

The Metal–Insulator Transition in Vanadium Oxide Nanofilms Enables Microkelvin-Resolution Thermometry

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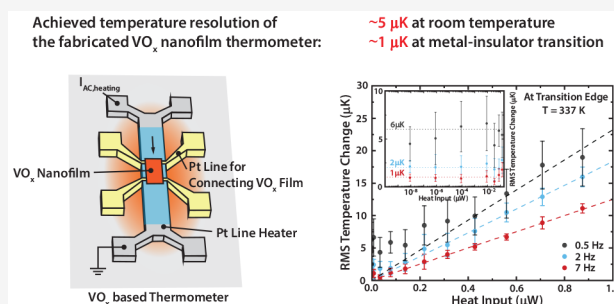
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Supporting Information

ABSTRACT: High-resolution thermometry is critical for probing nanoscale energy transport. Here, we demonstrate how high-resolution thermometry can be accomplished using vanadium oxide (VO_x), which features a sizable temperature-dependence of its resistance at room temperature and an even stronger dependence at its metal–insulator-transition (MIT) temperature. We microfabricate VO_x nanofilm-based electrical resistance thermometers that undergo a metal–insulator-transition at ~ 337 K and systematically quantify their temperature-dependent resistance, noise characteristics, and temperature resolution. We show that VO_x sensors can achieve, in a bandwidth of ~ 16 mHz, a temperature resolution of ~ 5 μK at room temperature (~ 300 K) and a temperature resolution of ~ 1 μK at the MIT (~ 337 K) when the amplitude of temperature perturbations is in the microkelvin range, which, in contrast to larger perturbations, is found to avoid hysteric resistance responses. These results demonstrate that VO_x -based thermometers offer a ~ 10 –50-fold improvement in resolution over widely used Pt-based thermometers.

KEYWORDS: thermometry, metal–insulator transition, phase change, high-resolution, microkelvin



Thermometry plays a critical role in a variety of research fields, including the study of heat transport in atomic and nanoscale systems,^{1–3} energy dissipation at the atomic scale,^{4,5} scanning thermal microscopy,^{6,7} and calorimetry of biological⁸ and inorganic systems.⁹ A number of state-of-the-art microcalorimeters used in nanoscale thermal transport studies employ resistance-based thermometers.^{10–14} In fact, a majority of these calorimeters employ Pt resistance-based thermometers that leverage the temperature dependent electrical resistivity of Pt for achieving a stable and relatively high-resolution resistive thermometer.^{7,15–19} A key metric that determines the sensitivity and resolution of a resistance thermometer²⁰ is the temperature coefficient of resistance (TCR), which is defined as

$$\text{TCR} = \frac{1}{R} \frac{dR}{dT} = \alpha \quad (1)$$

Platinum thin films feature TCR values of ~ 0.002 K^{-1} ($\sim 0.2\%$ /K) and have been incorporated into calorimeters to enable measurements with a resolution of ~ 200 μK in a bandwidth of ~ 16 mHz.²⁰ This temperature resolution, when combined with the typical thermal conductance of calorimeters, such as suspended SiN-based membranes and SiN-based cantilevers (~ 1 $\mu\text{W}/\text{K}$), results in a calorimetric resolution of ~ 200 pW.^{7,15,21} This impressive resolution has enabled a number of interesting studies and resulted in important new

insights into nanoscale thermal transport.^{15,16,21–23} However, there are many physical phenomena, e.g., thermal transport in 1D-polymer chains or single cell biocalorimetry, that require an even higher calorimetric resolution (on the order of ~ 1 –10 pW), which can be potentially achieved by developing thermometers with higher TCRs than that offered by Pt resistance thermometers.

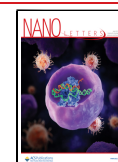
Vanadium oxide has been extensively studied^{24–28} due to its interesting phase transition properties that are attractive for novel optical,^{29,30} electrical^{31,32} and thermal applications.^{33–35} Specifically, VO_x films exhibit a strong temperature-dependence that is characterized by a monotonic resistance change of several orders of magnitude when the film is heated by a few degrees Kelvin. For example, past work³⁶ has shown a drop in resistance in excess of 3 orders of magnitude when VO_2 -films are heated from 340 to 344 K. This strong temperature-dependence of resistance results in very large values of TCR. In fact, past work^{36–38} has shown that VO_x films feature TCR values with a magnitude above 10%/K at the metal–insulator

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transition (MIT) temperature of ~ 340 K and feature TCR values of $\sim -2\%/K$ at room temperature. Further, past work³⁹ has also explored the possibility of using VO_x thin films as temperature sensors and has experimentally reported temperature resolution in the mK range (in a bandwidth of ~ 16 Hz). In spite of the interesting past work described above, the resolution limit that can be achieved using VO_x -based sensors remains poorly understood.

In this work we perform systematic studies to identify the temperature resolution limit of VO_x -based temperature sensors. Toward this goal, we first microfabricated a device that features both a Pt heater and a VO_x -based thin-film (100 nm thick) thermometer as shown in Figure 1a (for details of

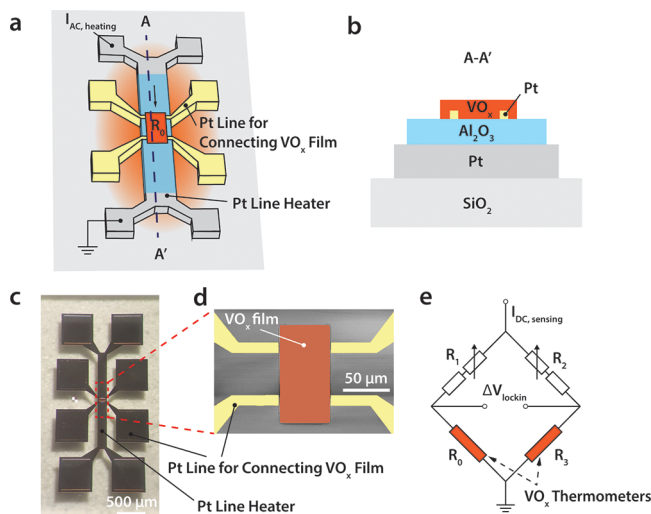


Figure 1. Schematic of the VO_x -devices studied in this work along with optical and SEM images and a schematic of the Wheatstone bridge employed for temperature measurements. (a) Schematic representation of a VO_x thermometer device (not drawn to scale) with an integrated Pt heater (gray) that is powered by the heater current $I_{AC, heating}$. The VO_x -thermometer (red) is deposited onto a Pt heater (gray) and electrically isolated from the Pt heater by a thin Al_2O_3 layer (blue) that maintains excellent thermal coupling. Electric connections for the readout of the VO_x -thermometer are made via separate, patterned Pt lines (yellow) to form a 4-wire connection. (b) Cross-section of a fabricated device along the dotted line A–A' in (a). (c) An optical image of the fabricated device showing the large wire bonding pads, the Pt heater line and the VO_x -thermometer. (d) A false colored scanning electron microscope (SEM) image shows the VO_x film with respect to the sensing Pt lines. (e) Schematic representation of the Wheatstone Bridge showing one heated VO_x -thermometer (the actual sensor in the reported measurements) and one unheated reference thermometer, labeled R_0 and R_3 , respectively, that are otherwise identical. In addition, resistors R_1 and R_2 , composed of potentiometers and fixed resistors, complete the bridge circuit. The two VO_x thermometers are mounted in close proximity (~ 4 mm) to enable excellent common mode rejection of residual thermal drift and electromagnetic interference noise. To characterize the temperature-resolution of the VO_x films, only one of the VO_x films (R_0) is heated.

the fabrication steps of the device see the Supporting Information (SI)). The cross-sectional profile of the fabricated device is shown in Figure 1b, and an optical image of the device along with all the electrical connections is shown in Figure 1c, including a false-colored scanning electron microscopy image (Figure 1d) of the region that has the VO_x film which makes contact with the four-probe leads. To conduct high-resolution temperature measurements we used

two, nominally identical VO_x -thermometer devices, which served as a sensor and a reference, and were integrated into a Wheatstone bridge circuit (see Figure 1e, where these VO_x resistors are labeled R_0 and R_3 , respectively). Further, two resistors, R_1 and R_2 , that each consist of a fixed 100 k Ω metal foil resistor (Vishay Foil Resistors Y0062100K000T9L200 with temperature coefficients of 1 ppm/ $^\circ\text{C}$) in series with a 100 k Ω potentiometer (TT Electronics 7286R100 KL.25, temperature coefficient of 20 ppm/ $^\circ\text{C}$), were included in the Wheatstone bridge as shown in Figure 1e.

We quantify the temperature resolution of the VO_x nanofilm thermometer based on the following approach. Briefly, a sinusoidal electrical current with amplitude $I_{AC, heating}$ and frequency f was supplied to the Pt heater in the sensor device (R_0) to generate an AC temperature modulation, ΔT_{AC} , at frequency of $2f$. At the same time, we also supplied a DC current, $I_{DC, sensing}$, to the Wheatstone bridge (with the VO_x sensors) shown in Figure 1e. The voltage output (ΔV_{2f}) at $2f$ from the Wheatstone bridge was recorded by first amplifying it via a voltage amplifier (SR 560, Stanford Research Systems (SRS)) with a gain ($G_v = 100$) and then demodulated with a lock-in amplifier (SR 830, SRS) by locking-in to the signal at $2f$. The measured ΔV_{2f} can be related to the temperature change (ΔT_{AC}) by the following expression (see details in the SI):

$$\Delta T_{AC} = \frac{\Delta V_{2f}(R_0 + R_1 + R_2 + R_3)}{G_v \alpha I_{DC, sensing} R_0 R_2} \quad (2)$$

where α is the TCR of the VO_x nanofilm at different operating temperatures. Below, we first describe how we systematically characterized α for our VO_x films.

We measured the temperature-dependence of the resistance of the VO_x nanofilm in a cryostat, where the temperature of the VO_x sample was varied in steps of 5 K (temperature changing rate is ~ 1 K/min) from 297 to 347 K and back to 297 K. At each temperature point, we waited for 50 min to thermally stabilize the cryostat temperature. Then, we measured the electrical resistance of the VO_x nanofilm using a standard four-probe measurement technique, where a 1 μA amplitude sinusoidal current ($I_{AC, 7\text{ Hz}}$) at a frequency of 7 Hz was applied to the VO_x thin film and the voltage across the thin film ($V_{AC, 7\text{ Hz}}$) was measured by a lock-in amplifier (SR 830, SRS) (see inset of Figure 2a for a schematic of the measurement approach) to obtain the temperature (T)-dependent electrical resistance ($R(T)$) via $R(T) = V_{AC, 7\text{ Hz}} / I_{AC, 7\text{ Hz}}$. The data from these experiments are shown in Figure 2a, where the measured electrical resistance is plotted as a function of temperature. These data indicate that the VO_x nanofilm undergoes a metal–insulator-transition (MIT) between 330 to 340 K and features thermal hysteresis with a heating and cooling cycle. In Figure 2b we present the TCRs of the VO_x film that are obtained using eq 1 and the resistance data presented in Figure 2a. These data indicate that the VO_x film features a TCR of $\sim -2.24\%/K$ at around room temperature (297–320 K) and a maximum TCR of $\sim -19\%/K$ at the MIT in the heating cycle.

We note that the observed thermal hysteresis, especially around the MIT, raises the question as to whether VO_x sensors can actually be employed for temperature sensing applications as linearity and a history-independent behavior are highly desirable, if not essential, for temperature sensing applications. Therefore, to understand if VO_x thermometers can be

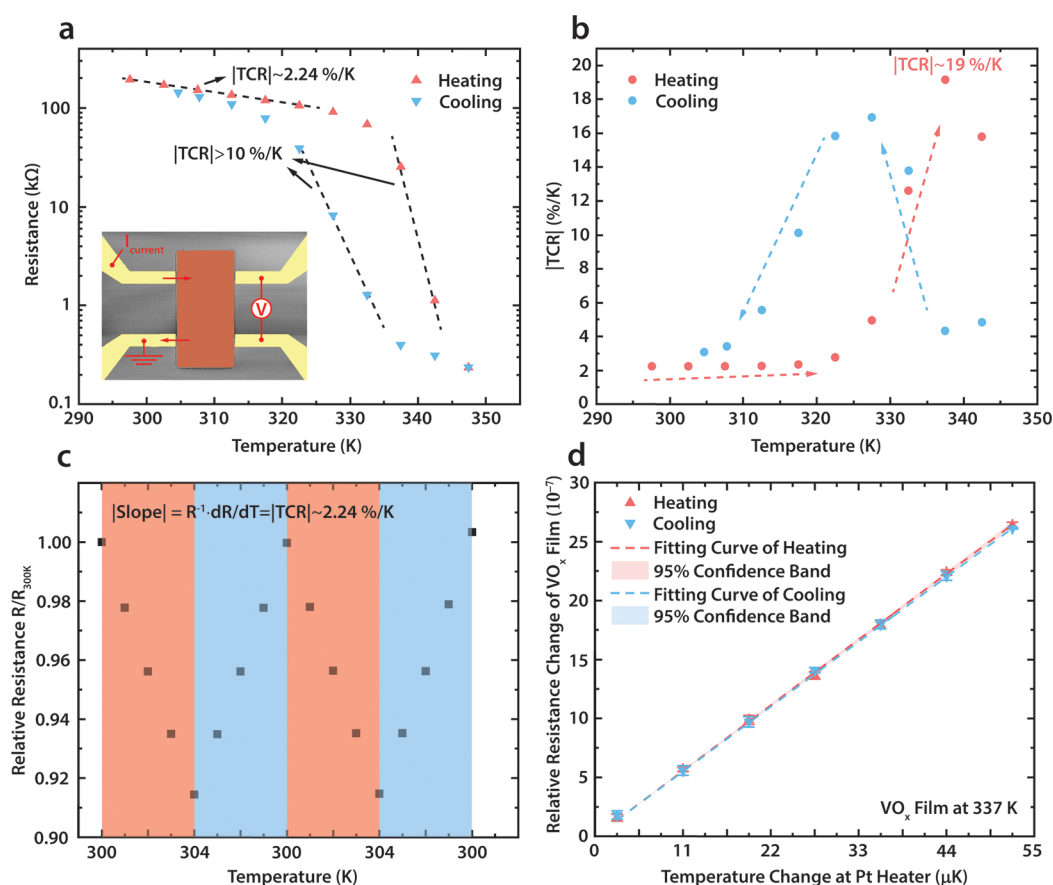


Figure 2. Characterization of VO_x nanofilm thermometers. (a) Measured resistance as a function of temperature during heating and cooling cycles. The VO_x nanofilm exhibited thermal hysteresis during the heating and cooling cycles and undergoes a metal–insulator transition (MIT) between 330 to 340 K during which its resistance changed by ~ 3 orders of magnitude. Inset shows the measurement configuration. (b) Magnitude of the temperature coefficient of resistance (TCR) as a function of temperature obtained from data shown in panel (a). The film's absolute TCR value is $\sim 2.24\%/K$ at room temperature and above $10\%/K$ at the MIT. The maximum observed TCR values were $\sim 19\%/K$ and $\sim 17\%/K$ during the heating and cooling cycles, respectively. (c) Temperature dependence of the electrical resistance of VO_x nanofilms when the temperature is swept from 300 to 304 K inside a cryostat. It can be seen that the electrical resistance changes reversibly as the temperature is cycled. The uncertainty of the plotted points in (a), (b) and (c), obtained from 5 min recorded data using the lock-in amplifier is smaller than the symbols. (d) Relative resistance change of the VO_x nanofilm thermometer as a function of the temperature change of the Pt heater, via Joule heating, in four consecutive heating and cooling cycles (at an environmental temperature of ~ 337 K). The symbols represent mean values, whereas the error bars represent the standard deviation across four cycles. The resistance change of the VO_x thin film was measured via the Wheatstone bridge shown in Figure 1e. Results indicate the resistance change of the VO_x nanofilm thermometer is linear and free of hysteresis with respect to the heat input (i.e., temperature change of the Pt heater).

employed for measuring small changes in temperature, we explored if they feature linear (i.e., without hysteresis) electrical resistance characteristics, upon small variations in temperature. First, we performed temperature sweep cycles at 300 K (i.e., at temperatures far from the MIT) by ramping the temperature up and down by 4 K. The data obtained from these experiments are shown in Figure 2c, where we show the relative resistance change ($R/R_{300\text{ K}}$) as a function of temperature. It can be seen that the resistance change is indeed linear without thermal hysteresis. Further, we find that the TCR of the VO_x thin-films under this condition is $\sim -2\%/K$, revealing that they have a relatively high TCR in comparison to Pt films.

Next, we performed similar experiments at 337 K (a temperature that corresponds to the MIT), which demonstrated that when the temperature is swept by 4 K (336 K – 340 K, rate of temperature change is ~ 1 K/min) the resistance change is highly nonlinear and features hysteresis (see data in Figure S4a in Supporting Information), a behavior which is

similar to that reported in past work due to the thermal hysteresis associated with the MIT.⁴⁰ In order to explore if the resistance change at the MIT is linear (i.e., free of hysteresis) for very small temperature changes (i.e., perturbations in the range of $\sim$ tens of μ K), we performed additional experiments using the *in situ* Pt heater to perturb the temperature of the VO_x film at the microkelvin level (see Figure S3 and Section 4 of the Supporting Information for details of how the temperature rise at the Pt heater is estimated). Results from such measurements in four consecutive heating and cooling cycles are shown in Figure 2d (rate of temperature change is ~ 0.8 μ K/min). These data show that, for small temperature changes at the Pt heater ($< \sim 55$ μ K), the resistance change of the VO_x film is linear (i.e., free of hysteresis). However, we find that, when the temperature perturbations at the Pt heater were increased to above ~ 75 μ K, the resistance change of the VO_x film was again nonlinear and featured hysteresis (see Figure S4b and Section 5 in Supporting Information for details of the measurement).

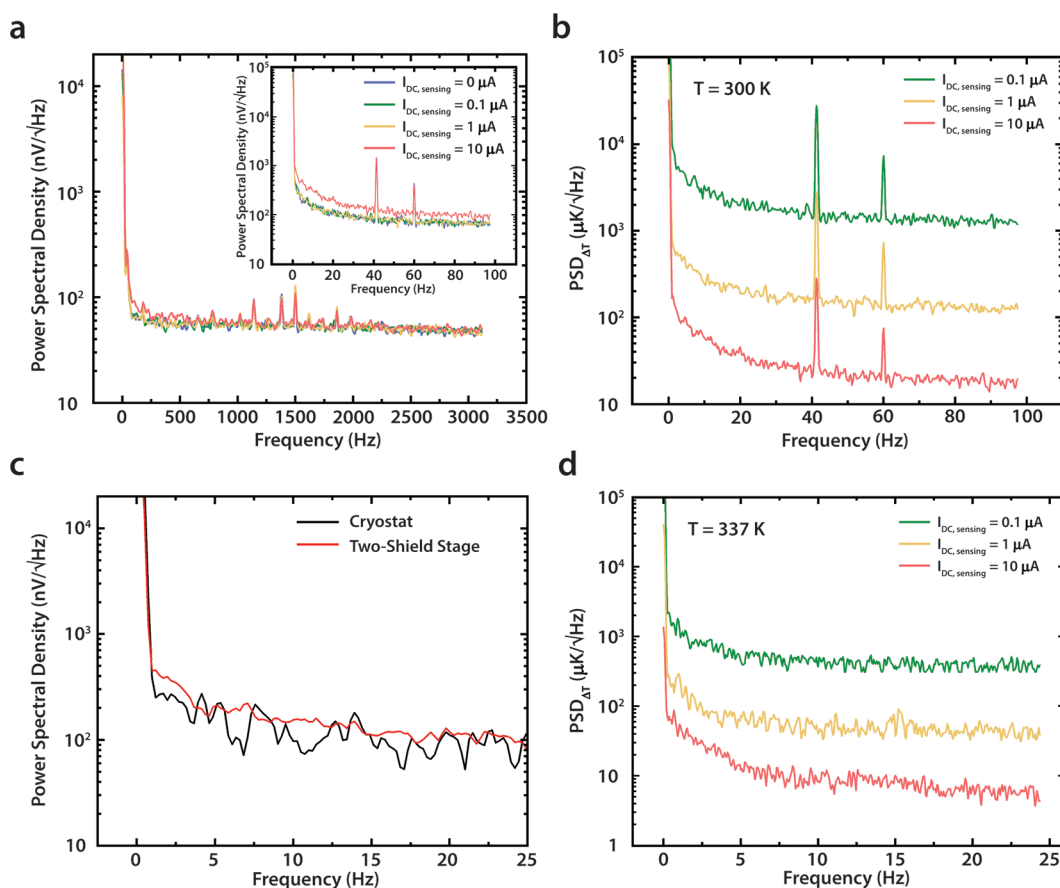


Figure 3. Measured power spectral density. (a) Measured power spectral density of noise (PSD_N) from ~ 20 Hz to 3 kHz for various sensing currents ($I_{DC,sensing} = 0 \mu A$, $0.1 \mu A$, $1 \mu A$ and $10 \mu A$) in a custom-built two-shield instrument at $300 K \pm 20 \mu K$. Inset shows the same PSD_N for a narrower frequency range (~ 1 to 97 Hz). (b) $PSD_{\Delta T}$ corresponding to the data shown in panel (a) for three different applied sensing currents, obtained using eq 3 and the data in panel (a). (c) Comparison of PSD_N obtained in measurements in a cryostat ($300 K \pm 5.5$ mK) and a two-shield instrument ($300 K \pm 20 \mu K$) with applied sensing current of $0 \mu A$. The obtained PSD_N results are almost identical indicating that noise caused by the temperature drift in the system is negligible. (d) The measured $PSD_{\Delta T}$ as a function of frequency (~ 0.3 to 24.4 Hz) at the metal–insulator transition ($337 K$) measured in the cryostat for various applied sensing currents ($0 \mu A$, $0.1 \mu A$, $1 \mu A$ and $10 \mu A$). The corresponding PSD_N plot is presented in the SI (see Figure S7). These results indicate that the temperature resolution is improved by ~ 5 times at the metal–insulator transition.

The above discussion suggests that VO_x sensors can indeed be employed at the MIT to track small (few μK to tens of μK level) temperature changes with a nearly 5-fold higher TCR than at room temperature. However, our data suggests that VO_x sensors cannot be used to measure temperature changes when the bottom Pt heater temperature change is larger than $\sim 75 \mu K$ due to the hysteresis described above. We note that, while our experiments provide first observation into how VO_x sensors can be employed to perform high resolution thermometry for measuring small temperature changes around the MIT, further studies are needed on how the linear range can be extended e.g., by controlling the rate of temperature change or the composition or the microstructure of the VO_x films.

Upon characterizing the TCR of the VO_x temperature sensors, we performed experiments to determine the noise power spectral density (PSD_N) associated with our thermometry scheme in order to estimate the noise floor of the thermometers described here. As discussed in past work,²⁰ PSD_N has contributions from Johnson noise of the resistors (see SI for an estimate of the total Johnson noise), shot noise of the current source, amplifier noise, $1/f$ noise at low frequency and noise associated with temperature drift in the

setup, the last of which we expect to be significantly attenuated due to common mode rejection offered by incorporating a reference thermometer (Figure 1e). These experiments were performed in a temperature-stabilized cryostat (temperature drift < 6 mK in 3 h, see SI for details) and in a custom-built two-shield instrument (temperature drift $< 20 \mu K$ in 5 h, see section 6 of the Supporting Information for details of the schematic of the custom-built two shield instrument). As described below, our experiments broadly support the conclusion that the noise floor in our measurements is not limited by environmental temperature fluctuations, consistent with the observation that the significantly larger temperature drift in the cryostat vs the custom-built instrument has negligible influence on the measured noise floor.

In Figure 3a, we present the PSD_N measured in the custom built two-shield instrument mentioned above (at a temperature $300 K \pm 20 \mu K$) for four different DC excitation currents of the Wheatstone bridge ($I_{DC,sensing} = 0.0, 0.1, 1.0, 10.0 \mu A$) using a commercial FFT analyzer (SR770, Stanford Research Systems). The inset of Figure 3a shows the PSD_N data on a log–linear scale for a smaller frequency range. The measured PSDs for all four cases show a $1/f$ dependence at low frequencies (especially for $f < \sim 10$ Hz) and a relatively weak

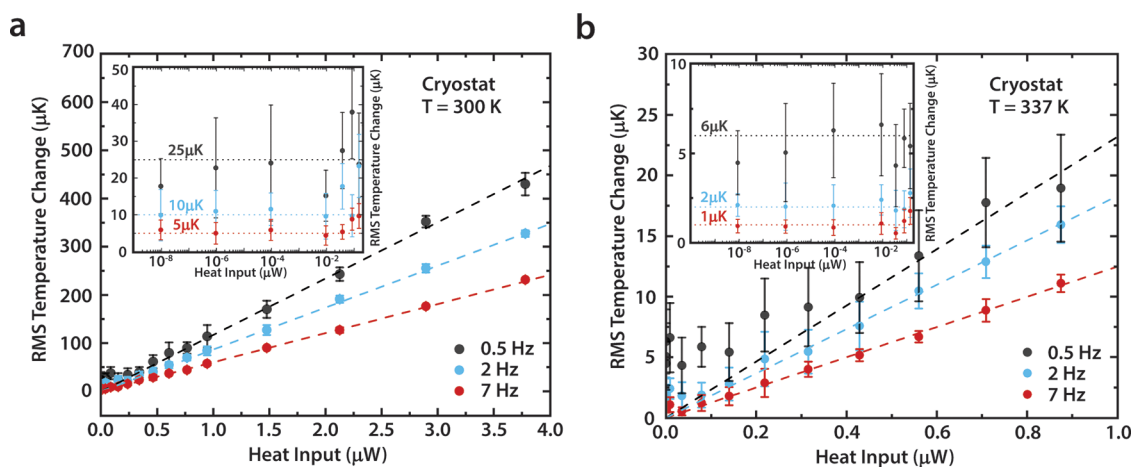


Figure 4. Characterization of the temperature resolution of a VO_x nanofilm thermometer. The RMS temperature change at (a) room temperature and (b) the metal–insulator transition temperature (337 K) for various heat inputs at frequencies (f) of 0.5 Hz, 2 and 7 Hz. The RMS temperature increases linearly with the magnitude of the heat input (dashed lines represent linear fits to the experimental data). The dotted lines in the inset, which zooms in on the same data at small heat inputs ($< \sim 0.2 \mu\text{W}$), indicate the temperature resolution of the VO_x thin film thermometer at the three frequencies. The temperature resolution at room temperature is approximately 25 μK , 10 μK and 5 μK for temperature modulations at frequencies ($2f$) of 1 Hz, 4 and 14 Hz, respectively. The temperature resolution at the MIT is about 6 μK , 2 μK and 1 μK for temperature modulations at frequencies ($2f$) of 1 Hz, 4 and 14 Hz, respectively. All measurements of the temperature resolution shown here were performed at a sensing current of 10 μA with a bandwidth of 16 mHz.

frequency dependence for frequencies above 20 Hz. It can also be seen that the measured power spectral densities in all four cases at frequencies above 500 Hz (where $1/f$ noise contribution is negligible) are rather similar indicating that shot noise makes negligible contributions to PSD_N , which is consistent with theoretical expectation for $\text{PSD}_{I,\text{Shot}} = \sqrt{2eI_s} \sim 1.26 \text{ pA}/\sqrt{\text{Hz}}$ (which translates to a voltage noise of $\sim 0.2 \text{ nV}/\sqrt{\text{Hz}}$ in our case). Moreover, it can be seen that at relatively high frequencies ($> 500 \text{ Hz}$) the measured PSD is $\sim 60 \text{ nV}/\sqrt{\text{Hz}}$, which is close to the theoretical estimate of Johnson noise $\text{PSD}_{V,\text{Johnson}} = \sqrt{4k_B T R} \sim 54.5 \text{ nV}/\sqrt{\text{Hz}}$, see SI for details), indicating that the resolution of our system is likely limited by Johnson noise. We note that the measured PSD_N at low frequencies shows a larger value than the estimate from Johnson noise and depends on the applied DC currents, which is likely due to the increase of the complicated $1/f$ noise generated from the semiconducting VO_x nanofilm and the noise induced by the current source (Keithley 6221) which increases with the applied DC current amplitude. Next, we compute the power spectral density of temperature ($\text{PSD}_{\Delta T}$) from the following expression:

$$\text{PSD}_{\Delta T} = \frac{\text{PSD}_N(R_0 + R_1 + R_2 + R_3)}{G_v \alpha I_{\text{DC,sensing}} R_0 R_2} \quad (3)$$

The obtained $\text{PSD}_{\Delta T}$ is shown in Figure 3b and reveals, as expected, improved temperature resolution for higher sensing currents. Similar experiments were also performed in the cryostat described above (at 300 K) and the obtained PSD_N values are nearly identical to those obtained in the two shield instrument (see Figure 3c), highlighting that the temperature drift in the cryostat poses no limitation to resolution. After establishing these facts, we also measured $\text{PSD}_{\Delta T}$ at the MIT in the cryostat described above. The data obtained from these experiments are shown in Figure 3d and indicate a lower $\text{PSD}_{\Delta T}$ (~ 5 fold lower), suggesting an ~ 5 fold better temperature resolution at the MIT. From the data shown in

Figure 3b and 3c, we estimated the temperature resolution in a 16 mHz bandwidth (this bandwidth was chosen to be consistent with past work²⁰) and found the expected resolution to be $\sim 5 \mu\text{K}$ at room temperature and $\sim 1 \mu\text{K}$ at MIT for an applied $I_{\text{DC,sensing}}$ of 10 μA and an AC current $I_{\text{AC,heating}}$ heating frequency (f) of 7 Hz (i.e., a temperature modulation frequency ($2f$) of 14 Hz).

To experimentally establish the above estimated temperature resolution of our VO_x thermometers, we took an approach where an AC current $I_{\text{AC,heating}}$ at various frequencies ($f = 0.5, 2, \text{ or } 7 \text{ Hz}$) was supplied to the Pt heater in device, while a 10 μA DC current $I_{\text{DC,sensing}}$ was supplied to the Wheatstone bridge. In Figure 4a we show the RMS temperature change detected (for measurements performed in the cryostat) by the VO_x nanofilm thermometer, as a function of the amplitude of heating power ($Q = I_{\text{AC}}^2 R_{\text{Pt}}/2$, see SI for details) at 300 K. As expected, the measured temperature increased linearly with the heating power when the detected temperature change was above the noise floor. As shown in Figure 4a and more clearly in the inset, the noise floor (i.e., the minimal detectable temperature change) at 300 K is 25 μK , 10 μK and 5 μK for heating frequencies (f) of 0.5 Hz, 2 and 7 Hz, respectively, in a bandwidth of $\sim 16 \text{ mHz}$. This measured temperature resolution is indeed consistent with the estimate presented above and represents a significant improvement over the resolution of Pt resistance thermometry, which has a resolution of 212 μK , 78 μK and 34 μK under similar conditions when the heating frequency (f) is 0.5 Hz, 2 and 20 Hz.²⁰

Next, we performed similar thermal resolution experiments at the MIT and the data obtained from these experiments is shown in Figure 4b. As expected, the temperature resolution is improved by ~ 5 fold at the MIT as can be clearly seen from data in the inset of Figure 4b, where we find a noise floor of 6 μK , 2 μK and 1 μK for electrical current frequencies (f) of 0.5 Hz, 2 and 7 Hz, respectively, in a bandwidth of $\sim 16 \text{ mHz}$. Curiously, while the measured data show a linear dependence on the input power, the absolute temperature rises at 300 K and at the MIT are different for the same input power. This

difference is likely because the VO_x film undergoes a first order phase transition,⁴¹ which attenuates the temperature rise of the Pt heater (see Figure S3 in Supporting Information).

In conclusion, we described a VO_x-based thin film thermometer that features a TCR of $\sim -2.24\%/K$ at room temperature and $\sim -15\%/K$ at the metal–insulator transition (MIT) at 337 K. We systematically determined the temperature resolution of VO_x thermometers and established a room temperature (300 K) resolution of $\sim 5\ \mu K$ for temperature changes modulated at 7 Hz with a bandwidth of $\sim 16\text{ mHz}$, which represents a 10-fold improvement compared to widely used Pt-based thermometers. We also demonstrated an improved temperature resolution of $\sim 1\ \mu K$ at the MIT temperature (337 K) when temperature perturbations are below $55\ \mu K$. However, for larger amplitude temperature changes the thermometer response was found to be nonlinear and featured hysteresis. Future work is necessary to examine how the thermometer response can be made to be linear even for larger perturbations around the MIT. We expect that incorporation of VO_x-based thermometers into scanning probes and calorimeters can significantly improve calorimetric resolution thus enabling novel studies of nanoscale thermal transport.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.4c01535>.

Details regarding the deposition of vanadium oxide films, fabrication of test devices, noise analysis, characterization of the temperature dependence of electrical resistance of vanadium oxide films, temperature stability of the experimental system, power spectral density of noise and data demonstrating repeatability across devices (PDF)

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Author Contributions

P.R. and E.M. conceived the work. Y.L., P.R., and E.M. wrote the manuscript, with input from all authors. Y.L., S.Y., J.G. and K.P. fabricated the microdevices and characterized the properties of the VO_x nanofilms. K.P., A.M. and R.M. built the two-shield instrument for testing the performance of the microdevices. Y.L. performed all the experiments and analyzed the data under the guidance of E.M. and P.R.

Notes

The authors declare no competing financial interest.

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