

Dynamical screening and plasmonic replica bands in SrVO₃

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SrVO₃ has long been considered a prototype for studying electronic correlations due to its simple electronic structure and coherent t_{2g} quasiparticle band. However, the origin of the incoherent satellite at 1.5 eV binding energy seen in photoemission remains enigmatic, having been attributed to Hubbard physics, oxygen vacancies, or plasmonic coupling. Here, we investigate the t_{2g} band and satellite feature in epitaxially strained thin films of SrVO₃ using angle-resolved photoemission spectroscopy. We observe an increase in the quasiparticle effective mass due to tensile strain as a result of the narrowing of the corresponding bandwidth. While a low-energy kink at 60 meV indicates additional electron-phonon interactions, no strain-dependent behavior is detected. The increased effective mass coincides with a decrease in the plasma frequency from reported ellipsometry measurements and theoretical calculations. From this, we associate the plasma frequency with the quasiparticle-satellite energy separation that follows the same strain-induced changes. This consistency affirms the identification of the satellite feature as a replica band due to coherent coupling between quasiparticles and low-energy plasmons and emphasizes the role of dynamical screening as a key organizing principle in the electronic structure of correlated oxides.

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I. INTRODUCTION

Owing to its simple electronic structure and moderate correlations, SrVO₃ has served as a benchmark quantum material for understanding the interplay between electron-phonon coupling, itineracy, electronic correlations, and coupling to collective excitations, as evidenced by numerous spectroscopic measurements [1–6], which have aided the development of advanced theoretical techniques [7–17]. Despite the abundance of calculations and experiments on SrVO₃, there still remain a number of open questions pertaining to features in its electronic structure. Foremost is the precise nature of a broad, incoherent satellite located ~ 1.5 eV below E_F seen with angle-resolved photoemission spectroscopy (ARPES) [2]. While initially identified as a lower Hubbard band [18], other recent work has suggested that this feature could arise either from coupling of the valence electrons to low-energy plasmons [15] or potentially from oxygen vacancies [19]. In particular, the recent interest in SrVO₃ as a transparent conducting oxide with high mobility [20,21] underscores the need

to understand the coupling between its low-energy quasiparticles, which govern its electrical transport [22], and collective charge excitations such as plasmons, which influence the material's optical properties [23,24].

A major challenge in determining the nature of this satellite is the coexistence of multiple interactions and excitations in SrVO₃ with nearly degenerate energy scales; the lower Hubbard band, oxygen-vacancy-induced states, and the plasma frequency, ω_p , are all believed to exist in a similar energy range of 1–2 eV. In order to distinguish between these possibilities, we have performed a systematic investigation of how epitaxial strain affects the ARPES spectra of the quasiparticle band and the satellite. We then compare these changes to the strain-dependent changes in ω_p , as measured by ellipsometry. The consistency between the strain-dependent changes in ω_p and the shift in the satellite energy with strain strongly signals the plasmonic nature of this satellite feature.

II. EXPERIMENTAL

Thin films of SrVO₃ with 20 nm thickness were grown on SrTiO₃ and NdGaO₃ substrates via pulsed laser deposition [21]. The films were transferred under continuous nitrogen flow conditions to the LOREA endstation at the ALBA synchrotron, where ARPES measurements were performed in ultrahigh vacuum with a photon energy of 93 eV at 20 K, after exposing the sample to the beam for several hours. This triggers physisorption of contamination species and

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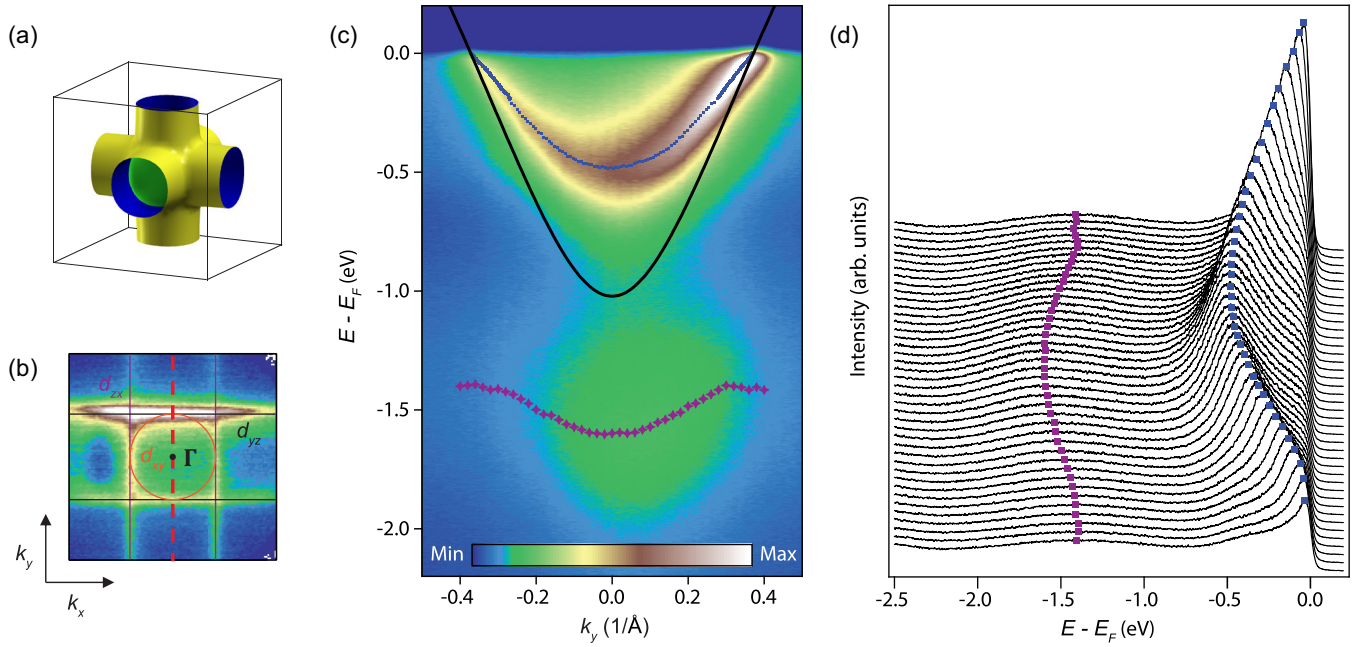


FIG. 1. Band structure and ARPES spectra of SrVO₃/NdGaO₃. (a) DFT-calculated Fermi surface of bulk SrVO₃. (b) 2D Fermi surface ARPES spectra of SrVO₃/NdGaO₃ with a schematic of the Fermi surface cross section at $k_z = 0$ overlaid. Position of the ARPES cuts taken inside the BZ is denoted by the dotted red line. (c) ARPES spectra of SrVO₃/NdGaO₃. Extracted dispersions for the quasiparticle band and incoherent satellite are plotted in blue squares and purple stars, respectively. DFT-calculated band structure is plotted in black. (d) Waterfall plot of the EDCs for the spectra in panel (c).

photoreduction of V⁵⁺ to V⁴⁺ [25,26], as shown in the Supplemental Material [27]. Low-energy electron diffraction measurements reveal slight film relaxation at the surface, indicating that the SrVO₃ film on SrTiO₃ experiences 1.0% biaxial tensile strain compared to its bulk lattice constant (3.842 Å), whereas the film on NdGaO₃ is fully relaxed (see the Supplemental Material [27]).

III. RESULTS AND DISCUSSION

SrVO₃ has a V 3d¹ electron configuration, with three degenerate t_{2g} bands crossing the Fermi level (E_F), forming three perpendicularly intersecting quasi-two-dimensional (quasi-2D) cylindrical tubes that hybridize to produce one jungle-gym-like structure and two other pseudocubic manifolds at the center of the Brillouin zone (BZ), as shown in Fig. 1(a). At $h\nu = 93$ eV and an inner potential of $V_0 = 17$ eV [6], the expected Fermi surface for $k_z \approx 0$ is shown in Fig. 1(b) in the k_x – k_y plane, consistent with previous measurements [6]. An E versus k momentum cut along the dashed red line in Fig. 1(b) for the SrVO₃/NdGaO₃ sample is shown in Fig. 1(c), showing the parabolic t_{2g} band that disperses from -0.5 eV to E_F ; the asymmetry in intensity along k_y is due to matrix element effects. The dispersion of the quasiparticle band is determined by fitting the peak positions of the momentum distribution curves for binding energies below ~ 0.2 eV and the energy distribution curves (EDCs) at higher binding energies. For comparison, the band dispersion calculated by density functional theory (DFT) for SrVO₃ is plotted in solid black in Fig. 1(c) (for details, see the Supplemental Material [27]). Scaling the dispersion by 2.02 results in excellent agreement with the experimentally

measured dispersion, indicative of electron-electron correlations, as previously reported [2]. We identify the well-reported broad satellite at a binding energy of approximately 1.5 eV [2]. Fitting the peak maxima of the EDCs [Fig. 1(d)] reveals that this satellite feature also exhibits a parabolic dispersion reminiscent of the quasiparticle band dispersion. Similar nondispersive or weakly dispersing satellite features in the 1.3–1.6 eV range have been observed in numerous other transition metal oxides such as SrTiO₃ [28], Pb(Zr,Ti)O₃ [29], and TiO₂ [30].

We first discuss the time dependence of the spectral features under beam exposure. In previous studies, it was argued that a peak at 1.5 eV binding energy, attributed to oxygen vacancies, can be generated by prolonged exposure to the intense synchrotron beam during measurements [19]. Instead, we observe the ratio of the quasiparticle to satellite peak (I_{QP}/I_{Sat}) grows with prolonged beam exposure over several hours (see the Supplemental Material [27]), opposite to the findings of Backes *et al.* [19]. This suggests that the primary change induced by the beam removes surface adsorbates that typically suppress the low-energy features in our *ex situ* loaded samples, without the formation of beam-induced oxygen vacancies. Since we do not see a time evolution of the satellite feature under beam exposure, any defect-related contribution must be constant, thus ruling out the formation of oxygen vacancies for our samples.

To assess how strain modifies the dispersion of the low-energy quasiparticle band, we compare the t_{2g} band dispersions for SrVO₃/NdGaO₃ and SrVO₃/SrTiO₃ in Fig. 2(a). A small but clear narrowing of the experimental bandwidth can be observed with increasing tensile strain. We compare with the DFT-calculated band structure by simulating

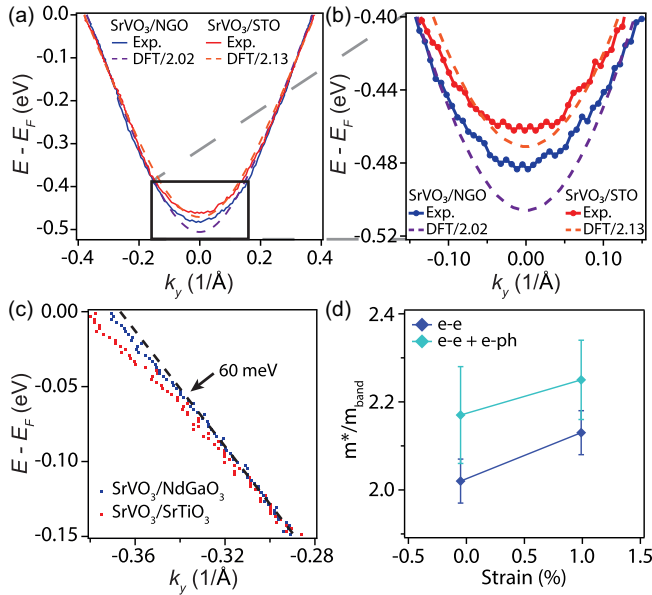


FIG. 2. (a) Extracted dispersion of quasiparticle band (b) near the band bottom for each substrate type. Renormalized DFT bands for each case are given by the dotted lines. (c) Dispersions near E_F demonstrating the low-energy kink in the 50–60 meV range. Black dotted line is a linear fit to the high binding energy region. (d) Strain dependence on the effective mass considering e-e contributions and e-e + e-ph contributions.

the equivalent strain for the corresponding films in our calculations (see the Supplemental Material for details [27]). The effective masses of the calculated bands are $0.42m_e$ for SrVO₃/NdGaO₃ and $0.45m_e$ for SrVO₃/SrTiO₃, based on parabolic fits to the near- E_F dispersions. The DFT calculations can be brought into quantitative agreement with the experimental measurements by rescaling the individual DFT dispersions by $2.02 \pm .05$ for SrVO₃/NdGaO₃ and $2.13 \pm .05$ for SrVO₃/SrTiO₃ [dotted lines in Figs. 2(a) and 2(b)]; here, the rescaled DFT band for each strain state was fitted to the corresponding experimental dispersion for $E_B > 100$ meV, where the dispersion is predominantly parabolic and unaffected by low-energy many-body effects (e.g., electron-phonon coupling). Parabolic fitting of the $E(k)$ dispersion in Fig. 2(b) yields a band minimum that lies 20 meV deeper for SrVO₃/NdGaO₃ than for SrVO₃/SrTiO₃, again consistent with a narrowing of the bandwidth with increasing in-plane lattice constant. These observations all clearly indicate that the strength of electronic correlations is enhanced with tensile strain.

Further renormalization of the energy dispersion occurs due to the presence of modest electron-phonon interactions that result in a kink in the low-energy band structure around 60 meV [5,22,31]. Figure 2(c) shows the near- E_F dispersion for both strain states, where the low-energy kink is apparent in both. The total coupling constants can be computed from $\lambda = v_F^0/v_F - 1$, where v_F^0 is the DFT Fermi velocity and v_F is the experimental Fermi velocity [32]. The electron-electron contribution can be subtracted out to obtain the electron-phonon coupling constants [33]: $0.15 \pm .11$ for SrVO₃/NdGaO₃ and $0.12 \pm .10$ for SrVO₃/SrTiO₃. To within the precision of

our experiment, we do not observe any appreciable strain-dependent change in the electron-phonon coupling constants or the kink energies. The strain-dependent changes in m^* are summarized in Fig. 2(d). The mass renormalization factor when only considering the electron-electron contributions is plotted in dark blue, with the increasing correlations likely due to the reduced in-plane V-O-V orbital overlap and subsequent band narrowing [34,35]. The effective mass renormalizations composed of both electron-electron and electron-phonon contributions are plotted in teal. Since there is little observable effect from the electron-phonon coupling, there is a near rigid shift between the two datasets. We now focus on the effects of strain on the incoherent satellite feature. Figure 3(a) shows EDCs of SrVO₃/NdGaO₃ centered on the satellite, showing a clear dispersive feature that mimics the parabolic nature of the quasiparticle band. Comparing the satellite dispersions of both samples [Fig. 3(b)] shows a noticeable energy difference between the band bottom energies, which are $1.53 \pm .01$ and $1.61 \pm .01$ eV for SrVO₃/NdGaO₃ and SrVO₃/SrTiO₃, respectively. While this strain-induced energy shift is reminiscent of the behavior of the low-energy quasiparticle band shown in Figs. 2(a) and 2(b), this shift is larger in the case of the satellite. This is illustrated by comparing the EDCs at $k = 0$ in Fig. 3(c), where $\Delta E_{\text{QP}} = 25 \pm 4$ and $\Delta E_{\text{Sat}} = 81 \pm 12$ meV are the strain-induced energy shifts between the band bottoms of SrVO₃/NdGaO₃ and SrVO₃/SrTiO₃, showing that ΔE_{Sat} is about three times larger than ΔE_{QP} . Defining Ω_p as the energy separation between the quasiparticle and satellite peaks, we find that $\Omega_p^{\text{NGO}} = 1.13 \pm 0.01$ eV while $\Omega_p^{\text{STO}} = 1.07 \pm 0.01$ eV.

Given the energy scale and strain dependence of Ω_p , we are able to relate it to the plasma frequency by comparing with other strain-dependent studies. Earlier ellipsometry measurements determined the bulk plasma frequency of SrVO₃ on different substrates to be in the range of 1.2–1.3 eV [36,37]. Both ω_p and Ω_p share a similar decrease in energy as biaxial strain increases, shown in Fig. 3(d). The same trend is likewise seen in DFT calculations of the plasma frequency as a function of strain in SrVO₃ [37] [orange squares in Fig. 3(d)]. These studies both attribute this strain dependence of the plasma frequency to an increase in the effective mass that we directly observe, as discussed above.

Taken as a whole, the shared behavior of Ω_p and ω_p , both from experiment and from first-principles calculations, point toward the high-energy satellite being a plasmonic replica of the main quasiparticle band. In addition, the satellite also exhibits a momentum-dependent parabolic dispersion reminiscent of the main quasiparticle band, and substantially loses intensity as soon as $k > k_F$. This “replica band” behavior has been observed by ARPES in a multitude of systems with strong electron-boson coupling, where the quasiparticles interact dynamically with a collective mode. For instance, dispersive replica bands have been observed in systems with strong electron-phonon coupling such as lightly doped SrTiO₃ [38], Sr₂TiO₄ [39], monolayer FeSe/SrTiO₃ [40], and TiO₂ [41], where the separation between the main quasiparticle band and the replica band is typically a high-energy, longitudinal optical mode associated with the out-of-plane motion of the oxygen atoms. Likewise, such replica bands can also be formed from the quasiparticles interacting dynamically with

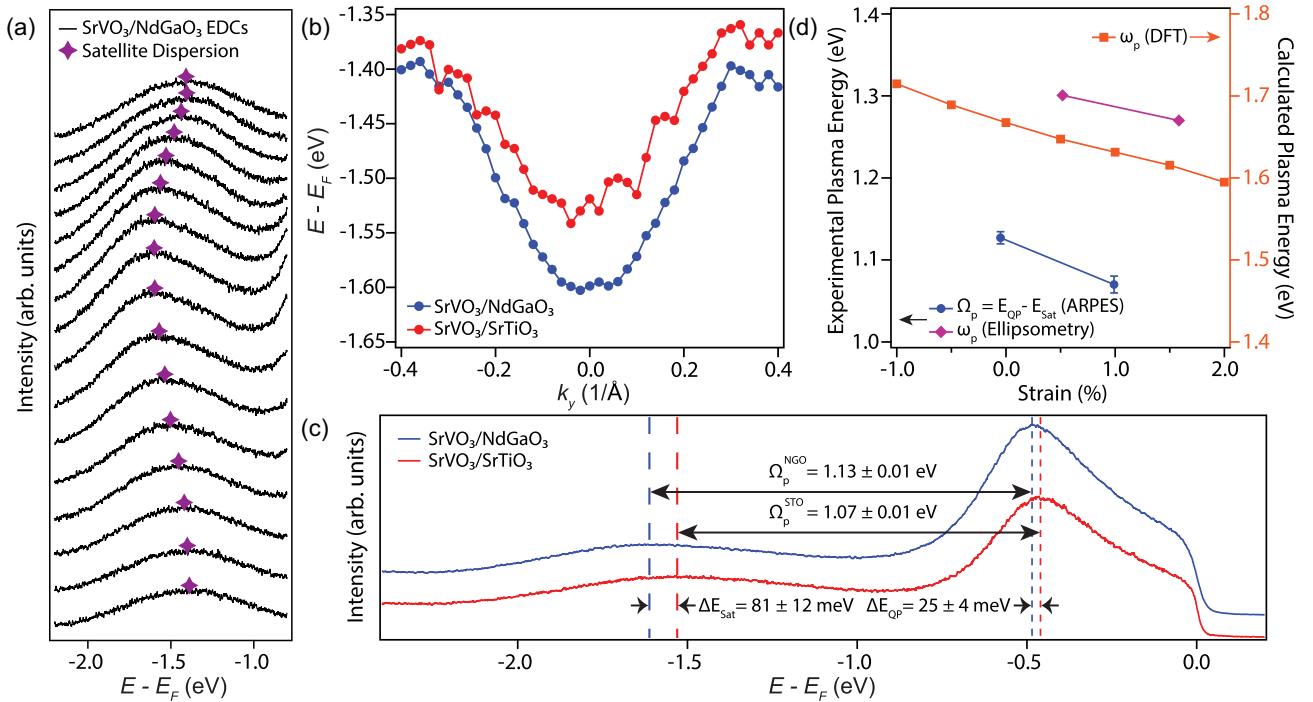


FIG. 3. (a) Waterfall plot centered on the satellite band. EDC peaks are marked with purple stars. (b) Extracted dispersions for the satellite. (c) Direct comparison of the EDCs at $k_y = 0$ between strain states. Ω_p is the energy separation between the QP and satellite band bottoms for a given sample. ΔE_{QP} and ΔE_{Sat} are the energy differences between strain states of the QP and satellite band bottoms, respectively. (d) Energy shift as a function of strain for the energy separation Ω_p , the plasma energy as determined by ellipsometry, and the screened plasma energy calculated by DFT.

a collective charge excitations of the electron gas, i.e., plasmons. The photoemission process generates a photohole that is screened by charge fluctuations of the remaining electrons, exciting a plasmon, and producing a shakeup satellite in the single-particle spectral function. The resulting replica band, displaced by the plasma energy and following the same dispersion as the quasiparticle, is thus a spectroscopic fingerprint of electron-plasmon coupling [42–44]. To our knowledge, plasmonic replica bands have only been reported primarily in low carrier density systems such as lightly doped semiconductors or semimetals, such as lightly doped EuO [45], SrIrO₃ [46], or doped van der Waals materials [47], where the plasma frequency is relatively low. In these situations, the plasmon energy can also overlap with phonons, or the plasmonic replicas can overlap with other bands, often obscuring their identification. In SrVO₃, the confluence of a simple d^1 electronic band structure, a clear separation between the V 3d and O 2p states, and a plasma frequency in the range of ≈ 1 eV makes the plasmonic replica feature particularly clear and robust.

While the evidence presented favors a plasmonic origin to this high-energy satellite, a contribution from a Hubbard-like feature cannot be entirely excluded. Strain-dependent DMFT calculations may be useful to compare the expected behavior of a Hubbard band. Additionally, detailed comparisons of 4d transition metal oxides such as SrNbO₃ or SrMoO₃ that likewise have plasma frequencies in the visible range but weaker electron correlations could provide clear and definitive evidence as to the prevalence of plasmonic replicas in ARPES spectra of quantum materials.

IV. CONCLUSIONS

In summary, we have studied the effects of epitaxial strain on the band structure of thin film SrVO₃ through ARPES. Our results support the view that quasiparticle excitations in SrVO₃ are dynamically screened by collective charge oscillations, and this plasmonic coupling manifests as a replica band at 1.5 eV below E_F . The observation of a dispersive replica band closely following the dispersion of the quasiparticle confirms that the plasmon couples coherently to the itinerant electrons forming the Fermi surface, rather than arising from localized (defects or impurities) or surface-confined charge oscillations and establishes momentum-resolved photoemission as a powerful probe for disentangling dynamical screening mechanisms—an approach that can be readily extended to other correlated and plasmonic oxides.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [48].

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