Dynamical screening in strongly correlated metal SrVO$_3$

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Abstract – The consequences of dynamical screening of Coulomb interaction among correlated electrons in realistic materials have not been widely considered before. In this letter we try to incorporate a frequency-dependent Coulomb interaction into the state-of-the-art \textit{ab initio} electronic structure computing framework of local density approximation plus dynamical mean-field theory, and then choose SrVO$_3$ as a prototype material to demonstrate the importance of dynamical screening effect. It is shown to renormalise the spectral weight near the Fermi level, to increase the effective mass, and to suppress the $t_{2g}$ quasiparticle bandwidth apparently. The calculated results are in accordance with very recent angle-resolved photoemission spectroscopy experiments and Bose factor ansatz calculations.

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Introduction. – Over the last years, tremendous progresses have been made in the studies of strongly correlated materials [1–3], which typically contain partially filled and relatively localized 3$d$, 4$f$ or 5$f$ orbitals. The low-energy physics of these materials can be generally described by an effective Hamiltonian which models the electronic Coulomb interaction among the correlated orbitals. In general, the electronic Coulomb interaction can be parameterized by the Hubbard $U$. Usually $U$ is assumed to be static, but in principle $U$ is dynamical, \textit{i.e.}, frequency dependent. The charge density fluctuation in correlated orbitals will induce electric fields, which will be screened by the higher- or lower-lying states in the systems. Then the screening effect will result in a frequency-dependent renormalization for the Coulomb interaction \cite{4–6}. On the one hand, it is realized that at high frequency the screening effect becomes weaker and eventually the interaction approaches the large bare Coulomb value, which is an order of magnitude larger than the static screened value. On the other hand, it is generally believed that the dynamical screening effect may play a crucial role in understanding the subtle electronic structures of strongly correlated materials \cite{7–9}, such as the plasmon satellites and spectral weight transfer in the photoemission spectra of hole-doped BaFe$_2$As$_2$ \cite{10}, which are less emphasized before. Thus, how to take the dynamical screening effect into account is one of the major challenges in studying the intriguing properties of strongly correlated materials.

In a realistic approach to strongly correlated materials, the frequency-dependent interaction $U = U(\omega)$ can be evaluated directly at the random phase approximation (RPA) level, which allows for the accurate evaluations of the matrix elements of Hubbard $U$ and its frequency dependence \cite{4–6}. So far the static screened value $U_0 = \Re U(\omega = 0)$, has been widely used in the local density approximation plus static Coulomb interaction (LDA + $U$) method \cite{11,12} and the local density approximation combined with dynamical mean-field theory (LDA + DMFT) approach \cite{1–3,13–15}. Since the dynamical screening has been completely neglected or empirically taken into account by adjusting the static value $U_0$ or reducing the effective bandwidth of correlated orbitals \cite{7}, very little is known on the consequences of frequency-dependent Coulomb interaction in the electronic structures of strongly correlated materials before. In order to include the dynamical screening effect into the LDA + DMFT framework, the hardest obstacle has been the lack of a reliable and efficient quantum impurity solver for the general impurity model with a frequency-dependent Coulomb interaction $U(\omega)$ \cite{8}.

Fortunately, it seems that this obstacle has been overcome by the recent development of strong coupling
The computational scheme used in the present work can be viewed as a straightforward extension of the traditional LDA + DMFT method [1–3]. In a Hamiltonian formulation, the multi-orbital model with dynamically screened interactions can be written as

$$H = H_{LDA}^{TB} - H_{DC} + H_I + H_S. \tag{1}$$

Here $H_{LDA}^{TB}$ is the effective single-particle tight-binding Hamiltonian. To generate $H_{LDA}^{TB}$, the ground-state calculations by using the pseudopotential plane-wave method as implemented in the ABINIT package [21] have been conducted at first. For the ground-state calculations, the projector augmented wave type pseudopotentials [22] for the Sr, V, and O species are constructed by ourselves, the cutoff energy for the plane-wave expansion is 20 Ha, and the $k$-mesh for Brillouin zone integration is $12 \times 12 \times 12$. These pseudopotentials and computational parameters are carefully checked and tuned to ensure the numerical convergences. Then $H_{LDA}^{TB}$ is obtained by applying a projection onto a set of maximally localized Wannier functions [23] including all the V-$t_{2g}$ and O-$2p$ orbitals, which results in a $12 \times 12$ tight-binding Hamiltonian. As for the double-counting term $H_{DC}$, the around mean-field scheme [12] is used, which is especially suitable for the strongly correlated metal system. The Coulomb interaction term $H_I$ is taken into consideration merely among the three V-$t_{2g}$ orbitals. In the present work, we choose $U = 4.0$ eV and $J = 0.65$ eV, which are close to previous estimations [13,15]. The dynamical screening term $H_S$ reads (here $i$ denotes the site index, and $\alpha$ the orbital index)

$$H_S = \sum_i \int d\omega \left[ \lambda_\omega (b_\omega^\dagger b_\omega) + \omega b_\omega^\dagger b_\omega \right], \tag{2}$$

which describes the coupling of electronic degrees of freedom (occupation number $n$) to bosonic modes (bosonic operators $b^\dagger$ and $b$) and the frequency-dependent coupling strength $\lambda_\omega = \sqrt{3U(\omega)}/\pi$. The bosonic modes represent the screening of the charge density fluctuations on the strongly correlated $V$-$t_{2g}$ orbitals.

The multi-orbital lattice problem (eq. (1) and eq. (2)) is studied using the DMFT method [1,2], which maps it to a self-consistent solution of a three-orbital quantum impurity model. Then the obtained impurity model (with frequency-dependent interactions $U(\omega)$) is solved using the CT-HYB impurity solver [8,16–18] which based on a stochastic diagrammatic expansion of the partition function in the impurity-bath hybridization. Indeed, the effect of $U(\omega)$ is to dress the fermionic propagators with a bosonic propagator $\exp[-K(\tau)]$, where $K(\tau)$ is the so-called twice-integrated retarded interaction. In terms of $3U(\omega)$ and a factor $B(\tau, \omega) = \cosh[(\tau - \omega)\omega]/\sinh[(\omega^2)\omega]$ with bosonic symmetry, $K(\tau)$ can be expressed by [8,10]

$$K(\tau) = \int_0^\infty \frac{d\omega}{\pi} \frac{3U(\omega)}{\omega^2} [B(\tau, \omega) - B(0, \omega)]. \tag{3}$$

Thus, the diagrammatic weight for Monte Carlo stochastic sampling is supplemented with an additional bosonic term,
which originates from the contribution of $K(\tau)$. The implemented details of the CT-HYB impurity solver improved with recently developed orthogonal polynomial representation algorithm [24] can be easily found in the literature [8,16,17]. In each LDA + DMFT iterations, typically $4 \times 10^9$ Monte Carlo samplings have been performed to reach sufficient numerical accuracy and the results are divided into 1000 data bins. All the calculations are carried out at the inverse temperature of $\beta = 10$ or $\beta = 30$. At last, the maximum entropy method [25] is used to perform analytical continuation to obtain the impurity spectral functions of the degenerate $V$-$t_{2g}$ states.

**Results and discussion.** – SrVO$_3$, a prototype of correlated metal, usually used to benchmark the new theories and tools describing the strongly correlated compounds [9]. Due to its undistorted perovskite structure, the band structure of SrVO$_3$ is relatively simple, resulting in the occupation of one electron in the threefold degenerate $V$-$t_{2g}$ bands crossing the Fermi level. The Oxygen 2$p$ bands are quite well separated from the $t_{2g}$ levels, such that the $3 \times 3$ $V$-$t_{2g}$ tight-binding Hamiltonian is a minimal model required for a correct description of the essential physics of SrVO$_3$. Thus, on the one hand SrVO$_3$ has been the testing case for various LDA + DMFT implementations [13–15,26–29]. And on the other hand, SrVO$_3$ has been the subject of numerous experimental studies, especially by means of the angle-resolved photoemission spectroscopy (ARPES) [30–34]. Therefore, SrVO$_3$ is an ideal system to verify our newly developed implementation of LDA + DMFT computing framework incorporated with the dynamical screening interaction.

To include the dynamical screening interaction within the LDA + DMFT computing scheme, one of the major bottle-necks is the determination of the frequency-dependent Coulomb interaction $U(\omega)$ and corresponding twice-integrated retarded interaction $K(\tau)$. Recently, different approaches such as the constrained density functional theory [11], GW-inspired methods [4], and RPA-based schemes [5,6] are used to extract these quantities. All these calculations are very tedious and tricky. Fortunately, the realistic frequency-dependent $U(\omega)$ for SrVO$_3$ has been computed before [5] based on the constrained RPA (cRPA) approach, so that we can extract $U(\omega)$ directly from the reference as a mandatory input to evaluate $K(\tau)$ in the preprocessing stage. The obtained $U(\omega)$ and $K(\tau)$ are shown in fig. 1, respectively. As is clearly seen in fig. 1(a), $3U(\omega)$ features prominent peaks at 14 eV, 26 eV and 33 eV, respectively. From the peaks of $3U(\omega)/\omega^2$, the energies of plasmon satellite structures in the $V$-$t_{2g}$ density of states can be easily speculated. The real part of $U(\omega)$ ranges from the static screened value $U_0 = 4.0$ eV to the bare unscreened value of about 12 eV at large $\omega$, and its frequency dependence resembles a typical $U(\omega)$ in strongly correlated metals. In a standard LDA + DMFT calculation without dynamical screening effect, the relatively small value of static screened value $U_0$ would lead to a rather weakly correlated picture.

Now let’s focus on the spectral properties of correlated $V$-$t_{2g}$ states. Two different models for SrVO$_3$ are taken into consideration, one contains only the $V$-$t_{2g}$ states (number of Wannier orbitals $N_{\text{wann}} = 3$), the other contains both the $V$-$t_{2g}$ and O-$2p$ states (number of Wannier orbitals $N_{\text{wann}} = 12$). Then we try to solve these models in the framework of LDA + DMFT at $\beta = 10$ and $\beta = 30$ with dynamical-$U$ and static-$U$ cases, respectively. The calculated results are illustrated in the upper panel of fig. 2. Obviously, the results obtained in the dynamical-$U$ and static-$U$ calculations display remarkable differences. Firstly, in the dynamical-$U$ calculations, the spectral weights are shifted strongly to higher frequencies, which leads to a stronger reduction in weight at low energies, compared with the static-$U$ results. Secondly, the quasiparticle resonance peaks around the Fermi level are intensively renormalized, not only the width but also the height of them shrink apparently as $U(\omega)$ is taken into account. Thirdly, in the dynamical-$U$ calculations, some shoulders near $\omega = 0$ are smeared out. Thus, the explicit treatment of the Coulomb repulsion at large frequencies has a substantial effect, even on the low-energy properties of the system [8–10].

We note that recently Casula et al. [9] have studied the spectral properties of SrVO$_3$ by using their BFA-DALA method. Since both the BFA-DALA method and our CT-HYB scheme intend to deal with the strongly correlated systems with frequency-dependent Coulomb interaction, it should be meaningful to justify the calculated results obtained by these two different methods. Comparison of the calculated spectral functions of $V$-$t_{2g}$ states are shown in the lower panel of fig. 2. Both the spectral functions exhibit significant three-peak structures, which are prominent for strongly correlated metal systems [1]. Clearly, the spectral function obtained by our CT-HYB scheme is consistent with that obtained by the BFA-DALA method roughly, besides a slightly spectral weight.

![Fig. 1](Color online) Frequency-dependent Coulomb interaction for SrVO$_3$ from constrained random phase approximation. (a) The real and imaginary parts of the average intra-orbital Coulomb interaction as a function of frequency. The data shown in this plot are taken from reference [5] directly. (b) The twice-integrated retarded interaction function $K(\tau)$ at finite temperatures $\beta = 10$ and $\beta = 30$ calculated by eq. (3).
After careful analytical continuation, the self-energy function of V-t_{2g} states on real axis is obtained and plotted in fig. 3(b). For the real part of the self-energy function \( \Re \Sigma(\omega) \), there are two extrema at the energies \( \omega \sim \pm 1.0 \text{ eV} \), originating from the crossover from the central dynamic \( U \) model, to compare the accuracy and reliability of our method and the BFA-DALA method, more calculations and analyses are needed.

In fig. 3 the calculated self-energy function of V-t_{2g} states are plotted. Let’s first concentrate our attention to the imaginary part of self-energy function at Matsubara frequency axis. In the dynamical-\( U \) model, \( -\Im \Sigma(\omega \rightarrow 0) \) is remarkable larger than that of the corresponding static-\( U \) model. According to the well-known Eliashberg equation: \[ [1] \ Z^{-1} \approx 1 - \frac{\pi}{2} \Im \Sigma(\omega \rightarrow 0) \], the quasiparticle weight \( Z \) can be approximately evaluated. Thus, the dynamical screening effect results in a smaller value of \( Z \), namely a larger value of effective mass \( m^* \). Indeed, for the realistic dynamical-\( U \) model we found a \( Z \approx 0.43 - 0.48 \), which gives an effective mass renormalized by 2.1–2.3 with respect to the LDA band structure. On the other hand, for the corresponding static-\( U \) model we obtained a value of \( Z \approx 0.6 \), which underestimates the electronic correlation by a factor of 1.67, and so the value of \( m^* \).

Recent ARPES experimental data yielded an effective mass \( m^* \approx 2m_0 \) [30–32], which is in good agreement with our findings for the realistic dynamical interaction. It is worthy noting that a static-\( U \) model with a larger instantaneous \( U_0 \) [26–28] can be used to fit the experimental mass renormalization artificially. But the experimentally observed lower Hubbard band (peaked at \( -1.5 \text{ eV} \)) cannot be explained by such a static-\( U \) model. The difficulty here is to reproduce both the effective mass and the position of the lower Hubbard band within the same model. With the dynamically screened interaction model, both the effective mass (\( \sim 2.1m_0 \)) and the lower Hubbard band peaked at \( \sim -1.5 \text{ eV} \) can be reproduced correctly, as is seen in both fig. 3 and fig. 2.

After careful analytical continuation, the self-energy function of V-t_{2g} states on real axis is obtained and plotted in fig. 3(b). For the real part of the self-energy function \( \Re \Sigma(\omega) \), there are two extrema at the energies \( \omega \sim \pm 1.0 \text{ eV} \), originating from the crossover from the central
quasiparticle resonance peak to the lower and upper Hubbard bands, respectively. In the energy regime of the quasiparticle resonance peak ranging from about $-0.5$ eV to $0.5$ eV, $\Re\Sigma(\omega)$ can be roughly fitted by a straight line and the slope of it can be used to evaluate the quasiparticle weight $Z$ as well. Not surprisingly, the quasiparticle weight $Z$ calculated with $\Re\Sigma(\omega)$ is almost identical with that calculated with $\Im\Sigma(i\omega)$. In this energy range the Landau Fermi liquid theory is valid. Strictly speaking, the Fermi liquid regime with $\Re\Sigma(\omega) \sim -\omega$ only extends from $-0.2$ to $0.2$ eV, which is in accord with previous LDA + DMFT calculations [26]. Next to this Fermi liquid regime, there are pronounced “kink”-like structures in $\Re\Sigma(\omega)$ around $\omega = \pm 0.25$ eV as are highlighted by coloured arrows. Apparently, the “kink”-like structures are enhanced by the dynamical screening effects. These “kink”-like structures can be regarded as a fingerprint of strongly correlated systems [36] and become important in the context of quasiparticle dispersion and electronic specific heat. We note that in the previous LDA + DMFT calculations [26] and ARPES experiments [30,34] the “kink”-like structures in SrVO$_3$ have been already identified.

With the knowledge of self-energy function on the real axis, we are now in the position to calculate the $k$-resolved spectral functions or quasiparticle dispersion $A(k, \omega)$. The LDA + DMFT quasiparticle dispersions for the static-$U$ and dynamical-$U$ models along predefined high symmetry lines in the Brillouin zone are shown in fig. 4, respectively. The LDA band structure is presented in this figure as a useful comparison as well. As for the static-$U$ model, the quasiparticle band structure displays strongly band renormalization with a factor $\sim 1.7$ with respect to the LDA band dispersion. For example, we can see from the upper panel of fig. 4 that the bottom of the quasiparticle band is located at approximately $\omega = -0.6$ eV, in contrast to the LDA value of $\omega = -1.0$ eV. At $\omega = \pm 0.3$ eV along the $R$-$\Gamma$ and $M$-$\Gamma$ lines there exist less-discernible “kink”-like structures as indicated by two green arrows [26,30,34], which stem from the shoulders in the real part of the self-energy function (see fig. 3(b)). As for the dynamical-$U$ model, its quasiparticle band structure exhibits similar features with respect to that of the static-$U$ model. Around the Fermi level, the quasiparticle band structure of the dynamical-$U$ model is more renormalized with a factor $\sim 2.1$ and has less intensity, and the bottom of the quasiparticle dispersion is located at $\omega = -0.5$ eV. The “kink” structures in the quasiparticle band structure are more discernible than those of the corresponding static-$U$ model. Previously, the $k$-resolved spectral functions for BaFe$_2$As$_2$ have been calculated by Werner and Casula et al. [7,10] with or without dynamical screening effect, similar trend was observed.

**Conclusion.** In this letter, we try to incorporate the dynamical screening effect into the modern LDA + DMFT computing framework and choose powerful

![Fig. 4: (Color online) Angle-resolved spectral functions $A(k, \omega)$ for SrVO$_3$. Upper panel: static Coulomb interaction case. Lower panel: frequency-dependent Coulomb interaction case. The coloured dash lines denote the LDA band structures and the green arrows indicate the “kink”-like structures. In these LDA + DMFT calculations, only the three V-3$d^2$ bands are taken into considerations and the inverse temperature $\beta$ is fixed to be 10.](image)

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