

Effects of strain on the stability of the metallic rutile and insulating M1 phases of vanadium dioxide

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(Received 29 October 2025; accepted 9 March 2026; published 1 April 2026)

We present a systematic density-functional theory study of the effects of strain on the structural and electronic properties in vanadium dioxide (VO_2), with particular emphasis on its effect on the relative stability of the metallic rutile and the insulating monoclinic M1 phases. We consider various strain conditions that can be related to epitaxial strain present in VO_2 films grown on different lattice planes. Our calculations confirm the dominant role of c -axis strain, i.e., along the direction of the V-V dimerization in the M1 phase. Our analysis suggests that this effect stems primarily from the weakening of the lattice stiffness, with the hopping amplitude along the c axis playing a minor role. We also confirm that, in strain scenarios that deform the basal plane, the c -axis strain still has a dominant effect on the phase stability.

DOI: [10.1103/rhdm-rdgd](https://doi.org/10.1103/rhdm-rdgd)

I. INTRODUCTION

Vanadium dioxide (VO_2) is a prototypical system undergoing a metal-insulator transition (MIT). The proximity of the MIT temperature, T_c , to room temperature makes VO_2 a particularly promising material for a wide range of potential applications [1,2]. For most proposed device concepts, a controlled tuning of T_c and other characteristics of the MIT is highly desirable [3]. Such tuning can be achieved, for example, via chemical doping [4] or through the application of strain [5–7], the latter method having the potential advantage of not producing chemical disorder. Although the dependence of the MIT on strain has been demonstrated, precise and effective tuning requires understanding of the underlying physical mechanisms.

During the MIT, chains of vanadium dimers form along the c direction of the underlying high-temperature metallic rutile (R) structure, resulting in the low-temperature insulating monoclinic M1 structure [8] [see Figs. 1(a) and 1(b), respectively]. The main physics behind this transition was first suggested by Goodenough [9] and assumes a singlet formation within the vanadium-vanadium dimers, due to the pairing of the electrons occupying the lowest-lying d orbital of the nominal d^1 vanadium cations. Defining a local coordinate system with the local x axis along c and the local z axis along the apical vanadium-oxygen bond [8], this lowest-lying orbital is the $d_{x^2-y^2}$ orbital [Fig. 1(c)]. The large bonding-antibonding splitting resulting from the vanadium-vanadium dimerization, together with a depletion of the d_{xz} and d_{yz} orbitals [Figs. 1(d)

and 1(e)] associated with a zigzag distortion perpendicular to c , gives rise to insulating behavior. The simplicity of this picture has, however, been called into question in part due to the presence of a Mott-insulating monoclinic M2 phase [10], and thus the exact origin of the electronic and structural transition in VO_2 has been a topic of controversy for decades [11–13]. Currently, the consensus characterizes the material as a correlation-assisted Mott-Peierls insulator [14–16].

Large changes to the MIT temperature, T_c , have been observed in VO_2 samples under strain. Strain has been applied to VO_2 through bending [5,6], pressure [17,18], nanowire engineering [19–22], or growth of thin films on substrates with a certain lattice mismatch and different surface orientations [23–30], giving rise to coherent epitaxial strain along different crystallographic directions. In particular, it has been shown that growth with the rutile c axis oriented either perpendicular to [Fig. 1(f)] or within [Fig. 1(g)] the substrate surface plane of a TiO_2 substrate suppresses and enhances T_c , respectively [23,27]. This observation has been rationalized by the different orientation and sign of the strain relative to the direction of dimerization, which is strongly linked to the hopping amplitude between the Peierls-active $d_{x^2-y^2}$ orbitals [23]. It has thus been suggested that elongating c leads to an increase in T_c , while compressing c decreases T_c . However, the role of strain along other crystallographic directions is still debated [27]. For example, it has been suggested that, as the basal lattice parameters contract, the relative oxygen apical and equatorial distances change, and the filling of the $d_{x^2-y^2}$ orbital is affected [25,30]. Computationally, the importance of the bandwidth change and the corresponding change in hopping amplitudes along the dimerization direction have been highlighted [31]. However, a systematic computational study detailing both structural and electronic effects responsible for the changes in the observed phase stability between the R and M1 VO_2 phases under a wide range of strain conditions is lacking.

In part, the absence of such a study to date stems from the difficulty in describing the different phases of VO_2 quantitatively using a single consistent first-principles-

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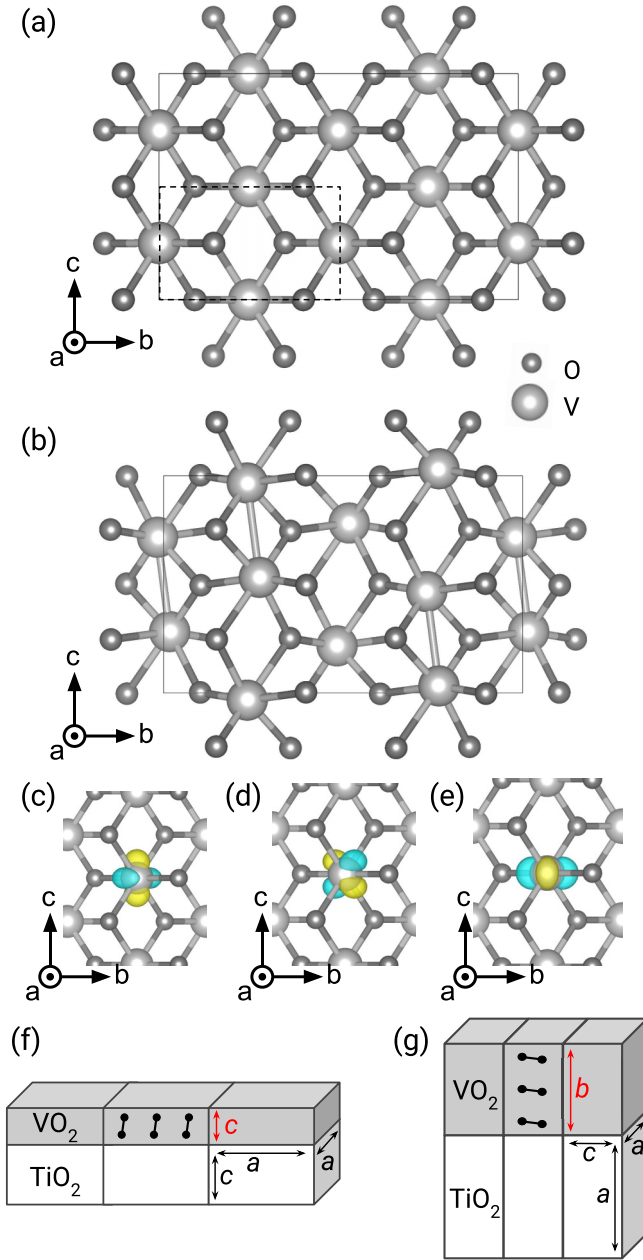


FIG. 1. Supercell used in the calculations to describe both the (a) R and (b) M1 structures. The dashed lines in panel (a) indicate the underlying primitive rutile cell. Arrows indicate the definition and orientation of the corresponding orthorhombic axes. V (O) atoms shown in light (dark) gray. (c)–(e) The three V t_{2g} orbitals, $d_{x^2-y^2}$, d_{xz} , and d_{yz} , respectively, highlighting their orientation with respect to the local environment. Yellow (cyan) colors indicate positive (negative) phase of the orbitals. The two epitaxial strain orientations considered in this work, corresponding to growth along the (f) [001] and (g) [010] directions, with VO₂ shown in gray and the substrate (e.g., TiO₂) shown in white. The dimerization direction of VO₂ is indicated by little pictograms. The VO₂ axes that are free to relax are shown in red, while the VO₂ axes shown in black are fixed by the substrate.

based approach. In particular, conventional density-functional theory (DFT) studies with standard semilocal exchange-correlation functionals predict an incorrect energetic ordering of the R and M1 phases [32,33] and fail to open a gap in the

insulating M1 phase [8,12]. More advanced meta-generalized gradient approximation functionals, e.g., strongly constrained and appropriately normed (SCAN) functional, also exhibit shortcomings such as needing a local magnetic order not present in experiment to obtain the correct value of the band gap, and underestimation of structural dimerization when such magnetic order is imposed [34]. Likewise, the widely used DFT + U method, which incorporates an explicit on-site Hubbard-like interaction term, does not capture the physics of VO₂ correctly, either opening a band gap in the metallic phase or causing a loss of structural dimerization depending on the choice of U [35]. Even more complex approaches such as DFT + dynamical mean-field theory (DMFT) work best with different single-site and cluster constructions for the R and M1 phases, respectively [14,16], although a unified approach based on the use of bond-centered orbitals has been suggested recently [36].

In the present work, we use our previously established approach [37], based on the DFT + V method [38], to mimic a static intersite self-energy. This approach builds on previous DFT + DMFT works that have shown that, in the dimerized M1 phase of VO₂, the on-site interactions give rise to a largely frequency-independent intersite self-energy [14], which can be well approximated by adding a static potential acting between neighboring V-V pairs [39–43]. In our recent work [37], we used this idea to consistently describe both R and M1 phases, as well as the transition between them, using the framework of the DFT + V method as a means to apply such an empirical static intersite self-energy correction. This offers a simple and computationally cheap approach that correctly describes the structural dimerization, the band-gap opening in the M1 phase, and the energy balance between the two phases. It also shows good agreement with previous works using more advanced (but also computationally more demanding) methods [14,16,36].

Here, we build on our previous work and extend it to investigate how different externally applied strain conditions affect the energy balance between the R and M1 phases and related structural and electronic properties. The resulting insights will allow for a more targeted strain tuning of the MIT toward possible device applications. We note that a static correction to the intersite self-energy is not expected to allow for a description of the more exotic monoclinic M2 phase, which hosts a Mott-insulating state [16], and we therefore restrict our discussion to the R and M1 phases.

In particular, we focus on strain corresponding to epitaxial growth on the (001) plane of the rutile structure, but we also exploit the flexibility of our computational approach to gain more systematic insights by varying the in-plane and out-of-plane strains independently from each other. We analyze the resulting changes in different microscopic electronic parameters, such as orbital energy levels and hopping amplitudes, to explain the observed behavior. Finally, we briefly explore the effect of applying different strains along the two orthogonal directions within the basal plane, corresponding, e.g., to epitaxial growth on the rutile (010) plane, which breaks the tetragonal symmetry of the underlying rutile structure.

Our calculations confirm that the phase stability of VO₂ is primarily governed by the strain along the dimerization direction, while the strain within the basal plane represents

only a secondary effect. Strain along the dimerization direction directly modifies the distance between nearest-neighbor vanadium atoms and, consequently, affects both the hopping amplitudes and the stiffness of the underlying crystal structure opposing the dimerization. Our analysis suggests that the decrease of the structural stiffness under elongation is the dominating factor favoring the M1 phase.

II. METHODOLOGY

To simulate different strain conditions in VO₂, we embed both the R and M1 structures in the same unit cell formed from four primitive cells of the underlying rutile structure, corresponding to a $1 \times 2 \times 2$ supercell [see Figs. 1(a) and 1(b), with the primitive rutile cell indicated by dashed lines in panel (a)]. In order to allow for a systematic comparison between the different phases, we use a metrically orthorhombic cell for both structures; i.e., we fix the angles between the corresponding lattice vectors to 90°, thereby neglecting the small monoclinic strain in the M1 structure. Note that the corresponding cell nevertheless can host the full monoclinic distortion of the (internal) atomic coordinates of the M1 phase, including both dimerization and zigzag distortion. We define c along the dimerization direction, aligning a , b , c with x , y , z of the global coordinate system.

In Sec. III A, we consider only strain conserving the tetragonal symmetry of the R phase, i.e., we keep the $a:b$ ratio of the supercell lattice vectors fixed to 1:2, applying $\epsilon_{xx} = \epsilon_{yy}$ strain, and observing the change in c by relaxing the σ_{zz} stress component. In Sec. III B, we also keep the tetragonal symmetry but now vary both $\epsilon_{xx} = \epsilon_{yy}$ and ϵ_{zz} strains independently. Finally, in Sec. III C, we investigate the orthorhombic strain regime by imposing $\epsilon_{xx} \neq \epsilon_{yy}$ and ϵ_{zz} .

We define the applied strain as $\epsilon_{xx} = (a - a_0)/a_0 \times 100\%$, where a is the strained lattice parameter and $a_0 = 4.630$ Å is its unstrained reference defined by the fully relaxed rutile R structure within DFT + V and using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [44], so that the definition of strain is not affected by the typical small over- and underestimation of the a and c lattice parameters within PBE compared to experiment. We then fix the lattice parameters to specific strain values while relaxing the internal positions of all atoms starting from both R and M1 internal positions, monitoring the resulting changes in energy as well as electronic and structural properties. We specify the fully relaxed lattice parameters for the R phase and the partially relaxed ones for the M1 phase in Table I. In the latter case, the lattice parameters are partially constrained, such that $a = b$, and all angles are set to 90°.

To account for the strong intersite effects along the dimerizing vanadium chains, we utilize the DFT + V machinery as presented in Ref. [37] and implemented in Ref. [45]. The DFT + V method [38] is motivated from the extended Hubbard model [46] and adds the following term to the standard Kohn-Sham potential:

$$\hat{V}_V = - \sum_{I,J} V_{IJ} \sum_{m,m'} n_{m'I}^{II} \hat{P}_{mm'}^{IJ}, \quad (1)$$

where I and J refer to atomic sites that interact through the interatomic Coulomb interaction V_{IJ} , $n_{m'I}^{II}$ denotes the

TABLE I. Relaxed lattice parameters of the R and M1 phases, and of the R phase strained according to the TiO₂ strain condition (i.e., with only b relaxed). All values are recast relative to the R unit cell and are given in Å. The M1 structure is partially constrained, such that $a = b$, and all angles are 90°. The particular strain orientation on the TiO₂ substrate is discussed in more detail in Sec. III C.

	a (Å)	b (Å)	c (Å)
R	4.630	4.630	2.765
M1	4.606	4.606	2.820
R on (010)-TiO ₂	4.670	4.550	2.865

interatomic occupation, also indexed by the local orbital indices m , m' , and $\hat{P}_{mm'}^{IJ}$ is the corresponding projection operator. Thus, the interatomic potential in Eq. (1) leads to an effective renormalization of the interatomic hopping [37], which depends on both the imposed interaction strength, V_{IJ} , and the interatomic occupation, $n_{m'I}^{II\sigma}$. The latter can be viewed as a measure of the strength of the hybridization between orbitals m and m' on sites I and J , respectively. As a result, the $+V$ term enhances the bonding-antibonding splitting on the dimerized vanadium-vanadium pairs, while it has little effect on the longer vanadium-vanadium bonds or in the R phase, where the corresponding interatomic hybridization is weak.

As outlined in Sec. I, and discussed in more detail in Ref. [37], we employ the $+V$ correction to mimic an empirical intersite static self-energy necessary to enhance electronic dimerization in the M1 phase [39]. In line with our previous findings [37], and unless otherwise noted, we fix $V = 2$ eV on both short and long bonds (SB and LB), obtaining the correct energetic ordering of the unstrained R and M1 phases, and the correct size of the band gap in the M1 phase.

We perform the DFT + V calculations using the QUANTUM ESPRESSO (v7.3) package [47,48] within the generalized gradient approximation using the PBE functional [44]. We use the ultrasoft pseudopotentials from the Garrity-Bennett-Rabe-Vanderbilt (GBRV) library [49], including the $3s$ and $3p$ semicore states in the valence manifold of the vanadium atoms. We use a plane-wave kinetic energy cutoff of 70 Ry and 12×70 Ry for the charge density. We use a Γ -centered $8 \times 5 \times 6$ k -mesh, and converge the change in total energies between consecutive iterations to be smaller than 5×10^{-8} eV, all force components to be smaller than 10^{-3} eV/Å, and all free components of the stress tensor below 0.5 kbar.

For further microscopic electronic analysis, we construct a localized basis set using WANNIER90 (v3.1.0) [50,51]. We include the set of bands around the Fermi level to encompass the whole $\{d_{x^2-y^2}, d_{xz}, d_{yz}\}$ manifold, and the resulting Wannier bands closely reproduce the underlying DFT band structure (for the DFT and Wannier bands, see, e.g., Fig. 2 in Ref. [36]). We use this basis to extract the hopping amplitudes between all pairs of orbitals and their respective on-site energies.

III. RESULTS AND DISCUSSION

A. (001)-oriented epitaxial strain

We first discuss the behavior of the R and M1 phases both embedded in the same supercell under a strain of $\epsilon_{xx} = \epsilon_{yy}$,

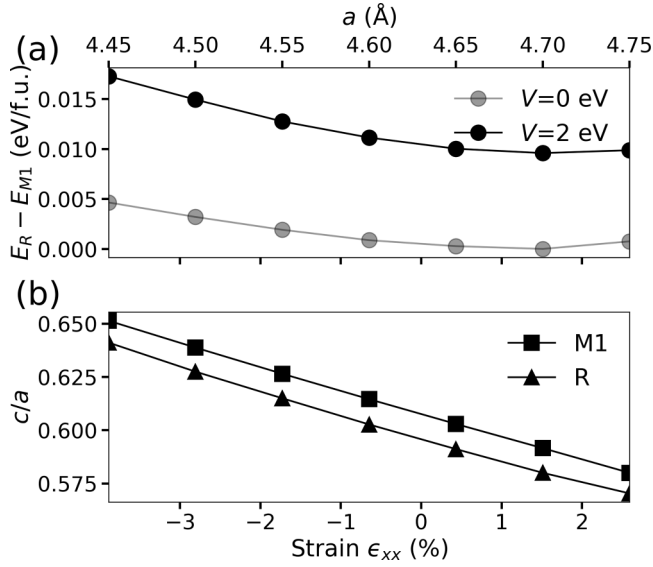


FIG. 2. (a) Energy difference between the R and M1 structures as a function of strain, $\epsilon_{xx} = \epsilon_{yy}$, calculated with (without) the $+V$ correction in black (gray). (b) The c/a ratio as a function of strain for the R and M1 phases marked by triangles and squares, respectively. Strain is defined with respect to the relaxed R phase lattice parameter, $a_0 = 4.630$ Å.

i.e., maintaining the quasitetragonal symmetry of the supercell lattice vectors, $b = 2a$, and allowing the cell to relax along the c axis, mimicking the epitaxial strain conditions of a thin film grown on a (001)-oriented square-lattice substrate, e.g., TiO_2 [see Fig. 1(f)]. This orients the vanadium chains along the growth direction, perpendicular to the in-plane orientation of the film.

In Fig. 2(a), we show the difference in total energies of both phases as a function of epitaxial strain in an experimentally relevant range. We find that the M1 phase is lower in energy than R for all strains, with the energy difference increasing under compressive and decreasing under moderate tensile strain, with a minimal energy difference at around $\epsilon_{xx} = 2\%$ (given in reference to the R phase). We also verify this result using standard DFT calculations without an intersite correction, i.e., $V = 0$ eV, for which the energy difference between the two phases is significantly smaller but shows an identical trend with strain. This highlights the robustness of the trends observed in this work, where although different values of V or different functionals may give quantitatively different results, the qualitative trends are expected to hold.

Indeed, if we interpret the relative phase stability as a proxy for the transition temperature, T_c , our results imply that increasing the basal plane lattice constant, a , decreases the stability of the M1 phase, matching the experimentally observed reduction in T_c under tensile epitaxial strain observed in thin films grown along (001) [23,25,26]. We discuss this effect in more detail in Sec. III B.

Additionally, as shown in Fig. 2(b), increasing the in-plane lattice parameter a leads to a shortening along c . Notably, the M1 phase always exhibits a larger c/a ratio than the R phase. Therefore, naturally, such tensile in-plane strain results in an interplay of two effects: not only the direct impact of the

increase in in-plane lattice parameter a , but also the shortening of c due to the Poisson effect. Previously, it has been suggested that it is the latter that is the main factor determining the change in T_c [23,26]. In the following, in order to clearly identify the relevant control parameter, we hence consider these two strains independently.

B. General tetragonal strain

To facilitate a deeper insight into the effect of epitaxial strain and to disentangle the effects caused by changes in different lattice directions, we now discuss the behavior of the R and M1 phases in VO_2 under a general tetragonal strain (i.e., with $b = 2a$) but treating the a and c lattice parameters as independent variables (i.e., independently applying $\epsilon_{xx} = \epsilon_{yy}$ and ϵ_{zz}). Although difficult to achieve experimentally, such strain variation allows us to disentangle the effects of the a and c lattice parameters individually.

In Fig. 3(a), we show the energy difference between the R and M1 phases as a function of a and c . We show a range of values around the equilibrium a and c of the R and M1 phases (triangular and square markers), also including the c values corresponding to the minimal energy for a given a (indicated by the white stripes). In this whole strain range, and for this choice of exchange-correlation functional and V value, the M1 phase is always lower in energy compared to R. With increasing c , the M1 phase becomes more energetically favorable for all a [lighter color for increasing c in Fig. 3(a)]. This is consistent with the observation of a larger c lattice parameter of the M1 phase compared to the R phase [as seen in Fig. 2(b) and indicated by the white stripes showing the same data, i.e., $c(a)$, in Fig. 3(a)]. Notably, we also observe the M1 phase becoming more favorable at both larger and smaller values of a than its equilibrium value [Fig. 3(a), lighter color at both large and small a values], although this effect appears weaker compared to the dependence on c .

To understand the origin of these trends, we now examine two different cuts through the a - c diagram shown in Fig. 3(a) for fixed values of $a = 4.6$ Å and $c = 2.85$ Å, respectively [Cuts 1 and 2 in Fig. 3(a)]. This analysis allows us to disentangle the two effects previously combined in Sec. III A, and to study the corresponding changes in structural and electronic properties in more detail. We first consider the nearest-neighbor V-V distance [Fig. 3(b)] and the ratios of these distances to c [Fig. 3(c)].

Along Cut 1, i.e., variation along the dimerization axis, c , VO_2 shows an increase in all nearest-neighbor V-V distances, also for the SB distance in M1 [Fig. 3(b), left, all lines increasing as c increases]. However, although the absolute V-V distances increase, the M1 SB bond length decreases relative to c , i.e., $r_{\text{SB}} = d_{\text{SB}}/c$ decreases [Fig. 3(c), left, square markers trending down as c increases], suggesting a strengthening of dimerization with increasing c .

Along Cut 2, i.e., basal plane strain at fixed c , the effects are more subtle. The nearest-neighbor distance remains largely unchanged [Fig. 3(b), right, all lines remain constant], except for a very weak upward trend in d_{LB} and r_{LB} related to an increase of the zigzag distortion (not shown). This is caused by the expansion in the basal plane and affects mainly the long bond.

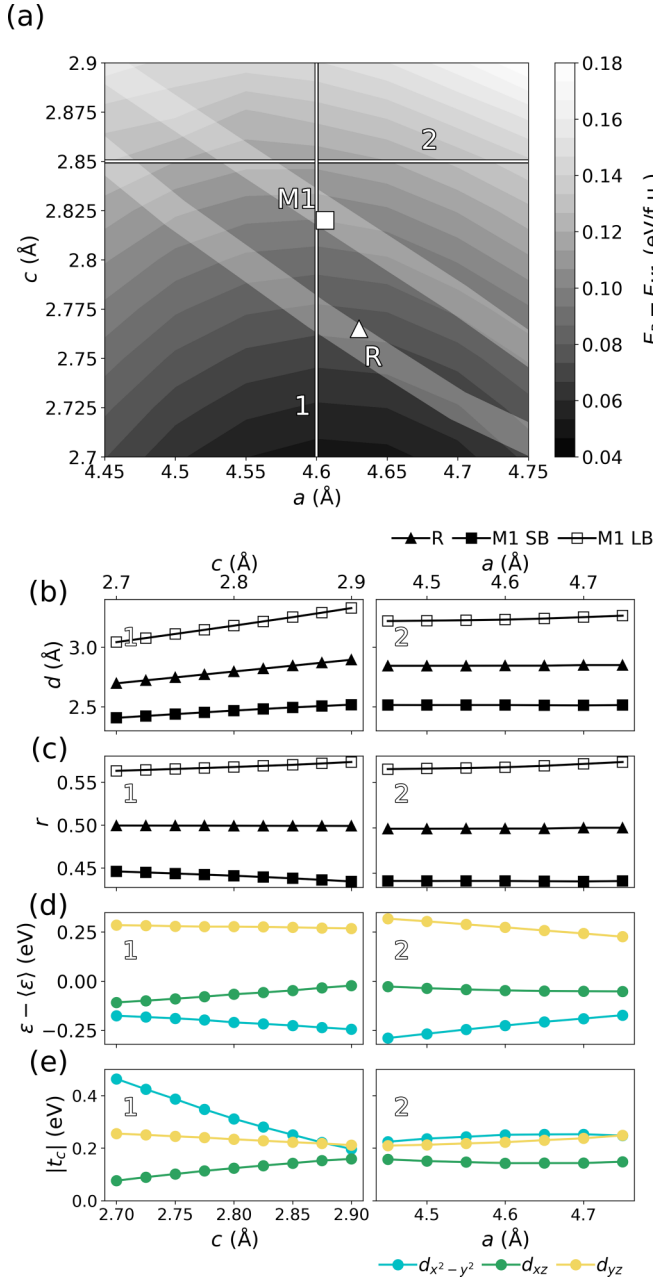


FIG. 3. (a) Energy difference between the R and M1 phases. Triangle (square) indicates the relaxed R (M1) lattice parameters with white stripes indicating the calculated $c(a)$ shown previously in Fig. 2(b). Panels (b)–(e) show different quantities obtained along Cut 1 (left panels) and Cut 2 (right panels) as indicated in panel (a). (b) Nearest-neighbor V-V distance, d , as a function of c or a lattice parameter, and (c) ratio of the nearest-neighbor V-V distance to c , $r = d/c$. Triangular markers indicate R and filled (empty) square markers indicate M1 SB (LB). (d) Energy levels, ϵ , relative to the average energy level, $\langle \epsilon \rangle$, and (e) nearest-neighbor hopping amplitudes along c , t_c , both corresponding to $d_{x^2-y^2}$ (cyan), d_{xz} (green), and d_{yz} (yellow) orbitals, as function of a or c in the R phase.

To rationalize the energetic competition between the R and M1 phases, we now draw on a simplified Peierls-like model, where a periodic structural displacement modulates the hopping amplitude between neighboring sites, leading to

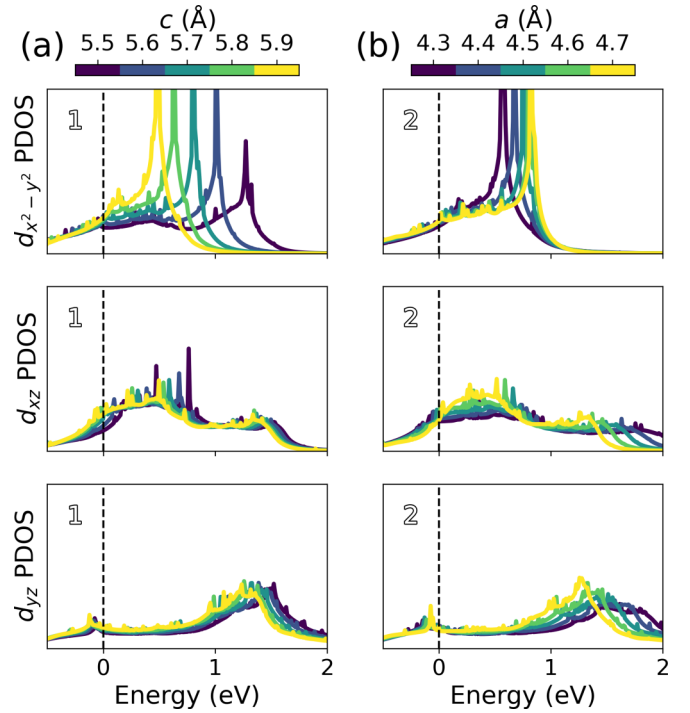


FIG. 4. Evolution of the PDOS of the different t_{2g} orbitals within the R phase (a) along Cut 1 and (b) along Cut 2. Color gradient indicates increasing lattice parameter. The vertical dashed line indicates the Fermi level.

an energy gap and a reduction in total energy. The key ingredients thereby are the hopping amplitude between neighboring atoms and the stiffness of the underlying crystal lattice. In VO_2 , the quasi-one-dimensional character of the $d_{x^2-y^2}$ orbital along the c axis makes this framework a useful approximation.

Whether the distortion becomes energetically favorable depends on the balance between two effects [52]. An energy gain arises from electronic stabilization due to gap formation related to the difference in the hopping amplitudes of the SB and LB, which can be related to the slope of the hopping with respect to interatomic distances. On the other hand, the distortion also leads to an energy cost that depends on the stiffness of the phonon mode related to the structural dimerization. Furthermore, in the multiband case realized in VO_2 , the small deviation from half-filling of the dimerizing $d_{x^2-y^2}$ orbital is expected to be relevant. To gain insight into these effects, we track the microscopic electronic observables, particularly the energy levels of the different t_{2g} orbitals [Fig. 3(d)], the corresponding hopping amplitudes, t_c , between nearest neighbors along the dimerization direction [Fig. 3(e)], and the projected densities of states (PDOS) (Fig. 4), all in the undimerized R phase.

Along Cut 1, the increasing nearest-neighbor V-V distances cause an energy lowering of the $d_{x^2-y^2}$ orbital compared to the other two orbitals [Fig. 3(d) left, cyan line coming down in energy]. This can be rationalized by the fact that the $d_{x^2-y^2}$ orbital points directly in between the oxygen ligands along c , and thus moving these ligands further away by increasing c is expected to lower its energy. A similar, albeit much weaker, energy lowering can also be expected for the d_{xz} orbital, which

can appear as an increase if taken relative to the average orbital energy. The lowering of the $d_{x^2-y^2}$ orbital can support the depletion of the other two orbitals in favor of a half-filling of the $d_{x^2-y^2}$ orbital, and thus further stabilize the M1 phase, even though the PDOS of the R phase [Fig. 4(a), left] does not indicate any noticeable changes in the orbital fillings for increasing c . On the other hand, the hopping amplitude between the $d_{x^2-y^2}$ orbitals along c decreases strongly with increasing c [Fig. 3(c), left]. This effect is also visible as a narrowing of the corresponding PDOS [Fig. 4(a), left]. If one makes the reasonable assumption that a smaller hopping amplitude also results in a smaller slope with respect to the V-V distance (e.g., if one assumes a typical power-law dependence), one would in fact expect a smaller electronic energy gain from the dimerization at larger c , and thus a disfavoring of the M1 phase. However, the decrease of the hopping amplitude with c in Fig. 3(c) appears close to linear, and thus its slope remains nearly constant, suggesting that this effect might be weak. Furthermore, an increase in c is also expected to weaken the interionic forces and thus to reduce the elastic stiffness opposing the structural dimerization. We thus conclude that the observed stabilization of the M1 phase for an elongated c lattice parameter results from the interplay between different electronic and structural effects, with the softening of the structural stiffness potentially being the decisive factor.

Along Cut 2, changing the basal plane lattice parameter a for constant c also shifts the energy of the $d_{x^2-y^2}$ orbital relative to the other two orbitals, but now the $d_{x^2-y^2}$ orbital is lowered for decreasing a [Fig. 3(d), right, cyan line]. Regarding the hopping amplitudes, while the hopping amplitudes in the basal plane (not shown) show the expected behavior (increasing with decreasing a), there is only a very weak effect on the hopping amplitudes along c , even though we observe a subtle decrease in t_c corresponding to the $d_{x^2-y^2}$ orbital for small a [Fig. 3(c), right, cyan line]. A potential explanation for the stabilization of the M1 phase for small a could therefore be related to the lowering of the $d_{x^2-y^2}$ orbital, while the stabilization for large a could again be due to a general softening of the structural stiffness, in this case affecting mostly the zigzag component of the structural distortion. However, these trends are weak and thus also more subtle effects not considered here could come into play.

Based on this analysis, we can now also interpret the epitaxial strain results discussed in Sec. III A. Under tensile strain, we observe that the increase in a is accompanied by a contraction in c [Fig. 2(b)]. This shortening of the c axis increases the hopping amplitude, which could, in principle, favor the Peierls distortion, but it also increases the stiffness opposing the Peierls distortion. The stabilization of the M1 phase under c -axis compression is thus governed by the interplay of two opposing tendencies, whereby the softening of the stiffness appears to dominate. Furthermore, the observed reduction in the energy difference between R and M1 under tensile strain primarily reflects the effect of c -axis compression rather than the in-plane expansion, even though both effects are interdependent.

C. Orthorhombic strain

Finally, we also consider a strain condition that breaks the tetragonal symmetry of the in-plane lattice parameters,

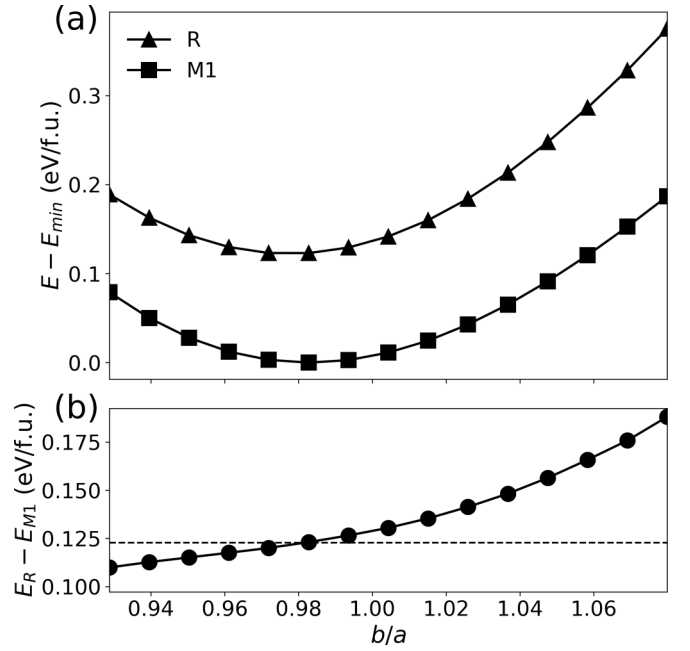


FIG. 5. (a) Energies of the R and M1 phases as a function of the b/a ratio, indicated by triangular and square markers, respectively, with a and c strained to values corresponding to a (010)-oriented TiO_2 substrate. (b) Energy difference between the two phases, with the dashed line indicating the corresponding energy difference at $c = c(\text{TiO}_2)$ and the corresponding equilibrium a .

corresponding to VO_2 epitaxially grown with (010) orientation, i.e., with the vanadium dimerization axis lying in plane [see Fig. 1(g)]. This approach complements our previous results by generalizing them to a less symmetric scenario with $\epsilon_{xx} \neq \epsilon_{yy}$ and ϵ_{zz} , aiming to verify the simplified picture above.

To make this strain setup experimentally relevant, we perform the analysis at the strain values imposed by a (010)-oriented TiO_2 substrate, a commonly used rutile substrate for VO_2 [23,27]. Thus, we fix the a and c lattice parameters of our supercell to strains of 0.863% and 3.62%, respectively, corresponding to the nominal lattice mismatch of VO_2 grown on the (010) surface of TiO_2 [23], and we then vary the b lattice parameter. The corresponding strained a and c lattice parameter values as well as the b value minimizing the total energy under this constraint for the R phase are listed in Table I.

In Fig. 5(a), we show the energy of the R and M1 phases as a function of b/a in the above-described strain regime. Due to the tensile strain in both c and a , both curves feature a minimum at $b/a < 1$. The positions of the two minima almost coincide around $b/a = 0.98$, with the R phase minimum located at a slightly smaller b/a as that of the M1. In Fig. 5(b), we also show the energy difference between the two structures as a function of b/a . Similar to what we observed previously for tetragonal symmetry, the M1 phase is always lower in energy.

To quantify the effect of the symmetry breaking due to the different strain values along a and b , we also include the energy difference between R and M1 obtained under the

tetragonal strain condition from Sec. III B taken at the same c -axis strain of 3.62% and the values of a that minimize the total energy for that c (which are nearly identical for R and M1). We observe that this energy difference [dashed line in Fig. 5(b)] matches almost exactly that of the orthorhombic strain at $b/a = 0.98$ [dashed line crosses black line at exactly 0.98 in Fig. 5(b)]. This indicates that even in the lower orthorhombic symmetry, with different strains along the two basal plane directions, the energy difference between R and M1 is mainly determined by the c -axis strain, validating the assumptions made in, e.g., Refs. [23,27].

IV. SUMMARY AND OUTLOOK

In this work, we have presented a systematic study of how strain affects the relative energetic stability of the metallic R and insulating M1 phases of VO₂, considering both structural and electronic effects arising from different strain geometries, including strain along the dimerization direction as well as strain within the basal plane. Strain represents an important tuning parameter for the transition in VO₂, and the DFT + V methodology [37] allows us to treat the R and M1 phases on the same footing and obtain energy differences between them as a function of strain.

We first examine the effects of coherent epitaxy within the (001) plane. We show that increasing in-plane strain leads to a reduction of the c/a ratio, and a decreased stability of the M1 phase. This behavior, driven predominantly by the shortening of the c axis, reflects the suppression of the Peierls-distorted phase and, if considered as proxy for the transition temperature, aligns with the experimental trends of a lower T_c under tensile (001) strain.

We then explore the effect of varying the a and c lattice parameters independently, observing that the M1 phase is energetically favored at large c , and at both large and small a . We analyze the changes in the nearest-neighbor V-V distances and the R phase electronic structure, and discuss several factors that can favor or disfavor the Peierls-distorted phase. While

the M1 phase stabilization at large c depends on an interplay between the $d_{x^2-y^2}$ orbital filling, the c -direction hopping amplitude, and the stiffness of the underlying lattice, our results indicate that the former two effects are weak, and it is the structural stiffness that is the decisive factor. Additionally, the fact that we observe an effect of the basal plane strain at all highlights the multidimensional character of VO₂.

Finally, we also investigate a symmetry-breaking case corresponding to an epitaxial constraint along a and c , resulting in $a \neq b$. We note that we do not observe large differences compared to the tetragonal strain case. In particular, we confirm that this regime is also mainly dominated by the strain in the c direction (now oriented in plane).

From our analysis of the three different strain scenarios, we can thus conclude that the strain along the dimerization axis c indeed plays the central role in tuning the relative stability of the R and M1 phases. While strain that changes a also affects the phase stability, its main effect is mediated through the resulting change in c , particularly in the epitaxial case. Altogether, our results highlight that although the basal plane strain also seems to affect the structural dimerization to some extent, hinting at the three-dimensional character of VO₂, both structurally and electronically, it is the c -axis strain, whether applied directly or indirectly, that plays the dominant role in tuning the MIT in VO₂. Strain engineering strategies should thus target the dimerization direction of the material, even at the cost of imposing nonessential basal plane strains.

ACKNOWLEDGMENTS

The authors thank A. M. Ionescu and his group for valuable discussions. This work was supported by ETH Zürich and the Swiss National Science Foundation (Grant No. 209454). Calculations were performed on the ETH Zürich Euler cluster.

DATA AVAILABILITY

The supporting data for this article are openly available on the Materials Cloud Archive [53].

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- [1] Z. Yang, C. Ko, and S. Ramanathan, Oxide electronics utilizing ultrafast metal-insulator transitions, *Annu. Rev. Mater. Res.* **41**, 337 (2011).
 - [2] K. Liu, S. Lee, S. Yang, O. Delaire, and J. Wu, Recent progresses on physics and applications of vanadium dioxide, *Mater. Today* **21**, 875 (2018).
 - [3] R. Shi, N. Shen, J. Wang, W. Wang, A. Amini, N. Wang, and C. Cheng, Recent advances in fabrication strategies, phase transition modulation, and advanced applications of vanadium dioxide, *Appl. Phys. Rev.* **6**, 011312 (2019).
 - [4] Y. Xue and S. Yin, Element doping: A marvelous strategy for pioneering the smart applications of VO₂, *Nanoscale* **14**, 11054 (2022).
 - [5] J. Cao, Y. Gu, W. Fan, L. Q. Chen, D. F. Ogletree, K. Chen, N. Tamura, M. Kunz, C. Barrett, J. Seidel, and J. Wu, Extended mapping and exploration of the vanadium dioxide stress-temperature phase diagram, *Nano Lett.* **10**, 2667 (2010).
 - [6] J. M. Atkin, S. Berweger, E. K. Chavez, M. B. Raschke, J. Cao, W. Fan, and J. Wu, Strain and temperature dependence of the insulating phases of VO₂ near the metal-insulator transition, *Phys. Rev. B* **85**, 020101(R) (2012).
 - [7] Z. Shao, X. Cao, H. Luo, and P. Jin, Recent progress in the phase-transition mechanism and modulation of vanadium dioxide materials, *NPG Asia Mater.* **10**, 581 (2018).
 - [8] V. Eyert, The metal-insulator transitions of VO₂: A band theoretical approach, *Ann. Phys.* **514**, 650 (2002).
 - [9] J. B. Goodenough, The two components of the crystallographic transition in VO₂, *J. Solid State Chem.* **3**, 490 (1971).
 - [10] J. P. Pouget, H. Launois, J. P. D'Haenens, P. Merenda, and T. M. Rice, Electron localization induced by uniaxial stress in pure VO₂, *Phys. Rev. Lett.* **35**, 873 (1975).
 - [11] A. Zylbersztein and N. F. Mott, Metal-insulator transition in vanadium dioxide, *Phys. Rev. B* **11**, 4383 (1975).

- [12] R. M. Wentzcovitch, W. W. Schulz, and P. B. Allen, VO₂: Peierls or Mott-Hubbard? A view from band theory, *Phys. Rev. Lett.* **72**, 3389 (1994).
- [13] T. M. Rice, H. Launois, and J. P. Pouget, Comment on “VO₂: Peierls or Mott-Hubbard? A view from band theory”, *Phys. Rev. Lett.* **73**, 3042 (1994).
- [14] S. Biermann, A. Poteryaev, A. I. Lichtenstein, and A. Georges, Dynamical singlets and correlation-assisted Peierls transition in VO₂, *Phys. Rev. Lett.* **94**, 026404 (2005).
- [15] C. Weber, D. D. O’Regan, N. D. M. Hine, M. C. Payne, G. Kotliar, and P. B. Littlewood, Vanadium dioxide: A Peierls-Mott insulator stable against disorder, *Phys. Rev. Lett.* **108**, 256402 (2012).
- [16] W. H. Brito, M. C. O. Aguiar, K. Haule, and G. Kotliar, Metal-insulator transition in VO₂: A DFT + DMFT perspective, *Phys. Rev. Lett.* **117**, 056402 (2016).
- [17] Y. Chen, S. Zhang, F. Ke, C. Ko, S. Lee, K. Liu, B. Chen, J. W. Ager, R. Jeanloz, V. Eyert, and J. Wu, Pressure-temperature phase diagram of vanadium dioxide, *Nano Lett.* **17**, 2512 (2017).
- [18] Y. A. Birkhölzer, K. Sotthewes, N. Gauquelin, L. Riekehr, D. Jannis, E. van der Minne, Y. Bu, J. Verbeeck, H. J. W. Zandvliet, G. Koster, and G. Rijnders, High-strain-induced local modification of the electronic properties of VO₂ thin films, *ACS Appl. Electron. Mater.* **4**, 6020 (2022).
- [19] S. Zhang, J. Y. Chou, and L. J. Lauhon, Direct correlation of structural domain formation with the metal insulator transition in a VO₂ nanobeam, *Nano Lett.* **9**, 4527 (2009).
- [20] J. Cao, E. Ertekin, V. Srinivasan, W. Fan, S. Huang, H. Zheng, J. W. L. Yim, D. R. Khanal, D. F. Ogletree, J. C. Grossman, and J. Wu, Strain engineering and one-dimensional organization of metal-insulator domains in single-crystal vanadium dioxide beams, *Nat. Nanotechnol.* **4**, 732 (2009).
- [21] J. H. Park, J. M. Coy, T. S. Kasirga, C. Huang, Z. Fei, S. Hunter, and D. H. Cobden, Measurement of a solid-state triple point at the metal-insulator transition in VO₂, *Nature (London)* **500**, 431 (2013).
- [22] H. Asayesh-Ardakani, A. Nie, P. M. Marley, Y. Zhu, P. J. Phillips, S. Singh, F. Mashayek, G. Sambandamurthy, K.-b. Low, R. F. Klie, S. Banerjee, G. M. Odegard, and R. Shahbazian-Yassar, Atomic origins of monoclinic-tetragonal (rutile) phase transition in doped VO₂ nanowires, *Nano Lett.* **15**, 7179 (2015).
- [23] Y. Muraoka and Z. Hiroi, Metal-insulator transition of VO₂ thin films grown on TiO₂ (001) and (110) substrates, *Appl. Phys. Lett.* **80**, 583 (2002).
- [24] T.-H. Yang, R. Aggarwal, A. Gupta, H. Zhou, R. J. Narayan, and J. Narayan, Semiconductor-metal transition characteristics of VO₂ thin films grown on c- and r-sapphire substrates, *J. Appl. Phys.* **107**, 053514 (2010).
- [25] N. B. Aetukuri, A. X. Gray, M. Drouard, M. Cossale, L. Gao, A. H. Reid, R. Kukreja, H. Ohldag, C. A. Jenkins, E. Arenholz, K. P. Roche, H. A. Dürr, M. G. Samant, and S. S. P. Parkin, Control of the metal-insulator transition in vanadium dioxide by modifying orbital occupancy, *Nat. Phys.* **9**, 661 (2013).
- [26] N. F. Quackenbush, J. W. Tashman, J. A. Mundy, S. Sallis, H. Paik, R. Misra, J. A. Moyer, J.-H. Guo, D. A. Fischer, J. C. Woicik, D. A. Muller, D. G. Schlom, and L. F. J. Piper, Nature of the metal insulator transition in ultrathin epitaxial vanadium dioxide, *Nano Lett.* **13**, 4857 (2013).
- [27] N. F. Quackenbush, H. Paik, M. J. Wahila, S. Sallis, M. E. Holtz, X. Huang, A. Ganose, B. J. Morgan, D. O. Scanlon, Y. Gu, F. Xue, *et al.*, Stability of the M2 phase of vanadium dioxide induced by coherent epitaxial strain, *Phys. Rev. B* **94**, 085105 (2016).
- [28] D. Lee, J. Lee, K. Song, F. Xue, S.-Y. Choi, Y. Ma, J. Podkaminer, D. Liu, S.-C. Liu, B. Chung, W. Fan, S. J. Cho, W. Zhou, J. Lee, L.-Q. Chen, S. H. Oh, Z. Ma, and C.-B. Eom, Sharpened VO₂ phase transition via controlled release of epitaxial strain, *Nano Lett.* **17**, 5614 (2017).
- [29] S. Fischer, J.-O. Krispeneit, M. Foerster, L. Aballe, J. Falta, and J. I. Flege, Massively strained VO₂ thin film growth on RuO₂, *Cryst. Growth Des.* **20**, 2734 (2020).
- [30] D. Lee, T. Min, J. Kim, S. Song, J. Lee, H. Kang, J. Lee, D.-Y. Cho, J. Lee, J. H. Jang, and S. Park, Octahedral symmetry modification induced orbital occupancy variation in VO₂, *J. Phys. Chem. Lett.* **13**, 75 (2022).
- [31] B. Lazarovits, K. Kim, K. Haule, and G. Kotliar, Effects of strain on the electronic structure of VO₂, *Phys. Rev. B* **81**, 115117 (2010).
- [32] H. Lu, S. Clark, Y. Guo, and J. Robertson, Modelling the enthalpy change and transition temperature dependence of the metal-insulator transition in pure and doped vanadium dioxide, *Phys. Chem. Chem. Phys.* **22**, 13474 (2020).
- [33] P. Mlkvik, C. Ederer, and N. A. Spaldin, Influence of germanium substitution on the structural and electronic stability of the competing vanadium dioxide phases, *Phys. Rev. Res.* **4**, 043129 (2022).
- [34] Y. Zhang, D. Ke, J. Wu, C. Zhang, L. Hou, B. Lin, Z. Chen, J. P. Perdew, and J. Sun, Challenges for density functional theory in simulating metal-metal singlet bonding: A case study of dimerized VO₂, *J. Chem. Phys.* **160**, 134101 (2024).
- [35] B. Stahl and T. Bredow, Critical assessment of the DFT + *U* approach for the prediction of vanadium dioxide properties, *J. Comput. Chem.* **41**, 258 (2020).
- [36] P. Mlkvik, M. E. Merkel, N. A. Spaldin, and C. Ederer, Single-site DFT + DMFT for vanadium dioxide using bond-centered orbitals, *Phys. Rev. Res.* **6**, 033122 (2024).
- [37] L. Haas, P. Mlkvik, N. A. Spaldin, and C. Ederer, Incorporating static intersite correlation effects in vanadium dioxide through DFT + *V*, *Phys. Rev. Res.* **6**, 043177 (2024).
- [38] V. Leiria Campo, Jr. and M. Cococcioni, Extended DFT + *U* + *V* method with on-site and inter-site electronic interactions, *J. Phys.: Condens. Matter* **22**, 055602 (2010).
- [39] J. M. Tomczak and S. Biermann, Effective band structure of correlated materials: the case of VO₂, *J. Phys.: Condens. Matter* **19**, 365206 (2007).
- [40] J. M. Tomczak, F. Aryasetiawan, and S. Biermann, Effective bandstructure in the insulating phase versus strong dynamical correlations in metallic VO₂, *Phys. Rev. B* **78**, 115103 (2008).
- [41] J. M. Tomczak and S. Biermann, Optical properties of correlated materials: Generalized Peierls approach and its application to VO₂, *Phys. Rev. B* **80**, 085117 (2009).
- [42] A. S. Belozarov, M. A. Korotin, V. I. Anisimov, and A. I. Poteryaev, Monoclinic M1 phase of VO₂: Mott-Hubbard versus band insulator, *Phys. Rev. B* **85**, 045109 (2012).
- [43] Z. He and A. J. Millis, Photoinduced phase transitions in narrow-gap Mott insulators: The case of VO₂, *Phys. Rev. B* **93**, 115126 (2016).

- [44] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [45] I. Timrov, N. Marzari, and M. Cococcioni, HP—A code for the calculation of Hubbard parameters using density-functional perturbation theory, *Comput. Phys. Commun.* **279**, 108455 (2022).
- [46] J. Hubbard, Electron correlations in narrow energy bands, *Proc. R. Soc. Lond. Ser. Math. Phys. Sci.* **276**, 238 (1963).
- [47] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, *et al.*, QUANTUM ESPRESSO: A modular and open-source software project for quantum simulations of materials, *J. Phys.: Condens. Matter* **21**, 395502 (2009).
- [48] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunau, M. Buongiorno Nardelli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni, N. Colonna, I. Carnimeo, A. Dal Corso, S. de Gironcoli, P. Delugas, R. A. DiStasio, A. Ferretti, A. Floris, G. Fratesi, G. Fugallo, *et al.*, Advanced capabilities for materials modelling with Quantum ESPRESSO, *J. Phys.: Condens. Matter* **29**, 465901 (2017).
- [49] K. F. Garrity, J. W. Bennett, K. M. Rabe, and D. Vanderbilt, Pseudopotentials for high-throughput DFT calculations, *Comput. Mater. Sci.* **81**, 446 (2014).
- [50] A. A. Mostofi, J. R. Yates, G. Pizzi, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, An updated version of wannier90: A tool for obtaining maximally-localised Wannier functions, *Comput. Phys. Commun.* **185**, 2309 (2014).
- [51] G. Pizzi, V. Vitale, R. Arita, S. Blügel, F. Freimuth, G. Géranton, M. Gibertini, D. Gresch, C. Johnson, T. Koretsune, J. Ibañez-Azpiroz, H. Lee, J.-M. Lihm, D. Marchand, A. Marrazzo, Y. Mokrousov, J. I. Mustafa, Y. Nohara, Y. Nomura, L. Paulatto, *et al.*, Wannier90 as a community code: new features and applications, *J. Phys.: Condens. Matter* **32**, 165902 (2020).
- [52] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Soliton excitations in polyacetylene, *Phys. Rev. B* **22**, 2099 (1980).
- [53] P. Mlkvik, L. Geistlich, N. A. Spaldin, and C. Ederer, Effects of strain on the stability of the metallic rutile and insulating M1 phases of vanadium dioxide, Materials Cloud Archive (2026), <https://doi.org/10.24435/materialscloud:bt-ah>.