapidly around the metal-insulator transition, $r_R^c$ $\sim 1.06\text{ A}$, but a small value survives into the weak insulating regime [37]. The FM $T_c$ is $\sim 80\text{ K}$ for large $r_R$ and drops sharply near $r_R^c$. The state for $r_R \lesssim r_R^c$ is a spin glass, with $T_{SG} \sim 20\text{ K}$.

Figure 2(b) shows our result over the window $U/t \sim 5$–$10$. For our parameter calibration the metal-insulator transition (MIT) at $T = 0$ occurs at $U_c \sim 7.6t$, and we present our results in terms of $U_c/U$. At the right end, where $U \sim 0.7U_c$, the ground state is metallic, double exchange (DE) dominated, and an almost saturated ferromagnet. This is also a weakly “ferro-orbital” state. With increasing $U$ the orbital moment grows and leads to a splitting of the $e_g$ band.

What drives the metal-insulator transition? i) Increasing $U$ increases the splitting $\Delta \sim |J(U)|$ between the electronic levels on Mo. This becoming comparable to the bandwidth would lead to a Mott transition (the correlation...
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Fig. 2: (Colour online) (a), (b): phase diagram of the molybdates — experiment [11,16] and theory. The experimental ground state changes from ferromagnetic metal (FM) to spin glass metal (SGM) and then spin glass insulator (SGI) with reducing $R$. Within theory the FM transforms to a “spin liquid” rather than to a spin glass. Panels (c), (d) show the ferromagnetic moment at low $T$ as the system is taken through the MIT. Within both experiment [37], (c), and theory, (d), a small moment survives in the insulator. In (b) we have cut off $T_c$ at the point where the $T_c=0$ magnetisation drops below 10%.

aspect). ii) With increasing $U$, the growing orbital moment suppresses the electron kinetic energy. This weakens DE. The competing AF superexchange reduces the magnetisation and increases the extent of spin disorder in the ground state. We call this the “Anderson” (disorder) aspect of the problem. It depends crucially on the presence of $J_{AF}$. $U_c$ is determined by a combination of the Mott and Anderson effects opening a gap in the electronic spectrum. Since magnetic disorder plays a role in the MIT, one can affect the transition by applying a magnetic field [21]. Note that interaction effects are crucial in our problem in driving a metal-insulator transition. So we cannot have a purely noninteracting “Anderson transition” without extrinsic disorder.

Figures 2(c) and (d) show the low-$T$ magnetisation in the molybdates and in our scheme. The dependence is very similar and a small magnetization survives beyond the MIT.

Resistivity. We demarcate the finite $T$ metal-insulator boundary based on the temperature derivative of resistivity $d\rho/dT$: “metal” if $d\rho/dT > 0$, “insulator” if $d\rho/dT < 0$. We compute $\rho(T)$ via the Kubo formula for changing $U/t$ [36].

Figure 3(a) shows experimental resistivity [10] while fig. 3(b) shows the theory result for parameter values set by the calibration. Even the limited $R$ variation in the experiments can be thought to represent three “regimes”. i) For $R = \text{Nd}$, the “high $T_c$” FM, $\rho(T)$ has traditional metallic behaviour, $\rho(0) < 1 \text{m}\Omega\text{cm}$ and $d\rho/dT > 0$ all the way to 400 K. ii) For $R = \text{Gd, Dy, Ho}$, the system is insulating at all $T$, with $\rho(0) \to \infty$. The behaviour is clearly activated for Dy and Ho while Gd seems to be weakly insulating. iii) $R = \text{Sm (and Eu, not shown)}$ represents the most interesting case, with $\rho(0) \sim 3 \text{ m}\Omega\text{cm}$ and a nonmonotonic $T$-dependence [10,12]. Any theory would have to capture the obvious regimes i) and ii) and also the peculiar large $\rho(0)$ and nonmonotonicity in iii).

Our results, fig. 3(b), show the following: i) For $U \ll U_c$, the itinerant $e_g$ electrons see a DE dominated ferromagnetic background, as well as an orbital-ferro state. The $T = 0$ state is ideally clean, and finite $T$ resistivity from spin and orbital fluctuations generate an approximate linear $T$ behaviour (see fig. 3(c)). ii) For $U \gg U_c$, there is a distinct gap $\Delta$ with $\rho(T) \sim \rho(0)e^{\Delta/T}$ as $T \to 0$ and $d\rho/dT < 0$ over the entire temperature range. iii) For $U \lesssim U_c$, the residual resistivity $\rho(0)$ is finite. This arises from a combination of depleting DOS at the Fermi level (due to the increasing orbital moment), and the magnetic disorder due to weakening DE. The behaviour of $\rho(0)$ is shown in the inset to fig. 3(b). Increasing $T$ does lead to a linear
behaviour, with a large slope, but the resistivity peaks at a scale $T_{\text{peak}}(U)$ and falls thereafter. Figures 3(c), (d) highlight this trend. As $U \rightarrow U_c$, $T_{\text{peak}} \rightarrow 0$, finally merging with the insulating behaviour in ii).

**Optical spectral weight.** Figure 4(a) shows the experimentally estimated optical spectral weight $n_{\text{eff}}(\Omega) = (2m_0/\pi e^2) \int_0^\Omega \sigma(\omega)d\omega$ for different $R$ and varying temperature at $\Omega = 0.5\text{ eV}$ [10]. It shows the expected trend of $n_{\text{eff}}$ growing with $T$ in the insulating, low $\tau_R$ side as the Mott gap is slowly filled, and reducing on the metallic side as weight gets transferred to high energy as coherence is lost.

We calculated the same quantity for different cutoff frequencies, $\Omega_c$, as $n_{\text{eff}}(\Omega) = \int_0^\Omega \sigma(\omega)d\omega$. Figures 4(b), (c) show our result for $\Omega = 0.3\text{ eV}$ and 0.5 eV, respectively. Figure 4(d) shows just $\sigma_{dc}$ to contrast the features in optical weight to the nonmonotonicity of the dc conductivity itself.

Our result at $\Omega = 0.3\text{ eV}$, roughly 2/3 of the experimental cutoff, has the same features as the experimental data. At $\Omega = 0.5\text{ eV}$, however, our data reveal a weak nonmonotonicity in the $T$-dependence when $U > U_c$. This arises because $\sigma(\omega)$ gains weight at low frequency, as in fig. 4(b), but loses more around $\omega \sim 0.5\text{ eV}$. The success in capturing the dc resistivity, fig. 3, does not translate to a similar success in capturing the high-energy optical conductivity. It is possible that some of the simplifying assumptions regarding band structure and coupling constants, i.e., $J_H$, affect this result.

The nonmonotonicity in our $\Omega = 0.5\text{ eV}$ spectral weight (fig. 4(c)) is distinct from the dc conductivity behaviour shown in fig. 4(d). Figure 3(b) shows that bad $T = 0$ metals, for $U \lesssim U_c$, become more resistive with increasing $T$ and beyond a $T_{\text{peak}}$ become less resistive again. We suggest that a detailed conductivity map, on materials like Gd$_2$–Sm$_3$Mo$_2$O$_7$ or Gd$_2$–Eu$_3$Mo$_2$O$_7$ could reveal this nonmonotonicity.

**Density of states.** We computed the single-particle density of states (DOS), $D(\omega) = \frac{1}{V} \sum_n \delta(\omega - \epsilon_n)$, for the studied interaction and temperature window. Figure 5(a) shows the dependence of $D(\omega)$ on $U/U_c$ as the system is driven across the Mott transition at $T = 0$. The DOS has its tight-binding form up to $U \sim 0.7U_c$ beyond which the presence of the orbital moment shows a visible depletion in the DOS around $\omega = 0$. This dip becomes a gap for $U > U_c$ which grows in the insulating phase. At $T = 300\text{ K}$, fig. 5(b), the systems with $U < U_c$ lose weight near $\omega = 0$, while those with $U > U_c$ gain weight. Figure 5(c) quantifies these trends by calculating $\int_{-\Omega'}^{\Omega'} D(\omega)d\omega$, where $\Omega' = 0.15\text{ eV}$ (to make a comparison with fig. 4(b)). We suggest that the optical behaviour observed experimentally has an analog in the single-particle spectral weight transfer as well.

Figures 5(d)–(f) show the thermal evolution of the DOS at three representative $U/U_c$. i) In fig. 5(d), for $U \sim 0.6U_c$, the ground state is a nearly saturated ferromagnet with a small orbital moments $\Gamma_i$’s and has finite DOS at $\omega = 0$. Thermal growth and fluctuations of $\Gamma_i$’s decrease the DOS at $\omega = 0$ resulting in a small dip at high temperature. ii) For $U = 1.1U_c$, fig. 5(f), there is significant spin disorder in the ground state (the ferromagnetic moment is $\sim 0.1$) and $\Gamma_i$’s are large, $\sim 1$, at all sites. A remnant of the atomic gap $\sim U|\Gamma|$ survives despite the presence of small orbital moments $\Gamma_i$’s and has finite DOS at $\omega = 0$. Thermal growth and fluctuations of $\Gamma_i$’s decrease the DOS at $\omega = 0$ resulting in a small dip at high temperature. ii) For $U = 1.1U_c$, fig. 5(f), there is significant spin disorder in the ground state (the ferromagnetic moment is $\sim 0.1$) and $\Gamma_i$’s are large, $\sim 1$, at all sites. A remnant of the atomic gap $\sim U|\Gamma|$ survives despite the presence of
hopping. The DOS shows a Mott gap. With the increase in temperature, the angular fluctuations of $\Gamma_i$'s result in a slight smearing of the gap edge and an increase in “low-energy” weight. iii) For $U = 0.9U_c$, fig. 5(e), the magnetic state has magnetisation, $M \sim 0.5$ and $\Gamma_i$'s are moderately large. As a result there is only a loss in weight around $\omega = 0$ but no hard gap. This is a pseudogap state.

**Discussion.** – There are some issues where our modeling differs from the experimental results. The origin of these differences is obvious, and we briefly touch upon them: i) *Temperature scales:* We have used a model with Hund’s coupling $J_H \gg t$ for convenience, and obtain $T_c \sim 160$ K for Nd. In reality $J_H \sim 5t$ [22], and as earlier results show [38] this would reduce $T_c$ by $\sim 50\%$ to about 80 K, close to the actual value for Nd. ii) *Spin freezing:* Within our scheme we do not find any spin freezing, so no $T_{SG}$. Our magnetic state for $U > U_c$ is a spin liquid (SL), rather than a spin glass, with weak ferromagnetism. To obtain freezing, and the correct $T_{SG}$, it seems that one requires significant bond disorder [39] (or bond distortions [40–43]). iii) We have also not touched upon the experimental $P_{\Gamma R}$ phase diagram, the field-driven IMT in Gd$_2$Mo$_2$O$_7$ [21], and the anomalous Hall effect (AHE) observed in Nd$_2$Mo$_2$O$_7$ [16–20].

**Conclusion.** – We provide the first study of the Mott transition in the pyrochlore molybdates, in a real space framework, retaining the double exchange, superexchange and correlation effects. Our phase diagram captures the transition from a ferromagnetic metal to a spin disordered insulator, as in experiments, with reasonable thermal scales. Our transport and optical results capture major features of the molybdate data, reveal unexplored non-monotonicities, and, we predict, correlate with the single-particle spectral weight. Ongoing work will address the field-driven Anderson-Mott transition.

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We acknowledge the use of the HPC clusters at HRI.

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