Electrical oscillations induced by the metal-insulator transition in VO$_2$

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We systematically investigate the characteristics of an electrical oscillation observed in two-terminal vanadium dioxide (VO$_2$) devices. These oscillations are observed at room temperature in a simple electrical circuit without inductive components. The circuit is composed only of a dc voltage source, the VO$_2$ device, and a standard resistor connected in series with the device. We explain why the observed oscillations are a result of the percolative metal-to-insulator transition (MIT) of VO$_2$ and the coexistence of the metal and insulating phases. Specifically, oscillations are attributed to the construction and destruction of capacitive regions composed of regions of the semiconducting phase, (as dielectric material) and metallic phase electron carriers, induced by the MIT (as capacitor electrodes). Since the coexistence of these phases—and thus the capacitive regions—is destroyed by elevated temperature, the MIT oscillation is not explained in terms of significant heat input but rather in terms of a voltage-triggered effect. It is also discussed whether the current jump at the onset of the oscillations is driven by Mott physics or by Peierls physics relying on a structural phase transition. Furthermore, the electrical parameter space surrounding these oscillations is explored, and a generation window is identified. Within this generation window, the oscillation frequency can be continuously tuned by adjusting applied voltage or by an external circuit component, such as resistor or added capacitor. The frequency of oscillations can be increased up to >1 MHz.


I. INTRODUCTION

Mott$^1$ predicted in 1949 that a first-order discontinuous metal-insulator transition (MIT) could arise from the removal of electrons in a strongly correlated electron system (or Mott insulator). The critical driving parameter in this transition is the on-site repulsive Coulomb interaction $U_C$. This concept has been derived by the extension of the Brinkman–Rice picture$^{3,4}$ and is based on inhomogeneity. The removal of electrons, regarded as doping of hole carriers, causes a breakdown of $U_C$ in a Mott insulator with a hole concentration below 0.01% and induces the breakdown of an energy gap formed by $U_C$ through impact ionization;$^{5,7}$ this is the hole-driven MIT generated at a critical hole concentration, $n_c$, not the metallic minimum carrier density in the Mott criterion.$^{1,4}$ In this case, both a metal phase with electron carriers and a semiconductor system with hole carriers can be formed. The metal phase has a strong correlation between electrons.$^{2,8}$ The semiconductive system is a remnant component of the parent Mott insulator which is an extrinsic compound semiconductor with disorder, impurities, dislocation, etc. The Mott system (or insulator) has two kinds of carriers$^{3,9}$ and is intrinsically inhomogeneous.$^{10,11}$

In strongly correlated system, there are several explanations besides Mott’s first-order theory: a Hubbard MIT,$^{12}$ which undergoes a continuous transition from metal to insulator following the increase in $U$, and a Peierls continuous MIT$^{13}$ driven by electron-phonon interactions, such as charge density wave (CDW), which induce a structural phase transition (SPT)$^{14}$.

In the context of the above MIT theories, VO$_2$ ($3d^1$) is a very interesting material because it is a representative insulator with both an energy gap of 0.6 eV and the metallic electronic structure of half filling. It undergoes a large resistance change (regarded as the MIT) near critical temperature $T_C \approx 67$ °C and a SPT between the monoclinic structure (below $T_C$) and the tetragonal rutile structure above $T_C$.$^{15}$

The insulator structure of VO$_2$ was known as the monoclinic one, $M_1$, at enough low temperatures compared to $T_C$ and at room temperature. With increasing temperature, $M_1$ changed to $M_2$ just below $T_C$ via an intermediate triclinic monoclinic structure $T$, and finally was transformed to the rutile R tetragonal structure of the metallic phase above $T_C$. $M_2$ is composed of two distorted structures; one-half of the V chain pairs but does not twist (this forms charge ordering of CDW) and the other half twists but does not pair (this is an equally spaced V zigzag chain and forms the Mott–Hubbard insulator).$^{16,18}$ $M_1$ is defined by a superposition of two distorted structures in $M_2$.$^{16,17}$ For V$_{1-x}$Cr$_x$O$_2$, with doping concentration x, the $M_1$ structure of the insulator phase at room temperature was transformed into $M_2$ via $M_3$.$^{17}$ Moreover, for VO$_2$ nanorods with the semiconducting characteristic below and over 190 K, a small jump in conductivity and photocurrent decay near 190 K was interpreted as a first-order SPT between $M_2$ and T structures.$^{19}$

Researchers on the MIT of VO$_2$ have mainly focused on the mechanism responsible for the SPT between the mono-
clinic insulating phase and the tetragonal rutile metal phase. However, Kim et al. concentrated researches on the presence of a resistivity change as MIT, which is not accompanied by any SPT, and the observation of a strong correlation. The presence of a MIT independent of any SPT (a metal phase with the monoclinic structure) was experimentally observed by several experiments such as Raman scattering with temperature, scattered by monoclinic and tetragonal structures occurs, prohibit such high-frequency oscillations.

In earlier studies, electrical oscillations similar to those we observed, but with a sinusoidal waveform, were observed in GaAs, 47 K0.3MoO3, 48 and θ-(BEDT-TTF)2CsCo(SCN)4. 49,50 The Gunn oscillation in GaAs was explained in terms of the negative differential resistance (NDR) caused by mobility change from (100) to (110). 51 The NDR characteristic is similar to the thyristor one. 52 For K0.3MoO3, the sliding motion of the CDW undergoing the SPT was suggested as the oscillation mechanism, and the oscillation frequency fosc could be changed from 10 to 17 kHz by varying the applied voltage at 17 K. 48 The 40 Hz oscillation in θ-(BEDT-TTF)2CsCo(SCN)4 was also observed at 4.2 K. 49 The current jump in the organic material was explained by melting of charge ordering with a charge difference between nearest neighbors. The charge difference is regarded as the CDW. The melting of the charge ordering is similar to the sliding of the CDW in K0.3MoO3 and indicates the melting of the structure distorted by electron-phonon interaction. This analysis is acceptable for organic and CDW materials. However, as far as a correlated material such as VO2, the mechanism of the oscillation is different from that in CDW materials. The physical reasons for these differences will be discussed in Sec. IV.

In this paper, we investigate the characteristics of a high-frequency room-temperature electrical oscillation observed in the VO2-based two-terminal devices with S-type NDR characteristics. We demonstrate the generation mechanism of this oscillation by analyzing experimental data based on inhomogeneity. We show that increasing applied source voltage to the device causes percolation and decreases the oscillation frequency fosc by effectively decreasing the device capacitance.

Values for the capacitance of the device are found by fitting exponential charging curves. Furthermore, an overall generation window for the MIT oscillation is found in terms of the external resistance and source voltage. In particular, the effect of the external circuit components, such as a voltage source, resistor, and capacitor, on fosc is systematically investigated. Thus, we reveal that fosc can be designed to be increased up to >1 MHz and controlled by simple adjustment of some external circuit components.

II. EXPERIMENTS

In order to fabricate the two-terminal VO2 device shown in Fig. 1(a), VO2 thin films were grown on sapphire (Al2O3) substrates by employing a sol-gel method. 53 Note that VO2 films could be deposited by several deposition methods. 54 The films were patterned into a line shape to make a current channel with photolithography processes and a rf ion-beam milling technique. A Ni electrode layer was deposited on the etched VO2 layer using a rf magnetron sputter deposition
technique and was patterned with a lift-off method. For direct microcontact electrical measurements, square-shape gold electrodes of $50 \times 50 \mu m^2$ were patterned on Ni electrodes of the device. The width of the VO$_2$ patch was designed to be narrower than that of the electrodes to avoid any delayed transition of VO$_2$ outside the electrodes. The final dimensions of the exposed film patches in the fabricated devices were $10 \times 10 \mu m^2$ (device I) and $5 \times 5 \mu m^2$ (device II). The thickness of the VO$_2$ films was approximately 100 nm.

The schematic diagram of the experimental setup to measure $I$-$V$ characteristics of the device is shown in Fig. 1(b). For the $I$-$V$ measurements, we used a microprobe station (Micromanipulator) with tungsten probes (5 $\mu$m in diameter) and a parameter analyzer (Agilent B1500A). The Ohmic contact resistance of the devices was below 1 $\Omega$, which is negligible compared with the device resistance $R_D$ at room temperature. The $I$-$V$ characteristics of the devices were obtained by using the $V$-mode and $I$-mode of the parameter analyzer. The $V$-mode and $I$-mode are the measurement modes to detect the current and voltage across the device, $I_D$ and $V_D$, as a function of the applied voltage ($V$-mode) and current ($I$-mode), respectively. Moreover, the stability of the MIT current-jump threshold point, responsible for the observed switching, was confirmed.$^{35}$

III. MEASUREMENT OF MIT ELECTRICAL OSCILLATION

A. Oscillation measurement and investigation of its generation mechanism

Figure 1(a) shows an optical microscope plane-view image of the fabricated two-terminal VO$_2$ device (device I) and the surface morphology of the VO$_2$ film used in the device (a rms roughness of ~5 nm). The film surface was comprised of grains of several different sizes. At room temperature, the VO$_2$ film has almost insulating VO$_2$ grains (high resistance dielectric components). At a critical temperature (~340 K), high resistance insulating grains begin to turn into low resistance metallic ones, and most insulating grains have transitioned into metallic ones just after the critical temperature.$^{27,28}$ Any void between grains (which will have large resistance) and non-VO$_2$ phases in the film such as V$_2$O$_5$ can affect $R_D$ throughout the entire process of the voltage-induced MIT.$^{23}$ Recently, it was observed that the MIT begins percolation near grain boundaries or void regions, suggesting that these have a strong role in the MIT transition dynamics.$^{11}$

Figures 1(c) and 1(d) show the $I$-$V$ hysteresis loops measured at room temperature for device I and device II, respectively. Below the threshold of any current-jumps [in region A of Figs. 1(c) and 1(d)], device I and device II exhibit semi-
conducting behavior with hole charges. There exist two threshold instability voltages \((V_{t1}, V_{t2})\) where abrupt current changes happen. At \(V_{t1}\), a critical carrier density inducing the jump, \(n_c\), is reached.\(^4\) In \(V\)-mode traces, \((V_{t1} \approx 9.7 \text{ V}, V_{t2} \approx 4.0 \text{ V})\) in device I and \((6.6 \text{ V}, 3.1 \text{ V})\) in device II were measured. The currents at \(V_{t1}\) and \(V_{t2}\) are designated as \(I_{t1}\) and \(I_{t2}\), respectively. The \(V\)-mode traces have large hysteresis loops with black and green jumps corresponding to negative and positive jumps in resistance, respectively. In the \(I\)-mode traces of both Figs. 1(c) and 1(d), a typical feature of the S-type NDR is shown between instability \(P_1(V_{t1}, I_{t1})\) and instability \(P_2(V_{t2}, I_{t2})\). The NDR behavior arises from the restriction of current and has little hysteresis width. This indicates that \(I\)-mode generates little heat and no SPT.

Anomalous behavior—decreasing voltage with increasing current—was observed in a semiconductor phase between instabilities \(P_1\) and \(P_2\) (S section in region B). This behavior indicates that the MIT occurs gradually, resulting in a gradual decrease in resistance (gradual increase in electrical conductivity). This observation is interpreted as evidence for a percolative transition\(^8\) and is evidence of inhomogeneity in the VO\(_2\) film. It is also observed by comparing device I with device II that the increase in the device dimension causes both threshold voltages and corresponding currents to increase and thus the NDR region to expand as well. Despite the dimension change, however, no noticeable variation was observed in the differential resistance \((dV/dI)\), estimated as approximately \(\sim 500 \text{ \Omega}\) from the measurement results. Furthermore, the oscillation is affected by external parameters such as a kind of electrodes, lattice-film mismatch, and film thickness. When electrical characteristics of a device are changed by the external parameters, current near zero voltage (leakage current), \(V_{t1}\), and \(I_{t2}\) vary,\(^5\) for example, a large contact resistance between film and a metal electrode with a low conductivity, a large lattice-substrate mismatch, and a more thick film induce a large leakage current, a reduced \(V_{t1}\), and an increased \(I_{t2}\).

Moving on to the experiments involving electrical oscillation, Fig. 2(a) shows a schematic diagram of the electrical circuit used for generating the MIT electrical oscillation and controlling \(f_{\text{osc}}\). The single-loop circuit was composed of a VO\(_2\) device (device I or device II), a voltage source of a dc voltage \(V_e\), and an external resistor of a resistance \(R_E\). In order to reduce the generation of an excess Joule heat, both a function generator (Agilent 33120A) and a dc power supply were used for generating rectangular voltage pulses with a variety of dc offsets. The oscillatory electrical responses were recorded by a digital oscilloscope (HP 54810A). An external capacitor of a capacitance \(C_E\) in the dashed box can also be connected in parallel to the external resistor giving further control of \(f_{\text{osc}}\). In fact, all circuit parameters \((V_e, R_E, C_E)\) can act as frequency tuning controls, and related experimental results are provided in Figs. 4 and 5.

Figure 2(b) shows the oscillatory electrical traces of \(V_e\) (blue diamonds) and \(I_p\) (red circles) measured with device I at room temperature, when a single voltage pulse with a peak value of \(V_p(V_p)\) of 13.8 V (including a dc offset of 10 V) and a pulse width of 25 \(\mu\text{ s}\) is applied \((R_E=8 \text{ k}\Omega)\). The voltage across the external resistor \(R_E\) is directly measured, and \(V_e\) and \(I_p\) are obtained by \((V_e-V_E)\) and \((V_E/R_E)\), respectively.

The peak-to-peak amplitudes of the oscillation waveforms of \(V_e\) and \(I_p\) were measured as \(\sim 5.7 \text{ V}\) and \(\sim 0.7 \text{ mA}\), respectively. The fundamental component of \(f_{\text{osc}}\) was measured as \(\sim 0.525 \text{ MHz}\). Note that a device without the NDR shown in region B of Figs. 1(c) and 1(d) did not generate any oscillation.

Considering one period of the oscillation in Fig. 2(b), the exponential buildup and decay of \(V_e\) and \(I_p\), respectively, are driven by the carrier charging process in a series \(RC\) circuit—whose electrical response time is restricted by a capacitive time constant \(\tau_0\) given by \(R_E C_D\) where \(C_D\) is a device capacitance. Within one period of the oscillation, the charging curve responses of \(V_e\) and \(I_p\), before the MIT threshold jump is triggered, can be fitted by the following expressions derived based on the \(RC\) circuit analysis:

\[
V_e(t) \approx V_p[1 - \exp(-t/\tau_0)],
\]

\[
I_p(t) \approx V_p[1 + (R_D/R_E)\exp(-t/\tau_0)].
\]

FIG. 2. (Color online) (a) Schematic diagram of the electrical circuit used for the generation of the MIT electrical oscillation. In order to control \(f_{\text{osc}}\), an external capacitor in a dashed box can be employed and connected to an external resistor in parallel. (b) Electrical waveforms of \(V_e\) (blue diamonds) and \(I_p\) (red circles) measured at room temperature when a voltage pulse with \(V_p=13.8 \text{ V}\) (including a dc offset of 10 V) and a pulse width of 25 \(\mu\text{ s}\) are applied \((R_E=8 \text{ k}\Omega)\).
tallic ones, and metallic electron carriers appear (region B); a capacitor is formed, which consists of a dielectric semiconducting component (S in region B) and the electron carriers. In conjunction with the swift decrease in $R_D$, $V_D$ instantaneously drops down below $V_{t1}$, and $I_D$ rapidly increases. Then, $V_D$ is restored to $V_{t2}$ by some repulsive force—designated as the elastic restoring force by Sakai. Behavior of $V_D$ after this moment is determined by $I_D$ affected directly by $R_D$ and $R_E$.

When we select an appropriate $R_E$ that forces $I_D$ to be less than $I_{t2}$, the device can return to its insulating state. The cycle can repeat with $V_D$ increasing again. In this condition, the temporal buildup of $V_D$ [Eq. (1)] proceeds to occur until $V_D$ reaches $V_{t1}$. After $V_D$ reaches $V_{t1}$, the MIT jump occurs, rapidly discharging $V_{tm}$, and the same process described above repeats as long as $V_{t2}$ is large enough to make $V_D$ become $V_{t1}$. In this manner, the MIT oscillation is maintained between both instabilities $P_1(V_{t1}, I_{t1})$ and $P_2(V_{t2}, I_{t2})$, as indicated in Figs. 1(c) and 1(d).

In the case of $R_E$ selected such that $I_D$ to be less than $I_{t2}$, just after the MIT [region C of Figs. 1(e) and 1(d)], most insulating VO2 grains have become metallic ones, and the film has few dielectric regions. The decreased dielectric (capacitive) component of the film causes a reduced number of charges to build on the VO2 device and prevents $V_D$ from increasing to $V_{t1}$ again. In other words, the disappearance of the capacitive component of the film makes it impossible for the VO2 device to recover its insulating state (high resistance state) and perform oscillations.

B. Nanoscale percolation: Explanation of forming capacitor

An explanation on the presence and absence of the capacitive component in the VO2 device after the MIT ($V_D > V_{t1}$) is now given, as shown in Fig. 3(a). A VO2 film composed of many grains [Fig. 1(a)] can be thought of as being comprised of two components: any VO2 phase (green or red grains) and any non-VO2 material such as vacuum or V2O3 (gray bottom). The amount of the non-VO2 phase may be negligible in a high-quality film. Looking only at the VO2 material, when $V_D < V_{t1}$ [plot I of Fig. 3(a)], the film is fully filled with green grains (insulating state) without red grains (metallic state). When $V_D$ starts to exceed $V_{t1}$ [plot II of Fig. 3(a)], one part of the VO2 phase turns into a metallic state, and the other part still remains an insulating state due to the minute compositional homogeneity between VO2 grains. Green and red grains may act as dielectric components and metal electrodes, respectively. Averaged across the entire film, they build a capacitor across the device. This intermediate state corresponds to the region B in Figs. 1(c) and 1(d). When $I_D$ exceeds $I_{t2}$, the film is occupied with primarily red grains resulting in vanishing capacitive component of the device [plot III of Fig. 3(a)].

This simple explanation is supported by midinfrared near-field images.8 Figure 3(b) shows the images of the near-field scattering amplitude over the same $4 \times 4$ µm$^2$ area obtained by using a scattering scanning near-field infrared microscope operating at the infrared frequency of 930 cm$^{-1}$.

These images are displayed for representative temperatures (341.0, 342.4, 342.6, 342.8, 343.0, and 343.6 K) in the MIT regime of VO2 and show the percolative nature of the MIT in progress. The metallic regions (green and red colors) give higher scattering near-field amplitude compared with the insulating phase (dark blue color), e.g., the region surrounded by the yellow dotted line in Fig. 3(b). These images directly show that the insulating and metallic phases coexist in VO2 over a finite temperature range in the transition region. Thus, insulating and metallic phases in the region surrounded by the white dotted lines in Fig. 3(b) may act as dielectric components and metal electrodes, respectively, resulting in the formation of a capacitor across the device. Further experimental evidence for the construction and destruction of capacitance in VO2 comes from observing the infrared resonance frequency of a hybrid-metamaterial comprised of splitting-resonators and VO2.

C. Applied voltage dependence of $f_{osc}$

In order to investigate the dependence of $f_{osc}$ on applied voltage (for a fixed value of $R_E$ with 1 kΩ step), the electrical responses of $V_D$ and $I_D$ were measured for increasing $V_{t2}$. Among the measurement results, Figs. 4(a) and 4(b) show
the life cycles of the MIT oscillation, measured in \( I_D \), for device I and device II (measured at \( R_E \) of 8 and 10 k\( \Omega \) with respect to various values of \( V_S^p \)). In device I and device II, \( V_S^p \)s in subplots I, II, III, IV, V, and VI correspond to 12.4, 12.8, 13.2, 14.4, 15.6, and 15.8 V and 7.5, 7.7, 9.0, 11.0, 13.0, and 15.5 V, respectively.

In plot I of Figs. 4(a) and 4(b), current pulses with peak values of \( \sim 0.40 \) and \( \sim 0.19 \) mA and pulse widths of 25 and \( \sim 16.67 \) \( \mu s \) are observed without any \( I_D \) oscillation. This implies that \( V_D \) does not reach \( V_{it} \) yet. In plot II of Figs. 4(a) and 4(b), current pulses with impulsive peaks whose values are \( \sim 1.10 \) and \( \sim 0.58 \) mA are observed, respectively, and the \( I_D \) oscillation is not yet observed. This is because \( V_S^p \) is not large enough to make \( V_D \) decreased after the MIT reach \( V_{it} \). In plots III, IV, and V of Figs. 4(a) and 4(b), periodical waveforms composed of impulsive peaks of \( I_D \) are observed, i.e., the \( I_D \) oscillation is observed. This suggests that \( V_S^p \) is large enough to make \( V_D \) decreased after the MIT reach \( V_{it} \). In these three plots (III, IV, and V), we observe that \( f_{osc} \) increases in proportion to \( V_S^p \), and a detailed discussion on the relationship between \( f_{osc} \) and \( V_S^p \) will be provided in Sec. III D. In plot VI of Figs. 4(a) and 4(b), rectangular pulses with average values of \( \sim 1.35 \) and \( \sim 1.26 \) mA, respectively, emerge without the periodical waveform. This extinction of the MIT oscillation is caused by the collapse of the capacitive component of the device [disappearance of dark blue region in Fig. 3(b)] due to Joule heat generated by a large value of \( I_D \) induced by \( V_S^p \) increase.

In order to find a region of \( R_E \) and \( V_S \) within which the MIT oscillation can be generated, the oscillation characteristics of the VO2 devices were measured with several values of \( R_E \) and \( V_S^p \). A specific region of \( R_E \) and \( V_S^p \), where the MIT oscillation is generated, can be determined in a two-dimensional domain defined with \( R_E \) and \( V_S^p \) as shown in Fig. 4(c). The left and right subplots in Fig. 4(c) indicate the generation window of the MIT oscillation in device I and device II, respectively. The lower and upper limits of \( V_S^p \) where the MIT oscillation can be generated for each \( R_E \) are plotted with blue squares and red circles, respectively. The solid lines are linear fits of the lower and upper limits of \( V_S^p \), and the yellow region enclosed by solid lines thus illustrates the generation window of the MIT oscillation for our two devices. This oscillation window can also be predicted from both relationships: \( V_D \approx V_{it} \) and \( I_D \approx I_{osc} \). The graphical intersection of these two one-dimensional inequalities with two variables \( (R_E \) and \( V_S^p \) creates a unique region in the two-dimensional domain of \( R_E \) and \( V_S^p \). It can be verified from simple graphical analysis that this calculated region is in good agreement with the measured oscillation window [yellow region of Fig. 4(c)].

### D. Analysis of oscillation waveform

In order to further investigate the dependence of \( f_{osc} \) on \( V_S \), oscillation waveforms of \( V_D \), measured at \( R_E=8 \) k\( \Omega \) for three values of \( V_S^p \) (13.8, 14.6, and 15.4 V), were superimposed, as shown in Fig. 5(a). As mentioned earlier, abrupt drops of \( V_D \) from points A, C, and E to points B, D, and F labeled in the figure are induced by the MIT of the VO2 film. As seen from the figure, we can recognize two important features: the temporal intervals of traces A-B, C-D, and E-F labeled in the figure are nearly the same (\( \sim 0.36 \) \( \mu s \)) regardless of \( V_S^p \). The time required for dropped \( V_D \) to recover to \( V_{i2} \) (by some elastic restoring force\(^{27}\)) is also nearly the same (\( \sim 0.14 \) \( \mu s \)) regardless of \( V_S^p \) with respect to each trace B-B’, D-D’, and F-F’. Thus, the time consumed on the exponential growth of \( V_D \) from \( V_{i2} \) to \( V_{it} \), e.g., \( t_1 \) at \( V_S^p=15.4 \) V (or \( t_2 \) and \( t_3 \) at \( V_S^p=14.6 \) and 13.8 V, respectively), dominates contrib-
For a given V_{S} and a fixed V_{T}, the exponential buildup of V_{D} in Eq. (3) is observed when \tau_{0} is ignored, i.e., \tau_{D} increases with V_{P}^{S}. This means that f_{osc} can be tuned by varying an external circuit parameter such as \tau_{0}. When external parameters are independent of \tau_{0}, the tuning sensitivity, defined as (\Delta f_{osc}/\Delta V_{P}^{S}), was calculated as 99.18 kHz/V. These results suggest that the VO_{2} device can be utilized as a voltage-controlled oscillator, which consists of only two circuit components and is operated by only one voltage source.

E. Effect of external parameters

We now consider controlling f_{osc} using passive external circuit components. f_{osc} is determined by 1/(T_{0} + T_{1}), where T_{0} and T_{1} indicate the time required for V_{D} to increase from V_{t2} to V_{t1} and required for V_{P} to drop from V_{t1} to V_{t2}, respectively. f_{osc} is mainly dependent on T_{0} = \tau_{0} \ln[1 + (V_{P}^{S} - V_{t2})/(V_{A} - V_{t1})], which is obtained by Eq. (3), because T_{1} is nearly the same (~0.50 \mu s) regardless of V_{P}^{S}. T_{0} depends not only on V_{P}^{S} but also on \tau_{0}. This means that f_{osc} can be tuned by varying an external circuit parameter such as R_{E}. Also, when an external capacitor of a capacitance C_{E} is connected in parallel with the external resistor, as shown in the dashed box of Fig. 2(a), it becomes another f_{osc} tuning parameter because \tau_{0} is proportional to (C_{E} + C_{D})R_{E}.

Based on the above discussion, external control of f_{osc} was demonstrated through the adjustment of R_{E} without external capacitor and the adjustment of C_{E} with R_{E} fixed (10 k\Omega). This experiment was done in device II, which was especially designed to obtain an enhanced frequency response. This frequency improvement is achieved because the reduced dimension of the VO_{2} patch in device II reduces (V_{t1} - V_{t2}), but with C_{D} maintained as nearly the same value as that of device I. Thus, T_{0} and f_{osc} in device II become smaller and larger compared with those in device I, respectively.

FIG. 5. (Color online) (a) Superimposed waveforms of V_{D} measured in device I serially connected with R_{E} = 8 k\Omega with respect to three values of V_{P}^{S} (13.8, 14.6, and 15.4 V). (b) Measured waveforms of I_{D} oscillation. (c) Their FFT amplitude spectra with respect to various values of V_{P}^{S} (only three cases including 13.8, 14.6, and 15.4 V are displayed) at R_{E} = 8 k\Omega, when the rectangular voltage pulse with a pulse width of 25 \mu s is applied to device I. The inset plot shows the functional relationship between f_{osc} and V_{P}^{S}.

utes to the determination of f_{osc}. We can fit this exponential growth section from V_{t2} to V_{t1} with the function

\[ V_{D}(t) = (V_{A} - V_{t2})[1 - \exp(-t/\tau_{0})] + V_{t2}. \]  

(3)

This exponential buildup of V_{D} in Eq. (3) means the buildup of charge on the capacitor in a series RC circuit, which is predicted in Eq. (1). This means that \tau_{0} in Eq. (3) limiting the maximum f_{osc} is the same as that in Eq. (1), and \tau_{0} is proportional to C_{D}R_{E}. \tau_{0}’s of the fitting curves evaluated from the traces were ~0.840, ~0.801, and ~0.765 \mu s for V_{P}^{S}’s of 13.8, 14.6, and 15.4 V, respectively. This indicates that \tau_{0} decreases when V_{P}^{S} increases, i.e., C_{D} decreases with the increase in V_{P}^{S}. When the effect of R_{D} dependent on V_{P}^{S} on \tau_{0} is ignored, C_{D} can easily be estimated as ~105.0, ~100.1, and ~95.6 pF (for V_{P}^{S}’s of 13.8, 14.6, and 15.4 V, respectively).

When we postulate the circuit model of the VO_{2} device as the parallel connection of C_{D} and R_{D}, \tau_{0} is given as R_{E}C_{D}R_{D}/(R_{D} + R_{E}), and thus C_{D} becomes larger in general cases compared with the above values (~105.0, ~100.1, and ~95.6 pF) and goes to the minimum in a special case such as R_{D} \to \infty, which is the same situation as the above case that \tau_{0} is independent of R_{D}. In addition to the relationship between \tau_{0} and V_{P}^{S}, the asymptotic voltage V_{A} in Eq. (3) also increases with V_{P}^{S}. As V_{P}^{S} increases, therefore, the exponential curve of V_{D} in Eq. (3) will be expanded due to the increase in V_{A}, as shown in red, green, and blue solid lines of Fig. 5(a). This will give rise to a faster arrival of V_{D} at V_{t1}, resulting in higher f_{osc}.

Figures 5(b) and 5(c) show the waveforms of I_{D} oscillation and their fast Fourier transform (FFT) amplitude spectra measured with respect to various V_{P}^{S}’s at R_{E} = 8 k\Omega (only three cases including V_{P}^{S}’s of 13.8, 14.6, and 15.4 V are displayed), respectively, when the rectangular voltage pulse is applied to device I with a pulse width of 25 \mu s. The inset plot in Fig. 5(c) shows the functional relationship between f_{osc} and V_{P}^{S}, and the blue circles indicate the measured values of f_{osc} with respect to the various values of V_{P}^{S}. The green solid line indicates the curve fitted by a logarithmic function determining f_{osc} with respect to V_{P}^{S}, which is obtained by manipulating Eq. (3) in consideration of the oscillation period. It is observed that the curve initially shows a logarithmic response but asymptotically shows a linear response after V_{P}^{S} exceeds 14 V. In an approximately linear region, the tuning sensitivity, defined as (\Delta f_{osc}/\Delta V_{P}^{S}), was calculated as 99.18 kHz/V. These results suggest that the VO_{2} device can be utilized as a voltage-controlled oscillator, which consists of only two circuit components and is operated by only one voltage source.
Figure 6(a) shows the measured waveforms of the $I_D$ oscillation with respect to various external resistances (three cases $R_E$=10, 12, and 14 kΩ at $C_E$=0 are displayed) and various external capacitances (three cases $C_E$=10, 51, and 100 pF at $R_E$=10 kΩ are displayed). As before, a rectangular voltage pulse is applied with $V_0$ of 10 V (no dc offset) and a pulse width of 25 μs. Figure 6(b) shows the FFT amplitude spectra of the corresponding $I_D$ oscillation shown in Fig. 6(a).

From the figure, it is clearly observed that $f_{osc}$ decreases with increasing $R_E$ and $C_E$. A remarkable difference is that the mean value of the peak current and peak-to-peak amplitude have relatively strong dependence on $R_E$ compared with that on $C_E$. This is caused by the fact that the amplitude of $I_D$ includes a term of $(V_sR_d/R_E)$ directly affected by $R_E$. The upper and lower inset plots of Fig. 6(b) show the linearity of the measured values of $f_{osc}$ with respect to $R_E$ and $C_E$, respectively. In each inset, the circular points indicate the measured values of $f_{osc}$ with respect to $R_E$ and $C_E$, and the solid lines indicate their linear fits. The tuning sensitivities, defined as $(\Delta f_{osc}/\Delta R_E)$ and $(\Delta f_{osc}/\Delta C_E)$, were calculated as 87.89 kHz/kΩ and 3.76 kHz/pF, respectively. The standard deviations of the linear fits in upper and lower insets were calculated as 0.0267 and 0.0282, respectively. These results suggest that a VO$_2$ two-terminal device can be incorporated as a simple oscillator whose frequency is tuned by only passive circuit components such as resistors and capacitors. In addition, the sensitivity of $f_{osc}$ on $V_s^p$ makes this device a good candidate for a voltage-controlled oscillator. Previous experimental evidence that light can photoinduce the MIT transition in VO$_2$ suggests that this is another possible external control parameter for these oscillations.

IV. COMPARISON ANALYSIS OF MOTT MIT AND PEIERLS (CDW) MIT

A. The Mott MIT as evidenced by the observed current jump

The observed MIT-driven electrical oscillation occurs between instabilities $P_1(V_{11},I_{11})$ and $P_2(V_{12},I_{12})$ at the $I$-mode trace, and begins at $P_1$ in Figs. 1(c) and 1(d). Since $I$-mode limits a current flow, the jump appears as a jump in voltage instead of a current jump. This jump has a very small hysteresis, as shown in Figs. 1(c) and 1(d). The small hysteresis indicates that the generated heat is small and does not reach the temperature needed to induce a SPT. This was directly proved by simultaneous measurements of micro-x-ray diffraction and $I$-$V$ measurements. The current jump in the $V$-mode trace in Figs. 1(c) and 1(d), viewed as the MIT, is thus independent of any SPT. A jump in resistance not accompanied by a SPT was also revealed by observing the temperature dependence of coherent phonons and the pressure dependence of phonons. It was also disclosed that current jump near the SPT temperature disappears. Hole carriers were observed at the semiconductor side near 67 °C.

As shown in plot VI’s of Figs. 4(a) and 4(b), heat generation caused by an increase in current through our device makes the MIT-driven electrical oscillation disappear. The available experimental results thus draw a decision that the electrical oscillation does not occur above a certain high temperature. It is deduced that this oscillation is below 77 °C based on the experimental results. Thus, the microscopic physics responsible for inducing the observed electrical oscillation is that of the hole-driven Mott MIT.
B. The MIT analysis in view of CDW

Since it has been suggested that the MIT oscillation is related to CDW or charge ordering for $K_{0.3}$MoO$_3$ and an organic material, recent work15–17 has focused on the MIT in the context of the Peierls MIT.13,17,18 The CDW energy is given by $V_{\text{CDW}} = \frac{b^2}{2} (q_i - q_j)^2$, where $b$ is a damping coefficient produced by charge difference between nearest-neighbor sites (i and j) of $\Delta q = (q_i - q_j)$, $k$ is the spring constant of an oxygen atom, and $x_o$ is a distortion length of a lattice from an equilibrium position. As an example of CDW,13 when we set two electrons at site i and no electron at the nearest-neighbor site j, the charge ordering (or charge difference) between nearest-neighbor sites is $\Delta q = (q_i - q_j) = (2 - 0) = 2$.

The melting of charge ordering indicates the Peierls MIT where $V_{\text{CDW}}$ and $x_o$ approach zero due to $\Delta q = 0$. This suggests that the distortion of the structure continuously lessens and finally disappears. Further, the Peierls CDW MIT should exponentially occur with carrier doping or temperature.13,17,18 Thus, since it has been known that the abrupt MIT cannot be explained in terms of CDW or charge difference28,29 for $K_0.3$MoO$_3$ and an example of CDW,13,17,18 when we set two electrons at site $i$ and no electron at the nearest-neighbor site $j$, the charge ordering (or charge difference) between nearest-neighbor sites is $\Delta q = (q_i - q_j) = (2 - 0) = 2$.

Furthermore, the qualitative and quantitative analyses of experimental results suggest that this MIT oscillation, whose frequency can be tuned by active or passive circuit components, can be beneficially applied to fields of power electronics including inverters, high-voltage dc transmission systems, and static synchronous compensators.58

V. CONCLUSION

In conclusion, we have demonstrated sustained electrical oscillations in a simple series circuit comprised of a voltage source, a resistor, and our VO$_2$ device. These oscillations are the result of the MIT in VO$_2$. It is shown that these MIT oscillations are attributed to the creation and extinction of a capacitor composed of the semiconducting and metal phases of VO$_2$. These two phases—coexisting at a nanoscale in VO$_2$ during the MIT—can be regarded as the dielectric material and electrodes of many nanocapacitors. Since these capacitive components disappear with input heat energy, the MIT oscillation is best explained not in terms of Joule heating inducing a SPT but in terms of hole carriers (or electronic carriers) driving a Mott MIT. Thus, the presence of the MIT oscillation in the VO$_2$ devices becomes the evidence of the Mott MIT.

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When Kim as an author of this paper wrote Ref. 5 on current jump in 2003, he predicted that because the current jump is a change of resistance and not driven by heat, when an appropriate resistance is connected to the MIT device, the MIT oscillation can be generated and be evidence of the Mott MIT. In 2003, since the MIT oscillation was not observed, the research on the MIT oscillation was not inserted into Ref. 5. After then, Kim’s researchers tried and discussed to observe the MIT oscillation in VO₂, but they could not succeed. Lee as an author of this paper unexpectedly observed a photo-assisted electrical oscillation and successively found the MIT electrical oscillation. It took over 4 years.


