Electrically tunable sign of capacitance in planar W-doped vanadium dioxide micro-switches

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Abstract

Negative capacitance (NC) in a planar W-doped VO₂ micro-switch was observed at room temperature in the low-frequency range 1 kHz–10 MHz. The capacitance changed from positive to negative values as the W-doped VO₂ active layer switched from semiconducting to metallic state under applied voltage. In addition, a capacitance–voltage hysteresis was observed as the applied voltage was cycled from −35 to 35 V. These observations suggest that NC results from the increase of the electrically induced conductivity in the active layer. This NC phenomenon could be exploited in advanced multifunctional devices including ultrafast switches, field-effect transistors and memcapacitive systems.

Keywords: thermochromic, phase transition, vanadium dioxide, negative capacitance, switching

1. Introduction

Vanadium dioxide (VO₂) exhibits a semiconductor-metal phase transition (SMT) at a relatively low transition temperature \( T_t \approx 68 \, ^\circ\text{C} \) [1]. This SMT is accompanied by a drastic change of electrical and optical properties in the infrared region. The VO₂ electrical resistivity decreases by several orders of magnitude from the semiconducting (high resistance) state to the metallic (low resistance) state. In addition, whereas VO₂ is transmitting light in the semiconducting state (at \( T < T_t \) ), it becomes more reflective and opaque in the metallic state (at \( T > T_t \)). This optical contrast switching is especially important in the infrared range. The SMT is easily induced by external stimuli such as temperature, photo-excitation, electric field, carrier injection and pressure. The \( T_t \) can be controlled by doping VO₂ films with donor-like or acceptor-like centers. In particular, the value of \( T_t \) can be decreased to room temperature by judiciously controlling the W doping concentration. Furthermore, the optical and electrical hysteresis can be completely eliminated by co-doping VO₂ with W and Ti [2]. The SMT characteristics of VO₂ can thus be exploited in various technological applications including all-optical switches, electro-optical switches, uncooled IR micro-bolometers, smart windows, etc. Studying and understanding the optical and electrical properties of this fascinating smart material are of great importance for successfully exploiting the SMT in multifunctional devices. The optical properties and electrical resistivity of VO₂ are largely reported in the literature. However, studies reporting capacitance–voltage characteristics (C–V) of VO₂ devices are scarce. Sclag and Scherber [3] investigated the temperature dependence of C–V of a VO₂-based metal–insulator–semiconductor device at 1 and 10 kHz using photo-resist as insulating layer. They observed that at any frequency, the capacitance increased with temperature while the state changed from semiconducting to metallic. On the other hand, in the semiconducting state, the capacitance decreased when the frequency was increased from 1 to 10 kHz. In contrast, the capacitance of the metallic state was frequency independent. Bugaev et al [4] studied the temperature dependence of the capacitance of Al-mica-VO₂ sandwich structures at 1 MHz. They observed abrupt changes in the capacitance when the VO₂ layer thermally switched from semiconducting to metallic state.
More recently, Ko and Ramanathan [5] investigated the capacitance and ac conductance of a planar Pd–VO₂–Si structure. They observed that the capacitance decreased when temperature was increased from room temperature up to 100 °C.

In this work, we show that the capacitance of a W-doped VO₂/Al₂O₃ planar micro-switch can be electronically tuned at room temperature from positive to negative values in the low-frequency range 1 kHz–10 MHz. The capacitance becomes negative when the applied electric field induces the SMT of the W-doped VO₂ layer.

2. Experimental details

High-quality thermochromic W(1.4 at%)–doped VO₂ layers (150 nm thick) were synthesized on c-Al₂O₃(0001) substrates by reactive pulsed laser deposition. The details of deposition conditions and the switching characteristics of these layers were reported in previous publications [6, 7]. Standard photolithography followed by plasma etching was used to pattern the layers into planar micro-switches (100 µm wide by 1000 µm long). To form electrical contacts, a NiCr layer (150 nm thick) was integrated over the micro-switch by lift-off process. The fabricated planar micro-switch device has a mirror symmetry as shown in inset of figure 1(a).

Recently [8], using such a structure [W(1.4 at%)-doped VO₂/c-Al₂O₃], we have demonstrated that the SMT can be exploited for the fabrication of planar micro-optical switch driven by a relatively low external voltage of ≈28 V. The temperature dependence of electrical resistance of that device showed clearly an SMT at Tᵣ = 36 °C. A reversible transmittance switching (on/off) as high as 28 dB was achieved at λ = 1.55 µm. In addition, transmittance switching modulation was demonstrated at λ = 1.55 µm by controlling the SMT with superposition of dc and ac voltages. The device was switched reversibly on and off during about 10,000 cycles without any degradation of its performance (i.e. the transmittance switching modulation was completely reversible and reproducible).

The dc current–voltage (I–V) curve of the fabricated micro-switch was recorded at room temperature using a semiconductor parameter analyzer (HP 4145A). The dependence of the capacitance on both dc voltage and ac frequency as well as the micro-switch conductance were measured at room temperature with a low-frequency impedance analyzer (HP 4192A) at an oscillating voltage of 50 mV. Note that the micro-switch device was directly connected to the HP measurement systems without using any external load electrical resistance. The choice of the W-doped VO₂ as active layer for the fabrication of micro-switch devices is motivated by its lower electrical resistance as compared to undoped VO₂. This enables an easy control of its SMT with a relatively low external voltage [8].

3. Results and discussion

Figure 1(a) shows the typical dc I–V characteristics of the W-doped VO₂ planar micro-switch. The current step was about 0.5 mA. The voltage monotonously increases with current until it reaches a maximum value Vₘ of about 23.5 V at a current Iₘ of 13 mA. Beyond this current, the voltage decreases while I further increases indicating a negative differential resistance [9]. The negative resistance effect occurs when the W-doped VO₂ layer is in the metallic state. Figure 1(b) shows the variation of the electrical resistance as a function of the applied current, revealing that the W-doped VO₂ switches from the semiconducting state (high resistance) to the metallic state (low resistance).

Figures 2(a) and (b) show the frequency dependence (from 1 kHz up to 10 MHz) of both conductance (figure 2(a)) and capacitance (figure 2(b)) of the W-doped VO₂ micro-switch for the semiconducting state (at 0 V) and the metallic state (at 35 V). Figure 3 shows the frequency dependence of the capacitance in the range 1–100 kHz in the semiconducting state (at 0 and 1 V) and the metallic state (at 35 V). It reveals that the semiconducting state has a similar dependence of the capacitance at higher frequencies (f > 8 kHz), while at low frequencies the capacitance is slightly dependent on the bias voltage. Note that Wang et al [23] observed a similar dependence of the capacitance on both frequency and bias voltage (0–3 V) in a La₀.₅Sr₀.₅MnO₃/Nb-doped SrTiO₃ heterojunction. As can be seen, the behavior of conductance and capacitance...
Figure 2. (a) Frequency dependence of the conductance of W-doped VO$_2$/Al$_2$O$_3$ planar micro-switch for semiconducting (at 0 V) and metallic (at 35 V) states. (b) Corresponding frequency dependence of capacitance at bias voltages of 0 and 35 V.

Figure 3. Dependence of the capacitance of W-doped VO$_2$/Al$_2$O$_3$ planar micro-switch for semiconducting (at 0 and 1 V) and metallic (at 35 V) states in the low frequency range (1–100 kHz). The inset shows the frequency dependence of capacitance at a bias voltage of 1 V.

differs depending whether the state is semiconducting or metallic. Overall, as expected, the metallic state is more conducting than the semiconducting state (see figures 2(a)

and 3). However, the metallic state is characterized by a negative capacitance (see figure 2(b)), while it is always positive in the semiconducting state. A detailed analysis of figure 2(b) shows that at low frequencies, the capacitance of the semiconducting state decreases abruptly. It then reaches a broad minimum and increases slowly at higher frequencies. An opposite behavior is observed for the metallic state—the capacitance increases rapidly with frequency, reaches a broad maximum, and slowly decreases at higher frequencies. Above 1 MHz, the capacitance switching contrast, defined as the capacitance difference between the two states, is about 10 pF. To confirm that the negative capacitance is due to the SMT of the W-doped VO$_2$ layer, we measured the C–V characteristics of various standard capacitors as a function of frequency and found that the capacitance of these capacitors was constant and positive.

In order to investigate in details this intriguing NC effect, we measured the capacitance at three different frequencies as a function of the bias voltage (from −35 up to 35 V). The applied switching sequence was chosen in such a way that the system changes from metallic (at −35 V) to semiconducting (at 0 V) and back to metallic state (at 35 V). Figure 4 compares the measured C–V characteristics at 1 kHz, 100 kHz and 10 MHz. The bias voltage step was about 1 V. At 1 kHz, the capacitance is initially negative and constant and starts to increase at about −15 V to reach a positive value at about −10 V. Beyond this voltage, the capacitance
is also due to the switching history of the device, as shown in figure 5:

- The C–V hysteresis memory effect of W-doped VO$_2$/Al$_2$O$_3$ planar micro-switch at 1.5 MHz. The measurements were carried out sequentially as indicated by the four C–V curves labeled 1, 2, 3 and 4.

The hysteresis width is typically 6–8 V. This result confirms that NC could appear if the conductivity is inertial (i.e. current lags behind voltage) [25].

Complex electrical circuits generating NC are currently used to improve the performance of various devices such as RF active band-pass filter [27], electrostatic actuators [28], piezoelectric actuator [29], sound-shielding systems [30], etc. More recently, it was proposed that NC in ferroelectric layers could considerably improve the gain of field-effect transistors (FETs) [31–34]. The electrical circuits generating NC are complex and require judicious choice of the electrical components and the control of the flowing current. Therefore, devices requiring NC could be considerably improved by replacing NC-electrical circuits by simple VO$_2$-negative-capacitor devices. The advantages of using VO$_2$-NC devices include simplicity and easy control of the SMT (i.e. control of the capacitance) by various external stimuli such as temperature, photo-excitation, electric field, carrier injection and pressure. These VO$_2$-NC capacitors can be used to reduce the subthreshold swing in FETs as simulated by Salahuddin and Datta [32] for a ferroelectric-based FET device with negative capacitance. In addition, the ultrafast phase transition of VO$_2$ [35] can be exploited for the fabrication of innovative ultrafast capacitor sensors.

Such VO$_2$-device can also be combined with standard capacitors to fabricate advanced tunable capacitor devices exhibiting a C–V hysteresis memory effect with positive capacitance. For example, figure 6 shows the positive C–V hysteresis as measured for a standard capacitor (C = $1.59 \times 10^{-10}$ F) in parallel with the VO$_2$-negative capacitor device. The hysteresis width is about 5 V. This result confirms that the negative capacitance is not due to the response of the HP device.

It is well known that external electric field induces formation of conducting filaments or current channels at the surface of VO$_2$ [36–38]. Recently Okimura et al. [39] reported that the formation of the current channels is responsible for the multistep resistance switching observed in the I–V characteristics of a planar VO$_2$/c-Al$_2$O$_3$ device. In our case, the observed NC and the variation of the conductance (see figure 2) cannot be uniquely explained by the formation of a current channel under the applied switching voltage. Indeed, our experimental results show clearly that the NC is directly

continues to slightly increase, reaches a maximum at $-7$ V and then decreases slowly up to 20 V and faster beyond this value. A somewhat similar behavior is observed at 100 kHz even though the return towards negative values takes place at a lower voltage (15 V rather than 20 V). At 10 MHz, the capacitance is initially slightly negative and decreases significantly to reach a minimum value at $-15$ V. It subsequently increases, crossing the zero line at about $-12$ V, remains positive up to $\sim$20 V and decreases again to negative values. Overall, the sign of the capacitance is correlated with the semiconducting state when it is positive and with the metallic state when it is negative. Note that the capacitance switching contrast decreases with increasing frequency. For example, it is about 6 nF at 1 kHz and 5.7 pF at 10 MHz. In addition, the corresponding conductance (not shown here) behaves oppositely to the capacitance as it is larger for the metallic state when it is negative. In addition, the ultrafast phase transition of VO$_2$ [35] can be exploited for the fabrication of innovative ultrafast capacitor sensors.

| Capacitance (F) | Bias voltage (V) |
|----------------|-----------------|
| $-6 \times 10^{-12}$ | -30 |
| $-4 \times 10^{-12}$ | -20 |
| $-2 \times 10^{-12}$ | -10 |
| $-1 \times 10^{-12}$ | 0 |
| $2 \times 10^{-12}$ | 10 |
| $4 \times 10^{-12}$ | 20 |
| $6 \times 10^{-12}$ | 30 |

The C–V hysteresis was obtained by measuring the capacitance of the device when the bias voltage cycle was alternatively reversed between $-35$ and 35 V (see figure 5). These capacitance measurements were reproducible as shown by the four C–V curves labeled 1, 2, 3 and 4, which were recorded sequentially. The curve 1 was recorded when the active layer was switched to its metallic state at $-35$ V (for this first curve, the bias voltage was changed from 0 V to $-35$ V), while the successive curves (2, 3 and 4) were recorded when the SMT of the active layer was controlled gradually by the bias voltage. The switching history of the active layer [10] can explain the small difference observed in the metallic region around $-35$ V (see curve 1). Note that the asymmetric change observed in the C–V characteristics of figure 4 is also due to the switching history of the device, that is metallic $\rightarrow$ semiconducting $\rightarrow$ metallic state. However, note the symmetric hysteresis of curves 2–4 in figure 5: the C–V curve obtained for increasing voltage is more or less the mirror image of that resulting from decreasing voltage. The hysteresis width is typically 6–8 V. This C–V hysteresis memory effect can be used in the fabrication of advanced memcapacitive systems exploiting the SMT of VO$_2$ [11].

NC has been observed in several materials and devices, such as gallium nanoparticles embedded in a dielectric matrix [12], PhS nanocrystals embedded in conducting polymers [13], In$_2$O$_3$Ge$_2$Sb$_2$Te$_2$ thin films [14], hydrogen-doped amorphous barium titanate device [15], GaN/AlGaN heterojunction dual-band detectors [16], GaAs homojunction far-infrared detectors [17], nanocomposite and polycrystalline solar cells [18], conducting polymer nanowires [19], organic semiconductor devices [20], metal-a-C$_1$–$N_2$–H-metal devices [21], porous TiO$_2$ layers [22], La$_{0.5}$Sr$_{0.5}$MnO$_3$/Nb-doped SrTiO$_3$ heterojunctions [23], etc. Its origin has been attributed to many factors including minority carrier flow, interface states, slow transition time of injected carriers, charge trapping and space charge [24–26]. It was also shown that NC could appear if the conductivity is inertial (i.e. current lags behind voltage) [25].

The C–V hysteresis memory effect of W-doped VO$_2$/Al$_2$O$_3$ planar micro-switch at 1.5 MHz. The measurements were carried out sequentially as indicated by the four C–V curves labeled 1, 2, 3 and 4.

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linked to the electrically induced increased conductivity in the active layer. In addition, Okimura and Sakai [40] investigated the time-dependent characteristics of electric-field-induced phase transition in a planar VO$_2$/c-Al$_2$O$_3$ structure. They observed a marked change of the differential conductance that indicates an increase of carrier density (hence of conductivity) in the applied electric field that alters the density of states near the Fermi level.

The frequency dependence of capacitance can be derived from Fourier analysis as [25]:

$$C(\omega) = C_0 + \frac{1}{\omega \delta V} \int_0^\infty \left[-\frac{d\delta I(t)}{dt}\right] \sin \omega t \, dt,$$

where $\omega$ is the angular frequency, $\delta I(t)$ is the transient current resulting from the application of a small voltage variation $\delta V$ superimposed to the dc bias voltage $V$ at $t = 0$, and $C_0$ is the geometric capacitance.

The NC effect may occur when the time derivative of the transient current $[d\delta I(t)/dt]$ is positive or non-monotonous with time. Penin [41] studied the NC effect in homogeneous semiconductor structures. He demonstrated that NC arises when the conductivity is inertial and that the reactive component of the current is larger than the displacement current. In this case, the transient current is related to the dc conductivity ($\sigma$). The capacitance can thus be expressed as a function of $\sigma$ as [42]:

$$C(\omega) = C_0 - \frac{A4\pi\tau\sigma}{d(1+\omega^2\tau^2)},$$

where $\tau$ is the dielectric relaxation time, $A$ the area of the semiconductor, and $d$ its thickness.

At very high frequency [$\omega \rightarrow \infty$], the second term of both equations (1) and (2) becomes negligible. The capacitance is therefore positive and tends towards the geometric capacitance $C_0$. However, at low frequency, the second term of equations (1) and (2) can become higher than $C_0$, hence resulting in NC.

As shown in figure 1(a), the $I$–$V$ curve significantly changes in the region where SMT occurs. This feature is characterized by the onset of a negative differential resistance at a threshold $V_{th}$ as already mentioned. It can be expected to be accompanied by an increase of the charge density to a critical value $N_C$ [9], resulting in a conductivity increase (i.e. decrease of electrical resistance with increasing current). In these conditions, one can empirically describe $\sigma$ by an exponential law:

$$\sigma (V) = \sigma_0 \exp \left[\frac{-(V_{th} - V)E_a}{V_{th}kT}\right],$$

where $\sigma_0$ is the conductivity at $V_{th}$, $k$ is the Boltzmann constant, $T$ is temperature and $E_a$ is the activation energy, i.e. the minimum energy required to initiate the conductivity change.

Combining equations (2) and (3) provides the dependence of the capacitance on both $\omega$ and $V$:

$$C(\omega, V) = C_0 - \frac{A4\pi\tau}{d(1+\omega^2\tau^2)}\sigma_0 \exp \left[\frac{-(V_{th} - V)E_a}{V_{th}kT}\right].$$

Relation (4) clearly indicates that the capacitance could be negative if the exponential is large enough, which may occur when $V$ is larger than $V_{th}$. As mentioned earlier, the conductance measurements indicate that the W-doped VO$_2$ becomes more conductive as the switching voltage increases as shown in figure 2(a). Therefore, the NC phenomenon observed in our device can be related to the electrically induced enhancement of $\sigma$.

4. Conclusions

We have shown that NC occurs in a planar W-doped VO$_2$ micro-switch at room temperature when SMT is tuned by an external electric field. This onset of NC can be explained by the increase of electrical conductivity that occurs when W-doped VO$_2$ switches from its semiconducting to metallic state. In addition, a relatively large C–V hysteresis occurs when the bias voltage is cyclically switched from $-35$ to $35$ V. The results of this investigation open new perspective for the future exploitation of SMT of VO$_2$ in advanced multifunctional devices such as ultrafast switches, FETS and memcapacitive systems.

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