

Current-induced successive structural phase transitions beyond thermal equilibrium in single-crystal VO₂

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In solids, nonequilibrium systems driven by external energy sources, such as electric current, host unexplored physics; however, phase transitions beyond thermal equilibrium remain elusive. Here, we show that electric current induces structural phase transitions in single-crystal VO₂. Synchrotron x-ray diffraction at room temperature reveals that a current density of 6.5 A/cm² disrupts V-V dimers, producing a monoclinic-to-tetragonal insulator-to-metal transition independent of Joule heating. Further increasing the current to 10 A/cm² triggers a discontinuous isotropic lattice expansion, stabilizing a tetragonal structure absent in thermal equilibrium. Optical microscopy and Raman spectroscopy further reveal dynamic domain motion and metastable phases. These results demonstrate direct access to hidden phases and symmetry changes beyond thermal equilibrium.

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Introduction. Nonequilibrium systems can generate macroscopic order sustained by continuous energy fluxes and entropy production. While equilibrium frameworks have provided deep insights into quantum phases in solids, the study of nonequilibrium phenomena remains limited [1]. A key question is whether crystalline solids driven far from thermal equilibrium can transition into steady nonequilibrium phases accompanied by symmetry breaking and dynamic dissipative structures. Electric current provides a direct route to induce such states, with nonlinear responses serving as hallmarks of nonequilibrium behavior. However, despite extensive studies reporting nonlinear conductivity and structural changes in materials such as organic salts [2,3] and transition-metal oxides [4–9], clear experimental evidence of current-induced structural phase transitions beyond thermal effects has remained elusive.

Vanadium dioxide (VO₂) is a prototypical strongly correlated electron system, renowned for its insulator-to-metal transition near 340 K upon heating [10–17], in which the breaking of V-V dimers drives a symmetry change from a monoclinic to a tetragonal structure. The mechanism of the insulator-to-metal transition remains controversial, involving molecular orbital formation [18–22], electron correlation

[23–25], and electron-lattice interaction [26–28]. Despite its simple composition and structure, VO₂ exhibits diverse electronic properties under chemical substitution [29,30] and external fields [31–41]. At room temperature, dramatic structural and electronic changes occur under light irradiation [31–35] and high magnetic fields [36], suggesting the disappearance of V-V dimers. However, these effects manifest in transient excited states rather than in steady states. Another intriguing phenomenon is nonlinear conduction under applied electric fields [37–39] and currents [40,41]. Previous studies have reported an electric-field-induced monoclinic-to-tetragonal transition in single-crystal [37], thin-film [39,42–44], and nanobeam samples [45,46]. However, since these transitions are driven by strong electric fields ($E \gg 10$ kV/cm), it was difficult to rule out the effect of Joule heating, raising doubts about whether the transitions are genuine nonequilibrium phase transitions.

Recent studies employing direct current have unveiled a distinct insulator-to-metal transition in single-crystal VO₂ [41], revealing a novel metallic state fundamentally different from Joule-heating-induced metallic phase under an electric field [37–39,42–46]. Notably, because Joule heating diminishes as resistance decreases under constant current, this approach mitigates thermal artifacts and opens a new avenue to explore genuine nonequilibrium phase transitions. A crucial question thus arises: Does this current-induced transition involve an intrinsic change in crystal symmetry, signaling the emergence of a truly nonequilibrium structural phase?

In this Letter, we report current-induced structural phase transitions beyond thermal equilibrium in single-crystal VO₂, observed via x-ray diffraction (XRD), optical microscopy, and microscopic Raman spectroscopy. Our findings demonstrate

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that these transitions occur independently of Joule heating and shed light on the intrinsic structure of nonequilibrium phases in crystalline solids.

Methods. Single crystals of VO₂ were grown by melting V₂O₅ at 950 °C under N₂ gas flow conditions (4 l/min) for 3 days by modifying the growth method in Ref. [47]. The single crystals used in this study were the same as those in Ref. [41]. A single crystal with dimensions of $2.51 \times 0.177 \times 0.129$ mm³ was used for the XRD experiments. The synchrotron XRD experiments were performed on BL02B1 at SPring-8, Japan [48]. The diameter of the incident x-ray beam was approximately 0.2 mm. An N₂-gas-blowing device was employed to control the sample temperature. A two-dimensional detector CdTe PILATUS was used to record the diffraction pattern. The x-ray wavelength was $\lambda = 0.30868$ Å. The lattice parameters and crystal orientations were calculated using the CrysAlisPro program [49]. The resistivity along the c_t axis was measured using the standard four-probe dc method. Several external currents were applied using a current source (KEITHLEY 6221). Note that we used dc currents instead of dc voltages for the control parameter to avoid thermal runaway associated with nonlinear conduction. To reduce contact resistance, which may be an extrinsic origin of nonlinear conduction due to Joule heating, we deposited a thick gold layer (~100 nm) on the sample surface before bonding the gold wires by using Ag paste. Raman spectra were measured using a Renishaw inVia microscope with a 633 nm He-Ne laser in a backscattering geometry. The incident light was polarized perpendicular to the c_t axis, with a spot size of ~2 μm and a power of 40 μW, which did not affect the sample temperature or domain structure.

Results and discussion. Figure 1 shows the measurement points in the current density versus temperature (J – T_0) phase diagram and schematic representations of the crystal structure in each phase, obtained from our synchrotron XRD experiments. Here, T_0 indicates the temperature at which the current is zero. We identified two current-induced phases, shown as the pink and green regions. Figures 2(a)–2(e) show the representative XRD data, with reciprocal lattice schematics in Fig. 2(f). At $T_0 = 300$ K and $J = 0$ A/cm², there are several Bragg peaks attributed to the M1 phase in Fig. 2(c). As the temperature increases, the number of Bragg peaks is reduced to half at 345 K [Fig. 2(e)] compared to the M1 phase, indicating a monoclinic-to-tetragonal transition to the high- T R phase with a halved unit-cell volume. It is noted that weak diffuse scattering intensity is observed along the $a_t^* + c_t^*$ direction in the high- T R structure, as shown in Fig. 2(e) and Fig. S8 in the Supplemental Material [50], corresponding to the short-range order of the V displacements along the $\langle 111 \rangle$ axes [22], as shown in Fig. 1(c).

Applying current along the c_t axis at $T_0 = 300$ K produces new peaks above $J = 6.5$ A/cm² [Fig. 2(b)], appearing only near Bragg positions of the high- T R phase. This indicates disappearance of V-V dimers and an insulator-to-metal transition. In this J region, monoclinic and tetragonal structures coexist as domains. We designate this R structure under applied current as the low- J R structure. Unlike the high- T R phase, this structure shows no pronounced diffuse scattering (Fig. S8 in the Supplemental Material [50]), suggesting weaker short-range order. Although the temperature was not

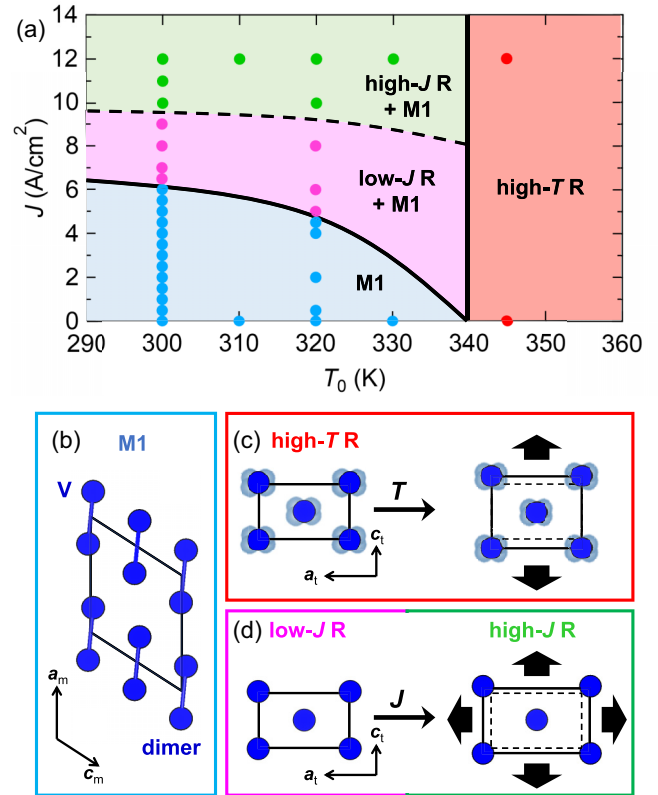


FIG. 1. (a) Current density vs temperature (J – T_0) phase diagram identified through single-crystal XRD experiments. Circles represent measurement points, with red, green, pink, and blue areas indicating different phases. (b) Schematic of the low-temperature M1 structure, in which V atoms form dimers. (c) Schematic showing the change in the crystal structure in the high- T R structure with increasing temperature. (d) Schematic showing the change in the crystal structure from the low- J R to high- J R structures under applied current. The unit-cell axes of each phase have approximate relationships of $a_m = 2c_t$, $b_m = b_t$, and $c_m = -a_t - c_t$, where the subscripts t and m in the axis labels indicate the tetragonal (R) and monoclinic (M1) lattices, respectively.

measured *in situ* during the XRD experiments, postexperiment thermography showed that the maximum sample temperature (325 K) remained below the equilibrium transition temperature $T_C = 340$ K, excluding Joule heating as the origin of the transition (Fig. S1 in the Supplemental Material [50]).

When a higher current density of $J \geq 10$ A/cm² was applied, the peaks corresponding to the low- J R structure disappeared, and new peaks emerged, as shown in Fig. 2(a) and Fig. S6 in the Supplemental Material [50], defining a high- J R structure with the same tetragonal symmetry. In Fig. 2(a), a weak residual intensity is visible near the position corresponding to the $9\ 10_R$ reflection observed in Fig. 2(b). This suggests that a small fraction of the low- J R structure persists even in the high- J region, although its contribution is minor. This phase transition at higher current densities was not observed in nonlinear conduction measurements [41], as the resistance was sufficiently low at these high current densities. At $T_0 = 345$ K and $J = 12$ A/cm², the Bragg peaks corresponding to the M1, low- J R, and high- J R structures

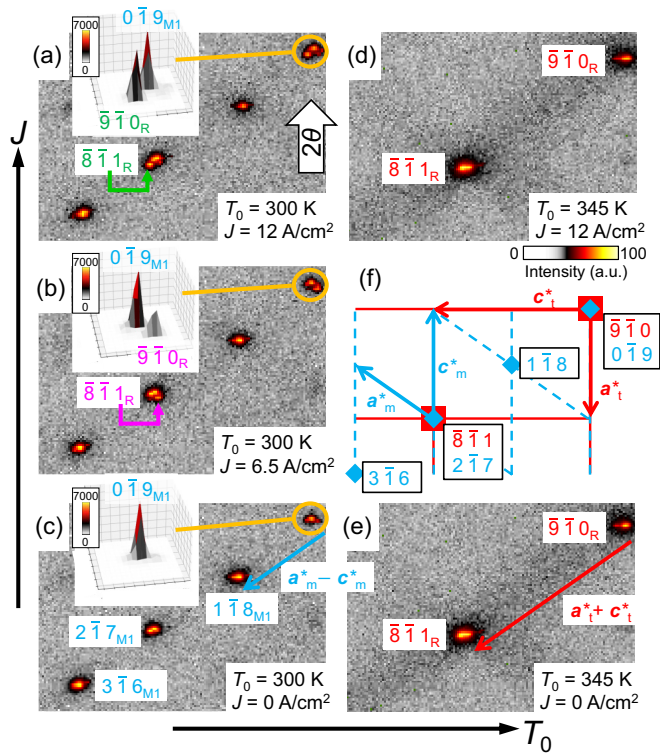


FIG. 2. (a)–(e) XRD images at various temperatures T_0 and current densities J . (f) Schematic of the reciprocal lattice points of the R and M1 lattices. Red squares and light blue diamonds denote the positions of the Bragg peaks for the R and M1 lattices, respectively.

disappear [Fig. 2(d)], resulting in a pattern almost identical to that of the single high- T R structure [Fig. 2(e)].

From the peak positions of the tetragonal lattice in each XRD dataset, we investigate the current density dependence of the lattice parameters at $T_0 = 300$ K, as shown in Figs. 3(a)–3(d). The a_t and c_t parameters of the low- J R structure (black circles in the $J = 6.5$ – 9 A/cm² range) are comparable to those in the high- T R structure at $T_0 = 345$ K (red squares), as shown in Figs. 3(a) and 3(b). In contrast, the a_t and c_t parameters of the high- J R structure are larger than those in both the high- T R and low- J R structures. Furthermore, the cell volume V_t increases sharply by 0.28(14)% during the phase transition from the low- J R to high- J R structures around $J = 10$ A/cm² [Fig. 3(c)], while the a_t/c_t ratio remains nearly unchanged [Fig. 3(d)], indicating isotropic volume expansion [Fig. 1(d)]. In contrast, temperature-driven expansion of the high- T R phase is anisotropic, with elongation along the c_t axis [Figs. 3(e), 3(f), and 1(c)]. This qualitative difference demonstrates that the high- J R structure represents a nonequilibrium phase inaccessible through heating alone.

XRD measurements indicate that current-induced R domains account for roughly 10% of the sample volume (Fig. S6 in the Supplemental Material [50]). While this does not by itself exclude filamentary Joule-heating scenarios [38,39,44–46], the observed structural response cannot be explained by simple local heating. In particular, the high- J R phase exhibits a discontinuous and isotropic lattice expansion, in contrast to the anisotropic thermal expansion of the equilibrium rutile phase (Fig. 3). Such an isotropically expanded

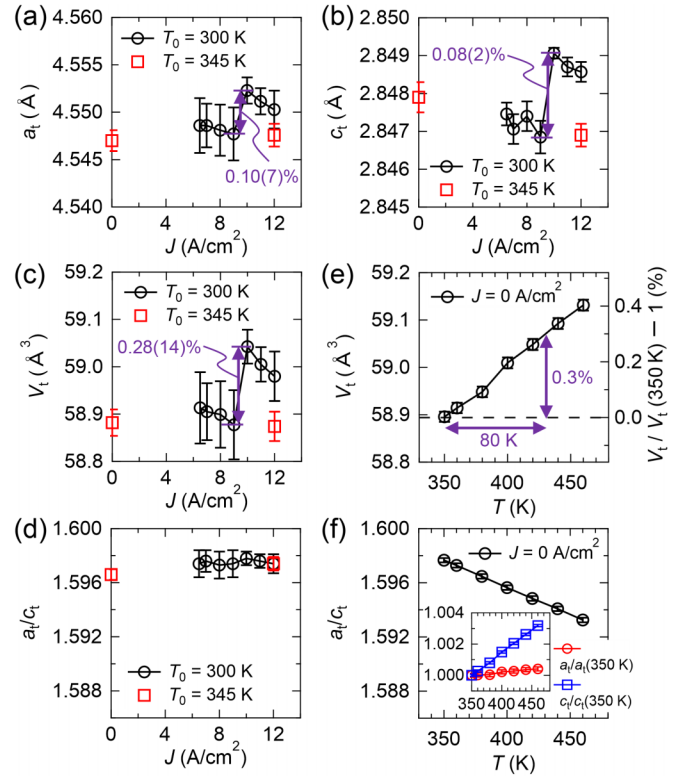


FIG. 3. (a)–(d) Current density dependence of the tetragonal lattice constants a_t , c_t , volume V_t , and the ratio of a_t/c_t . (e), (f) Temperature dependence of the tetragonal lattice parameters V_t and a_t/c_t of the high- T R structure. The inset of panel (f) shows the lattice constants a_t and c_t , normalized to 350 K. The volume expansion rate of 0.28(14)%, resulting from the low- J R to high- J R transition, corresponds to approximately 80 K in terms of thermal expansion with temperature.

tetragonal structure is not realized at any temperature in thermal equilibrium.

To track domain evolution in real space, we performed optical microscopy observations [Figs. 4(a)–4(c)]. With increasing current, dark green metallic domains emerge above $J \approx 5$ A/cm² and coexist with light green insulating domains [Fig. 4(b)], assigned to the low- J R and M1 structures, respectively. The orientation of the phase-separation boundaries appears to be unrelated to any crystallographic axis of VO₂. Although the optical microscopy observations probe only the surface state, the order of the domain fraction is consistent with that obtained from bulk-sensitive XRD measurements (Fig. S6 in the Supplemental Material [50]). This indicates that such complex domain formation is unlikely to occur only at the crystal surface. Typical domain sizes are 10–100 μ m, consistent with transport-derived correlation lengths [41]. This size range is 10–100 times larger than what is observed in thin films [38,44] and nanobeam samples [45,46].

Surprisingly, in the intermediate current range ($J = 5$ – 8.5 A/cm²), metallic domains slide along the current direction (Movie S1 in the Supplemental Material [50]). Near $J \approx 8.5$ A/cm², domain patterns become finer and more distorted [Fig. 4(c)]. In the intermediate low- J regime, the striplike domains evolve continuously in time, whereas at higher current densities they become immobilized. This behavior suggests

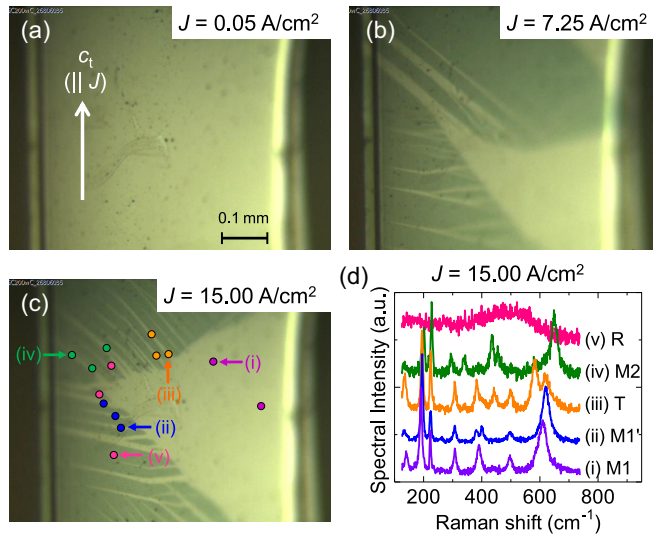


FIG. 4. (a)–(c) Current density dependence of optical microscope images at room temperature under applied currents of 0.05, 7.25, and 15.00 A/cm^2 . (d) Position dependence of the Raman spectra under an applied current of 15.00 A/cm^2 .

the presence of a thresholdlike current scale associated with the structural transition from low- J R to high- J R structures with volume expansion. The pattern appears to separate into a large insulating-phase domain and striped domains consisting of metallic and insulating phases. Domain motion ceases at higher currents, and the sample returns to the insulating state after current removal. These behaviors resemble dissipative structures in nonequilibrium systems. While current-driven domain patterns are also reported in Ca_2RuO_4 [6] and K-TCNQ [3], VO_2 shows two-step structural transitions and more complex domain morphologies, highlighting distinct nonequilibrium behavior.

To investigate the structure of each domain observed from optical micrographs, we performed Raman spectroscopy using a light spot size of 2 μm . Figure 4(d) shows the position dependence of the Raman spectra under an applied current of 15.00 A/cm^2 . Each spectrum, labeled (i)–(v) in Fig. 4(d), corresponds to positions (i)–(v) shown in Fig. 4(c). Note that points of the same color in Fig. 4(c) exhibit almost identical spectra (Fig. S10 in the Supplemental Material [50]). We observed five distinct spectra: M1, M1', T, M2, and R, with each structure assigned based on previous Raman spectra measurements [30]. In the broad areas colored light green and dark green, spectra corresponding to the insulating M1 and metallic R structures are observed, respectively, which is consistent with the electronic states predicted from the surface colors.

It is noteworthy that in the light green area, sandwiched between the dark green areas, the M1', M2, and T structures were observed. Here, the M1' corresponds to the M1 structure where the polarization direction of the light is slightly shifted from $E_i \perp c_i$. The M2 corresponds to another monoclinic structure, distinct from the M1 structure, while the T corresponds to the triclinic structure. The M2 and T structures in VO_2 have been identified as resulting from element substitution [29,30] and uniaxial stress [51], consistent with reports that applied

current can mimic pressure effects in VO_2 [52]. The presence of multiple nearly degenerate metastable states may explain the complex domain patterns compared to other nonlinear conductors such as Ca_2RuO_4 [6] and K-TCNQ [3].

The possible existence of a metallic monoclinic phase in VO_2 remains under debate, with reports based on micro-XRD of thin films [43], ultrafast photoexcitation [33], high-pressure experiments [53], and electrochemical gating [54]. In the present measurements under applied current, the emergence of metallic regions observed by optical microscopy correlates with tetragonal reflections in the XRD data, indicating that the metallic state is predominantly associated with tetragonal symmetry within the present resolution.

Despite these insights, the microscopic mechanism of the current-induced structural transition remains an open question. Several possibilities may be considered, including electric-field-driven electronic instabilities, electromigration, and current-induced stress or nonequilibrium electronic states. The present results provide some constraints on these scenarios. In particular, the transition occurs at a relatively low threshold electric field ($E_c \sim 10\text{--}10^2 \text{ V/cm}$) [41], which is comparable to that reported for Ca_2RuO_4 [4] and much smaller than in systems such as K-TCNQ [3]. Such a weak electric field is generally insufficient to directly modify the electronic structure of a strongly correlated system.

Instead, the results point to a mechanism associated with the flow of current itself. The emergence of a discontinuous transition at a well-defined current threshold, together with the isotropic lattice expansion distinct from equilibrium behavior, suggests a collective nonequilibrium response of the coupled electronic and lattice degrees of freedom. Possible scenarios include current-induced modifications of electron-phonon coupling or momentum transfer from carriers to the lattice under steady current. Further experimental and theoretical studies are required to clarify the microscopic origin of this phenomenon.

Overall, these results demonstrate that electric current induces successive symmetry changes and stabilizes nonequilibrium structures beyond thermal equilibrium. The observed structural phase transitions in VO_2 represent an unprecedented phenomenon that cannot be readily explained within the conventional framework of nonequilibrium thermodynamics. Continued investigation along this direction promises to deepen our understanding of the complex interplay among electronic correlations, lattice dynamics, and external driving forces in nonequilibrium phases.

In summary, we demonstrate that electric current induces two successive structural phase transitions in single-crystal VO_2 , revealing nonequilibrium phases beyond thermal equilibrium. Synchrotron XRD, optical microscopy, and Raman spectroscopy show a transition from the monoclinic M1 phase to a nonequilibrium low- J R structure, followed by a high- J R structure with isotropic lattice expansion. The observed domain formation and motion highlight symmetry breaking and spatial inhomogeneity driven by current, providing direct access to hidden phases and new insights into nonequilibrium transitions in correlated materials.

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Data availability. The data that support the findings of this article are openly available [55].

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