Metal-insulator transition and lattice instability of paramagnetic V_2O_3

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We determine the electronic structure and phase stability of paramagnetic V_2O_3 at the Mott-Hubbard metal-insulator transition (MIT) by employing a combination of an *ab initio* method for calculating band structures with dynamical mean-field theory. The structural transformation associated with the MIT occurs upon a slight expansion of the lattice volume by ∼1.5%, in agreement with experiment. Our results show that the structural transition precedes the MIT, implying a complex interplay between electronic and lattice degrees of freedom. The MIT is found to be driven by a strong correlation-induced, orbital-selective renormalization of the V t_{2g} bands. The effective electron mass of the e_g^π orbitals diverges at the MIT. Our results show that full charge self-consistency is crucial for a correct description of the physical properties of V_2O_3.

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I. INTRODUCTION

The question concerning the nature of a Mott metal-insulator phase transition (MIT) poses one of the most fundamental problems in condensed-matter physics. It becomes an even greater challenge in the case of materials with a strong interplay between electronic correlations and lattice degrees of freedom. The most famous example is V_2O_3, which has generally been considered to be the classical example of a material with a Mott-Hubbard MIT [1]. Under ambient conditions, V_2O_3 is a paramagnetic metal with a corundum crystal structure (space group $R\bar{3}c$) [2,3]. Upon doping with Cr it undergoes a phase transition to a paramagnetic Mott-Hubbard insulator without change of crystal symmetry [4]. The transition from the paramagnetic insulator (PI) to the paramagnetic metal (PM) can also be triggered by doping with Cr, which essentially amounts to the application of a negative pressure. The MIT is intimately linked with an abrupt expansion of the lattice volume by $\frac{\Delta V}{V} \sim 1.3\%$ and a reduction of the c/a lattice parameter ratio by ∼1.4% across the MIT, indicating a strong coupling between electronic and lattice degrees of freedom [4]. In spite of intensive research over several decades, an explanation of this mutual influence of electronic structure and phase stability of paramagnetic V_2O_3 at the MIT is still missing. Therefore investigations of V_2O_3 continue with a high level of activity [5,6].

A realistic description of V_2O_3 requires taking into account the interplay of electronic correlations and lattice degrees of freedom on a microscopic level. State-of-the-art methods for the calculation of the band structure, which allow for *ab initio* computations of the electronic and structural properties, can account for neither the MIT nor the paramagnetic insulating phase of V_2O_3 [7]. This obstacle can be overcome by employing the LDA/GGA+DMFT approach [8], a combination of band-structure methods in the local-density approximation (LDA) or the generalized gradient approximation (GGA) with dynamical mean-field theory (DMFT) of strongly correlated electrons [9]. Applications of LDA+DMFT have already provided a good quantitative description of the electronic structure and spectral properties of V_2O_3 [6,10–15]. In particular, it was found that in all phases the V^{3+} ions are in a $S = 1$ spin configuration, with a mixed (a_{1g}, e_g^π) and (e_g^π, e_g^π) orbital occupation. These calculations also allow one to perform a direct comparison of the calculated spectra with, e.g., photoemission, x-ray absorption, and optical conductivity measurements near the metal-insulator transition [6,10–15]. The LDA+DMFT results capture all generic aspects of a Mott-Hubbard metal-insulator transition, such as a coherent quasiparticle behavior, formation of the lower and upper Hubbard bands, and strong renormalization of the effective electron mass. In addition, these calculations reveal an orbital-selective behavior of the electron coherence and a strong enhancement of the crystal-field splitting between the a_{1g} and e_g^π bands, caused by electron correlations at the MIT [10,12]. Despite this success the coupling between electronic correlations and lattice structure at the Mott-Hubbard MIT in V_2O_3 is still poorly understood. We will address this problem in our investigation and thereby shed new light on the long-standing question regarding the origin of the structural changes in the vicinity of the MIT.

II. METHOD

In this paper we employ the GGA+DMFT computational approach [16] to explore the electronic and structural properties of paramagnetic V_2O_3 across the Mott-Hubbard MIT. In particular, we will explore the structural phase stability of paramagnetic V_2O_3, i.e., the influence of electronic correlations on the *structural* transition. We first compute the electronic structure of paramagnetic V_2O_3 within the nonmagnetic GGA using the plane-wave pseudopotential approach [17]. To investigate the structural stability, we use the atomic positions and the c/a lattice parameter ratio of V_2O_3 taken from experiment [4]. To this end, we adopt the crystal structure data for paramagnetic metallic V_2O_3 and insulating $(V_{0.962}Cr_{0.038})_2O_3$, respectively, and calculate the total energy as a function of volume. In Fig. 1 (inset) we show the results of the nonmagnetic GGA total-energy calculations, which agree with previous band-structure results [7]. In particular, we find

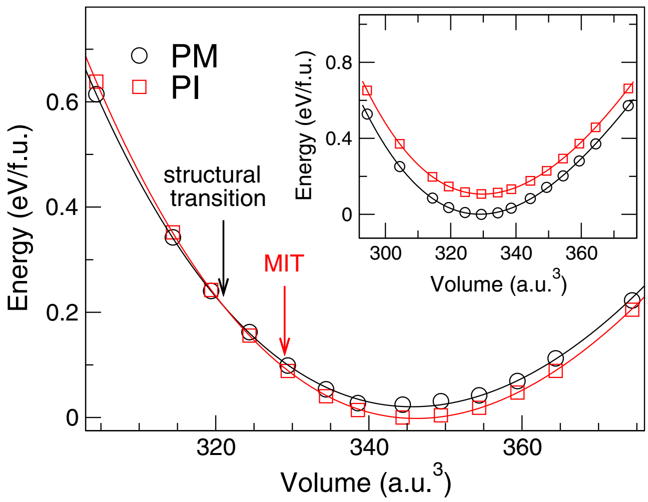

FIG. 1. (Color online) Variation of the total energy of paramagnetic V_2O_3 computed within GGA+DMFT for different volumes. The inset shows the results of the nonmagnetic GGA total-energy calculations.

equilibrium lattice constants $a = 4.923$ Å for the PM phase and 4.955 Å for the PI phase of V_2O_3. The calculated bulk moduli are $B \sim 252$ GPa for both phases. These results are in neither quantitative nor even qualitative agreement with experiment. Namely, they give a metallic solution with no structural phase transition between the PM and PI phases. Clearly, standard band-structure techniques cannot explain the properties of paramagnetic V_2O_3 since they do not treat electronic correlations adequately.

III. RESULTS AND DISCUSSION

A. Non-self-consistent calculations

Therefore we now compute the electronic structure and phase stability of V_2O_3 using the GGA+DMFT computational scheme [16]. For the partially filled V t_{2g} orbitals, which split into a_{1g} and e_g^π bands due to a trigonal distortion, we construct a basis of atomic-centered symmetry-constrained Wannier functions [18]. To solve the realistic many-body problem within DMFT, we employ the continuous-time hybridization-expansion quantum Monte Carlo algorithm [19,20]. The calculations are performed at the temperature $T \sim 390$ K, which is below the critical end point of $T_c \sim 458$ K [21]. We use values of the Coulomb interaction $U = 5$ eV and Hund's exchange $J = 0.93$ eV, in accordance with the previous theoretical and experimental estimations [6,10–12]. The U and J values are assumed to remain constant across the phase transition, which is consistent with recent hard x-ray photoemission experiments [22]. Furthermore we employ the fully localized double-counting correction, calculated from the self-consistently determined local occupancies, to account for the electronic interactions already described by the GGA.

In Fig. 1 (main panel) we display the calculated variation of the total energy of paramagnetic V_2O_3 as a function of lattice volume. Our results for the equilibrium lattice constant and bulk modulus, which now include the effect of electronic correlations, agree well with experiment [4]. The calculated equilibrium lattice constants are $a = 5.005$ and 5.035 Å for the PM and PI phases, respectively. The corresponding bulk moduli are $B = 202$ and 222 GPa. In agreement with previous studies [10,12], our results show an orbital dependence of the electron coherence, with coherent quasiparticle behavior for the a_{1g} orbital, while the e_g^π bands remain incoherent, with a large imaginary part of the self-energy at $T \sim 390$ K. We also evaluated the spectral function of paramagnetic V_2O_3 (not shown here) and determined the MIT phase boundary. The MIT is found to take place at a positive pressure of $p_{el} \sim 125$ kbar, implying a $\frac{\Delta V}{V} \sim 5\%$ reduction of the lattice volume. We find that both the a_{1g} and the e_g^π quasiparticle weights remain finite at the MIT; that is, there is *no* divergence of the effective electron mass at the transition as it would occur in a Brinkman-Rice picture of the MIT [23]. Thus we conclude that the MIT is driven by a strong enhancement of the a_{1g}-e_g^π crystal-field splitting caused by electron correlations [10,12].

The GGA+DMFT results are qualitatively different from those obtained with the nonmagnetic GGA and provide clear evidence for a structural phase transition. However, this transition is found to occur at a critical pressure of $p_c \sim 186$ kbar, corresponding to a large ($\sim$7%) reduction of the lattice volume. In addition, the results imply that the PM phase is energetically unfavorable at ambient pressure, i.e., thermodynamically unstable, with a total-energy difference with respect to the PI phase of $\Delta E \equiv E_{\rm PM} - E_{\rm PI} \sim 20$ meV/f.u. These features are in contrast to experiment. The origin of this discrepancy can be ascribed to the lack of charge self-consistency in the present calculations. Indeed, a strong enhancement of the a_{1g}-e_g^π crystal-field splitting will cause a substantial redistribution of the charge density and thereby influence the lattice structure due to electron-lattice coupling. All this makes charge self-consistency [24] particularly important at the metal-insulator transition.

B. Self-consistent calculations

For this reason we implemented a fully charge self-consistent GGA+DMFT method [24] within a plane-wave pseudopotential approach to compute the electronic structure and phase stability of V_2O_3. In Fig. 2 we present our results for the total energy for different volumes. The calculated pressure-volume equation of state is shown in Fig. 2 (inset). The PM phase is now found to be thermodynamically stable at ambient pressure, with a total-energy difference between the PM and PI phases of $\Delta E \sim 3$ meV/f.u. The structural transition takes place upon a slight expansion of the lattice, $\frac{\Delta V}{V} \sim 1.5\%$, at a negative critical pressure of $p_c \sim -28$ kbar, in agreement with experiment. The phase transition is accompanied by an abrupt increase of the lattice volume by $\sim$0.5% and a simultaneous change of the c/a ratio by $\sim$1.5%. This result is seen to differ significantly from that obtained with the non-charge-self-consistent GGA+DMFT scheme, according to which the PM phase is thermodynamically unstable at ambient pressure. The calculated equilibrium lattice constants and bulk moduli are now in remarkably good agreement with experiment [4]. In particular, we obtain $a = 4.99$ and 5.021 Å for the PM and PI phases, respectively, which is less than 1% larger than the experimental values. The calculated bulk moduli are $B = 219$ and 204 GPa, respectively. We note that the bulk modulus in the

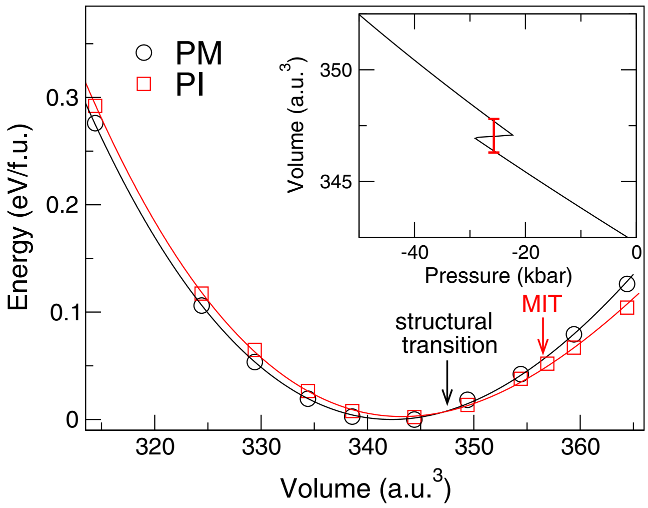

FIG. 2. (Color online) Variation of the total energy of paramagnetic V_2O_3 as a function of volume obtained by employing the fully charge self-consistent GGA+DMFT method. The inset shows the corresponding volume-pressure equation of state obtained as a derivative of the spline interpolation of $E(V)$. The calculated critical pressure $p_c \sim -28$ kbar and volume collapse by ~0.5% are marked by a red bar.

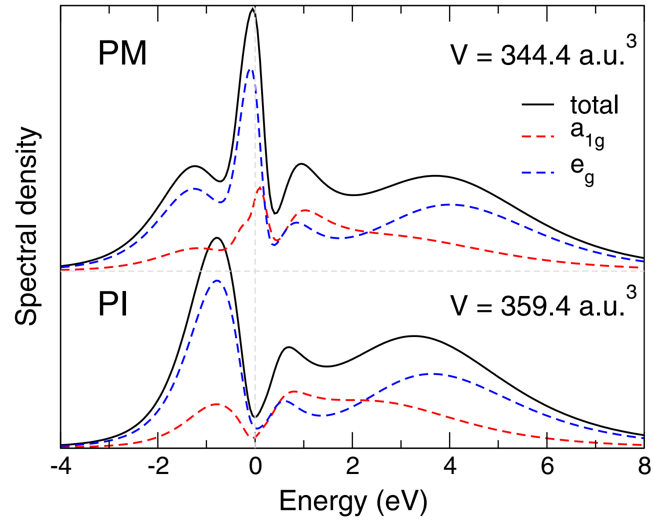

FIG. 3. (Color online) Spectral function of paramagnetic V_2O_3 across the MIT computed within GGA+DMFT.

PI phase is somewhat smaller than that in the PM phase, which implies an *enhancement* of the compressibility at the phase transition. Furthermore, the total-energy calculation results exhibit a weak anomaly near the MIT, which is associated with a divergence of the compressibility, in accordance with previous model calculations [25]. Overall, the electronic structure, the equilibrium lattice constant, and the structural phase stability of paramagnetic V_2O_3 obtained with the fully charge self-consistent GGA+DMFT scheme are in remarkably good agreement with the experimental data. Our calculations clearly demonstrate the crucial importance of electronic correlations and full charge self-consistency to explain the thermodynamic stability of the paramagnetic metal phase of V_2O_3.

C. Spectral function

In addition, we investigated the evolution of the spectral function of paramagnetic V_2O_3. In Fig. 3 we present our results obtained with the fully charge self-consistent GGA+DMFT approach for paramagnetic V_2O_3 across the MIT. Our calculations show that the V^{3+} ions are in a $S = 1$ spin configuration in all phases, with a predominant occupation of the e_g^π bands and substantial admixture of the a_{1g} orbital. The admixture is almost independent of changes of the lattice volume (deviations <5%), with a small decrease at the structural transition. Our results for the (a_{1g}, e_g^π) orbital occupations at the phase transition are (0.44, 0.78) for the PM phase and (0.42, 0.79) for the PI phase of V_2O_3. These findings are in good agreement with previous experimental estimations [26]. We note that the MIT takes place at a negative pressure $p_{el} \sim -66$ kbar, i.e., upon an expansion of the lattice volume by ~2%, above the structural phase transition. Thereby we conclude that the structural transition and the electronic transition are *decoupled*. Apparently, the structural transformation occurs as a precursor to the Mott-Hubbard MIT [27], implying an intricate interplay between electronic and lattice degrees of freedom at the transition.

D. Quasiparticle weight

Finally, we calculate the quasiparticle weight employing a polynomial fit of the imaginary part of the self-energy $\mathrm{Im}\Sigma(i\omega_n)$ at the lowest Matsubara frequencies ω_n. It is evaluated as $Z = [1 - \partial \mathrm{Im}\Sigma(i\omega)/\partial i\omega]^{-1}$ from the slope of the polynomial fit at $\omega = 0$. In Fig. 4 we present our results for the a_{1g} and e_g^π quasiparticle weights evaluated for the PI phase. Upon lattice volume expansion, both the a_{1g} and e_g^π quasiparticle weights monotonously decrease. Moreover, in the vicinity of the MIT the a_{1g} quasiparticle weight remains finite, while $Z = 0$ for the e_g^π orbitals, i.e., $\mathrm{Im}\Sigma(\omega)$ diverges for $\omega \to 0$. Therefore we conclude that the electronic effective

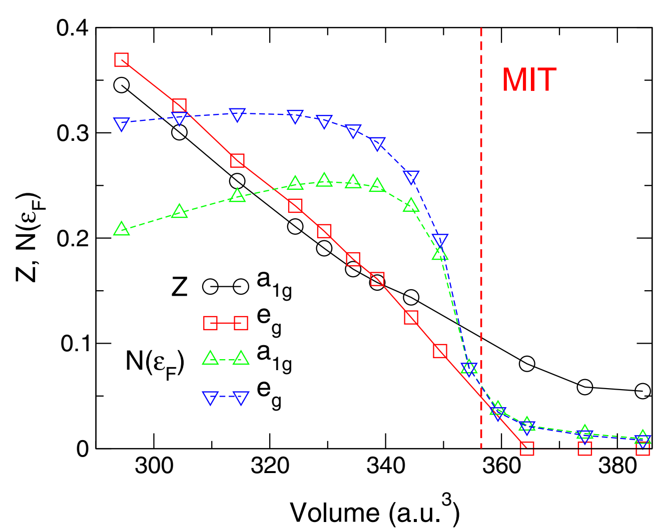

FIG. 4. (Color online) Quasiparticle weight Z and spectral weight at the Fermi level $N(\epsilon_F) = -\frac{\beta}{\pi}G(\tau = \beta/2)$ obtained for the a_{1g} and e_g^π orbitals across the MIT in the PI phase of V_2O_3. The MIT itself is indicated by a red dashed line.

mass of the e_g^π bands diverges at the MIT, in agreement with thermodynamic measurements of V_2O_3 [28]. We note that this divergence coincides with the drop of the spectral weight for the a_{1g} and e_g^π orbitals at the Fermi level shown in Fig. 4. The MIT is therefore driven by a strong orbital-selective renormalization of the V t_{2g} bands, in accordance with a Brinkman-Rice picture of the MIT. These results correct previous reasonings based on a correlation-induced enhancement of the crystal-field splitting of the V t_{2g} manifold, which results in a suppression of the hybridization between the a_{1g} and e_g^π bands [10,12]. On this basis, we conclude that an orbital-selective Mott transition is the only viable scenario for the MIT in V_2O_3.

IV. CONCLUSION

In conclusion, we employed the GGA+DMFT computational approach to determine the electronic structure and phase stability of paramagnetic V_2O_3 across the Mott-Hubbard metal-insulator phase transition. The calculated structural phase stability and spectral properties are in good agreement with experiment. Full charge self-consistency is found to be crucial to obtain the correct equilibrium lattice and electronic structure of V_2O_3 at ambient pressure. Upon lattice expansion, a structural phase transition is found to take place at $p_c \sim -28$ kbar, in agreement with experiment. The phase transition is accompanied by an abrupt lattice volume expansion by $\sim$0.5% and a simultaneous change of the c/a lattice parameter ratio by about 1.5%. Our calculations reveal an orbital-selective renormalization of the V t_{2g} bands caused by strong electron correlations. The electronic effective mass of the e_g^π orbitals diverges at the MIT, in accordance with a Brinkman-Rice picture of the MIT. However, the a_{1g} orbital exhibits a finite quasiparticle weight across the MIT. We therefore conclude that an orbital-selective Mott transition is the only viable scenario for the MIT in V_2O_3. Most importantly, we find that the structural transformation is decoupled from the electronic MIT. Our findings highlight the subtle interplay between electronic correlations and lattice stability across the Mott-Hubbard MIT.

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[1] N. F. Mott, Rev. Mod. Phys. **40**, 677 (1968); *Metal-Insulator Transitions* (Taylor and Francis, London, 1990); M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. **70**, 1039 (1998).

[2] D. B. McWhan and T. M. Rice, Phys. Rev. Lett. **22**, 887 (1969).

[3] Upon cooling below $T_N \sim 155$ K, V_2O_3 undergoes a first-order phase transition from a paramagnetic metal to an antiferromagnetic insulator, with a concomitant transformation to a monoclinic crystal structure ($I2/a$) [4].

[4] D. B. McWhan, T. M. Rice, and J. P. Remeika, Phys. Rev. Lett. **23**, 1384 (1969); D. B. McWhan and J. P. Remeika, Phys. Rev. B **2**, 3734 (1970); D. B. McWhan, A. Menth, J. P. Remeika, W. F. Brinkman, and T. M. Rice, *ibid.* **7**, 1920 (1973).

[5] Y. Ding, C.-C. Chen, Q. Zeng, H.-S. Kim, M. J. Han, M. Balasubramanian, R. Gordon, F. Li, L. Bai, D. Popov, S. M. Heald, T. Gog, H. K. Mao, and M. van Veenendaal, Phys. Rev. Lett. **112**, 056401 (2014); X. Deng, A. Sternbach, K. Haule, D. N. Basov, and G. Kotliar, *ibid.* **113**, 246404 (2014); L. Dillemans, T. Smets, R. R. Lieten, M. Menghini, C.-Y. Su, and J.-P. Locquet, Appl. Phys. Lett. **104**, 071902 (2014); J. S. Brockman, L. Gao, B. Hughes, C. T. Rettner, M. G. Samant, K. P. Roche, and S. S. P. Parkin, Nat. Nanotechnol. **9**, 453 (2014).

[6] D. Grieger, C. Piefke, O. E. Peil, and F. Lechermann, Phys. Rev. B **86**, 155121 (2012); P. Hansmann, A. Toschi, G. Sangiovanni, T. Saha-Dasgupta, S. Lupi, M. Marsi, and K. Held, Phys. Status Solidi B **250**, 1251 (2013); D. Grieger and F. Lechermann, Phys. Rev. B **90**, 115115 (2014).

[7] L. F. Mattheiss, J. Phys. Condens. Matter **6**, 6477 (1994).

[8] V. I. Anisimov, A. I. Poteryaev, M. A. Korotin, A. O. Anokhin, and G. Kotliar, J. Phys. Condens. Matter **9**, 7359 (1997); A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998); K. Held, I. A. Nekrasov, G. Keller, V. Eyert, N. Blümer, A. K. McMahan, R. T. Scalettar, Th. Pruschke, V. I. Anisimov, and D. Vollhardt, Phys. Status Solidi B **243**, 2599 (2006); G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. **78**, 865 (2006); J. Kunes, I. Leonov, M. Kollar, K. Byczuk, V. I. Anisimov, and D. Vollhardt, Eur. Phys. J. Spec. Top. **180**, 5 (2010).

[9] W. Metzner and D. Vollhardt, Phys. Rev. Lett. **62**, 324 (1989); A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996); G. Kotliar and D. Vollhardt, Phys. Today **57**(3), 53 (2004).

[10] K. Held, G. Keller, V. Eyert, D. Vollhardt, and V. I. Anisimov, Phys. Rev. Lett. **86**, 5345 (2001); G. Keller, K. Held, V. Eyert, D. Vollhardt, and V. I. Anisimov, Phys. Rev. B **70**, 205116 (2004).

[11] G. Panaccione, M. Altarelli, A. Fondacaro, A. Georges, S. Huotari, P. Lacovig, A. Lichtenstein, P. Metcalf, G. Monaco, F. Offi, L. Paolasini, A. Poteryaev, O. Tjernberg, and M. Sacchi, Phys. Rev. Lett. **97**, 116401 (2006).

[12] A. I. Poteryaev, J. M. Tomczak, S. Biermann, A. Georges, A. I. Lichtenstein, A. N. Rubtsov, T. Saha-Dasgupta, and O. K. Andersen, Phys. Rev. B **76**, 085127 (2007).

[13] L. Baldassarre, A. Perucchi, D. Nicoletti, A. Toschi, G. Sangiovanni, K. Held, M. Capone, M. Ortolani, L. Malavasi, M. Marsi, P. Metcalf, P. Postorino, and S. Lupi, Phys. Rev. B **77**, 113107 (2008); J. M. Tomczak and S. Biermann, J. Phys. Condens. Matter **21**, 064209 (2009); F. Rodolakis, J.-P. Rueff, M. Sikora, I. Alliot, J.-P. Itié, F. Baudelet, S. Ravy, P. Wzietek, P. Hansmann, A. Toschi, M. W. Haverkort, G. Sangiovanni, K. Held, P. Metcalf, and M. Marsi, Phys. Rev. B **84**, 245113 (2011).

[14] S.-K. Mo, J. D. Denlinger, H.-D. Kim, J.-H. Park, J. W. Allen, A. Sekiyama, A. Yamasaki, K. Kadono, S. Suga, Y. Saitoh, T. Muro, P. Metcalf, G. Keller, K. Held, V. Eyert, V. I. Anisimov, and

D. Vollhardt, Phys. Rev. Lett. **90**, 186403 (2003); S.-K. Mo, H.-D. Kim, J. D. Denlinger, J. W. Allen, J.-H. Park, A. Sekiyama, A. Yamasaki, S. Suga, Y. Saitoh, T. Muro, and P. Metcalf, Phys. Rev. B **74**, 165101 (2006).

[15] S. Lupi, L. Baldassarre, B. Mansart, A. Perucchi, A. Barinov *et al.*, Nat. Commun. **1**, 105 (2010).

[16] I. Leonov, N. Binggeli, Dm. Korotin, V. I. Anisimov, N. Stojić, and D. Vollhardt, Phys. Rev. Lett. **101**, 096405 (2008); I. Leonov, Dm. Korotin, N. Binggeli, V. I. Anisimov, and D. Vollhardt, Phys. Rev. B **81**, 075109 (2010).

[17] S. Baroni, S. de Gironcoli, A. Dal Corso, and P. Giannozzi, Rev. Mod. Phys. **73**, 515 (2001); P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car *et al.*, J. Phys. Condens. Matter **21**, 395502 (2009).

[18] V. I. Anisimov, D. E. Kondakov, A. V. Kozhevnikov, I. A. Nekrasov, Z. V. Pchelkina, J. W. Allen, S.-K. Mo, H.-D. Kim, P. Metcalf, S. Suga, A. Sekiyama, G. Keller, I. Leonov, X. Ren, and D. Vollhardt, Phys. Rev. B **71**, 125119 (2005); G. Trimarchi, I. Leonov, N. Binggeli, Dm. Korotin, and V. I. Anisimov, J. Phys. Condens. Matter **20**, 135227 (2008); Dm. Korotin, A. V. Kozhevnikov, S. L. Skornyakov, I. Leonov, N. Binggeli, V. I. Anisimov, and G. Trimarchi, Eur. Phys. J. B **65**, 91 (2008).

[19] P. Werner, A. Comanac, L. de'Medici, M. Troyer, and A. J. Millis, Phys. Rev. Lett. **97**, 076405 (2006); E. Gull, A. J. Millis, A. I. Lichtenstein, A. N. Rubtsov, M. Troyer, and P. Werner, Rev. Mod. Phys. **83**, 349 (2011).

[20] We here employ the density-density approximation of the local Coulomb interaction between the V t_{2g} electrons.

[21] A. Jayaraman, D. B. McWhan, J. P. Remeika, and P. D. Dernier, Phys. Rev. B **2**, 3751 (1970); P. Limelette, A. Georges, D. Jerome, P. Wzietek, P. Metcalf, and J. M. Honig, Science **302**, 89 (2003).

[22] H. Fujiwara, A. Sekiyama, S.-K. Mo, J. W. Allen, J. Yamaguchi, G. Funabashi, S. Imada, P. Metcalf, A. Higashiya, M. Yabashi, K. Tamasaku, T. Ishikawa, and S. Suga, Phys. Rev. B **84**, 075117 (2011).

[23] W. F. Brinkman and T. M. Rice, Phys. Rev. B **2**, 4302 (1970).

[24] For a review, see, e.g., L. V. Pourovskii, B. Amadon, S. Biermann, and A. Georges, Phys. Rev. B **76**, 235101 (2007); K. Haule, *ibid.* **75**, 155113 (2007); B. Amadon, F. Lechermann, A. Georges, F. Jollet, T. O. Wehling, and A. I. Lichtenstein, *ibid.* **77**, 205112 (2008); M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, *ibid.* **80**, 085101 (2009); H. Park, A. J. Millis, and C. A. Marianetti, *ibid.* **90**, 235103 (2014).

[25] G. Kotliar, S. Murthy, and M. J. Rozenberg, Phys. Rev. Lett. **89**, 046401 (2002); S. R. Hassan, A. Georges, and H. R. Krishnamurthy, *ibid.* **94**, 036402 (2005).

[26] J.-H. Park, L. H. Tjeng, A. Tanaka, J. W. Allen, C. T. Chen, P. Metcalf, J. M. Honig, F. M. F. de Groot, and G. A. Sawatzky, Phys. Rev. B **61**, 11506 (2000).

[27] P. Pfalzer, G. Obermeier, M. Klemm, S. Horn, and M. L. denBoer, Phys. Rev. B **73**, 144106 (2006); S. Populoh, P. Wzietek, R. Gohier, and P. Metcalf, *ibid.* **84**, 075158 (2011); J. Köndel, P. Pontiller, C. Müller, G. Obermeier, Z. Liu, A. A. Nateprov, A. Hörner, A. Wixforth, S. Horn, and R. Tidecks, Appl. Phys. Lett. **102**, 101904 (2013).

[28] D. B. McWhan, J. P. Remeika, T. M. Rice, W. F. Brinkman, J. P. Maita, and A. Menth, Phys. Rev. Lett. **27**, 941 (1971); S. A. Carter, T. F. Rosenbaum, P. Metcalf, J. M. Honig, and J. Spalek, Phys. Rev. B **48**, 16841 (1993).