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Synchronized charge oscillations in correlated electron systems

SUBJECT AREAS:

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Strongly correlated phases exhibit collective carrier dynamics that if properly harnessed can enable novel functionalities and applications. In this article, we investigate the phenomenon of electrical oscillations in a prototypical MIT system, vanadium dioxide (VO₂). We show that the key to such oscillatory behaviour is the ability to induce and stabilize a non-hysteretic and spontaneously reversible phase transition using a negative feedback mechanism. Further, we investigate the synchronization and coupling dynamics of such VO₂ based relaxation oscillators and show, via experiment and simulation, that this coupled oscillator system exhibits rich non-linear dynamics including charge oscillations that are synchronized in both frequency and phase. Our approach of harnessing a non-hysteretic reversible phase transition region is applicable to other correlated systems exhibiting metal-insulator transitions and can be a potential candidate for oscillator based non-Boolean computing.

Rhythmic activity is ubiquitous in the physical world; and the process of synchronization of coupled oscillatory systems can be observed in the circadian rhythms of organisms^{1–3}, the synchronization of an ensemble of fireflies⁴ and is a distinguishing feature in certain structures of our brain including the neocortex and the thalamus⁵. Participating elements of a coupled system have the fundamental ability to exhibit a non-linear response to forcing stimuli; and hence under certain conditions, a collectively ordered state emerges from the complex dynamics of such a non-linear system. This motivates the development of physical systems of synchronous oscillators which are energy efficient, compact, scalable and could potentially enable large scale biologically inspired computing algorithms such as associative computing for applications like real-time image and pattern recognition^{6–12}.

In this paper, we investigate the dynamics of self-sustained electrical oscillations in a prototypical correlated system vanadium oxide (VO₂) which undergoes an abrupt first order metal-insulator transition with up to five orders of change in conductivity. Further, we explore the possibility of realizing a coupled system of such VO₂ based relaxation oscillators by exploring their synchronization and coupling dynamics. Through experiment and circuit simulation, we demonstrate that a system of capacitively coupled VO₂ oscillators exhibits charge oscillations that are synchronized in frequency and phase (anti-lock), thereby allowing the opportunity to harness the non-linear dynamics associated with their synchronization in computational algorithms. Our work enables a unique application and design space for a system of coupled correlated oxide based oscillators particularly suitable for applications in the domain of non-Boolean computing⁶.

Results

Single Oscillator Dynamics. Electrical oscillations in VO₂ harness the first order metal-insulator phase transition, which in this case is triggered electrically. It is well accepted that these oscillations represent a triggering of the insulator-to-metal transition (IMT) followed by a resetting metal-to-insulator transition (MIT) leading to spontaneous oscillations¹³. However, the physical origin of the electrically driven phase transition and therefore, the fundamental origin of the electrical oscillations in VO₂ is widely debated as being electronic^{14–19} or electro-thermally driven^{20,21}. Researchers have also shown evidence of electric field induced

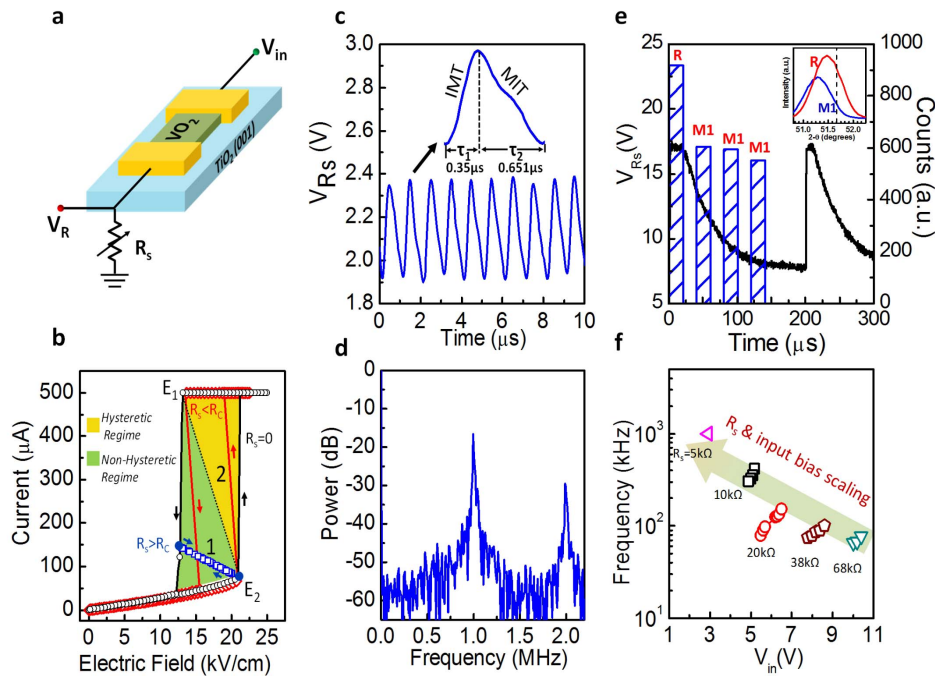


Figure 1 | Stabilizing a non-hysteretic and reversible phase transition in VO₂ and single oscillator dynamics. (a) Schematic of the circuit used to access the non-hysteretic transition regime in VO₂ using the negative feedback generated by the series resistor R_s . (b) R_s modifies the VO₂ phase transition dynamics through negative feedback. A non-hysteretic reversible (NHR) transition regime can be accessed using the appropriate R_s ($>R_C$). Electrical load lines for non-hysteretic (1) and hysteretic (2) switching are superimposed. (c) Time domain waveform of the VO₂ relaxation oscillator. (Inset) shows the waveform for one oscillation period with the two time constants ($\tau_1 = 0.35 \mu\text{s}$ and $\tau_2 = 0.65 \mu\text{s}$) associated with IMT and MIT. (d) Power (Mean Square Amplitude in dB) spectrum of a single oscillator. The fundamental frequency of the oscillator here is ~ 1 MHz. (e) Simultaneous measurement of the voltage and scattered x-ray diffraction intensity for an oscillating VO₂ device. Based upon the Bragg peaks, the structural phase of VO₂ is labeled. (Inset) shows the $[40\bar{2}]$ and $[002]$ Bragg peaks of the M1 and rutile phase of VO₂, respectively. The dashed line indicates the $\theta/2\theta$ angle ($=51.714^\circ$) at which this measurement was performed. Details of x-ray diffraction measurement are described in the Supplementary S4. (f) Frequency scaling of the VO₂ relaxation oscillator with R_s . The frequency increases as the external R_s is scaled down. The device resistance has also been scaled to further increase the output frequency.

nucleation²² as the origin of IMT in VO₂, similar to switching in chalcogenide glasses^{23,24}. Some reports elucidate the formation of conducting filaments^{20,25} that trigger the IMT. This debate is linked to the larger controversy over the fundamental driving physics of the phase transition in VO₂, particularly whether it is a Mott^{14,17,26,27} or a Peierls^{28–30} transition or a weighted contribution from both the mechanisms³¹. Despite the uncertainty over the unequivocal origin of the electrically driven transition in VO₂, our work, as well as other reports in literature^{32,33}, show that the dynamics of such relaxation oscillators can be modulated with temperature, total circuit resistance and capacitance.

To understand the single oscillator dynamics, we first explore the direct current (DC) characteristics of the oscillator circuit which consists of a two terminal VO₂ device (see Methods) with a series resistor R_s (Fig. 1a). The dimensions of the device used for measurements (except in experiments relating to Fig 2b, c) are $L_{\text{VO}_2} = 4 \mu\text{m}$; $W_{\text{VO}_2} = 10 \mu\text{m}$. The device size used in experiments relating to Fig. 1c, d is $L_{\text{VO}_2} = 4 \mu\text{m}$; $W_{\text{VO}_2} = 80 \mu\text{m}$. The series resistor provides a negative feedback and, therefore, modifies the transition dynamics. Unless stated otherwise, all electrical measurements reported are performed near room temperature (18°C).

When electrically driven across the phase transition with $R_s = 0 \Omega$ (Fig. 1b), the IMT and the MIT in VO₂ occur at critical fields, E_2 and E_1 , respectively. A current compliance is set to limit the metallic state current and prevent permanent device breakdown. While the exact magnitude of these electric fields depends on the origin of the phase transition (thermal/electronic), they represent a quantitative measure of the stimulus threshold that enables the IMT and the MIT to occur. If the transition is thermally driven then E_2 will represent the

electric field at which there is enough current density and corresponding thermal flux (most likely to be highly localized) to induce the IMT. Similar consideration applies to electronic case as well as to E_1 . Therefore, irrespective of the specific physical origin of the transition, the state of VO₂ can be described by knowing the electric field across the device. E_2 represents the field at which the conducting metallic phase can be stabilized to form a low resistance state while E_1 is the field at which the metallic phase can no longer be stabilized and the device returns to its high resistance insulating state. Even though the fundamental mechanism of electric field induced switching remains nontrivial, the delay time²² as a function of DC trigger voltage as well as the temperature dependence of these critical field points is shown in the Supplementary S1. These critical fields differ in magnitude giving rise to a hysteresis in the electrically driven phase transition.

In contrast to this hysteretic transition, a non-hysteretic transition could be induced by modifying the phase transition dynamics such that, triggering the IMT (by surpassing E_2) results in a negative differential resistance (NDR) characterized by an increase in conductivity and a simultaneous reduction in the field across the VO₂ device. If the electric field across the VO₂ now drops below E_1 , it makes the metallic phase unstable resulting in a spontaneous MIT. Representing this scenario through an electrical load line (Fig. 1b), a VO₂ device operating on load line ‘1’ will undergo a non-hysteretic transition, whereas a device operating on load line ‘2’ will go through a hysteretic transition.

While the NDR and the load lines described above cannot be achieved using the $R_s = 0 \Omega$ configuration, the addition of a simple R_s in series makes this feasible. R_s provides a stabilizing negative



feedback (see Supplementary S2) compensating the positive feedback associated with the abrupt change in conductivity. This negative feedback modulates the current-electric field characteristics across the VO₂ and establishes the load line relation:

$$E_{VO_2} = E_2 - \Delta I_{VO_2} R_s / L_{VO_2} \quad (1)$$

where $\Delta I_{VO_2} = I_{VO_2}(E_{VO_2}) - I_{VO_2}(E_2)$ and L_{VO_2} is the VO₂ channel length (see Supplementary S3 for derivation of equation (1) and (2)). From equation (1), it is evident that R_s tunes the VO₂ current-electric field characteristics and sets the criteria for non-hysteretic/hysteretic switching. The critical resistance, R_C , that enables the non-hysteretic regime of operation, ensures that the metallic phase is never completely stabilized and is given by:

$$R_C \geq \frac{L_{VO_2}}{W_{VO_2} t_{VO_2}} \left| \frac{E_2 - E_1}{\sigma_{sc} E_2 - \sigma_m E_1} \right| \quad (2)$$

where W_{VO_2} and t_{VO_2} are the VO₂ channel width and thickness, respectively, and σ_m and σ_{sc} are the equivalent insulating and metallic state conductivities, respectively.

The conductivity in the NDR regime as indicated by ΔI_{VO_2} can be tuned by the electric field (equation (1)) with conductivity increasing as the electric field drops (and vice-versa). Such a system can oscillate if the conductivity and electric field across the device modulate each other in a way that the restoring force enables the system to trace the same electrical trajectory periodically³⁴. This criterion is fulfilled only in the non-hysteretic operating regime and hence oscillations are observed only when $R_s > R_C$.

The time domain waveform of the VO₂ oscillator (Fig. 1c; single time period shown as inset) shows an exponential voltage buildup and decay (across R_s ; 5 k Ω in this case) associated with MIT and the IMT in VO₂ respectively and is characterized by two R-C time constants τ_1 and τ_2 (see inset Fig 1c) which primarily control the oscillation frequency. τ_1 (=0.35 μ s), associated with the IMT is smaller than τ_2 (=0.65 μ s). These time constants are much larger than the intrinsic electronic switching time³⁵ of VO₂ and therefore oscillation frequency can be scaled by reducing R and C through VO₂ device scaling. The corresponding power spectrum for the oscillations is shown in Fig. 1d. The exponential nature of the time domain waveform results in a gradual decay of the frequency harmonics and therefore the first harmonic is approximately 10 dB below the fundamental frequency.

In-situ nano X-ray diffraction (see Supplementary S4) performed on the VO₂ film stabilized in an oscillating state (Fig. 1e) confirmed that a structural transition occurs during each oscillation cycle similar to the DC electrically driven phase transition²⁵. The low resistance regime of VO₂ during the oscillation cycle is characterized by the rutile metallic phase, whereas the insulating regime is characterized by the monoclinic M1 phase. This is expected for the -0.9% compressively strained VO₂ films at room temperature³⁶ (see Methods).

We then study the oscillator frequency (f_{osc}) and dimensional scalability which is crucial to implementing dense, large scale VO₂ oscillator circuits. Since f_{osc} is set by the R-C time constant of the oscillator circuit, we explore f_{osc} scaling with R_s (Fig. 1f). As R_s is reduced, frequencies up to 1 MHz are experimentally realized. All experiments in this work are performed using discrete components (resistors) and eliminating the parasitic elements associated with the present wiring schemes through monolithic integration along with device scaling will further increase f_{osc} as observed with other correlated materials^{21,37}.

Further, we note that Nardone *et al.*³⁸ who demonstrated relaxation oscillators in chalcogenide phase change memory, have illustrated some of the stochastic features associated with such oscillations including the observation of multiple oscillatory patterns such as oscillations with decaying amplitudes. We have experimentally observed sustained stable oscillations over 2.5×10^9 cycles (see

supplementary S1) with no decaying amplitudes. Additionally, it is expected that the system of coupled VO₂ oscillators (described below) will have higher stability to stochastic variations due to mutual feedback induced stabilization³⁹.

Coupled VO₂ oscillators and their dynamics. The realization of inductor free and scalable non-linear oscillatory systems using VO₂ can be better exploited in the application domain if the oscillators can be synchronously coupled giving access to the large array of non-linear dynamics which are associated with their synchronization. Additionally, long range electrical charge based coupling is crucial to implementing large oscillator arrays compared to other state-variables (like spin), which may have limited coherence length at room temperature.

To investigate the coupling dynamics, we first explore 'injection locking' wherein a single oscillator is first excited with an external AC sinusoidal signal coupled through a coupling element which in this case is a capacitor C_C (Fig. 2a). The oscillator locks onto this external frequency only when it is close to the oscillator's natural frequency and within its lock-in range so that a large power output is observed at the input signal frequency (Fig. 2b). The ability of a single oscillator to lock onto an external AC signal implies that the two coupled

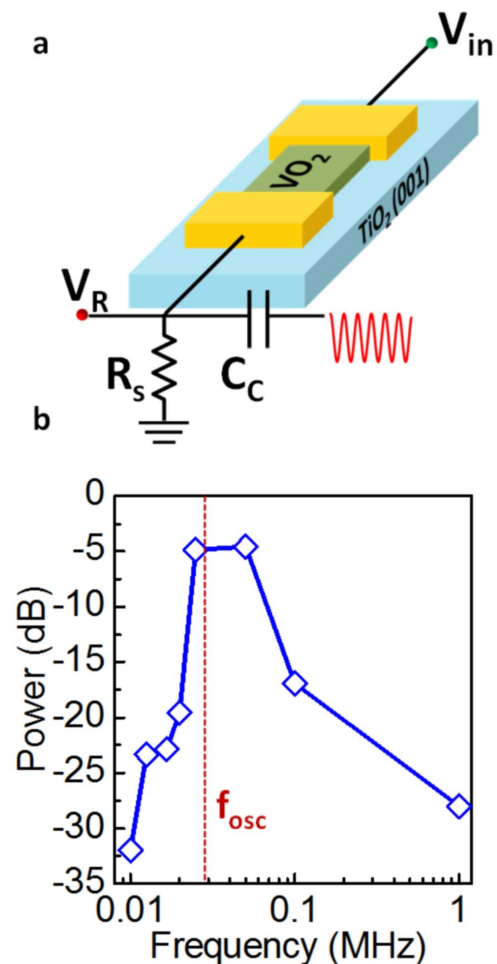


Figure 2 | Injection locking of VO₂ oscillators. (a) Schematic diagram of the injection locking circuit wherein an external AC signal is coupled to the oscillator through a coupling capacitor C_C . (b) Power output characteristics of a VO₂ relaxation oscillator ($R_s = 68$ k Ω ; $\sim 10R_C$) capacitively coupled to an external frequency signal ($C_C = 1.15$ nF) showing locking behavior for frequencies close to its natural oscillation frequency.

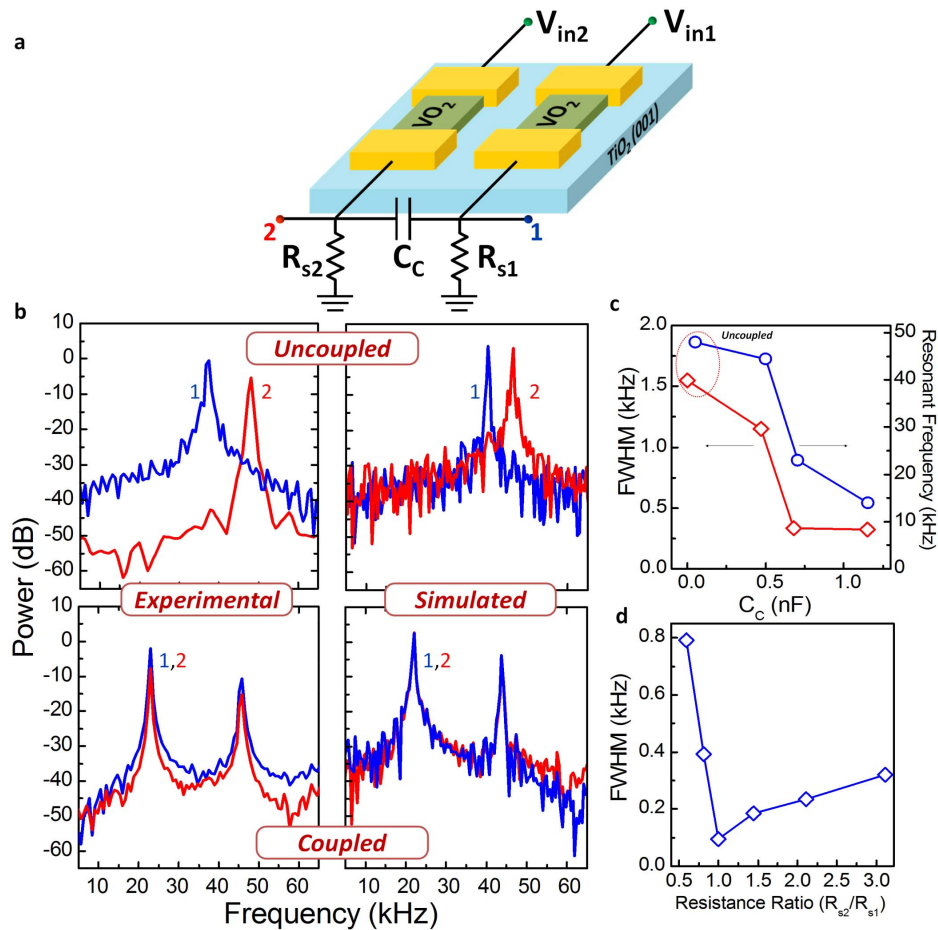


Figure 3 | Synchronized oscillations in VO₂ oscillators and their coupling dynamics. (a) Schematic of two capacitively coupled VO₂ oscillators. Individually, the oscillators are tuned to different oscillating frequencies using R_{s1} and R_{s2} of 38 kΩ (=6R_C) and 47 kΩ (=7.5R_C), respectively. (b) Experimental and simulated power spectrum of both oscillators before and after coupling. The individual oscillator frequencies lock to a new frequency when coupled capacitively (C_C = 680 pF). (c) Effect of C_C on the full width at half maximum (FWHM) of the spectral line width and resonant frequency of coupled VO₂ oscillators. Decreasing FWHM with increased C_C reflects the increased coupling strength. (d) Effect of external resistance symmetry on the coupling strength. The coupling strength is maximal when the oscillators are identical (equal R_s).

oscillators could mutually lock and exhibit synchronized oscillations as investigated further.

Fig. 3a shows a schematic of the coupled oscillator circuit. Prior to coupling, the two individual VO₂ oscillators have f_{osc} of 48.43 kHz and 37.27 kHz, respectively (Fig. 3b). f_{osc} is set by choosing appropriate R_s values: 38 kΩ (=6R_C) and 47 kΩ (=7.5R_C), respectively. After the oscillators are capacitively coupled (C_C = 680 pF), they frequency lock and converge to a single resonant frequency (22.95 kHz). This is accompanied by a sharp narrowing of the spectral line width (Fig. 3b). The reduced line-width after locking reflects the mutual feedback induced stabilization against noise³⁹, implying that the synchronized state is a stable configuration for the coupled system.

The use of capacitive coupling is motivated by the high pass filtering characteristics of the coupling configuration, which ensures that the individual oscillators can synchronize without mutually affecting their DC quiescent point. C_C modifies the frequency dynamics of each oscillator through an effective coupling term C_{eff}

$$C_{eff} = C_C C_1 + C_C C_2 + C_1 C_2 \quad (3)$$

where C₁ and C₂ are the net capacitances of each oscillator (see macro-model in supplementary). The C_C element stores and dynamically redistributes reactive (non-dissipative) power between the oscillators facilitating synchronization. Further, C_C also tunes the oscillator coupling dynamics and influences the strength of the coupling

interaction between them as indicated by the spectral line-width (full width at half maximum; FWHM) (Fig. 3c). Additionally, R_s, which tunes the natural frequencies of the individual oscillators, also affects the strength of the coupling between the two oscillators. In the symmetric oscillator configuration (equal R_s, identical frequencies for both oscillators), the strongest coupling is observed (Fig. 3d). The ability to tune the synchronization dynamics of the oscillators is crucial from an application standpoint. The experimentally observed frequency locking is also verified through circuit simulations using macro-models (see Supplementary S5), which describe the system's time evolution (Fig. 3b).

The synchronization dynamics of this coupled oscillatory system are analysed using the equivalent model shown in Fig. 4a. The interaction between the two oscillators, each of which traverses between the metallic (M) and the insulating (I) phases, is analysed through the resulting voltage flow diagrams. Figure 4b illustrates the flow diagrams of the coupled systems as they evolve over time. As the VO₂ device in each oscillator can be either metallic (M) or insulating (I), the resultant phase plane for the two VO₂ devices will consist of four quadrants where the oscillators can be in state MM, MI, IM or II. After coupling, the system will settle on one fixed orbit/trajectory, which may go through these four quadrants. As the device and circuit parameters are changed, the stable orbit will change indicating a different path in the phase plot. The blue arrows indicate a family of such orbits corresponding to the time evolution of the system

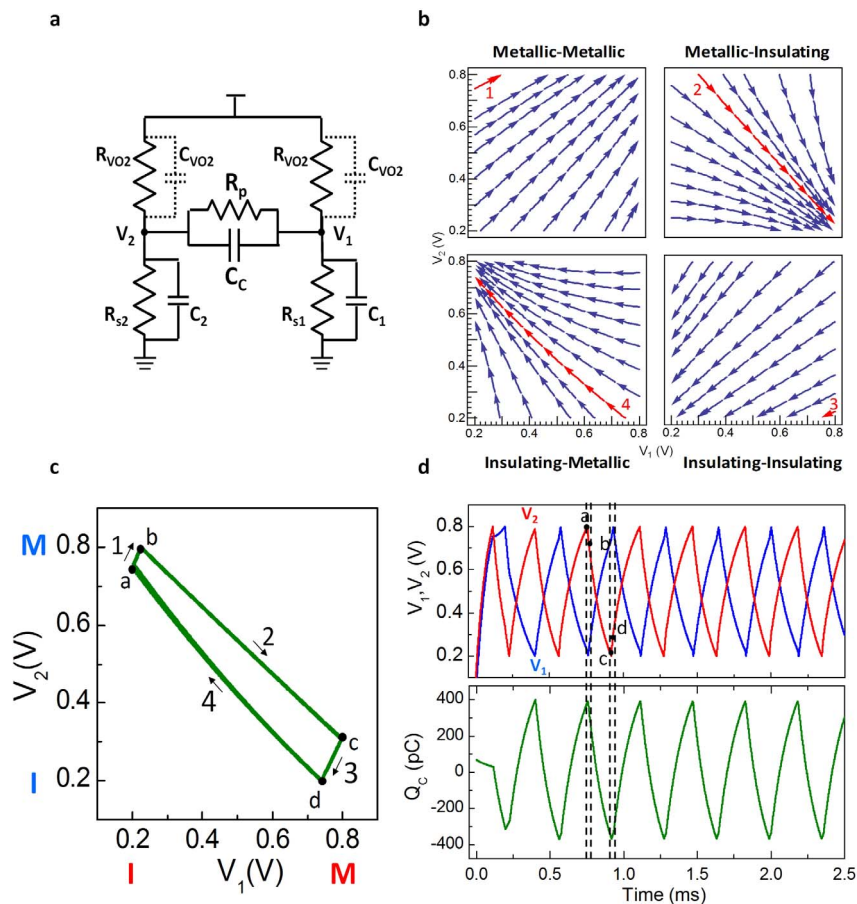


Figure 4 | Analyzing the coupling dynamics of VO₂ oscillators. (a) Equivalent circuit model for capacitively coupled VO₂ oscillators. C_1 and C_2 are the effective capacitances of each oscillator, which include the internal VO₂ capacitance associated with VO₂ film and the device (C_{VO2}) as well as parasitic capacitances. $R_p \rightarrow \infty$. The same circuit is used to simulate the experimentally observed frequency locking and has been verified with experimental data as shown earlier. (b) Voltage flow diagram in phase space for steady state coupled oscillations. 1–2–3–4 is the electrical trajectory of the capacitively coupled configuration. It reveals that the coupled oscillators primarily move in the M-I and I-M phase planes. (c) The electrical trajectory of the coupled oscillator configuration in phase space. (d) Simulated time domain waveform of the two VO₂ oscillators after coupling (top). The to and from movement of charge (Q_C) across the capacitor enables the two oscillators to synchronize (bottom).

shown in Fig. 4a. The stable orbit corresponding to our experiment has been shown in red. The stable orbit (red curve in Fig. 4b) of the system has been exclusively shown again in Fig. 4c as a phase plot (V_1 vs. V_2) along with the corresponding metallic and insulating phases for each VO₂ device (the numbers 1,2,3,4 show the related arm in the flow diagrams in Fig. 4b). The phase diagram in Fig. 4c reveals that the synchronized oscillators primarily move between the M-I and I-M planes such that as one VO₂ device transitions into the metallic phase, the other device evolves into the insulating phase (b \rightarrow c; d \rightarrow a). This is also marked by charging/discharging of the coupling capacitor C_C . The region a \rightarrow b; c \rightarrow d defines the short phase space trajectory where both the devices have the same phase and represents the time during the coupling capacitor C_C reverses polarity. The corresponding time domain waveform of the two oscillators after coupling is shown in Fig. 4d. As the oscillators move in the phase space, the periodic sourcing/drawing of charge (Fig. 4d) by the coupling capacitor C_C enables the oscillators to synchronize.

Discussion

The non-hysteretic transition dynamics that enable such spontaneous electrical oscillations in VO₂ inspires the exploration of other correlated systems for similar applications^{40–42}. The coupled oscillator dynamics demonstrated here provides an important milestone of experimental demonstration of coupled synchronous oscillations in correlated oxide materials and unravels new oppor-

tunities towards exploring large coupled oscillatory systems. We anticipate a wide array of dynamics in larger arrays of such coupled oscillators that could potentially be harnessed to develop a robust hardware platform capable of supporting novel computing schemes and algorithms.

Methods

The VO₂ two terminal devices are fabricated on a 17.7 nm thick VO₂ films. The films are epitaxially grown on a TiO₂ (001) substrate employing reactive oxide molecular-beam epitaxy using a Veeco GEN10 system. Due to epitaxial mismatch, the VO₂ films are biaxially strained in compression by $\sim 0.9\%$. First the electrodes are patterned using contact lithography followed by electron beam evaporation of Pt/Au (15 nm/80 nm) and lift-off in RemoverPG at 70°C. Next the channel width and isolation are defined by electron beam lithography followed by a CF₄ dry etch. Finally, the resist is stripped with RemoverPG at 70°C.

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Author contributions

N.S. performed the experiments and analysed the data. A.P. performed the simulations. E.F. fabricated the devices and H.P. grew the VO₂ films. E.F., G.S., H.W. and Z.C. performed the nano X-ray characterization. S.D. conceived and directed the experiment. V.N., V.G., R.E.-H., D.S. and A.R. supervised the study. N.S., S.D., V.G., A.R., R.E.-H. and D.S. wrote the manuscript. All authors discussed the results and commented on the manuscript.

Additional information

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