Dielectric Response Near the Density-Driven Mott Transition in infinite dimensions

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We study the dielectric response of correlated systems which undergo a Mott transition as a function of band filling within the dynamical mean field framework. We compute the dielectric figure of merit (DFOM), which is a measure of dielectric efficiency and an important number for potential device applications. It is suggested how the DFOM can be optimized in real transition metal oxides. The structures seen in the computed Faraday rotation are explained on the basis of the underlying local spectral density of the $d = \infty$ Hubbard model.

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The ac conductivity and dielectric tensor provides valuable information concerning the finite frequency, finite temperature charge dynamics of an electronic fluid in a metal. The potential for microwave devices, for e.g, is enormous, given the progress in synthesis of strongly correlated materials with electronic properties that are sensitive to external perturbations like applied electromagnetic fields, pressure(doping), etc \[2\]. Recent improvements in theoretical tools at our disposal allow controlled, physically meaningful calculations to be undertaken \[2\]. These advancements have, and will continue to, spur increased investigations to tap their full technological potential.

The dielectric figure of merit (DFOM) reflects the quality of the dielectric response of a material, and is a quantity of interest for potential microwave device applications. It is formally defined as

$$DFOM = \frac{|\varepsilon_{xy}(\omega)|}{2|Im \varepsilon_{xx}(\omega)|} (1)$$

and so one requires the full dielectric (ac conductivity) tensor to access this quantity. It also follows that it is linked to the magneto-optical response of the material under study. In weakly correlated metals, the details of the ac conductivity tensor, and hence of the dielectric and Hall response, are determined by the vagaries (shape and size) of the Fermi surface \[3\]. That such a connection is untenable for strongly correlated metals has been pointed out by Shastry et al \[4\], who show that the Hall constant, for e.g, is affected by contributions coming from the whole Brillouin zone, and may have nothing to do with Fermi surface effects. On the other hand, it has been observed that the physics of strongly correlated metals undergoing metal-insulator transitions is understandable in terms of spectral weight transfer over large energy scales \[3\]. The consequences for dielectric response and the DFOM have, however, not been studied at all.

In this letter, we address this issue. We are primarily interested in materials like $La_{1-x}Y_xTiO_3$ \[4\] which undergo Mott transitions with doping. We stress that 3D transition metal oxides are the most interesting candidates for the kind of effects we want to study, as filling driven insulator-metal transitions are realized in a variety of them in a wide range of parameters.

We consider the one-band Hubbard model \[3\],

$$H = -\sum_{ij\sigma} \tilde{t}_{ij} \tilde{c}_i^{\dagger} \tilde{c}_j + U \sum_i n_{i\uparrow} n_{i\downarrow} - \mu \sum_i n_{i\sigma} (2)$$

as a prototype model describing the electronic degrees of freedom in TM oxides. To study the 3D case, we employ the $d = \infty$ approximation, which is the best approximation possible at the present time \[3\]. Since this method has been extensively reviewed, we only summarize the relevant aspects. All transport properties, which follow from the conductivity tensor, are obtained from a $k$-independent self-energy in $d = \infty$; the only information about the lattice structure comes from the free band dispersion in the full Green fn:

$$G(k, \omega) = G(\epsilon_k, \omega) = \frac{1}{\omega + \mu - \epsilon_k - \Sigma(\omega)} (3)$$

To solve the model in $d = \infty$ requires a reliable way to solve the single impurity Anderson model(SIAM) embedded in a dynamical bath described by the hybridization fn, $\Delta(\omega)$. There is an additional condition that completes the selfconsistency:

$$\int d\epsilon \rho_0(\epsilon) G(\epsilon, i\omega) \rho_0(\epsilon) = \frac{1}{\omega + \mu - \Delta(i\omega) - \Sigma(i\omega)} (4)$$

where $\rho_0(\epsilon)$ is the free DOS ($U = 0$). In $d = \infty$, this is sufficient to compute the transport, because the vertex corrections in the Bethe Salpeter eqn. for the conductivity vanish identically in this limit \[3\]. Thus, the conductivity is fully determined by the basic bubble diagram made up of fully interating local GFs of the lattice model.

The optical conductivity and the Hall conductivity are computable in terms of the full $d = \infty$ GFs as follows \[3\]:

$$\sigma_{xx}(i\omega) = \frac{1}{i\omega} \int d\epsilon \rho_0(\epsilon) \sum_{iv} G(\epsilon, iv) G(\epsilon, iv + i\omega) (5)$$
and the Hall conductivity has been worked out by Lange \[7\], so we use the approach developed there. Explicitly, after a somewhat tedious calculation, the imaginary part of $\sigma_{xy}(\omega)$ is given by

$$
\sigma''_{xy}(\omega) = c_{xy} \int_{-\infty}^{+\infty} d\epsilon p_0(\epsilon) e \int_{-\infty}^{+\infty} d\omega_1 d\omega_2 A(\epsilon, \omega_1) A(\epsilon, \omega_2) \frac{1}{\omega} \left[ F(\epsilon, \omega; \omega) - F(\epsilon, \omega_2; \omega) \right] - (\omega \to -\omega) \quad (6)
$$

where

$$
F(\epsilon, \omega; \omega_1) = A(\epsilon, \omega_1 - \omega)[f(\omega_1) - f(\omega_1 - \omega)] \quad (7)
$$

and $A(\epsilon, \omega) = -\text{Im}[\omega - \epsilon - \Sigma(\omega)]^{-1}/\pi$ is the s.p spectral function in $\hbar \omega = \infty$.

Knowledge of $\sigma''_{xy}(\omega)$ permits us to use the analyticity properties to compute its real part via a Kramers-Krönig transform. The dielectric tensor is determined from,

$$
\epsilon_{xx}(\omega) = 1 + \frac{4\pi}{\omega} i \sigma_{xx}(\omega) \quad (8)
$$

and

$$
\epsilon_{xy}(\omega) = \frac{4\pi}{\omega} i \sigma_{xy}(\omega) \quad (9)
$$

The paramagnetic Faraday rotation is directly related to $\sigma_{xy}(\omega)$ via

$$
\theta_F(\omega) = C(n) i \frac{\sigma''_{xy}(\omega)}{\omega} \quad (10)
$$

The DFOM can readily be computed from the dielectric tensor, as determined above. So can the ac Hall constant and angle, as well as the Raman intensity, which has been discussed in detail elsewhere \[8,9\].

Before embarking on our results and their analysis, it is instructive to summarize what is known about the $d = \infty$ Hubbard model. At large $U/t$, and away from half-filling ($n = 1$), the ground state is a paramagnetic FL if one ignores the possibility of symmetry breaking towards antiferromagnetism, as well as disorder effects, which are especially important near $n = 1$. This can be achieved formally by introducing a nn hopping, which in $d = \infty$ leaves the free DOS essentially unchanged \[10\].

This metallic state is characterized by two energy scales: a low energy coherence scale $T_{coh}$, below which local FL behavior sets in \[11\], and a scale of $O(D)$, ($D$ is the free bandwidth) characterizing high energy, incoherent processes across the remnant of the Mott-Hubbard insulator at $n = 1$. At $T < T_{coh}$, the quenching of the local moments leads to a response characteristic of a FL at small $\omega < 2D$ (but with the dynamical spectral weight transfer with doping, a feature of correlations) \[12\], but at higher $T > T_{coh}$, the picture is that of carriers scattered off by effectively local moments, which makes the system essentially like a non-FL (note that the metal with disordered local moments is not a FL).

Armed with this information, we are ready to discuss our results. We choose a gaussian unperturbed DOS, and $U/D = 3.0$ to access the strongly correlated FL metallic state off $n = 1$, ignoring the possible instability to an AF-ordered phase. All calculations are performed at a low temperature, $T = 0.01D$. Fig. 1 shows the optical conductivity and the longitudinal dielectric constant. $\sigma_{xx}(\omega)$ agrees with calculations performed earlier in all the main respects; in particular, it clearly exhibits the low-energy quasicohherent Drude form, the transfer of optical spectral weight from the high-energy, upper-Hubbard band states to the low energy, band-like states with increasing hole doping, and the isosbectic point at which all the $\sigma_{xx}(\omega)$ curves as a fn. of filling cross at one point, to within numerical accuracy. It is interesting to point out that such features have also been observed in experimental studies \[11,12\]. Correspondingly, $\text{Im}\sigma_{xx}(\omega)$ also shows the isosbestic point, the explanation for which is identical to that provided recently by us for the case of $\sigma_{xx}(\omega)$ \[12\].

In Fig. 2, we show the DFOM obtained using eqn. (2) and the dielectric tensor calculated as above. We are mainly interested in the variation of the DFOM with hole doping, given here by $\delta = (1 - n)$. This fixes the chemical potential, and the FL resonance position, and the IPT describes the evolution of spectral features in good agreement with exact diagonalization studies \[12\]. In view of the ability of the IPT to reproduce all the qualitative aspects observed in $\sigma_{xx}(\omega)$, we believe that is a good tool in the present case.
The DFOM shows a sharp peak at low energy (around 0.05 ev) in the metallic state off half-filling with a maximum value of order 3 for $\delta = 0.3$. It also reaches a minimum value at around 1.8 ev for $\delta = 0.1, 0.2$; this is related to the frequency dependence of the optical conductivity tensor with filling. These features are understandable in terms of the dynamical transfer of optical spectral weight from the high-energy (upper-Hubbard band) incoherent states to low energy quasicoherent states upon hole doping the Mott insulator (at $n = 1$). The sharp peak is related to the fact that the action of the current operator on the lower-Hubbard band states creates well defined elementary excitations in a strongly correlated Fermi liquid, while the increase of the low energy weight is understood in terms of the increasing weight of the quasicoherent, itinerant part of the spectrum relative to that of the atomic like, incoherent part with increasing $\delta = (1 - n)$. Thus, the absolute DFOM is determined by the outcome of the competition between the itinerant and atomic parts of the spectrum. The above suggests that the DFOM in the IR and the mid-IR can be increased even further by enhancing the weight of the transitions within the lower-Hubbard band and those in the (lower-Hubbard band + central FL peak) manifold, by suitable materials engineering. In reality, a multi-orbital situation would be more favorable, since one might expect increased contributions to the mid-IR part coming from interorbital hopping, as well as from spin-orbit coupling terms present in multi-orbital systems. However, static disorder, for e.g., in $d = \infty$, would shift coherent spectral weight to higher energy and destroy low energy coherence, limiting the DFOM to modest values, suggesting that good sample quality is one of the prerequisites to increase the DFOM (something that maybe hard to limit in the 3d transition metal compounds undergoing doping/filling driven Mott transitions).

Next, we describe our results for the paramagnetic Faraday rotation, computed from eqn. \( \theta_F(\omega) \). $\theta_F(\omega)$ shows a monotonic fall-off with $\omega$ in the IR and the mid-IR region, before increasing around $\omega/D \approx 1$, whereafter, it passes through a broad maximum at $\omega/D \approx 1.4$. This is followed by another broad peak around $\omega/D = 3.0$. The origin of these features are linked to the nature of the transitions reflected in the interacting LDOS of the $d = \infty$ Hubbard model. We assign the central feature to the “quasiparticle peak” near $\omega = 0$, the maximum feature around $\omega/D \approx 1.4$ to the “$U/2$ peak”, corresponding to transitions between the QP peak and the upper- (or lower) Hubbard bands, and the broad feature at $\omega/D = 3.0$ to transitions between the lower- and the upper Hubbard bands.

The above suggests that strongly correlated metals near the borderline of the filling-driven Mott transition might be good candidates for dielectric device applications. A multi-orbital situation (plus spin-orbit couplings) would be more effective in enhancing the DFOM, while static disorder, which inevitably accompanies doping, would act to limit it. Consideration of real bandstructure would be desirable; in our approach, this simply entails replacing the free (gaussian) DOS used here by the actual bandstructure DOS as an input into the DMFT calculation. Effects of static disorder will be especially important near the doping induced Mott transition; these require the consideration of correlations and disorder on an equal footing, and is left for a future work. However, one expects that the destruction of low energy coherent spectral weight which accompanies static disor-
der will limit the DFOM to more modest values.

In conclusion, we have investigated the dielectric figure of merit (DFOM) and the paramagnetic Faraday rotation near the density driven Mott transition. We have shown how a consistent treatment of the interplay between the atomic and itinerant aspects inherent in strongly correlated systems leads to encouraging values for the DFOM. In practice, however, the estimate provided here would be an overestimate, since real bandstructure and disorder effects, as well as multi-orbital character of real $d$-band systems can well change our conclusions quantitatively. We have also provided a simple explanation for the origin of structure in the Faraday rotation; in real materials, the above effects will also affect $\theta_F(\omega)$. Nevertheless, we have provided a theoretical framework within which such calculations can be undertaken, and all of the above effects can be included in a suitable extension within the $d = \infty$ framework.

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