Anisotropy of Mott-Hubbard Gap Transitions due to Spin and Orbital Ordering in LaVO$_3$ and YVO$_3$

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Anisotropic optical spectra coupled with antiferromagnetic spin ordering (SO) and orbital ordering (OO) have been investigated for single crystals of LaVO$_3$ and YVO$_3$. The orbital-dependent Mott-Hubbard gap transitions are observed around 2 eV. The transitions composed of the two peaks show distinct anisotropy and selection rules, reflecting the respective SO and OO patterns. The temperature dependence of the anisotropic transitions clearly indicates the SO/OO correlation and the evolution of the order parameters.

KEYWORDS: orbital-dependent Mott-Hubbard gap transitions, spin and orbital ordering, LaVO$_3$, YVO$_3$

As a consequence of strong electron correlation, tight coupling among spin, charge and orbital degrees of freedom produces a variety of intriguing physical properties in $3d$ transition-metal oxides. An example of such a property is the colossal magnetoresistance and related phenomena for the hole-doped perovskite-type manganites, in which spin ordering (SO) and orbital ordering (OO) govern the charge dynamics and electronic structure. Vanadates, as well as manganites, are regarded as one of the prototypical systems that show spin-charge-orbital coupled phenomena. To investigate charge dynamics coupled with the spin and orbital correlations, we have revisited classic vanadium oxides, perovskite-type LaVO$_3$ and YVO$_3$.

LaVO$_3$ and YVO$_3$ are prototypical Mott-Hubbard type insulators with the V electron configuration of $3d^2$, and have a $Pbnm$ orthorhombic unit cell with $a \approx b \approx c/\sqrt{2}$ at room temperature. These compounds undergo a magnetic transition to an antiferromagnetic (AF) state as well as a structural phase transition due to the OO as temperature is decreased. The schematic structures of the SO and OO in both compounds are shown in Figs. 1(a) and 1(b). In LaVO$_3$, the magnetic transition occurs at 143 K, and the ordered spin configuration is $C$-type with an AF coupling in the $ab$-plane and a ferromagnetic one along the $c$-axis. This compound undergoes another phase transition, a first-order structural transition from an orthorhombic to a monoclinic form at 141 K.
Sawada et al. have theoretically demonstrated that LaVO$_3$ in the monoclinic phase has an OO with an alternative $d_{xy} d_{yz} / d_{zy} d_{zx}$ electron configuration (Fig. 1(a)). Here, we call this OO $G$-type by analogy to spin ordering. In YVO$_3$, which is attracting interest because of a temperature-induced magnetization reversal phenomenon, synchrotron X-ray diffraction, neutron and resonant X-ray scattering studies have confirmed the existence of two AF spin- and orbital-ordered states.

Between the first and second Néel temperatures ($T_{N1} = 115$ K $>$ $T > T_{N2} = 71$ K), this compound shows the $C$-type SO, and hence has been supposed to have SO and OO identical to those of the ground-state in LaVO$_3$ (Fig. 1(a)). Below $T_{N2}$, a distortion of octahedral VO$_6$ occurs due to the Jahn-Teller effect, and the SO pattern changes into the $G$-type with the antiferromagnetically arranged V$^{3+}$ spins in all three directions, while the OO turns into the $C$-type with the alternative $d_{xy} d_{yz} / d_{zy} d_{zx}$ electron configuration in the $ab$-plane and the identical one along the $c$-axis (Fig. 1(b)). The structures of the SO and OO in LaVO$_3$ and YVO$_3$ are thus highly anisotropic in spite of the almost cubic lattice structure. The SO and OO are expected to cause appreciable anisotropy in the electronic structure and charge dynamics through the spin-charge-orbital correlations. In this paper, we have investigated polarized optical conductivity spectra (electric field, $E \parallel c$-axis ($E \parallel c$) and $E \perp c$-axis ($E \perp c$)) in the perovskite-type LaVO$_3$ and YVO$_3$ single crystals, to clarify the anisotropic charge dynamics coupled with the SO and OO, as well as to probe the spin-orbital correlations.

Single crystals of LaVO$_3$ and YVO$_3$ were grown by a floating zone method in an atmosphere of Ar, as reported elsewhere. The melt-grown crystal of YVO$_3$ was confirmed to be a single-domain crystal by Laue reflection and single-crystal X-ray diffraction with a four-circle diffractometer. The results of the X-ray analyses indicated that the crystal of LaVO$_3$ was also detwinned in the $ac$- and $bc$-planes, although we did not confirm whether it was detwinned in the $ab$-plane because of the nearly identical lattice parameters for the $a$- and $b$-axes at room temperature. In the present experiment, this form of LaVO$_3$ crystal was sufficient for the optical reflectivity measurements, since the structures of the SO and OO can be viewed almost isotropic in the $ab$-plane. The crystals of both compounds with a (100) or (010) surface were polished to optical flatness with alumina powder. To remove the mechanical stress induced by surface polishing, we annealed the crystals at 1000 °C for 24 h in a flow of forming gas of Ar/H$_2$ (93/7%). Reflectivity measurements were carried out on the (100) or (010) surface between 0.06 and 40 eV. We used a Fourier-transform interferometer for the 0.06-0.8 eV region and grating spectrometers for the higher-energy region (0.6-40 eV). Measurements with varying temperature were performed between 0.06 and 5 eV. The synchrotron radiation source (UV-SOR) at the Institute for Molecular Science was utilized for spectroscopy above 5 eV at room temperature. Optical conductivity ($\sigma(\omega)$) spectra at various temperatures were obtained by Kramers-Kronig analysis of the respective reflectivity spectra combined with the room-temperature spectrum above 5 eV. For the analysis, we assumed a constant reflectivity below 0.06 eV and a $\omega^{-1}$-type extrapolation above 40 eV.
Figure 2 presents the $\sigma(\omega)$ spectra for $E \parallel c$ and $E \perp c$ in LaVO$_3$ and YVO$_3$ at various temperatures. In both vanadates, the $E \parallel c \sigma(\omega)$ spectra show large temperature ($T$) dependence, while the spectra for $E \perp c$ are almost independent of $T$. In LaVO$_3$, the spectral weight of the $\sigma(\omega)$ for $E \parallel c$ in a higher-energy region is rapidly transferred across the isosbestic (equal absorption) point at 2.9 eV to a lower-energy region with the decrease of $T$ around the magnetic and structural phase transition temperatures ($T_N$ and $T_S$). As a result of the spectral weight transfer, the peak intensity around 2 eV is markedly enhanced in the $C$-type spin- and $G$-type orbital-ordered state. In YVO$_3$, on the other hand, the spectral weight of the $c$-axis polarized $\sigma(\omega)$ is transferred from the region above 3.4 eV to a lower-energy region, and the peak intensity around 2 eV significantly increases with decreasing $T$ between $T_{N1}$ and $T_{N2}$. Moreover, the $\sigma(\omega)$ for $E \parallel c$ shows further accumulation of the spectral weight below 3.4 eV in the $G$-type spin- and $C$-type orbital-ordered state ($T < T_{N2}$).

In both compounds, the peak intensity around 2 eV in the $\sigma(\omega)$ is markedly enhanced only for $E \parallel c$ with decreasing $T$. The 2 eV band in the $\sigma(\omega)$ spectrum has been assigned to the Mott-Hubbard gap transition. In the AF phase, however, the peak around 2 eV in the $E \parallel c$ spectra shows a distinct shape, which has seldom been observed in other three-dimensional Mott-Hubbard insulators. Such an anisotropic and sharp spectral feature for the Mott-Hubbard gap transition reflects the anisotropic electronic structure due to the SO and OO with one-dimensional character, as argued in the following.

As seen in Fig. 2, the 2 eV band in the $\sigma(\omega)$ for $E \parallel c$ is composed of two peaks at 1.77 eV and 2.41 eV in LaVO$_3$, and at 1.88 eV and 2.37 eV in YVO$_3$. The $E \parallel c \sigma(\omega)$ spectra around 2 eV can be fitted with the Lorentz model, expressed by the following formula:

$$\sigma(\omega) = \sum_{i=1}^{3} f_i(\omega) = \sum_{i=1}^{3} \frac{S_i \omega_i^2 \gamma_i \omega^2}{(\omega_i^2 - \omega^2)^2 + \gamma_i^2 \omega^2}. \quad (0.1)$$

Here, the simulated spectra expressed by the functions ($f_i(\omega)$, $i = 1 - 3$) correspond to the lower-lying peak 1, the higher-lying peak 2 and the peak for the charge-transfer gap transition located above 4 eV. $\gamma_i$, $\omega_i$ and $S_i$ are width, excitation frequency and oscillator strength, respectively. The insets of Figs. 3 and 4 exemplify the results of the fitting at 10 K for the both vanadates. At the ground state (10 K), peak 1 bears much larger (smaller) intensity than peak 2 in LaVO$_3$ (YVO$_3$), as clearly seen in this spectral decomposition. In both vanadates, an electron can hop only between the $d_{yz}$ orbitals or the $d_{zx}$ ones on the neighboring V sites along the $c$-axis through the $\pi$-bonding with the O $2p_y$ or $2p_x$ state. The $d_{yz}$-$d_{yz}$ transition energy should be nearly equal to the $d_{zx}$-$d_{zx}$ one. In Figs. 1(c) and 1(d), we show the initial and possible final states of the $t_{2g}$ orbitals on the neighboring V sites along the $c$-axis in the optical Mott-Hubbard gap transitions in the respective cases of the SO and OO. In both cases, the $d_{zx}$ electron can be transferred to the $d_{zx}$ orbital on the nearest-neighbor V site along the $c$-axis. The energy of the $d_{zx}$-$d_{zx}$ excitation in the $C$-type SO and $G$-type OO is smaller than that in the $G$-type SO and $C$-type OO by an energy of the effective
Hund’s-rule coupling. Therefore, in the case of full polarization of the spin and orbital, the $\sigma(\omega)$ spectrum for $E_{\parallel c}$ would exhibit only one peak at $U' - K$ for the $C$-type SO and $G$-type OO, or otherwise at $U' + K$ for the $G$-type SO and $C$-type OO, where $U'$ and $K$ denote the energy of effective on-site Coulomb repulsion and Hund’s-rule coupling, respectively. Such orbital-dependent Mott-Hubbard gap transitions and their selection rules in the spin- and orbital-ordered state can explain heavily the unbalanced two-peak features in the respective ground-state (e.g., 10K) spectra of $\text{LaVO}_3$ and $\text{YVO}_3$.

Neutron scattering studies have revealed that the magnetic moments are approximately $1.3 \mu_B$ for $\text{LaVO}_3$, and $1.6 \mu_B$ below $T_{N2}$ and $1.0 \mu_B$ between $T_{N1}$ and $T_{N2}$ for $\text{YVO}_3$, while the pure spin value is $2 \mu_B$ for $S=\frac{1}{2}$. The excitation with the energy of $U' + K$ may be optically allowed by the imperfect polarization of the spin in the $C$-type SO even under the complete polarization of the orbital in the $G$-type OO. Moreover, the transition with the energy of $U' - K$ becomes weakly allowed in the $G$-type SO and $C$-type OO, if both orbital and spin show imperfect polarization. Therefore, the imperfect polarization of the spin and perhaps that of the orbital as well may relax the above-mentioned selection rule and produce the two-peak feature in the $E_{\parallel c} \sigma(\omega)$ spectra. The orthorhombic distortion in the actual crystal structure and/or the next-nearest-neighbor electron hopping should further relax the selection rule, although the large difference in the respective peak intensities should remain appreciable as observed. Thus assigning the two-peak feature to the orbital-dependent Mott-Hubbard transitions ($U' - K$ and $U' + K$), $U'$ and $K$ are estimated to be approximately 2.09 eV and 0.32 eV for $\text{LaVO}_3$, and 2.13 eV and 0.25 eV for $\text{YVO}_3$ on the basis of the localized electron picture. Incidentally, an electron can be transferred in the $ab$-plane not only between the $d_{xy}$ orbitals but also between the $d_{yz}$ or $d_{zx}$ ones on the neighboring V sites in both spin- and orbital-ordered states, and the energy of the excitations is $U' + K$. It is noted, however, that the $ab$-plane V-O bonds are longer than the out-of-plane ($c$-axis) ones even at high temperatures. Below $T_S$ and $T_{N2}$, the V-O bonds are further stretched due to the occupied $d_{yz}$ or $d_{zx}$ orbital, and the octahedral VO$_6$ units tilt (rotate) in the $ab$-plane. Consequently, the O 2p-V 3d transfer interaction is suppressed in the spin- and orbital-ordered state. This explains why the peak intensity in the $\sigma(\omega)$ for $E_{\perp c}$ remains low in the AF phase, in contrast to the $E_{\parallel c}$ case.

Figure 3 shows the $T$ dependence of the spectral weight ($N_{\text{eff},i}$) of each peak ($i = 1$ or 2) in $\text{LaVO}_3$ for $E_{\parallel c}$ that is defined as

$$N_{\text{eff},i} = \frac{2m_0}{\pi e^2 N} \int_0^\infty f_i(\omega) d\omega = \frac{m_0 S_i \omega_i^2}{e^2 N} \quad (i = 1, 2).$$

Here, $m_0$ is the free electron mass and $N$ the number of V atoms per unit volume. The spectral weight of peak 1 rapidly increases with decreasing $T$ below $T_N$ and $T_S$, while that of peak 2 is almost independent of $T$. As a result, the spectral weight of peak 1 is four times as large as that of peak 2 at the lowest $T$. The distinct enhancement of the spectral weight of peak 1 immediately
below \(T_N\) and \(T_S\) is consistent with the aforementioned SO/OO scenario and hence ensures the emergence of the \(C\)-type SO and \(G\)-type OO in LaVO\(_3\). Peak 1 shows a slight accumulation of the spectral weight even above \(T_N\) and \(T_S\) with the decrease of \(T\). The result implies the subsisting correlation of the \(C\)-type SO and \(G\)-type OO above \(T_N\) and \(T_S\). The correlation of the SO and OO is likely to cause the small anisotropy between the \(\sigma(\omega)\) spectra for \(E||c\) and \(E\perp c\) even around room temperature, as observed in Fig. 2(a).

In YVO\(_3\) (Fig. 4), the spectral weight of peak 2 is much larger than that of peak 1 at temperatures below \(T_{N2}\). This is quite consistent with the predicted low-\(T\) electronic state of YVO\(_3\), i.e., the \(G\)-type SO and \(C\)-type OO. In the intermediate region, \(T_{N2} < T < T_{N1}\), on the other hand, the \(T\) dependent behavior of the respective spectral weights shown in Fig. 4 cannot be explained as straightforwardly as expected from the proposed spin-orbital order structure, i.e., the \(C\)-type SO and \(G\)-type OO. The increase of the spectral weight for peak 1 below \(T_{N1}\) is not inconsistent with the simple prediction. However, much larger increase of the intensity of peak 2 is observed below \(T_{N1}\), and resultantly peak 2 bears a larger spectral weight than peak 1 in this intermediate \(T\) region.

The result suggests that the electronic state with the \(C\)-type SO realized in this intermediate \(T\) region considerably differs from the ground state of LaVO\(_3\) with a similar \(C\)-type SO. Since the relative intensity between peaks 1 and 2 is quite sensitive to the orbital order or correlation, the straightforward conclusion derived from the present observation is that even in the spin \(C\)-type state the \(C\)-type orbital correlation is present other (dominant rather) than the \(G\)-type correlation. As additional evidence of this, a recent neutron scattering study on the single-crystalline YVO\(_3\)\(^{24}\) has revealed that the SO at \(T_{N2} < T < T_{N1}\) is not simple \(C\)-type but appreciably affected by the \(G\)-type canting with a canting angle of \(\pm 10\) deg. Nevertheless, the hypothesis of the dominant \(C\)-type orbital correlation at \(T_{N2} < T < T_{N1}\) in YVO\(_3\) is not consistent with the results of the recent synchrotron X-ray diffraction studies,\(^{16}\) that clearly indicate the lattice modulation being consistent with the \(G\)-type OO in this \(T\) region.

As a possible scenario that may solve the apparent discrepancy, we may consider a role of the large orbital fluctuation.\(^{8}\) Khaliullin et al.\(^{8}\) have recently pointed out that the quantum fluctuation between the \(d_{yz}\) and \(d_{zx}\) orbitals (while \(d_{xy}\) being occupied on every V site) is possibly quite large and tends to stabilize the ferromagnetic interaction along the \(c\)-axis as realized in the spin \(C\)-type state. Thus, the orbital state in the \(C\)-type SO of YVO\(_3\) \((T_{N2} < T < T_{N1})\) might be better described by the orbital singlet correlation rather than by the simple \(G\)-type orbital order, in particular along the \(c\)-axis. This might allow the evolution of the \(G\)-type magnetic correlation that was observed to mix in the spin \(C\)-type order\(^{24}\) and the resultant \(C\)-type orbital correlation along the \(c\)-axis as observed in the present spectroscopic analysis. However, the spectral shape and the spectral weight ratio immediately above \(T_{N2}\) rather resemble those of the ground state, and such a significant feature remains puzzling and should be theoretically elucidated.
In summary, we have investigated the effect of the spin and orbital ordering (SO and OO) and their correlation on the optical gap transitions and their anisotropy. In LaVO$_3$ and YVO$_3$, the spectra for the E||c-axis show two peaks around 2 eV, which correspond to the orbital-dependent Mott-Hubbard gap transitions in the C-type SO and G-type OO, and those in the G-type SO and C-type OO, respectively. In the ground state of both vanadates, the spectral weight of the respective peaks and the anisotropy clearly reflect each type of the SO and OO.

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Fig. 1. (a, b) Schematic structures of the C-type spin ordering (SO) and G-type orbital ordering (OO), and the G-type SO and C-type OO in the perovskite-type LaVO$_3$ and YVO$_3$. Open arrows indicate spins and gray and black lobes indicate occupied $d_{yz}$ and $d_{zx}$ orbitals on the vanadium ions, respectively. The commonly occupied $d_{xy}$ orbitals are displaced for clarity. (c, d) Initial states (upper pictures) and possible final ones (lower ones) of $t_{2g}$ orbitals on the neighboring V sites (V$_1$ and V$_2$) along the c-axis in the optical $d_{xz}$-$d_{zx}$ transition in each type of SO and OO.

Fig. 2. Optical conductivity spectra for $E \parallel c$ and $E \perp c$ (black lines) in single crystals of (a) LaVO$_3$ and (b) YVO$_3$. For LaVO$_3$, red and blue lines indicate the spectra for $E \parallel c$ at temperatures (10-142 K) below the magnetic transition temperature ($T_N$), and at 150-293 K above $T_N$, respectively. For YVO$_3$, green, red and blue lines show the c-axis polarized spectra in the G-type spin- and C-type orbital-ordered state (at 10-67 K), in the C-type spin- and G-type orbital-ordered one (at 75-110 K) and in the paramagnetic one (at 120-293 K), respectively.

Fig. 3. Inset shows the fitting result with the Lorentz model for the c-axis polarized optical conductivity spectrum ($\sigma(\omega)$) at 10 K in LaVO$_3$. The lower- and higher-lying peaks are the Mott-Hubbard gap transitions, denoted “peak 1” and “peak 2”, respectively. The solid black and red, blue, violet and green broken lines indicate the experimental and fitted curves for peak 1, peak 2, the charge-transfer gap transition and total components, respectively. The main panel presents the temperature dependence of the spectral weight for peak 1 (solid circles) and peak 2 (open circles). (See text for definition.) The spectral weight of peak 1 rapidly increases with decreasing temperature below the magnetic and structural phase transition temperature ($T_N$ and $T_S$), i.e., in the C-type spin-ordered (C-type SO) and perhaps G-type orbital-ordered (G-type OO) state.
Fig. 4. Inset shows the fitting result with the Lorentz model for the c-axis polarized optical conductivity spectrum ($\sigma(\omega)$) at 10 K in YVO$_3$. The lower- and higher-lying peaks are the Mott-Hubbard gap transitions, denoted “peak 1” and “peak 2”. The solid black and red, blue, violet and green broken lines indicate the experimental and fitted curves for peak 1, peak 2, the charge-transfer gap transition and total components, respectively. The main panel presents the temperature dependence of the spectral weight for peak 1 (solid circles) and peak 2 (open circles). (See text for definition.) The spectral weight of peak 2 is much larger than that of peak 1 in the G-type spin- and C-type orbital-ordered state (G-SO and C-OO) below $T_{N2}$. 
Figure 1  S. Miyasaka et al.
Figure 2  S. Miyasaka et al.
LaVO$_3$ (E||c-axis)

Spectral weight

Temperature (K)

peak 1

C-type SO
(G-type OO)

peak 2

$T_S, T_N$

Figure 3  S. Miyasaka et al.
Figure 4  S.Miyasaka et al.