Systematic Development of the Spectral Function in the $3d^1$ Mott-Hubbard System $Ca_{1-x}Sr_xVO_3$

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We present an experimental manifestation of how the single-particle density of states around the Fermi level (E_F) changes near a Mott transition. In a metallic perovskite-type $3d^1$ Mott-Hubbard system $Ca_{1-x}Sr_xVO_3$, as one decreases x the spectral weight of the coherent band near E_F is gradually transferred to that of the precursor of the lower Hubbard band ~2 eV below E_F. The momentum dependence of the quasiparticle self-energy and hence the long-range exchange or correlation is shown to become progressively important as one approaches the Mott transition from the metallic side.

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From various viewpoints, attempts have been made to describe how the single-particle spectral density $\rho(\omega)$ behaves in a system close to a Mott transition. In the original treatment by Hubbard [1], a metal-insulator transition (MIT) is controlled by the relative magnitudes of the on-site Coulomb energy U and the one-electron bandwidth W. If the number of electrons per atomic site is an integer, the splitting between the lower and upper "Hubbard bands" (LHB and UHB) gradually increases with increasing U/W, and the MIT occurs at $U/W \sim 1$, where a Mott-Hubbard gap opens. This treatment, however, does not properly treat low-energy excitations and the Fermi-liquid properties in the metallic region [2]. In contrast, a Fermi-liquid description has been given by Brinkman and Rice [3] in their Gutzwiller treatment of the Hubbard model, but high-energy excitations are missing there. Fujimori *et al.* reported the ultraviolet photoemission spectroscopy (UPS) spectra of several Ti^{3+} and V^{4+} compounds and indicated that $\rho(\omega)$ consists of a band of quasiparticle excitations near the Fermi energy (E_F) and a high-energy satellite signaling the formation of the LHB. They also suggested that the spectral weight is transferred from the quasiparticle band to the precursor of the LHB with increasing U/W. Recent theoretical studies of the Hubbard model in the limit of infinite dimension have given similar illustrations of $\rho(\omega)$ [5–7].

In this Letter, we go one step further and investigate more closely how $\rho(\omega)$ actually changes as we systematically control U/W within a single Mott-Hubbard system, $Ca_{1-x}Sr_xVO_3$. Both $CaVO_3$ and $SrVO_3$ are metallic perovskite-type V^{4+} oxides, which have a nominally $3d^1$ configuration. $CaVO_3$ is near a MIT [8,9], while the transport and magnetic properties of $SrVO_3$ are those of a conventional Fermi liquid [10]. The difference between $CaVO_3$ and $SrVO_3$ is attributed to the different magnitudes of the U/W value, which are roughly estimated in the present study. If we replace the Sr^{2+} ions by Ca^{2+} ions, whose ionic radius is smaller than Sr^{2+}, the V-O-V bond angle θ is changed from 180° to ~160° [11]. The distortion reduces the overlap between neighboring $3d$ orbitals mediated by oxygen $2p$ orbitals, leading to a decrease in W by a factor of $|\cos\theta|$. Therefore, as one changes the Sr concentration (x) in $Ca_{1-x}Sr_xVO_3$, one can control U/W. We note that, since the average number of $3d$ electrons is always one per V atom, one can study changes in the electronic properties as functions of U/W.

Polycrystalline $Ca_{1-x}Sr_xVO_3$ samples were prepared as reported elsewhere [11]. By x-ray diffraction measurements, it was confirmed that each of the samples was in a single phase and that the lattice constants varied continuously as x was changed. Oxygen stoichiometry was determined by a thermogravimetric analysis, and was carefully controlled to become 3.00 ± 0.01. UPS experiments were performed using synchrotron radiation at the beam line BL-11D of the Photon Factory, National Laboratory for High Energy Physics. We obtained clean surfaces by scraping *in situ* with a diamond file in an ultrahigh vacuum (the operating pressure $\sim 2 \times 10^{-10}$ Torr). During the scraping and measurements, the sample temperature was maintained at ~80 K to avoid surface degradation [12].

Figure 1 (top) shows the density of states (DOS) $D(\varepsilon_k^0)$ of $CaVO_3$ and $SrVO_3$ calculated by the augmented-plane-wave (APW) method within the local density approximation (LDA) [13], where ε_k^0 is the energy of an electron with momentum $\boldsymbol{k}$. In this figure, the $D(\varepsilon_k^0)$'s have been convoluted with a Gaussian function of 0.3 eV FWHM which represents the instrumental resolution and was compared with the UPS spectra $\rho_{\mathrm{expt}}(\omega)$ near E_F [Fig. 1 (bottom)]. For simplicity, it has been assumed that both $CaVO_3$ and $SrVO_3$ have a cubic structure, so that there is no significant difference between the two calculations. However, if we consider the actual distortion of the crystal in the calculation, the bandwidth is multiplied by $|\cos\theta|$; hence, in the following analysis of the spectral function, we use the DOS thus narrowed. All the $\rho_{\mathrm{expt}}(\omega)$'s have been normalized to the area integrated up to ~3 eV, assuming an appropriate background [14]. The spectra of the Fermi cutoff of Au evaporated on the samples as shown in the figure were used as a reference of E_F and the instrumental resolution. Although LDA band-structure calculations often

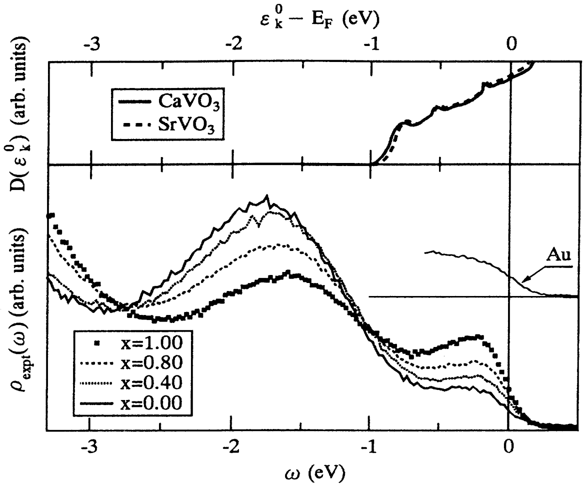

FIG. 1. Top: DOS $D(\varepsilon_k^0)$ of CaVO$_3$ and SrVO$_3$ calculated by the APW method. Bottom: measured photoemission spectra $\rho_{\text{expt}}(\omega)$ of Ca$_{1-x}$Sr$_x$VO$_3$ taken with $h\nu = 50$ eV. A spectrum of Au is also shown as a reference to E_F and the instrumental resolution.

give correct Fermi surfaces, these calculations do not consider electron correlation effects, which play an essential role in determining $\rho(\omega)$ in strongly correlated systems. Indeed, in Ca$_{1-x}$Sr$_x$VO$_3$, $D(\omega)$ and $\rho_{\text{expt}}(\omega)$ appear quite different. We recognize two well-defined structures in $\rho_{\text{expt}}(\omega)$: one is due to the quasiparticle excitations near E_F, and the other is the precursor of the LHB centered at ~1.7 eV. Both features were confirmed to be of V 3d origin, using resonant photoemission spectroscopy [15]. In going from SrVO$_3$ to CaVO$_3$, i.e., as we increase U/W, the spectral density of the quasiparticle band gradually decreases and the missing spectral weight is transferred to the high-energy part, consistent with the previous report on other $3d^1$ materials [4]. The observed systematic change of $\rho(\omega)$ illustrates how the Mott-Hubbard gap collapses as U/W decreases.

There is another feature in $\rho(\omega)$ that needs further attention. In the simplest Fermi-liquid picture where the self-energy of the quasiparticle is local, i.e., $\boldsymbol{k}$ independent, $\rho(E_F)$ would be independent of the interaction strength U/W, that is, $\rho(E_F)$ would be equal to $D(E_F)$ [16]. In particular, $\Sigma(\omega, \boldsymbol{k})$ is known to be $\boldsymbol{k}$ independent in infinite dimension, leading to the $\rho(E_F)$ value equal to $D(E_F)$ [17]. However, what we observed is that $\rho(E_F)$ is much smaller than $D(E_F)$, particularly for CaVO$_3$, in contrast to the Fermi-liquid picture with the $\boldsymbol{k}$ independent self-energy. There may be several explanations for the suppression of $\rho(E_F)$ compared to $D(E_F)$. First, we may suspect that we have overlooked a sharp peak at E_F because of the limited resolution. In that case, however, the peak should be so sharp compared to the resolution that the effective mass of the quasiparticles will become much larger than the relevant value as described below. Second, the effect of the finite temperature should also be considered. Georges and Krauth [7] described the suppression of the quasiparticle excitations when raising the temperature to values comparable to the quasiparticle bandwidth. However, no intrinsic temperature dependence was observed in our UPS spectra, probably because the temperature (~80 K) was low enough compared with the bandwidth of ~1 eV. Third, the suppression may be caused by the $\boldsymbol{k}$ dependence of the self-energy [18]; the $\boldsymbol{k}$ dependence can be caused by a pseudogap formation, as discussed in strongly couple electron-phonon systems [19] and in spin-fluctuation systems [20], or nonlocal exchange interaction neglected in LDA.

In the following, we give a phenomenological description of the single-particle spectral function by taking into account the $\boldsymbol{k}$ dependence of the self-energy. Since we observed Fermi surfaces in CaVO$_3$ by Shubnikov–de Haas measurement [21] as well as the clear Fermi cutoff in the UPS spectra of Ca$_{1-x}$Sr$_x$VO$_3$, we are convinced that low-energy excitations of this system can be considered within the Fermi-liquid description [22]. For simplicity, the self-energy $\Sigma(\omega, \boldsymbol{k})$ has been assumed to be of the form

$$\Sigma(\omega, \boldsymbol{k}) \equiv \Sigma(\omega) + \Sigma(\boldsymbol{k}). \tag{1}$$

Since $\Sigma(\omega, \boldsymbol{k}) = [\Sigma(\omega, \boldsymbol{k}) - \Sigma(0, \boldsymbol{k})] + \Sigma(0, \boldsymbol{k})$, Eq. (1) means that we have assumed $\Sigma(\omega, \boldsymbol{k}) - \Sigma(0, \boldsymbol{k})$ to be $\boldsymbol{k}$ independent. $\rho(\omega)$ is the $\boldsymbol{k}$ integral of $A(\omega, \boldsymbol{k}) = -\mathrm{Im}G(\omega, \boldsymbol{k})/\pi$, where $G(\omega, \boldsymbol{k}) = [\omega - \varepsilon_k^0 - \Sigma(\omega, \boldsymbol{k})]^{-1}$:

$$\rho(\omega) = -\frac{1}{\pi} \int d\varepsilon_k^0 \, D(\varepsilon_k^0) \, \mathrm{Im}G(\omega, \boldsymbol{k}). \tag{2}$$

As for $\Sigma(\omega)$, we adopted a model function

$$\Sigma(\omega) \equiv g \frac{\gamma \omega}{\omega + i\gamma} \frac{\Delta}{\omega + i\Delta},$$

which behaves as $\Sigma(\omega) \approx -a\omega - ib\omega^2$ near E_F, and satisfies the Kramers-Kronig (KK) relation in the whole energy range. Here, the quasiparticle energy $\omega = \varepsilon_k$ is the solution of $\varepsilon_k = \varepsilon_k^0 + \mathrm{Re}\Sigma(\varepsilon_k, \boldsymbol{k})$. Now we define the "$k$ mass" m_k and the "ω mass" m_ω [23] as

$$\frac{m_b}{m_k} \equiv \left| \frac{\partial \varepsilon_k^0}{\partial \boldsymbol{k}} \right|_{k=k_F} + \frac{\partial \,\mathrm{Re}\Sigma(\omega, \boldsymbol{k})}{\partial \boldsymbol{k}} \Bigg|_{k=k_F} \Bigg| \Bigg/ \left| \frac{\partial \varepsilon_k^0}{\partial \boldsymbol{k}} \right|_{k=k_F} \Bigg|$$

and

$$\frac{m_\omega}{m_b} \equiv 1 - \frac{\partial \,\mathrm{Re}\Sigma(\omega, \boldsymbol{k})}{\partial \omega} \Bigg|_{\omega=E_F},$$

where m_b is the band mass given by the band-structure calculation. m_ω/m_b describes the mass enhancement of the quasiparticle at E_F due to dynamical effects of electron correlation, electron-phonon interaction, etc., $m_k/m_b < 1$ causes the suppression of $\rho(E_F)$ through the $\boldsymbol{k}$ dependence of the self-energy. Then, we can express the effective mass ratio of the quasiparticle

$$\frac{m^*}{m_b} \equiv \left| \frac{\partial \varepsilon_k^0}{\partial \boldsymbol{k}} \right|_{k=k_F} \Bigg| \Bigg/ \left| \frac{d\varepsilon_k}{d\boldsymbol{k}} \right|_{k=k_F} \Bigg|$$

since $m^*/m_b = (m_\omega/m_b) m_k/m_b$. As for $\Sigma(\boldsymbol{k})$, we assume the simplest tractable form,

$$\Sigma(\boldsymbol{k}) \equiv (\alpha - 1)\varepsilon_k^0,$$

which means the uniform widening of the energy bands for $\alpha > 1$. Then the k mass is given by $m_k/m_b = 1/\alpha$.

We calculated $\rho(\omega)$ using Eq. (2) and compared it with the observed spectral density $\rho_{\text{expt}}(\omega)$ in the quasiparticle excitation (coherent) regime (-0.5 eV $< \omega < 0$). Adjustable parameters g, γ, Δ, and α were varied to obtain the best fit. The parameters may be related to U and W according to the second-order perturbation in U [24]. In $\text{Ca}_{1-x}\text{Sr}_x\text{VO}_3$, however, we observed such a large incoherent band that we should interpret U and W as the renormalized ones, U_{eff} and W_{eff}: In this case, $\sqrt{\gamma\Delta} \sim W_{\text{eff}}$ and $g\gamma\Delta/(\gamma + \Delta) \sim -\text{Im}\Sigma(-W_{\text{eff}})$, and g reflects U_{eff}, since m_ω/m_b is equal to $1 + g$. The shift of the precursor of the LHB as we change x (Fig. 1) may correspond to the changes in U_{eff}. The precursor of the LHB (i.e., the incoherent band) cannot be reproduced by the present form of $\Sigma(\omega, \boldsymbol{k})$, while we take into account the effect of the evolution of the incoherent band through the normalization of the spectra, i.e., the relative magnitudes of the intensity of the incoherent band I_{incoh} to that of the coherent band I_{coh} is related to the ω mass as $m_\omega/m_b \sim I_{\text{incoh}}/I_{\text{coh}} + 1$ [25].

Figure 2 shows the results of the analysis for SrVO_3 and CaVO_3. Open circles in the figure correspond to the UPS spectrum. The best fits of the calculated $\rho(\omega)$'s to the experimental spectra $\rho_{\text{expt}}(\omega)$'s within the energy range of $-0.5 < \omega < 0.3$ eV are shown in Fig. 2 for SrVO_3 [curve (a3)] and CaVO_3 [curve (c)]. Adjustable parameters $\alpha = 3.2$, $g = 3.1$, $\Delta = 12$ eV, and $\gamma = 0.31$ eV for SrVO_3 yield $m_k \cong 0.3m_b$, $m_\omega \cong 4.1m_b$, and $m^* \cong 1.3m_b$. For CaVO_3, $\alpha = 7.1$, $g = 19$, $\Delta = 8.0$ eV, and $\gamma = 8.0 \times 10^{-2}$ eV, which yield $m_k \cong 0.13m_b$, $m_\omega \cong 19m_b$, and $m^* \cong 2.6m_b$. Before we discuss the results in more detail, it is worthwhile to see how $\rho(\omega)$ was obtained from $D(\omega)$. The dash-dotted line in Fig. 2 (top) represents $D(\omega)$ of SrVO_3. $D(\omega)$ was used for the calculation of $\rho(\omega)$ via Eq. (2). The $D(\omega)$ in the figure has been normalized so that the area below E_F becomes equal to the integrated area of the d-band emission of the experimental spectrum, and has been convoluted with the instrumental resolution. First, we ignored the $\boldsymbol{k}$ dependence of the self-energy, i.e., $\alpha = 1$: $m_k = m_b$. Curve (a1) shows $\rho(\omega)$ for $m_k = m_b$ and $m_\omega = m^* \cong 4.1m_b$, which is multiplied by a Fermi distribution function. Then, in curve (a2), we took into account the instrumental resolution. It is obvious that $D(\omega)$ has become a sharp coherent band in $\rho(\omega)$ due to the mass enhancement of $m_\omega \cong 4.1m_b$, and the peak has not been suppressed enough to be fitted to the experimental spectrum solely by the instrumental broadening. Therefore, we switched on the $\boldsymbol{k}$ dependence of the self-energy [26]: We took $m_k \cong 0.32m_b$, which reduces the effective mass from $m^* = m_\omega \cong 4.1m_b$ to $m^* \cong 1.3m_b$, and the calculated $\rho(\omega)$ fits well to the experimental spectrum [curve (a3)]. Since m_b is around $1.5m_0$, where m_0 is the free electron mass, the obtained $m^* \sim 2m_0$ corresponds well to $m^* = 2.33m_0$ [27], deduced from the Pauli spin susceptibility $\chi = 9.5 \times 10^{-5}$ emu/mol [10]. Here we roughly estimate the U/W values. $U/2$ corresponds to the energy of the incoherent peak ($\sim$1.6 eV for SrVO_3 and $\sim$1.7 eV for CaVO_3), and the half bandwidth is $W/2 \sim (m^*/m_b)W_{\text{eff}}/2$ ($\sim$1.3 eV for SrVO_3 and $\sim$1.0 eV for CaVO_3) comparable to that of the LDA calculation. Accordingly, the U/W value becomes $\sim$1.2 for SrVO_3 and $\sim$1.7 for CaVO_3 [28].

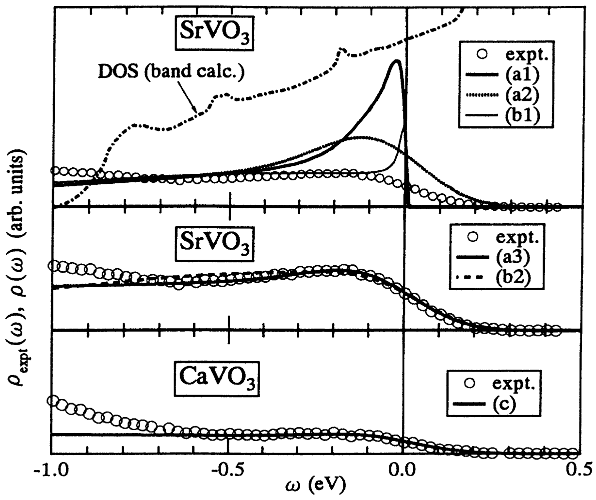

FIG. 2. Comparison between $D(\omega)$ (dash-dotted line) for SrVO_3, $\rho(\omega)$, and the UPS spectrum (open circles) for SrVO_3 and CaVO_3. For curves (a1), (a2), and (a3), $m_\omega \cong 4.1m_b$ and the spectra have been multiplied by a Fermi distribution function. See text for details.

As mentioned before, we may obtain an alternative fit without introducing the $\boldsymbol{k}$ dependence of the self-energy, as depicted by curves (b1) and (b2) in Fig. 2 for SrVO_3 [29], if we assume a strong ω dependence in $\Sigma(\omega)$, which leads to a very large m_ω [4]. For these curves, $m_k = m_b$, $m_\omega = m^* \cong 45m_b$, and the extremely sharp peak at E_F is obscured by the instrumental broadening. However, if this is the case, it is difficult to explain the moderately large m^* deduced from the spin susceptibility.

We have fitted the UPS spectra of the $\text{Ca}_{1-x}\text{Sr}_x\text{VO}_3$ with various x in the same way; the results are summarized in Fig. 3. The figure shows that as x decreases, m_ω increases monotonically up to $\sim 20m_b$, indicating a very large enhancement as U/W approaches $\sim$1. Nevertheless, the effective mass m^* is not so much enhanced, because of the decrease in m_k with decreasing x. In CaVO_3, the effective mass m^* is not so large, although the spectral weight at E_F is very small. We find $m^* \cong 2.6m_b \approx 4m_0$, which is comparable to $m^* \sim 4.9m_0$ deduced from the electronic specific heat coefficient $\gamma \sim 14$ mJ/mol K^2 [30]. The small m_k means that the self-energy shows a strong $\boldsymbol{k}$ dependence; therefore, it seems reasonable to advocate that, in the vicinity of a Mott transition, long-range interactions such as electron-optical-phonon interaction, spin fluctuations, or electron-electron interaction through the long-range Coulomb and exchange interaction become important, causing the suppression of $\rho(E_F)$, as well as preventing m^* from so extremely being enhanced. The microscopic origin of the small m_k remains to be clarified in future experimental and theoretical studies.

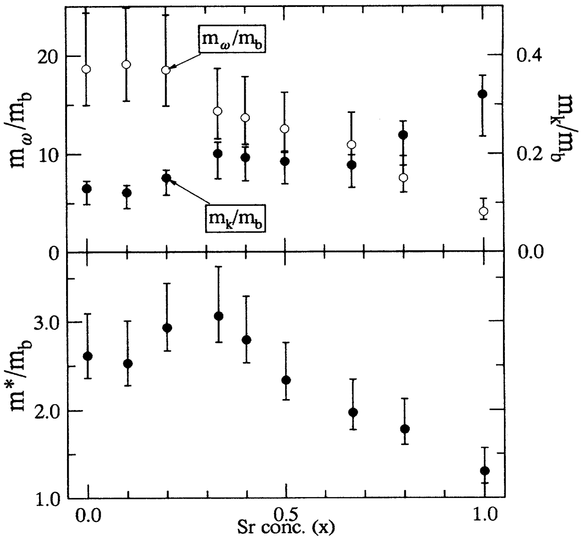

FIG. 3. Effective masses deduced from the UPS spectra of $Ca_{1-x}Sr_xVO_3$. As x decreases, m_ω shows a fairly large enhancement (top). Nevertheless, the effective mass m^* is not enhanced as much (bottom) because of the decrease in m_k.

In conclusion, we have presented a systematic evolution of $\rho(\omega)$ observed by the UPS measurement of $Ca_{1-x}Sr_xVO_3$. We have confirmed that, as we increase the magnitude of U/W, the spectral weight is transferred from the coherent band to the precursor of LHB. Phenomenological analysis of the suppression of $\rho(E_F)$ based on the Fermi-liquid picture indicates that the $\boldsymbol{k}$ dependence of the single-particle self-energy becomes important near the Mott transition.

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[12] Once we obtained fresh surfaces by scraping, the UPS spectra were never changed by the successive scraping; this will also help us confirm the adequate quality and homogeneity of the samples at least within the escape depth of the photoelectrons.
[13] In this Letter, LDA band structures are taken as the noninteracting ($U = 0$) limit.
[14] In Fig. 1, no background has been subtracted. But, for each $\rho_{\mathrm{expt}}(\omega)$ in Fig. 2, a background, i.e., a broad Gaussian function centered at ~5 eV, which shows that the tail of the O $2p$ band has been subtracted.
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[25] m_ω/m_b in $CaVO_3$ and $SrVO_3$ deduced by this simple relation [K. Morikawa *et al.* (unpublished)] are roughly equal to our results. If we add another term to $\Sigma(\omega,\boldsymbol{k})$ that reproduces the incoherent peak, the contribution of the term would be small at $\omega \sim 0$.
[26] We should note that our approach is purely phenomenological and that there is little first principle theoretical basis for it.
[27] If we consider the Wilson's ratio is equal to 2, the effective mass of $SrVO_3$ is estimated to be $\sim 1.2 m_0$.
[28] Since the system is metallic, U/W should be less than 1. This discrepancy is due to the naive estimation assuming that the $3d$ band is symmetric with regard to E_F.
[29] If we consider the $\boldsymbol{k}$ dependence of $\Sigma(\omega,\boldsymbol{k})$, as assumed in this Letter, all the adjustable parameters are determined within the error bars as shown in Fig. 3.
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