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Probing orbital fluctuations in $R\mathrm{VO}_3$ (R = Y, Gd, or Ce) by ellipsometry

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We study optical excitations across the Mott gap in the multiorbital Mott-Hubbard insulators $R\mathrm{VO}_3$. The multipeak structure observed in the optical conductivity can be described consistently in terms of the different $3d^3$ multiplets or upper Hubbard bands. The spectral weight is very sensitive to nearest-neighbor spin-spin and orbital-orbital correlations and thus shows a pronounced dependence on both temperature and polarization. Comparison with theoretical predictions based on either rigid orbital order or strong orbital fluctuations clearly rules out that orbital fluctuations are strong in $R\mathrm{VO}_3$. Both the line shape and the temperature dependence give clear evidence for the importance of excitonic effects.

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I. INTRODUCTION

Orbitals play a decisive role in the low-energy physics of a large variety of transition-metal oxides with strong electronic correlations.[1,2] The orbital occupation is crucial for, e.g., the metal-insulator transition in $\mathrm{V}_2\mathrm{O}_3$ (Ref. 3) and governs both the size and the sign of the exchange coupling between spins, paving the way for, e.g., low-dimensional quantum magnetism. In Mott-Hubbard insulators, orbital degeneracy gives rise to more exotic phases such as orbital liquids.[4] However, the orbital degeneracy typically is lifted by the crystal field, opening a gap for orbital excitations of a few hundred milli-electron volts or larger, strongly suppressing orbital fluctuations. The central task in the field of orbital physics still is to establish compounds in which the crystal field is *not* dominant. This can be achieved by, e.g., strong spin-orbit coupling such as in the $5d$ iridates[5] in which anisotropic exchange interactions may yield a realization of the Heisenberg-Kitaev model with exotic phases and excitations.[6] In $3d$ systems, superexchange interactions may dominate over the crystal-field splitting. Different groups have pointed out that orbital fluctuations may be strong in the orbitally ordered Mott-Hubbard insulators $R\mathrm{VO}_3$ (R = Y, rare earth), claiming for YVO_3 the observation of a one-dimensional orbital liquid, of an orbital Peierls phase, and of two-orbiton excitations.[7–14] Studies based on LDA + U and LDA + DMFT[15,16] (local density approximation + dynamical mean-field theory) find that orbital fluctuations are suppressed in YVO_3 by a sizable crystal-field splitting but support strong orbital fluctuations for larger R ions such as in LaVO_3.

The experimental determination of orbital fluctuations is a difficult task. Our approach involves the analysis of the optical conductivity $\sigma(\omega)$. Optical excitations across the Mott gap invoke microscopic hopping processes between adjacent sites. Thus, the spectral weights (SWs) of these excitations depend sensitively on nearest-neighbor spin-spin and orbital-orbital correlations and may show pronounced dependence on both polarization and temperature T.[10,11,17–22] For the spin-spin correlations, this has been demonstrated in the $3d^4$ manganites LaMnO_3 and $\mathrm{LaSrMnO}_4$.[17,18] There, the orbital occupation is independent of T due to the large crystal-field splitting of the e_g orbitals,[18] and the spin-spin correlations govern the spectral weight. For $R\mathrm{VO}_3$ it has been predicted that orbital fluctuations have a strong impact on the T dependence of $\sigma(\omega)$,[10] but the experimental data are still controversial. In LaVO_3, the spectral weight of the lowest peak in $\sigma_1(\omega)$ at 1.9 eV for polarization $E\|c$ shows a pronounced T dependence, which has been taken as strong evidence for orbital fluctuations.[10,20] However, the proposed multiplet assignment of the peaks fails to describe the T dependence of the higher-lying peaks. In addition, there is a striking disagreement concerning the data reported for the sister compound YVO_3 (Refs. 20,21, and 23) as none of the different data sets is in agreement with the theoretical predictions.[10,11,22]

Here, we report on a detailed analysis of the optical conductivity $\sigma(\omega)$ of YVO_3, GdVO_3, and CeVO_3 in the frequency range from 0.8 to 5.0 eV for temperatures from 20 K to 500 K. Our results clarify the striking discrepancies of the data reported in Refs. 20,21,23 for YVO_3. We derive a consistent description of the observed absorption bands in terms of the different upper Hubbard bands of these multiorbital compounds. The temperature and polarization dependences of the spectral weights are in excellent agreement with predictions for rigid orbital order. We firmly conclude that orbital fluctuations are only weak in $R\mathrm{VO}_3$ and propose that the lowest peak is caused by excitonic effects.

II. EXPERIMENT

Single crystals of $R\mathrm{VO}_3$ with R = Y, Gd, and Ce were grown by the traveling-solvent floating-zone method.[24] The purities, stoichiometries, and single-phase structures of the crystals were checked by x-ray diffraction and thermogravimetry. Ellipsometric data were obtained using a rotating-analyzer ellipsometer (Woollam VASE) equipped with a retarder between the polarizer and sample. The complex optical conductivity $\sigma_1^j(\omega) + i\sigma_2^j(\omega)$ for $j \in \{a,c\}$ was derived from a series of measurements with different orientations of a polished ac surface.

III. ORBITAL ORDER

At room temperature, $R\mathrm{VO}_3$ exhibits an orthorhombic crystal structure ($Pbnm$).[24–27] A phase transition to a monoclinic phase ($P2_1/b$) is observed at $T_{\infty} \approx 200$ K (R = Y), 208 K

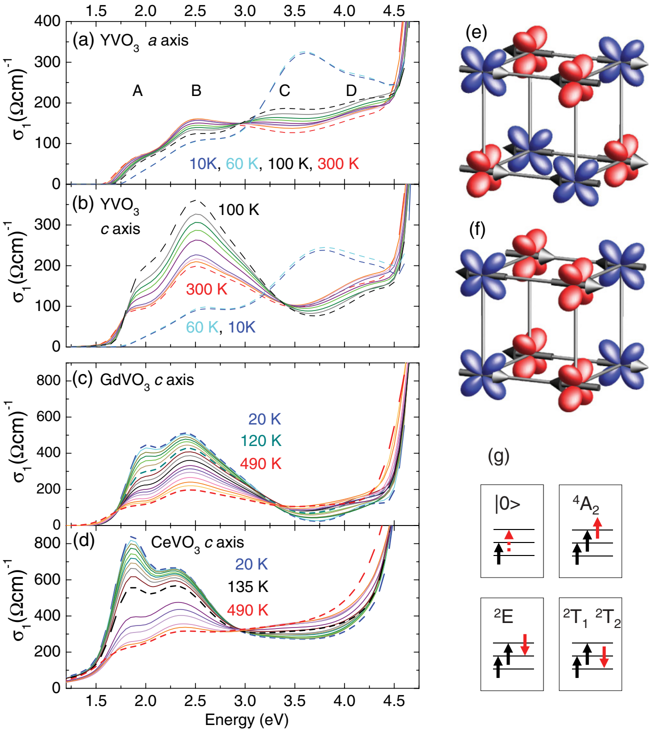

FIG. 1. (Color online) (a) Optical conductivity $\sigma_1^a(\omega)$ of YVO$_3$. (b)–(d) $\sigma_1^c(\omega)$ of YVO$_3$, GdVO$_3$, and CeVO$_3$, respectively. Temperature labels refer to the dashed lines. The absorption bands are labeled as peaks A (≈2 eV), B (≈2.5 eV), C (3.0–3.6 eV), and D (3.8–4.4 eV). (e) G-type OO with C-type SO. (f) C-type OO with G-type SO. In both, (e) and (f), the vertical axis corresponds to the c axis. (g) Sketch of t_{2g}^2 and t_{2g}^3 multiplets.

(Gd), and 154 K (Ce), and antiferromagnetic order sets in at $T_N \approx 116$ K (Y), 122 K (Gd), and 134 K (Ce).[24,26,28–30] YVO$_3$ shows a further phase transition at $T_S = 77$ K to a low-temperature orthorhombic phase,[24,25] which is absent for R = Gd and Ce. We use the same set of axes at all temperatures and neglect the small monoclinic distortion.[24] The undoped Mott-Hubbard insulators RVO$_3$ have two electrons in the 3d shell per V site. In the ground state, both electrons occupy t_{2g} orbitals with total spin 1. The t_{2g} manifold is split into d_{xy}, d_{xz}, and d_{yz} orbitals by the crystal field, and the total splitting is on the order of 0.1–0.2 eV.[14,16,31,32] In the orbitally ordered phases, the d_{xy} orbital is occupied by one electron at each V site. The occupation of d_{xz} and d_{yz} by the second electron can be viewed as a pseudospin, and both spins and pseudospins have been reported to show ordering patterns of either the G type (antiferro along all bonds, i.e., xz and yz alternate) or the C type (ferro along c, antiferro within the ab plane) [see Figs. 1(e) and 1(f)]. In YVO$_3$, one finds G-type spin order (SO) and C-type orbital order (OO) below $T_S = 77$ K [see Fig. 1(f)].[16,21,24,25,28,30,33] In the monoclinic phase, all compounds show C-type SO below T_N while the correct description of the orbitals is controversial (see the discussion in Ref. 14). Both, G-type orbital order[24,33] [see Fig. 1(e)] and strong orbital fluctuations[7–13] have been claimed. Here, we show that the optical data rule out strong orbital fluctuations.

IV. RESULTS

A. Optical conductivity and multiplet assignment

The overall behavior of $\sigma_1(\omega)$ is very similar for YVO$_3$, GdVO$_3$, and CeVO$_3$ [see Figs. 1(a)–1(d)]. The main difference is that YVO$_3$ shows the low-temperature phase with C-type OO below $T_S = 77$ K with a pronounced peak at 3.5 eV. In all three compounds, the Mott gap is about 1.6 to 1.8 eV in excellent agreement with infrared-transmittance data.[14] Above the gap we observe Mott-Hubbard (MH) excitations, i.e., excitations from a $|d_i^2 d_j^2\rangle$ ground state to a $|d_i^1 d_j^3\rangle$ final state where i and j denote different V sites and d^1 and d^3 refer to the lower and upper Hubbard bands, respectively. Above 4.5 eV, $\sigma_1(\omega)$ steeply rises up to roughly 1500 $(\Omega\,\mathrm{cm})^{-1}$ (not shown), corresponding to the onset of charge-transfer excitations from the O 2p band to the upper Hubbard band, $|d^2 p^6\rangle \rightarrow |d^3 p^5\rangle$. This general picture is well accepted.[20–23,34] The MH excitations show a multipeak structure (peaks A–D at roughly 2 eV, 2.5 eV, 3–3.6 eV, and 3.8–4.4 eV, respectively), which is expected to reflect the local d^3 multiplet structure of these multiorbital systems.[10,11,20–22] The splitting between the t_{2g} level and the e_g level amounts to 10 Dq = 1.9 eV in YVO$_3$.[35] For a discussion of the lowest excited states, we thus may neglect the e_g orbitals. For the sake of simplicity, we assume cubic symmetry and neglect the crystal-field splitting within the t_{2g} levels of roughly 0.1–0.2 eV.[14,16,31,32] In the ground state, the t_{2g}^2 configuration shows $^3T_1$ symmetry with spin 1. The excited states $|t_{2g}^1 t_{2g}^3\rangle$ have to be distinguished according to the t_{2g}^3 sector because the t_{2g}^1 configuration always has the same energy. The t_{2g}^3 multiplets exhibit energies of $U - 3J_H$ ($^4A_2$), U (^2E, $^2T_1$), and $U + 2J_H$ ($^2T_2$) [see Fig. 1(g)][10,11,21] with the on-site Coulomb repulsion $U \approx$ 4–5 eV (Refs. 16 and 36) and the Hund coupling $J_H \approx 0.55$–0.7 eV.[14]

We now focus on a consistent assignment of the MH excitations to the different t_{2g}^3 multiplets. The spectral weight of a given excitation depends on the spin-spin and orbital-orbital correlations between adjacent sites and thus depends strongly on polarization and temperature. The lowest multiplet $^4A_2$ is a high-spin state in which the xy, xz, and yz orbitals are occupied by one electron each [see Fig. 1(g)]. Due to the high-spin character, parallel spins on adjacent sites in the initial state give rise to a larger SW than antiparallel spins. In contrast, the other t_{2g}^3 multiplets, ^2E, $^2T_1$, and $^2T_2$, all are low-spin states; thus, the SW is larger for *antiparallel* spins. This yields the following clear predictions for the phase with C-type SO[10,11,22] in which spins are parallel along the c axis and antiparallel within the ab plane [see Fig. 1(e)]. (1) The SW of the excitation into the lowest multiplet $^4A_2$ is expected to be larger in σ_1^c than in σ_1^a. (2) With decreasing temperature T, spin-spin and orbital-orbital correlations are enhanced; thus, σ_1^c (σ_1^a) is expected to increase (decrease) for the lowest multiplet. (3) The *opposite* T dependence is expected for the higher multiplets. A comparison of these predictions with our data clearly shows that *both* peaks A and B at 2.0 and 2.5 eV, respectively, have to be assigned to the lowest $3d^3$ multiplet $^4A_2$. Peak C is located roughly $3J_H$ above peak A in σ_1^a in agreement with the expectations for the (^2E, $^2T_1$) multiplets. The SW of this excitation vanishes for parallel spins,[10,11,22] and therefore, it is absent in σ_1^c in the phase with C-type SO.

The dramatic changes observed at $T_S = 77$ K in YVO$_3$ unambiguously prove that our peak assignment is correct. At T_S, the nearest-neighbor correlations along c change from ferro to antiferro for the spins and vice versa for the orbitals; thus, two adjacent sites show the same orbital occupation below T_S with, e.g., xz occupied on both sites. In this case, an excitation to $^4A_2$ requires hopping from xz on one site to yz on a neighboring site, which is forbidden along c in cubic symmetry, explaining the spectacular suppression of peaks A and B. The finite SW at low T is due to deviations from cubic symmetry.[22] At the same time, the transition to the $^2T_1$ multiplet (contributing to peak C) strongly favors G-type SO and C-type OO, explaining the dramatic increase of peak C below 77 K.

The highest t_{2g}^3 multiplet $^2T_2$ is roughly expected at $U + 2J_H$, i.e., $5J_H > 2.7$ eV above the lowest peak. It is thus reasonable to assume that this excitation is located above the onset of charge-transfer excitations at 4.5 eV. Peak D at 3.8–4.4 eV lies about 10 Dq = 1.9 eV (Ref. 35) above peaks A and B and thus can be assigned to the lowest $t_{2g}^2e_g^1$ multiplet. In cubic symmetry, the excitation from a t_{2g} orbital on site i to an e_g orbital on a neighboring site is forbidden, but deviations from cubic symmetry yield a finite spectral weight. Accordingly, peak D is hardly visible in less-distorted CeVO$_3$.

B. Comparison with literature

Conflicting with our assignment, peaks A and B have been attributed to the two lowest multiplets $^4A_2$ and (^2E, $^2T_1$) in Refs. 20–23. Between these multiplets, a splitting of $3J_H > 1.5$ eV is expected, which is incompatible with the observed splitting between A and B of only 0.5 eV. In other words, the previous assignment of peaks A and B to two different multiplets yields a nonphysically small value of J_H.[22] Moreover, this scenario is inconsistent with the fact that the SWs of peaks A and B show the same T dependence (see Sec. IV A). Additionally, the data for YVO$_3$ are strikingly different.[20,21] In Ref. 21, the pronounced peak at 3.5 eV, characteristic of the low-temperature phase, is also observed above T_S whereas it is not seen at any temperature in Ref. 20. Both the incorrect assignment and the discrepancies of the data can be traced back to problems with the sample temperature. Samples of YVO$_3$ tend to break at the first-order structural transition at T_S, often leading to a loss of thermal contact.[23] We were able to avoid this problem by very slow cooling. A comparison of our data and the data of Ref. 23 shows that the seemingly contradictory data of Refs. 20 and 21 can be reconciled with each other by taking into account problems with the sample temperature across T_S. The data of Ref. 23 show the expected jump of the spectral weight at T_S, but both this jump and the T dependence above T_S are much smaller than in our data. We attribute this difference to the different experimental techniques. Reference 23 reports reflectivity data with a subsequent Kramers-Kronig analysis. In contrast, ellipsometry is a self-normalizing technique which is much better suited for a precise determination of the T dependence.[17,37]

C. Temperature dependence of the spectral weight: strength of orbital fluctuations

The correct assignment of peaks A and B to the lowest multiplet $^4A_2$ is crucial for the discussion of the role of orbital fluctuations. The spectral weight of the $^4A_2$ multiplet in σ_1^c depends sensitively on spin-spin and orbital-orbital correlations between adjacent sites.[10,11,22] Comparing a fully polarized ferromagnetic state ($T = 0$) with a disordered spin state ($T = \infty$), the SW in the latter case is reduced to 2/3 based on the nearest-neighbor hopping matrix element. The change from a fully ordered orbital state to a disordered one yields another factor of 1/2. Thus, in total one expects a reduction of the SW by a factor of 3 from low T to high T.[10,11] This is valid in any scenario, i.e., it applies to both rigid orbital order and strong orbital fluctuations. In order to determine the strength of orbital fluctuations, one has to study the detailed T dependence of the SW, which thus is the most interesting quantity. The inset of Fig. 2 shows the predictions for both strong orbital fluctuations (blue line, assuming $T_{\rm OO} = T_N$) and rigid orbital order (red line, assuming $T_{\rm OO} = \infty$). A near coincidence of the ordering temperatures $T_{\rm OO}$ and T_N for orbitals and spins is realized in CeVO$_3$. The red line for rigid orbital order shows only the reduction by a factor of 2, stemming from the spin part, because it assumes $T_{\rm OO} = \infty$. The key feature of this comparison is not the difference in absolute value but the T dependence above the ordering temperature. For rigid orbital order, the SW is nearly constant for $T > T_N$ and exhibits a clear kink right at T_N. In contrast, there is no pronounced effect at T_N for strong quantum fluctuations. The smoking gun for strong fluctuations is a strong T dependence far above T_N or $T_{\rm OO}$; most of the change occurs above the ordering temperature.

In Fig. 2, these predictions are compared with our results. The SW of a single absorption band is given by $N_{\rm eff} = (2mV/\pi e^2)\int_0^\infty \sigma_1(\omega)d\omega$, where m is the free-electron mass and V is the volume per magnetic ion. We used four Lorentz oscillators to describe peaks A–D. The total SW of peaks A and B is shown in Fig. 2. For all compounds we find nearly

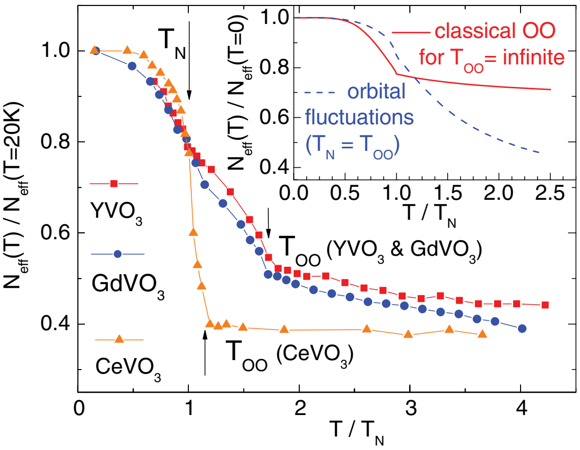

FIG. 2. (Color online) Sum of the spectral weights of peaks A and B in σ_1^c. Inset: Theoretical results for $N_{\rm eff}(T)$ of the lowest Mott-Hubbard excitation ($^4A_2$ multiplet) in σ_1^c for strong orbital fluctuations (blue, $T_N = T_{\rm OO}$) and rigid orbital order [red, $T_{\rm OO} = \infty$, i.e., only the reduction of the spin part to 2/3 is taken into account (see Refs. 10 and 11)].

constant spectral weights above T_{OO}, a clear kink at T_{OO}, and also a kink at T_N. These findings are in excellent agreement with the expectations for rigid orbital order. The fact that the changes above T_{OO} are much smaller than below rules out strong orbital fluctuations. Also, the total change of the SW is in excellent agreement with theory.

The claim that orbital quantum fluctuations are strong in pseudocubic $R\mathrm{VO}_3$ is based on the idea that superexchange interactions between t_{2g} orbitals are frustrated on a cubic lattice.[7] More precisely, orbital quantum fluctuations are blocked in the ab plane due to the occupied d_{xy} orbital, but they have been claimed to be strong along the c axis in the monoclinic phase where orbital fluctuations and ferromagnetic spin order may support each other.[7] However, distortions away from cubic symmetry give rise to both a crystal-field splitting of the t_{2g} orbitals and a reduction of superexchange interactions. The orthorhombic splitting $\varepsilon = (b-a)/(b+a)$ between the lattice parameters a and b amounts to 0.03 for $R =$ Y and Gd and only 0.003 for Ce[25,27,38] while the V-O-V bond angle increases from about 144° in YVO_3 to about 156° in LaVO_3.[24,27,39] Our results clearly show that orbital fluctuations are suppressed not only in strongly distorted YVO_3 but also for large R ions such as in pseudocubic CeVO_3.

D. Hubbard exciton

Finally, we address the double-peak structure A/B of the excitation into the lowest multiplet. Similar double- and multipeak structures of the lowest multiplet have been reported for YTiO_3 and LaMnO_3.[17,40] The peak splitting has been assigned to either excitonic or band-structure effects, which both have been neglected thus far in our discussion of local multiplets. We propose that peak A is an excitonic resonance, i.e., not a truly bound state below the gap but a resonance within the absorption band. Due to an attractive interaction between a d^3 state in the upper Hubbard band and a d^1 state in the lower Hubbard band, the energy of the resonance (peak A) is less than the energy of peak B where peak B reflects an excitation to $|d^1 d^3\rangle$ without attractive interaction. In order to substantiate this claim, we discuss results from photoemission spectroscopy (PES)[41,42] and from band-structure calculations.[16,22,34]

Using LDA + U, Fang *et al.*[22] calculated the optical conductivity of LaVO_3 and YVO_3 for different polarizations and for different ordering patterns of spins and orbitals. The two lowest peaks (called α and β in Ref. 22) correspond to the two lowest multiplets of our local approach, i.e., to the double peak A/B and peak C, respectively. Accordingly, the spectral weight of peak α (β) in σ_1^c decreases (increases) in YVO_3 across the phase transition from the intermediate phase with C-type SO to the low-T phase with G-type SO as observed experimentally for peak A/B (C). Peak α is the lowest peak, well separated from the higher-lying excitations, and clearly consists of a single peak only, both for YVO_3 and LaVO_3.[22] The experimentally observed splitting into peaks A and B is absent in the LDA + U results, which neglect excitonic effects. For the intermediate phase of YVO_3, Fang *et al.*[22] predict peaks α and β (with β observable for $E \perp c$ only) at about 1.7 and 2.9 eV, respectively, both significantly lower than peaks B and C in experiment, but in the LDA + U results, the peak energies depend sensitively on the particular choice of U. Considering only the energies, one may be tempted to assign peaks α and β to peaks A and B, but this is clearly ruled out by the dependence of the spectral weight on both temperature and polarization as well as by the value of J_H as discussed in Sec. IV A. Also the LDA + U study of Solovyev *et al.*[34] reports on the optical conductivity of LaVO_3. In agreement with the results of Fang *et al.*,[22] there is no splitting of the lowest excitation. Solovyev *et al.*[34] find the band gap at 0.7 eV and the charge-transfer gap at about 3.5 eV while the lowest absorption band is peaking at about 1.7 eV. Since both gaps are about 0.7–1 eV lower than in experiment, we assign the lowest peak from LDA + U at 1.7 eV to peak B in our data. As mentioned above, the peak energy depends sensitively on the choice of U. The LDA + DMFT study of De Raychaudhury *et al.*[16] does not report on the optical conductivity, but it shows the electron-removal and -addition spectra (as measured by PES and inverse PES) for LaVO_3 and YVO_3. For LaVO_3, the electron-removal spectrum shows contributions from all three t_{2g} orbitals, peaking at about 1.2–1.4 eV below the Fermi energy E_F.[16] The small splitting reflects the crystal-field splitting within the t_{2g} level. The first peak of the electron-addition spectrum lies at about 1.2 eV above E_F. Neglecting excitonic effects, one may thus expect the first peak in the optical conductivity at about 2.4–2.6 eV, which is in agreement with peak B. For YVO_3, the electron-removal and -addition spectra peak at about −1.4 to −1.5 eV and + 1.2 eV, respectively;[16] thus, the peak in the optical conductivity is expected at a slightly larger energy in YVO_3 than in LaVO_3, which is in agreement with our experiment. The calculated electron-removal and -addition spectra for YVO_3 show small shoulders at about −1.1 and +0.7 eV. However, similar features are absent in the calculated spectra of LaVO_3. In strong contrast, peak A in the optical conductivity is much more pronounced in LaVO_3 than in YVO_3. In summary, band-structure calculations do not provide any explanation for the observed splitting of about 0.5 eV between peaks A and B.

Experimental photoemission spectra of LaVO_3 and YVO_3 show a single peak lying about 1.5–1.8 eV below E_F.[41,42] For LaVO_3, the combination of PES and inverse PES has been reported by Maiti and Sarma.[41] The separation between the highest peak below E_F and the lowest peak above E_F amounts to roughly 3 eV, but the inverse-PES data were measured with a resolution of only 0.8 eV. These results are in agreement with the LDA + DMFT study of Ref. 16 discussed above. Neither band-structure calculations nor the PES data provide any explanation for the splitting of peaks A and B. Electron-removal and -addition spectra do not reflect excitonic effects in contrast to the optical conductivity. Altogether, this strongly supports an excitonic interpretation of peak A.

In simple band insulators, exciton formation is driven by a lowering of the Coulomb energy whereas the kinetic energy increases. The term *Hubbard exciton* refers to an exciton in a Mott-Hubbard insulator. Such Hubbard excitons are of particular interest because of the possible role of the *kinetic* energy for the attractive interaction.[43–47] In $R\mathrm{VO}_3$, the ratio $\mathrm{SW_A}/\mathrm{SW_B}$ of the spectral weights of peaks A and B in σ_1^c strongly increases from $R =$ Y via Gd to Ce. Interestingly, this ratio also depends sensitively on the temperature, but only below the orbital-ordering temperature T_{OO} (see Fig. 3).

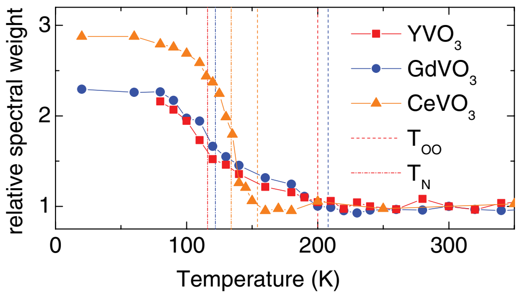

FIG. 3. (Color online) Ratio of the spectral weights of peaks A and B in σ_1^c, normalized to the value at 300 K.

Below T_{OO}, the SWs of both peaks A and B increase, but this increase is much more pronounced for the excitonic peak A. This clearly demonstrates the significant role of orbital order for exciton formation in Mott-Hubbard insulators. We propose that this influence of orbital order indicates the importance of the kinetic energy for Hubbard excitons in the case of antiferro-orbital order (here, along c) as discussed for YTiO_3.[40] Along c, the motion of an exciton is not hindered by the antiferro-orbital order whereas the hopping of either a single d^3 state or a single d^1 state leaves a trace of orbitally excited states (see Ref. 40 for a more detailed discussion). This results from the restriction that hopping is essentially only allowed within the same type of orbital, e.g., from xz on one site to xz on a neighboring site. Therefore, the exciton can hop on a larger energy scale than a single d^1 state or a single d^3 state, and exciton formation is equivalent to a *gain of kinetic energy*.[40] This scenario is supported by recent pump-probe measurements on YVO_3, which cover the frequency range of peaks A and B.[48]

V. SUMMARY AND CONCLUSIONS

In summary, we provide a consistent assignment of the Mott-Hubbard excitations and a quantitatively reliable T dependence of the spectral weights of YVO_3, GdVO_3, and CeVO_3. A comparison of our data with theoretical results[10,11] clearly rules out strong orbital fluctuations in $R\mathrm{VO}_3$. We propose that the line shape and the T dependence of the lowest absorption band reflect excitonic effects.

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