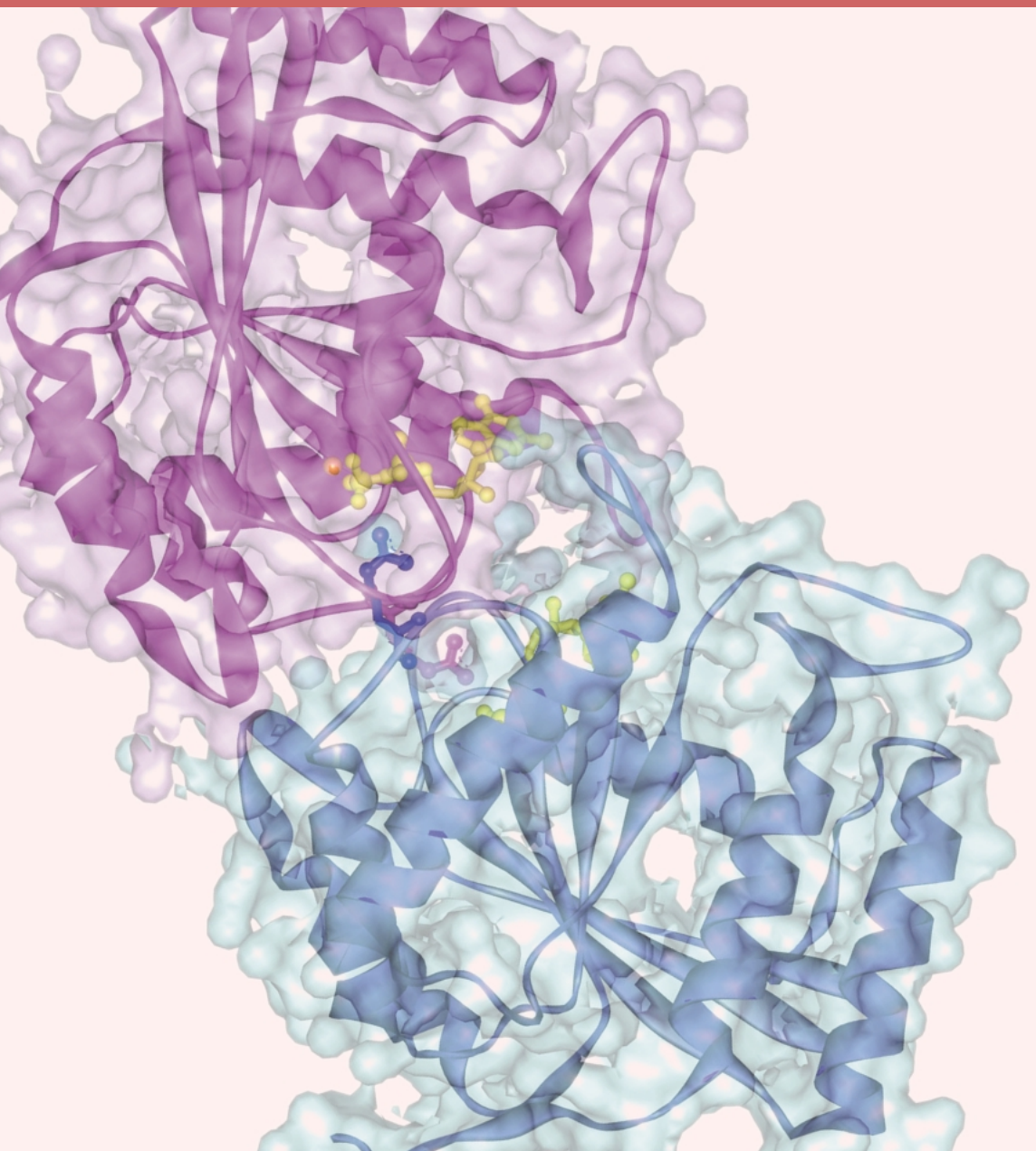
Highlights



11 Theory

11-1 Unified Theory on the Angle-Resolved Photoemission and Light Absorption Spectra of the Strongly Correlated Electron System

As experimental techniques have become more sophisticated to clarify precise electronic structures, we have faced a serious problem that the conventional band picture does not work well on the strongly correlated electron system.

According to the linear response theorem, it has been believed that a small external perturbation induces only small changes in the system, and that we can clarify the electronic structures of the ground and excited states of the objective materials by observing the response function. Even now, we only know the electronic structure through this response function, but the relationship between the perturbation and its response is quite complicated in the strongly correlated electron system. For example, the energy resolution of angle-resolved photoemission spectroscopy has now been reduced to less than 1 meV. Nevertheless, the angle-resolved photoemission spectrum (ARPES) of the copper-oxide compound only has quite a broad and structure-less peak of about 1 eV, which indicates that the ARPES is dominated by the incoherent component [1]. The conventional band picture cannot explain such an incoherence-dominated ARPES.

To overcome the present situation, we have to calculate the response functions as correctly as possible and explain different experiments in terms of the same model comprehensively.

In this study, we use the quantum Monte Carlo simulation to calculate optical response functions. We show that the one-dimensional (1-D) extended Hubbard model explains the light absorption spectrum (LAS) and the ARPES of $[Ni(chxn)_2Br]Br_2$ (chxn=1R, 2R-cyclohexane-diamine) consistently.

The crystal structure of the Ni-Br complex is schematically shown in Fig. 1. Each chain is separated by the ligand molecules and counter anions. Therefore, the electron state of this system is highly 1-D oriented. Furthermore, due to the tight hydrogen-bond network between the ligand and counter anion, the Ni-Br complex does not suffer the Peierls distortion and an equidistant lattice structure remains even at low temperature.

The LAS of the Ni-Br complex is shown in Fig. 2(a) [2]. The spectrum has a characteristic form constituted of a sharp and asymmetric peak at 1.28 eV and a long tail leading to a broad peak at 2.7 eV. We try to explain this LAS of the Ni-Br complex within the framework of the 1-D

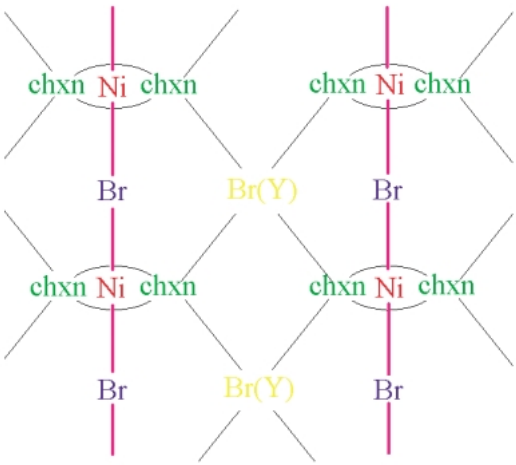


Figure 1
Schematic representation for the crystal structure of $[Ni(chxn)_2Br]Br_2$.

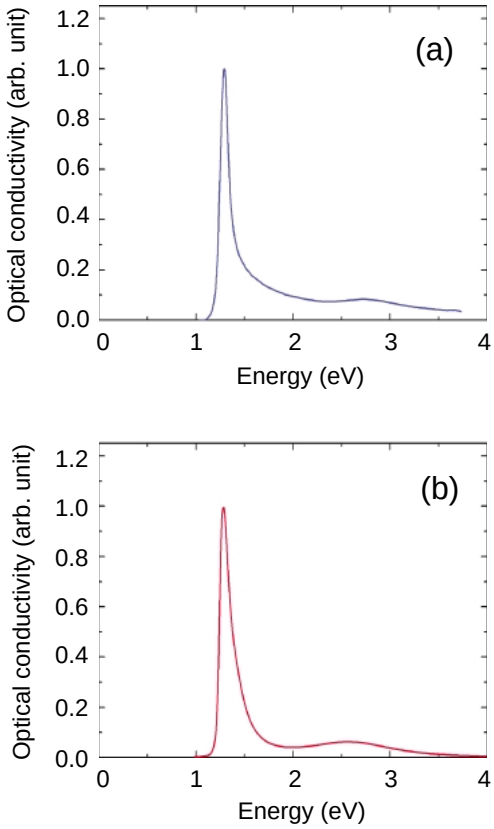


Figure 2
Experimentally observed LAS of the Ni-Br complex (a) and numerical results (b).

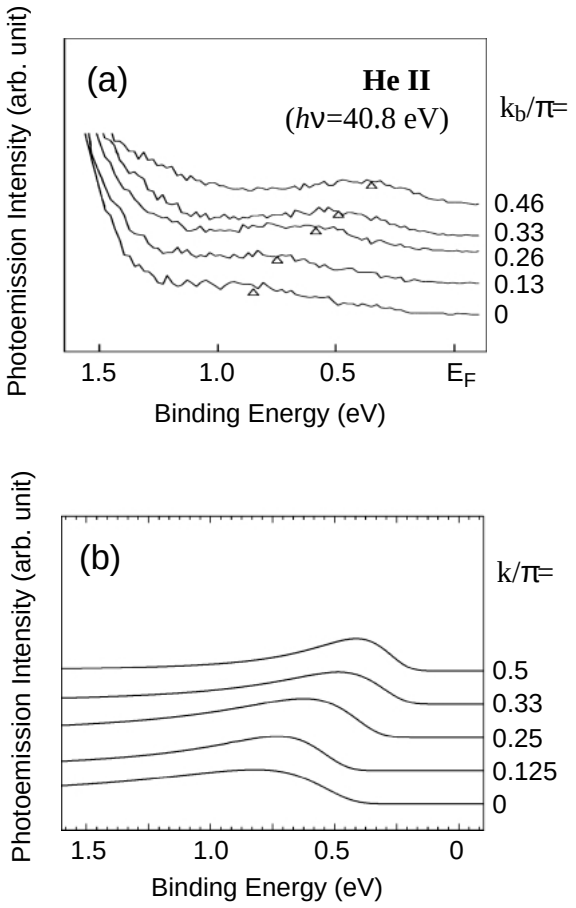


Figure 3
Experimentally observed ARPES of the Ni-Br complex (a) and numerical result (b).

extended Hubbard model. The Hamiltonian is represented by

$$H = -t\sum_{l=1}^{N}(a_{l\sigma}^{+}a_{l+1\sigma} + a_{l+1\sigma}^{+}a_{l\sigma}) + U\sum_{l=1}^{N}n_{l\alpha}n_{l\beta} + V\sum_{l=1}^{N}n_{l}n_{l+1}$$

The LAS of the 1-D extended Hubbard model is shown in Fig. 2(b). The parameters (*t,U,V*) are set at (0.22 eV, 2.4 eV, 1.1 eV) and a half-filled periodic ring with 48 electrons is considered. We can see that our numerical result reproduces the experimentally observed spectrum. The infrared divergence-like peak at 1.28 eV is caused by the nonlinear coupling between a charge transfer exciton and collective spin excitation [3]. On the other hand, a small peak at 2.7 eV originates from a multiple excitation of electron-hole pairs induced by the Coulomb interaction [3].

Recently the ARPES of the Ni-Br complex has been observed at room temperature [4]. As shown in Fig. 3(a), the ARPES has quite a broad peak at each wave-number. To check the validity of the 1-D extended Hubbard model for the Ni-Br complex, we calculate the momentum-resolved Lehmann spectrum of the one-body Green function with the same parameters and the same conditions as in the case of the LAS. The Lehmann spectrum of the one-body Green function directly corresponds to the photoemission spectrum. Figure 3(b) shows the momentum dependence of the Lehmann spectrum of the 1-D extended Hubbard model [5]. By comparing Fig. 3(a) and (b), we can conclude that both the peak shapes and peak positions of our numerical results agree well with those of the experimentally observed ARPES of the Ni-Br complex. Thus, we have verified that the LAS and ARPES of the Ni-Br complex can be consistently explained by the 1-D extended Hubbard model.

We believe that such a comprehensive explanation is a big advance for the true understanding of the strongly correlated electron system. We need as many experiments as possible to refine the model. Eventually, we will be able to find the best model for each material, and clarify its electronic structure appropriately.

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References

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11-2 Path-Integral Theory for Photoemission Spectra of Intermediately Correlated Systems

The nature of the electronic states around the Fermi level of perovskite-type compounds, such as $CaVO_3$ and $SrVO_3$, is a matter of great interest in recent years [1-3]. According to the most up-to-date experiments, a new peak structure has been discovered near the Fermi level (not on the Fermi level) of these crystals, in addition to the well-known Hubbard-band peaks, as shown in Fig. 4. The intensity is clearly suppressed just on the Fermi level, but enhanced slightly below this level. The origin of this new peak has become a debating point.

In this study, we calculate the Lehman spectrum of the one-body Green function of the one-dimensional (1-D) half-filled Hubbard model. The Hamiltonian is given by

$$H = -t\sum_{l=1}^{N}(a_{l\sigma}^{+}a_{l+1\sigma} + a_{l+1\sigma}^{+}a_{l\sigma}) + U\sum_{l=1}^{N}n_{l\alpha}n_{l\beta}$$

Especially, we are interested in the weak and intermediate correlation regime, because we have significant temperature-induced spectral changes in this correlation regime. In Fig. 5, we show the temperature-dependence of the Lehmann spectrum of the 1-D half-filled Hubbard model at U/t=2. At $t/(k_BT)=1$, the Lehmann

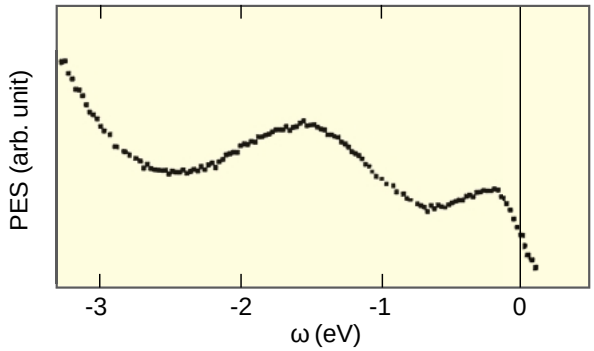

Figure 4
Photoemission spectrum of $SrVO_3$, taken from ref. 3.

spectrum shows the metallic state, which is quite fuzzy around the Fermi level because of the high temperature. At $t/(k_BT)=25$, the Lehmann spectrum has two narrow peaks near the Fermi level and two broad peaks outside the narrow peaks. We can see that there is still a finite spectral component at the Fermi level. At $t/(k_BT)=30$, the spectral intensity at the Fermi level moves to the narrow peaks, and the system becomes insulating.

Figure 6 shows the Lehmann spectra at U/t = 2.5, and 4, with fixed T, $t/(k_BT) = 18$. We can see that the Lehmann spectrum has no peak around the Fermi level in the case of U/t=4. Thus, we have clarified that the change of the Lehmann spectrum from the metallic state to the insulating one has intermediate steps. The Lehmann spectrum of the intermediate state has characteristic peaks around the Fermi level, in addition to the so-called Hubbard-band peaks.

Let us now qualitatively clarify the origin of these multi-peaked structures in connection with the infinite dimensional case. Since our system is half-filled, antiferromagnetic correlation dominates the electronic property. As is well-known, in the case of the infinite dimensional system, this antiferromagnetic order has no inter-site coherence. Thus, we can infer that the absence

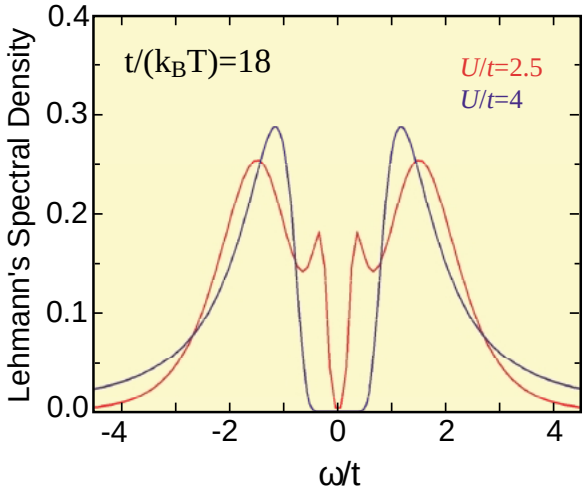

Figure 6
U-dependence of the Lehmann spectrum of the 1-D half-filled Hubbard model.

of the inter-site coherence is the origin of the new peak structure. Since our system is one-dimension, the real long-range order is absent from the beginning. However, in the present case, we should note that the strength of U is limited within intermediate (or small) values. Therefore, the spatial extent of the antiferromagnetic spin correlation is extremely short, compared to the large U regime. As a result, the antiferromagnetic order only has negligibly small inter-site coherence in the intermediate correlation regime. In this point of view, it becomes quite similar to the infinite dimensional systems. Therefore, we can understand that the characteristic peaks appear in the finite (1-, 2-, 3-) dimensional systems as well as in the infinite dimensional systems, if the correlation strength is intermediate. In fact, we have preliminarily calculated the cases of 2- and 3- dimensional systems with intermediate strengths, and have confirmed that the multi-peaked structures appear even in these cases.

We also show the Lehmann spectrum of the momentum specified one-body Green function in Fig. 7.

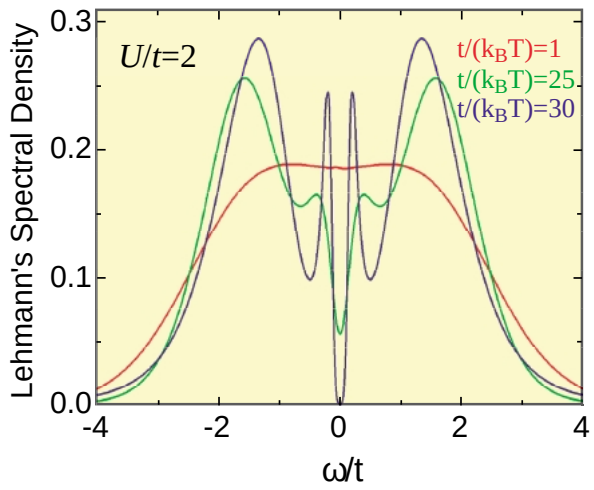

Figure 5
Temperature-dependence of the Lehmann spectrum of the 1-D half-filled Hubbard model (U/t=2).

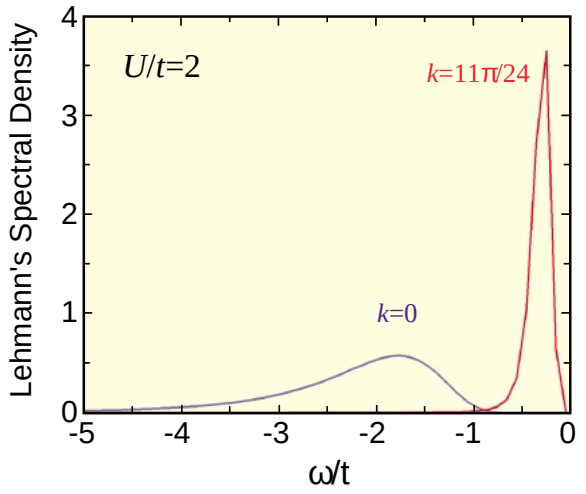

Figure 7
The Lehmann spectrum of the momentum-specified one-body Green function.

The spectrum has a sharp peak near the Fermi level, while it only has a broad peak near the band bottom. Therefore, we can say that the sharp peak near the Fermi level shows the quasi-coherent state, while the broad peak near the band bottom shows the incoherent state.

Our numerical results in the intermediate regime agree qualitatively with the recently observed photoemission spectra of $CaVO_3$ and $SrVO_3$. In contrast to the previous experiments shown in Fig. 4, the recent bulk-sensitive experiment, using soft X-ray synchrotron radiation, has revealed that the photoemission spectra of these materials have a much sharper peak just below the Fermi level in addition to the broad peak on the high-binding-energy side [1]. Therefore, this sharp peak is explained as the quasi-coherent state, while the broad peak is caused by the incoherent state. A more precise analysis of the experiments will be done in the near future.

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