

Resistive asymmetry due to spatial confinement in first-order phase transitions

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We report an asymmetry in the R vs T characteristics across the first-order metal-insulator transition (MIT) of V_2O_3 nanowires. The resistance changes in a few, large jumps during cooling through the MIT, while it does it in a smoother way during warming. The asymmetry is greatly enhanced as the width of the nanowire approaches a characteristic domain size. Our results, together with previous reports on VO_2 [W. Fan *et al.*, *Phys. Rev. B* **83**, 235102 (2011)] and $FeRh$ [V. Uhlř *et al.*, *Nat. Commun.* **7**, 13113 (2016)] imply that asymmetry is a generic feature of first-order phase transitions in one-dimensional systems. We show that this behavior is a simple, elegant consequence of the combined effects of the transition hysteresis and the temperature dependence of the insulating gap in this case (and generically, the order parameter relevant for the physical observable). We conclude that our proposed asymmetry mechanism is universally applicable to many electronic first-order phase transitions.

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I. INTRODUCTION

The dynamics of phase transitions in correlated materials is a topic of much current interest. Generally during first-order phase transitions two different phases coexist, giving rise to domain formation [1–7]. Using nanofabrication techniques it is possible to reduce the size of the devices to a scale comparable to that of the domains, confining the system and thus reducing its dimensionality and unveiling effects not observed in bulk or thin films [3,7–14]. One important such effect is the emergence of a resistive asymmetry across the first-order transitions, which had previously been observed in VO_2 [8] and $FeRh$ [9] nanowires. When confined to one dimension (1D), both materials show a R vs T curve which displays large jumps in the cooling branch of the hysteresis, while the heating branch is smoother. The two materials are very different: $FeRh$ is an alloy which undergoes a first-order antiferromagnetic metal to ferromagnetic metal transition at 360 K [9], while VO_2 is an oxide with a first-order nonmagnetic insulator to metal transition at 345 K [8]. Despite of this, the experimental phenomenology is very similar in both cases, which suggests a common underlying origin for this effect. However, these two papers [8,9] invoked very different, material-specific mechanisms to explain this phenomenon.

Here we report a similar asymmetry in yet another system: V_2O_3 nanowires. Unlike VO_2 and $FeRh$, V_2O_3 , has a first-order MIT between a paramagnetic metal and an antiferromagnetic insulator. We show that our findings strongly support the idea that the asymmetry is a universal feature of first-order phase transitions unrelated to detailed mechanisms of the transition.

V_2O_3 is a classic example of a correlated oxide, which has been a subject of intense research [2,15,16]. It features a metal-insulator transition (MIT), along with a magnetic and a structural transition close to 160 K [17]. The first-order nature of this transition can be identified due to its pronounced hysteresis [17] and marked peaks in the specific heat [18]. A recent study demonstrated phase separation during the MIT, with coexistence of metallic and insulating domains in V_2O_3 thin films and characteristic domain sizes in the range of hundreds of nanometers [2]. Our study shows that the asymmetry is greatly increased as the nanowire width approaches the domain size. Using a simple resistor network model, we show that this phenomenon arises from the inherent hysteresis of first-order transitions together with the temperature dependence of the insulating gap. This asymmetry mechanism should also apply to transitions where a gap does not necessarily fully open but there is a significant change in the density of states at the Fermi level. This makes it generically applicable to most electronic first-order transitions in materials spatially confined at the nanoscale.

A. Experiment

We fabricated V_2O_3 nanowires using 100-nm-thick films grown on r -cut sapphire substrates with rf magnetron sputtering from a V_2O_3 target. Details about the growth conditions can be found elsewhere [19]. We used e -beam lithography and negative resist (MA-N2405) to pattern a hard mask on top of the V_2O_3 film. A subsequent reactive ion etching ($Ar + Cl_2$ plasma) was used to produce the nanowires. In order to study the effect of confinement, their width was varied from 1300 to 80 nm, keeping the length constant at 5 μm . Ti/Au electrodes were prepared using optical lithography and e -beam evaporation for the transport measurements (see top inset in Fig. 1). Measurements were carried out on a Lake Shore TTPX probe station, using a Keithley 6221 current source and a

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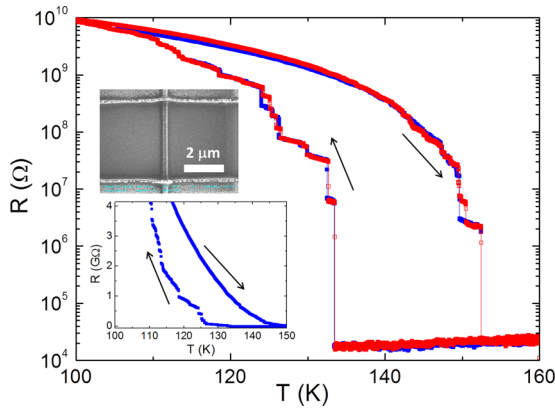


FIG. 1. R vs T of an 80-nm-wide V_2O_3 nanowire, scanning the temperature at a rate of 0.5 K/min (red empty squares) and 2 K/min (blue squares). Lower inset shows the 0.5 K/min scan in linear scale. Top inset: SEM image of a 200-nm-wide V_2O_3 nanowire on the sapphire substrate. The top and bottom gold electrodes are visible.

Keithley 2182A nanovoltmeter. The resistance R as a function of temperature T of the wires shows a MIT transition of over five orders of magnitude (Fig. 1) indicating that the quality of the oxide does not degrade after the fabrication process. A probing current of 10 nA was used to avoid any heating effects.

Figure 1 shows the R vs T of an 80-nm-wide nanowire. A clear feature appears: The resistance changes in discrete, discontinuous jumps. Such jumps had been previously observed in micron-sized devices, and correspond to individual domain nucleation and growth events [11,13,14]. Although jumps are present both in the heating and cooling branches of the hysteresis, there is a clear qualitative difference between the two cases. The jumps are significantly larger in the cooling branch of the hysteresis, causing an asymmetry that is very noticeable if the same curve is plotted in linear scale (lower inset in Fig. 1). The two curves plotted in Fig. 1 correspond to two different temperature sweeping rates: 0.5 and 2.0 K/min. The two curves plotted in Fig. 1 correspond to two different temperature sweep rates: 0.5 and 2.0 K/min. The effect is very reproducible and most jumps can be observed in both curves, independent of the sweep rate.

Figure 2(a) shows a histogram of the number of jumps as a function of their size. The cooling branch shows an evident shift towards larger jump sizes, with several jumps more than an order of magnitude larger than those in the heating branch, making the transition more discontinuous. To quantify this asymmetry we use the ratio $\Delta R_{\text{Jumps}}/\Delta R_{\text{MIT}}$, where ΔR_{MIT} is the total resistance change across the MIT and ΔR_{Jumps} the sum of resistance changes due to large jumps, bigger than 1% ΔR_{MIT} . Figure 2(b) shows $\Delta R_{\text{Jumps}}/\Delta R_{\text{MIT}}$ for the heating and cooling branches as a function of the nanowire width. The asymmetry becomes larger as the width is reduced due to confinement to one dimension. However, it should be noted that this increment is gradual; even for the widest nanowire there is asymmetry in the transition. This suggests that asymmetry is not an intrinsic property of 1D systems, and it might be present in thin films or bulk samples. Asymmetry is only revealed when the size is reduced and the smoothing effect of percolation is eliminated.

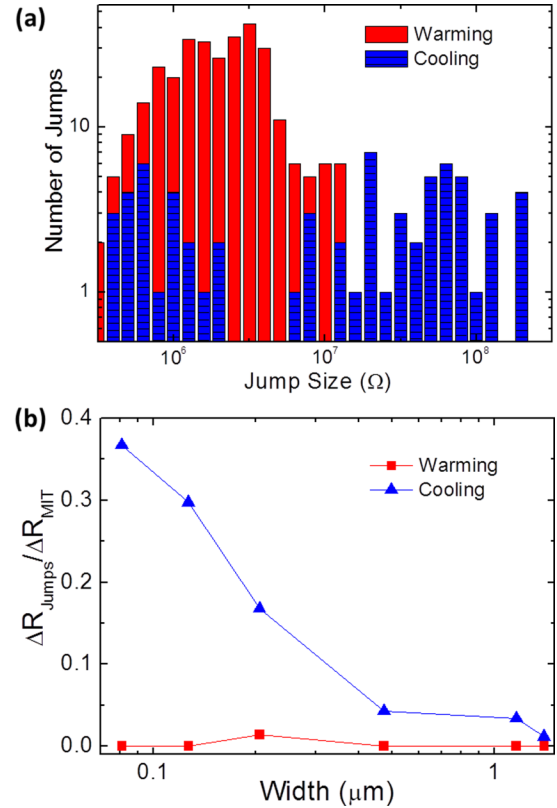


FIG. 2. (a) Jump distribution as a function of the jump size of an 80-nm-wide nanowire for the heating (red bars) and cooling (blue striped bars) branches of the hysteresis. (b) $\Delta R_{\text{Jumps}}/\Delta R_{\text{MIT}}$ as a function of the nanowire width for the heating (red squares) and cooling (blue triangles) branches of the hysteresis. Only large resistance jumps, bigger than $0.01\Delta R_{\text{MIT}}$, were considered for the calculation.

In order to reach this nonpercolative limit, only one domain should fit within the width of the nanowire. An indication that this is indeed the case here for the thinnest nanowire is present in the $R(T)$ curve (Fig. 1). The MIT onset is abrupt, with the resistance increasing more than one order of magnitude in a single jump, as opposed to the gradual increase observed in other micrometer-sized devices [11,13]. This indicates that the sample immediately loses percolation as it enters the coexistence (hysteresis) region upon cooling, in contrast to what is expected in a two-dimensional (2D) system. In this quasi-1D regime, during cooling from the metallic state the first domain that becomes insulating breaks the conduction path. As a consequence this produces a sudden resistance increase onset. It has been shown that domain formation and its characteristic size is determined by strain in the film, which is dependent on the thickness and coupling to the substrate [8,2,20]. Strain is accommodated by alternating metallic and insulating domains, with a characteristic size that increases with thickness [20]. This implies that quasi-1D behavior will be observed in wider nanowires as the film thickness is increased. We must note that our inferred domain size (~ 100 nm) is in very good agreement with previously reported VO_2 [8,20,21] and V_2O_3 [2] films of comparable thickness.

II. DISCUSSION

A resistive asymmetry across a first-order phase transition was previously reported in FeRh nanowires [9] and VO₂ microbeams [8], although their width dependence was not studied. These two previous studies invoked different mechanisms to explain the phenomenon. Fan *et al.* [8] gave an explanation based on the crystal structure while Uhlřř *et al.* [9] proposed a mechanism involving the magnetic nature of FeRh. Fan *et al.* argue that in VO₂ this asymmetry might originate from the presence of structural twin walls in the insulating phase. Such walls may serve as a catalyst for the nucleation of metallic domains, producing a smoother transition when changing from insulator to metal. However, this mechanism cannot apply to the FeRh experiment by Uhlřř *et al.* [9]. In fact, their films have an orthorhombic structure with fourfold symmetry in the plane for both the high- and low-temperature phases, and thus twin walls are not possible. Instead, Uhlřř *et al.* [9] proposed an alternative mechanism induced by the stronger electronic correlations of the high-temperature ferromagnetic (FM) phase compared to the low-temperature antiferromagnet (AF). This should result in residual ferromagnetic domains at low temperatures that serve as seeds for the AF to FM transition, thus making it smoother. However, this mechanism could not work in the VO₂ case of Fan *et al.* [8], since they observe homogeneous, single-crystal phases both at high and low temperatures. Our observation of a qualitatively similar experimental asymmetry in another very different material, V₂O₃, indicates that this behavior is a universal feature of first-order phase transitions in very different quasi-1D systems. Thus, it is important to find a generic mechanism that could explain this phenomenon for a broad number of different systems.

An aspect of phase transitions in correlated materials, generally overlooked, is the evolution of the electronic density of states with the temperature. For instance, in a Mott-Peierls transition a gap opens at the Fermi level; however, its magnitude is not constant but evolves with temperature, especially in the vicinity of the transition temperature T_{MIT} . This has been revealed by transport measurements in high-quality V₂O₃ crystals [22], and by optical measurements in Sr₂IrO₄, VO₂, and V₂O₃ thin films [23–26]. Using focused infrared spectroscopy, McLeod *et al.* [2] were able to probe the optical response of insulating V₂O₃ domains across the MIT, finding a strong temperature dependence of the gap. This variation greatly affects the temperature dependence of electronic properties such as resistivity, which depends exponentially on the gap size, i.e., $\rho \propto e^{\Delta(T)/kT}$. This temperature dependence, together with the hysteresis of the transition, implies a natural asymmetry between heating and cooling, as schematically depicted in the top inset of Fig. 3. The hysteresis region is enclosed between two spinodals, which denote the points where one of the phases is no longer metastable. At the high-temperature spinodal T_2 (insulator \rightarrow metal) the gap will be Δ_2 , while it will be $\Delta_1 > \Delta_2$ in the low-temperature spinodal T_1 (metal \rightarrow insulator). This way, the magnitude of the resistance jumps is different at T_1 and T_2 : i.e., $e^{\Delta_1/kT}$ is bigger than $e^{\Delta_2/kT}$. The magnitude of this asymmetry depends on (i) the variation of Δ between T_1 and T_2 , and (ii) the magnitude of Δ compared to the temperature, i.e., the Δ/kT

ratio. In the particular case of bulk V₂O₃, Δ is on the order of 0.7 eV, much larger than the thermal energy $kT_C \approx 13$ meV at which the transition takes place, yielding a ratio $\Delta/kT_C \approx 50$. This implies that even a 10% change in Δ between T_1 and T_2 might lead to an asymmetry over two orders of magnitude in the resistance jumps.

In order to test whether this mechanism could induce an asymmetry consistent with our observations, we simulate the R vs T for nanowires of different widths. We consider a 2D resistor network model, in which each resistor is assumed to represent one domain of size ~ 80 nm \times 80 nm. These domains can be either in the metallic ($\rho \propto \text{const.}$, i.e., $\Delta = 0$) or in the insulating state ($\rho \propto e^{\Delta(T)/kT}$). In order to emulate the hysteretic nature of a first-order transition, we associate Δ with an order parameter, whose T dependence can be described by a conventional free-energy functional for first-order phase transitions [27]:

$$F(\Delta, T) = \frac{a}{2}(T - T_1)\Delta^2 - \frac{b}{3}\Delta^3 + \frac{c}{4}\Delta^4.$$

This functional has a coexistence region between the spinodals T_1 and $T_2 = T_1 + b^2/4ac$, in which two states are (meta)stable: $\Delta = 0$ and $\Delta = f(T) \neq 0$. Parameters a , b , and c were chosen to provide a hysteresis width similar to the experimental one. This functional naturally produces a temperature-dependent gap as depicted in the top inset of Fig. 3. The state (metallic or insulator) of each site is determined by this free-energy functional, so each domain has an intrinsic hysteresis. In order to mimic the intrinsic disorder of the material, we assign each domain a different T_1 value, following a Gaussian distribution with a width $\sigma = 20$ K. The only effect of disorder is to broaden the transition, and it does not participate in the width of the hysteresis. The simulation is carried out as follows: In the initial state all domains start in the same phase, either insulating (for the heating curve) or metallic (cooling curve). The temperature is then increased (decreased) in steps, and the energy functional is evaluated for each domain. When the insulating (metallic) phase is no longer stable, the domain turns metallic (insulating). That happens at the spinodal points. Our functional represents a first-order phase transition, so the insulator-to-metal and metal-to-insulator spinodals are at different temperatures (T_2 and T_1), yielding the characteristic hysteresis.

Figure 3(a) and its lower inset show the calculated R vs T in logarithmic and linear scale, respectively. The similarity to Fig. 1 is remarkable, reproducing most of the features observed experimentally. Figure 3(b) shows the simulated $\Delta R_{\text{jumps}}/\Delta R_{\text{MIT}}$ for the heating and cooling branches as a function of the nanowire width. Remarkably this also reproduces qualitatively the experimental results.

This shows that the increased asymmetry with decreasing width is due to percolation. As the number of transversal domains increases, the size of the jumps decreases, as does the apparent asymmetry although it does not fully disappear. We must note that, necessarily, an asymmetry will always be present even in macroscopic systems: the hysteresis forces the cooling branch to have a higher slope.

Other physical properties, which are not affected by percolation, such as magnetization, may reveal an asymmetry in systems which are not confined to 1D. That could be the case of

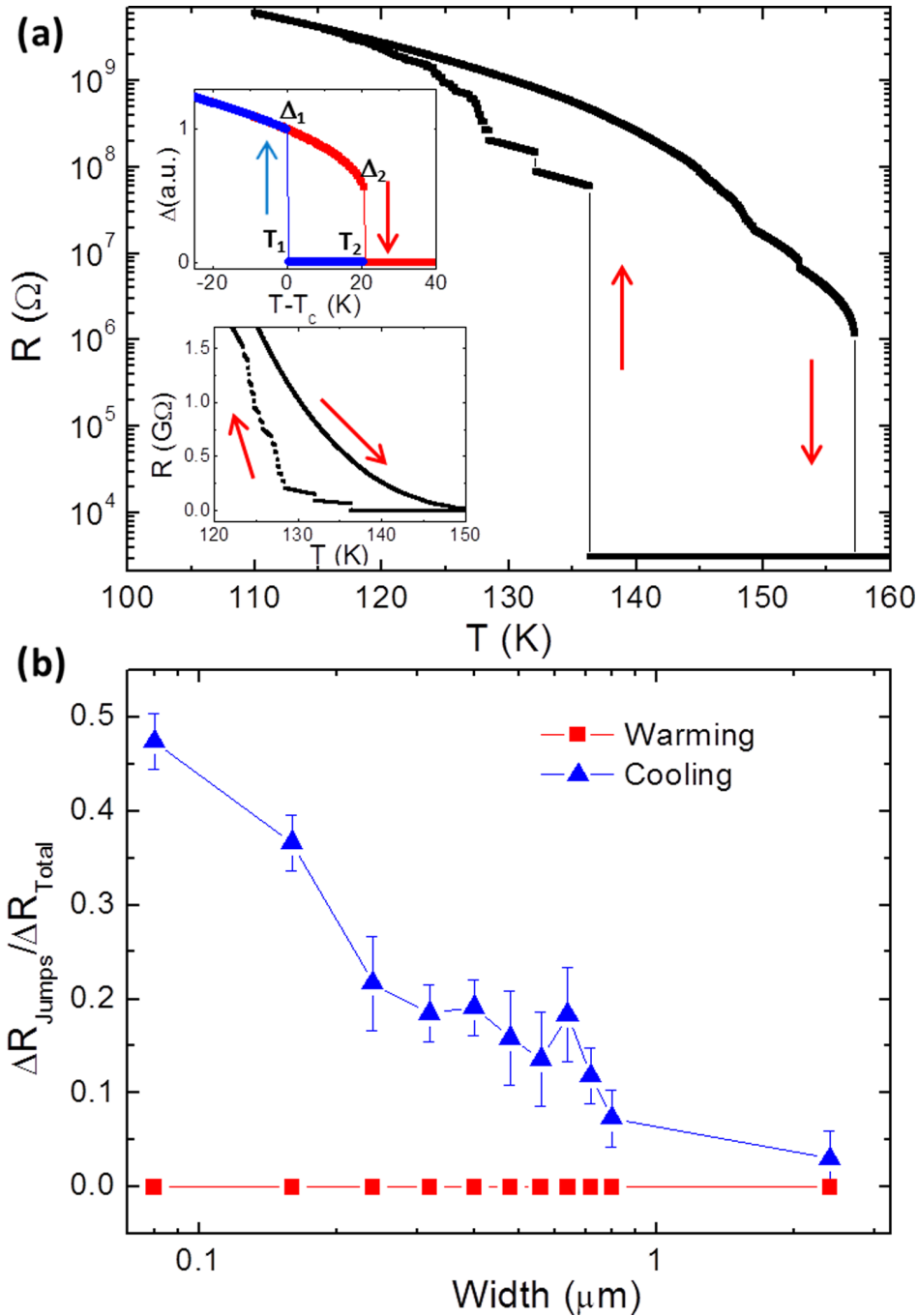


FIG. 3. (a) Simulated R vs T of an 80-nm-wide V_2O_3 nanowire. Lower inset shows the same plot in linear scale. Top inset: temperature dependence of the gap given by the used free-energy functional (see text). T_1 and T_2 are the low- and high-temperature spinodals. (b) Simulated $R_{\text{jumps}}/\Delta R_{\text{MIT}}$ as a function of the nanowire width for the heating (red squares) and cooling (blue triangles) branches of the hysteresis.

Ru- or Si-doped $CeFe_2$, which shows a metamagnetic phase transition from AF to FM, similar to that of $FeRh$ [28]. In fact, experiments conducted in bulk samples have shown a clear asymmetry between cooling and warming in this system [28,29]. Even more surprisingly, the transition is sharper when going from low (AF) to high (FM) temperature, contrary to our results and to those reported by Fan *et al.* [8] and Uhlir *et al.* [9]. However, within our present model this feature has a natural interpretation: Since the order parameter (magnetization in this case) increases with increasing temperature, then the asymmetry is reversed.

We must note that our results do not rule out the possibility that differences in domain nucleation between heating and cooling may contribute to this asymmetry. Different

nucleation mechanisms in the metallic and insulating phases could produce similar effects. However, our recent study [2] in V_2O_3 thin films has shown no difference in the domain pattern formation between heating and cooling. This makes differences in domain formation an unlikely mechanism for the asymmetry we observe here.

III. CONCLUSIONS

In conclusion, we have studied the R vs T of V_2O_3 nanowires of varying widths. We observe a large asymmetry in which the resistance changes in a smoother way in the heating branch of the hysteresis compared to the cooling branch. We show this asymmetry increases as the width of the

wire approaches the domain size. Our observation, together with similar reports in FeRh [9] and VO₂ [8] nanowires, implies that this is a universal feature of first-order phase transitions in spatially confined quasi-1D devices. We propose that the temperature dependence of the gap, together with the intrinsic hysteresis of first-order phase transitions, leads to a universal origin for the asymmetry. The proposed mechanism is further supported by resistor network simulations, which reproduce the experimental results very well. This mechanism is applicable to systems where a full gap opens (like in a MIT), or where there is a big change in the density of states around the Fermi level, like in FeRh [30]. Even more generally, this scenario should apply in transitions where the order parameter

controlling the physical observable has a monotonic behavior as a function of the temperature. These general considerations imply that this mechanism is applicable to a large number of different electronic first-order phase transitions.

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