Optical conductivity in doped manganites with planar $x^2$-$y^2$ orbital order

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We investigate a planar model for the ferromagnetic (FM) phase of manganites, which develops orbital order of $e_g$ electrons with $x^2$-$y^2$-symmetry at low temperature. The dynamic structure factor of orbital excitations and the optical conductivity $\sigma(\omega)$ are studied with help of a finite-temperature diagonalization method. Our calculations provide a theoretical prediction for $\sigma(\omega)$ for the 2D FM state and are of possible relevance for the recently found A-type phase of manganites at high doping which consists of FM layers coupled antiferromagnetically. In the $x^2$-$y^2$ ordered regime $\sigma(\omega)$ shows both a Drude peak and a gapped incoherent absorption due to a gap in the orbital excitations.

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Recently Kawano et al. [1] and Akimoto et al. [2] observed a new ground state in heavily doped 3D manganites $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ and $(\text{La}_{1-x}\text{Nd}_x)_{1-x}\text{Sr}_2\text{MnO}_3$ (e.g. $x = 0.54$ and $z = 0.0 - 1.0$), which surprisingly show the same antiferromagnetic A-structure known from the undoped LaMnO$_3$ where ferromagnetic (FM) planes are coupled antiferromagnetically. It was proposed that the occupied orbitals in this phase have $x^2$-$y^2$ symmetry [2] in contrast to the alternating orbital order in LaMnO$_3$ [3], and as a consequence the conductivity should be quasi-2D and the magnetic coupling between the layers small. This broken symmetry state has to be distinguished from the orbital structure of the 3D FM state realized at lower doping, where orbital correlations are expected to be liquid like [4]. The latter FM state shows a highly anomalous form of the orbital conductivity [4] with a large incoherent absorption extending up to $\omega \sim 1$eV. It has been argued that the anomalous incoherent absorption is related to the orbital degree of freedom [4] and characteristic for an orbital liquid [2]. However there are also alternative scenarios which attribute the incoherent feature to Jahn-Teller polarons [2].

Comparative optical studies in the high doping regime where the orbital ordered A-phase is the stable ground state have not yet been reported. The purpose of this work is to present a theoretical analysis of the possible outcome of such experiments. The frequency dependence of $\sigma(\omega)$ is interesting since it is expected to reveal the orbital excitations in the doped $x^2$-$y^2$ ordered phase, and may allow to distinguish between the orbital excitation and the Jahn-Teller polaron scenario.

The evolution of $x^2$-$y^2$ orbital order upon doping was also found in the study of planar models derived from the degenerate Kondo lattice model in the fully polarized FM phase [10]. Therefore we believe that this model is an appropriate starting point to investigate these questions. The $t_{2g}$-spins of the Mn$^{3+}$ and Mn$^{4+}$ ions as well as the $e_g$-electron spins of Mn$^{3+}$ are aligned globally in the FM-phase and can be integrated out. What remains is the orbital degree of freedom since there are two nearly degenerate $e_g$ orbitals per site. The large intra-atomic repulsive interactions prevent double occupancy of $e_g$ orbitals at the same site, which leads to a strong correlation problem and to nontrivial charge and orbital dynamics.

For the saturated FM state the restriction to configurations without double occupancy leads to the orbital $t$-$J$ model [11,10]

$$H_{\text{arb}} = - \sum_{\langle ij \rangle, ab} (t_{ij}^{ab} \tilde{d}_{ia}^{\dagger} \tilde{d}_{jb} + H.c.) + H_{\text{int}}. \quad (1)$$

with $\tilde{d}_{ia}^{\dagger} = d_{ia}^{\dagger} (1 - n_{ia})$. Here we use $a$ and $b$ ($\alpha$ and $\beta$) as orbital-pseudospin indices; while $\bar{a}$ denotes the orthogonal $e_g$ orbital with respect to orbital $a$. A convenient basis is $| \uparrow \rangle = d_{x^2-y^2}$ and $| \downarrow \rangle = d_{z^2-r^2}$. The transfer matrix elements are then given by

$$t_{ij}^{ab} = t \left( \begin{array}{cc} 3 & \mp \sqrt{3} \\ \mp \sqrt{3} & 1 \end{array} \right) \quad (2)$$

which allows for inter-orbital hopping in the $xy$-plane. The $\mp$-sign distinguishes hopping along $x$ and $y$ direction. Here $t/4$ is the matrix element between $d_{z^2-r^2}$ orbitals in the $xy$-plane. For one electron per site this model describes a Mott insulator. The orbital interaction $H_{\text{int}}$ follows as a consequence of the elimination of doubly occupied sites with energy of an effective $U$ [2], where we assume that because of the large Hund-splitting the high-spin intermediate states are most important.

$$H_{\text{int}} = - \sum_{\langle j u \rangle} \sum_{\alpha \beta} \sum_{a \beta} \frac{t_{\alpha \beta j}^{ab} \tilde{d}_{j a}^{\dagger} \tilde{d}_{j u}^{\dagger} \tilde{d}_{j u}^{\dagger} \tilde{d}_{j a}^{\dagger}}{U} \left[ \delta_{\beta, \bar{\beta}} \delta_{\bar{\beta}, \bar{\beta}} d_{j a}^{\dagger} d_{j a}^{\dagger} d_{j a}^{\dagger} d_{j a}^{\dagger} + u.a. 

- \delta_{\beta, \bar{\beta}} d_{j a}^{\dagger} d_{j a}^{\dagger} d_{j a}^{\dagger} d_{j a}^{\dagger} + u.a. \right]. \quad (3)$$

Here $u, u' = (\pm a, 0)$ or $(0, \pm a)$ are lattice unit vectors. The orbital interaction $H_{\text{int}} = H_{\text{int}}^{(2)} + H_{\text{int}}^{(3)}$ consists of two types of contributions: (i) 2-site terms ($u = u'$), i.e. similar to the Heisenberg interaction in the standard $t$-$J$ model, yet more complex because of the nonvanishing off-diagonal $t_{ij}^{ab}$, and (ii) 3-site hopping terms ($u \neq u'$) between second nearest neighbors. In the half-filled case
only the 2-site interaction is operative and may induce some orbital order. In the presence of hole doping, i.e., for less than one $e_g$-electron per site, both the kinetic energy and the 3-site contributions in $H_{int}$ lead to propagation of the holes and to a frustration of the orbital order preferred by the orbital interaction $H_{int}^{(2)}$. When neglecting the 3-site terms our Hamiltonian is equivalent to the orbital model studied by Ishihara et al. [11]. Since the spectral shape of $\sigma(\omega)$ is only weakly affected by these terms [10], we do not consider them in this study. For the calculation of correlation functions (CF) we use a generalization of the exact diagonalization technique for finite temperature developed by Jaklič and Prelovšek [13].

There is a clear manifestation of long-range orbital correlations below $N_{h}/N=10/16$ and $U = 16t$. The inset gives a schematic view of the orbital correlations for $T = 0.05t$ (strength proportional to radius).

Figure 1 monitors the evolution of $x^2-y^2$ orbital correlations as function of distance and temperature for a 16-site cluster [11]. Here we use the pseudospin operator notation $T_i^z = \frac{1}{2}(n_{i\uparrow} - n_{i\downarrow})$, $T_i^+ = d_{i\uparrow}^d d_{i\downarrow}^\dagger$ and $T_i^- = d_{i\downarrow}^d d_{i\uparrow}^\dagger$. There is a clear manifestation of long-range orbital correlations below $T = 0.5t$ which saturates below $T = 0.2t$. Although $\langle T_i^z T_{i+R}^z \rangle \sim 0.02$ appears small, it is close to the maximal value $\sim 0.03$ for this CF given the electron concentration $6/16$. This CF has a minimum for nearest-neighbors (see inset of Fig.1), which is partially due to the evolution of an exchange-correlation hole in the charge CF $\langle n_{i\uparrow} n_{i+1\uparrow} R \rangle$. In addition $\langle n_{i\uparrow} n_{i+1\uparrow} R \rangle$ shows further neighbor density modulations, which disappear with the orbital order. Usually such charge correlations near quarter-filling are attributed to strong nearest-neighbor (n.n.) repulsion. Since such interactions are not contained in (1), charge correlations appear here as pure correlation effect. Results are shown for $U/t = 16$, corresponding to $t \sim 0.3$ eV and $U \sim 4.5$ eV [12].

![Figure 1](image1.png)

**FIG. 1.** Evolution of orbital correlations $\langle T_i^z T_{i+R}^z \rangle$ as function of distance $R$ for various temperatures $T$. The ratio of hole number and system size is $N_h/N=10/16$ and $U = 16t$. The inset gives a schematic view of the orbital correlations for $T = 0.05t$ (strength proportional to radius).

![Figure 2](image2.png)

**FIG. 2.** Transverse orbital excitation spectra $\langle T_i^+ T_{-q}^- \rangle$ (a) in the orbital ordered low-temperature regime for various temperatures and (b) in the high-temperature regime with gapless orbital excitations for same parameters as in Fig. 1. Spectra are broadened using $\Gamma = 0.01t$. In (a) a comparison with results from the bandpicture is included (dash-dotted lines).

The orbital excitations are crucial for the understanding of the optical conductivity of manganites in the FM
phase. The form of the orbital excitation spectra changes significantly with doping. In the undoped case with alternating orbital order due to the interaction (3) the orbital excitation spectra ($T^+_q T^-_{-q}$) are essentially described by two collective modes \[ \omega \] , while at low doping with the emergence of $x^2$-$y^2$ orbital order this changes into a single collective orbital mode which is gapped. In the high doping case, $x \sim 0.6$, studied here (Fig. 2a) the single mode features have almost disappeared and the small cluster results show the features of a continuum with spectral weight distributed up to energies of about $\sim 4t$. We note, however, that the dominant peaks in these spectra correspond to the collective excitations observed for systems at low doping.

In the low and moderate doping regime a slave fermion approach provides a satisfactory description of the orbital excitations. In such a treatment holes are described by spinless fermions $h_i$ and the orbital pseudospins by Schwinger boson operators $b_{ia}$ leading to the kinetic energy Hamiltonian $H_t = -\sum_{\langle ij \rangle ab} t_{ij}^{ab} b_i^\dagger b_j + H.c.$. Subsequent mean-field treatment, i.e. assuming condensation of $b_{1\uparrow}$, plus Gaussian fluctuations leads to $H \sim \sum \omega_q b_i^\dagger b_j$ with $\omega_q = 3xt(1 - \gamma_q/3)$, where $\gamma_q = (\cos q_x + \cos q_y)/2$. This result tells us that the energy scale of orbital excitations is proportional to the doping concentration $x$, which is consistent with the diagonalization data. The scaling of $\omega_q$ with $x$ is due to the fact that $x^2$-$y^2$ orbital order is induced by the kinetic energy of the holes which overcomes the superexchange interaction (3). Further boson-fermion coupling terms not included in this consideration lead to deviations from single mode behavior and will be discussed elsewhere.

In this work we provide a comparison with an analytical approach based on the band picture which was proposed by Shiba and coworkers \[16\]. This Fermi-liquid approach assumes that $U/t$ is not too large in sytmes like La$_{1-x}$Sr$_x$MnO$_3$. Moreover the approach is expected to be useful in the dilute ($e_g$ electron density) limit. In this case the constraint in (1) may be neglected and correlation effects can be included in a perturbative way. Thus the kinetic energy can be diagonalized, leading to two tight-binding (TB) bands. For the planar model the energies are $E_{\pm}/t = 2\gamma_k \pm \sqrt{\gamma_k^2 + 3\eta_k^2}$, where $\mu$ is the chemical potential and $\eta_k = (\cos k_x - \cos k_y)/2$. Results for the transverse orbital excitation spectrum ($T^+_q T^-_{-q}$) are shown in Fig. 2a for $T = 0$ and compared with the diagonalization data. The TB-spectra for the planar model, i.e. neglecting correlation effects, show a broad continuum with several van Hove singularities. The pronounced structures in the diagonalization data around $\omega \sim 3xt$, reminescent of the collective orbital waves at lower doping, are not reproduced.

At temperatures $T > 0.2$ thermal fluctuations destroy the orbital order (Fig. 1). In this regime the orbital excitation gap changes into a pseudo gap with increasing low energy spectral weight (Fig. 2b). The appearance of low energy excitations in the orbital liquid phase has a dramatic effect on the optical conductivity as we shall see in the following.

The frequency dependent conductivity consists of two parts \[10\], $\sigma_0(\omega) = 2\pi e^2 D_c \delta(\omega) + \sigma(\omega)$. The $\delta$-function contribution is proportional to the charge stiffness $D_c$, which vanishes in insulators. The finite frequency absorption (or regular part) $\sigma(\omega)$ is determined by the current-current correlation function:

$$\sigma(\omega) = \frac{1 - e^{-\omega T}}{N\omega} \text{Re} \int_0^\infty dt e^{i\omega t} \langle j_x(t) j_x \rangle. \quad (4)$$

where the $x$-component of the current operator is given by $j_x^{(1)} = -ie \sum_{j+uab} t_{j+uab} u_x d_{j+uab}^\dagger d_{j+uab}$.

![FIG. 3. Optical conductivity (a) in the $x^2$-$y^2$ orbital ordered low-temperature phase for $T = 0.05$, 0.1 and 0.2t and (b) in the high-temperature regime with gapless orbital excitations. Parameters as in Fig. 1 and $\Gamma = 0.1t$. The inset in (a) shows the Drude weight $D_c$, the sum rule $S$ and the incoherent spectral weight $S_u$ versus temperature. Inset (b): Optical conductivity in the band model for the hole concentration $x = 0.6$ for various temperatures ($\Gamma = 0.02t$).](3)
Figure 3a shows the regular part $\sigma(\omega)$ at low temperature in the orbital ordered phase (the Drude peak $\sim D_{\sigma}(\omega)$ is not shown). The conductivity is given in dimensionless form $\rho_0\sigma(\omega)$, where $\rho_0 = \hbar^2/e^2$. A typical value is $\rho_0 \sim 0.2 \cdot 10^{-3}$ $\Omega$cm. The three data sets for $T = 0.05t, 0.1t$ and $0.2t$ almost coincide, consistent with the saturation of orbital order below $T = 0.2t$ (Fig. 1). The finite frequency absorption $\sigma(\omega)$ for $x = 0.6$ in Fig. 3a shows a well developed gap $\omega_{opt} \sim 2t$, which is due to the pronounced gap in the orbital excitation spectra. We note that in the $x^2-y^2$ ordered phase at low doping studied in Ref. [11] there is also a pseudogap in $\sigma(\omega)$, which is however smaller in size and only observable at low temperatures, because of the smaller orbital excitation gap. The inset in Fig. 3a shows the temperature dependence of the optical sum rule $S = -\frac{1}{\pi} \int_{-\infty}^{\infty} \sigma(\omega) d\omega$ and the Drude weight $D_{\sigma}$. The latter quantity is determined from the sum rule $S = D_{\sigma} + S_{\sigma} [10]$. The weight of the Drude peak strongly increases below $T = 0.5t$ and saturates below $T = 0.2t$, i.e. similar to the evolution of orbital order in Fig. 1. At low temperature about 60% of the spectral weight is in the Drude peak, which may however depend somewhat on the cluster size.

There is a dramatic shift of spectral weight in $\sigma(\omega)$ at higher temperatures when entering the orbital liquid phase (Fig. 3b). The conductivity is characterized by a broad peak centered at $\omega = 0$, reminiscent of a conventional Drude peak with a natural broadening $\Gamma \sim 0.5t$. We attribute this intrinsic broadening to the finite density of low-energy orbital excitations in the orbital liquid phase (Fig. 2b). There is still some structure in $\sigma(\omega)$ near $\omega \sim 2t$ close to the onset of absorption in the ordered low-$T$ phase.

The conductivity for the planar model calculated using the band picture is shown in Fig. 3b (inset). In this approach the finite frequency absorption is due to interband scattering, i.e. a consequence of the off-diagonal hopping $t'$. At low temperature the results for $\sigma(\omega)$ obtained by neglecting correlation effects are not very different from the exact diagonalization result. Both methods yield a clear gap and the main absorption is close to $\omega \sim 2t$. The onset of absorption in the band picture is near $\omega \sim 1t$, i.e. about $0.5t$ lower than in the cluster calculation. The temperature dependence in the band picture is, however, quite distinct. The filling in of spectral weight at low energy occurs only gradually, i.e. even at temperatures as high as $T = 1t$ there is only little low-energy spectral weight. The temperature scale of the transition from the $x^2-y^2$ ordered phase to the orbital liquid is a consequence of strong correlations, and not contained in the band picture, where the natural energy scale is much larger, i.e. given by the band width $W = 6t$.

Planar $x^2-y^2$ correlations are favored by the kinetic energy of holes, since the partially filled $x^2-y^2$ band has the largest overlap. The motion is quasi-2D since the hopping along the c-direction vanishes by symmetry. Moreover the antiferromagnetic coupling between layers in the A-phase in combination with double exchange would prevent coherent c-axis motion.

In a cubic system there are now two possibilities: (i) Since $x^2-y^2$ correlations are not preferred compared to $y^2-z^2$ and $z^2-x^2$ correlations in the corresponding perpendicular planes, the system remains in a disordered quantum liquid state. This is the orbital liquid state proposed by Nagaosa and coworkers [1]. (ii) Another possibility is spontaneous symmetry breaking selecting one particular set of planes, e.g. the formation of $x^2-y^2$ long-range order. We note that in the planar model the cubic symmetry is explicitly broken, therefore case (i) does not occur for this geometry. In the planar model the orbital liquid develops at higher temperature due to thermal fluctuations. The orbital ordering temperature $T^{(c)}_c$ increases with doping.

We believe that in cubic manganites both cases are realized in different doping regimes. At moderate doping concentration the orbital liquid state appears to be the proper ground state in combination with the isotropic ferromagnetic spin structure. This is consistent with the broad absorption in $\sigma(\omega)$ observed by Okimoto et al. and others. These spectra can be compared with our result for the orbital liquid in the planar model (Fig. 3b) which is stabilized by thermal fluctuations.

The observation of the A-phase at high doping concentration by Kawano et al. [1] and Akimoto et al. [2] seems to be a realization of case (ii) where symmetry is broken spontaneously. Our prediction for this phase are gapped orbital excitations with energy $\sim 3xt$. The optical conductivity of this quasi-2D metallic state is expected to consist of a large Drude peak, while the finite frequency absorption shows a pronounced gap due to the gapped orbital excitations.

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