A hybrid optoelectronic Mott insulator

Cite as: Appl. Phys. Lett. 118, 141901 (2021); https://doi.org/10.1063/5.0044066 Submitted: 13 January 2021 . Accepted: 19 February 2021 . Published Online: 05 April 2021

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ABSTRACT

The coupling of electronic degrees of freedom in materials to create "hybridized functionalities" is a holy grail of modern condensed matter physics that may produce versatile mechanisms of control. Correlated electron systems often exhibit coupled degrees of freedom with a high degree of tunability which sometimes lead to hybridized functionalities based on external stimuli. However, the mechanisms of tunability and the sensitivity to external stimuli are determined by intrinsic material properties which are not always controllable. A Mott metal-insulator transition (MIT) is technologically attractive due to the large changes in resistance, tunable by doping, strain, electric fields, and orbital occupancy but not, in and of itself, controllable with light. Here, an alternate approach is presented to produce optical functionalities using a properly engineered photoconductor/strongly correlated hybrid heterostructure. This approach combines a photoconductor, which does not exhibit an MIT, with a strongly correlated oxide, which is not photoconducting. Due to the intimate proximity between the two materials, the heterostructure exhibits giant volatile and nonvolatile, photoinduced resistivity changes with substantial shifts in the MIT transition temperatures. This approach can be extended to other judicious combinations of strongly correlated materials.

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The physical properties of strongly correlated materials appeal to a broad scientific community because of the versatility and tunability of their electronic responses via internal and/or external perturbations. However, the number of ways of controlling a single correlated material is limited by the available internal degrees of freedom. This presents a difficult challenge when a device concept requires a specific mechanism of control that is not accessible within a material. Here, we present a simple, general solution to this limitation by judiciously designing a heterostructure that hybridizes the functionalities of two seemingly unrelated materials: a strongly correlated Mott insulator with a photoconducting semiconductor. This provides exquisite control of a metal-to-insulator transition (MIT) via optical means. With its striking simplicity, our methodology can be expanded to create hybridized functionalities across, e.g., semiconductors, strongly correlated, magnetic, and topological materials.

The Mott MIT in vanadium oxides—a hallmark of strong correlations—exemplifies behavior that is attractive for its fundamental interest and potential applications in modern technologies.^{2,4–10} This transition is characterized by an abrupt change in the electrical

resistivity by several orders of magnitude and can be controlled by modulating the temperature, doping, pressure, electric field, and to a small extent with laser light. $^{11-24}$ Some materials, such as $\rm VO_2$, also feature large changes in their optical properties across the MIT, making them attractive for optoelectronic applications. $^{25-27}$ Earlier studies 28,29 used $\rm VO_2$ to modify the boundary conditions in heterostructures and this way to implement electrically controlled optical devices. The reverse effect, i.e., controlling the Mott insulating transition by applying light, remains difficult because the effect of light is limited to small changes in the MIT. 30

We have developed a highly sensitive optoelectronic Mott insulator, by heterostructuring vanadium oxide with a well-known semiconducting photoconductor. In this geometry (Fig. 1), light strongly affects the MIT through proximity effects, due to changes in the interfacial Schottky barrier, carrier doping, and/or light-induced interfacial catalytic reactions. We used the archetypical photoconductor, cadmium sulfide (CdS), one of the most studied chalcogenide semiconductor materials which has a high concentration of donor carriers. When exposed to visible light, CdS changes its conductivity

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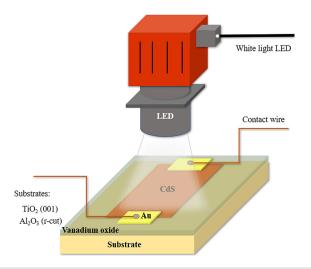


FIG. 1. Schematic device and experimental setup. A CdS (80 \pm 1 nm) film was deposited over vanadium oxide thin films (10 \pm 1 nm both). The CdS/V₂O₃ and CdS/VO₂ heterostructures were illuminated with a white light LED. CdS/V₂O₃ heterostructures were directly grown on Al₂O₃ (R-cut) substrates, and CdS/VO₂ were grown on a TiO₂ (001) substrate for optimal growth. The bilayer conductivity was measured with two probes contacted by two Au pads.

substantially by generating electron and hole carriers ³³ with electrons as the main carriers in the photoexcited state. Moreover, when the CdS interacts with oxygen and light, this material can behave as a photocatalyst. ^{34,35} To maximize the effect of the interface on the transport properties, the thickness of the VO_X was reduced to 10 nm. We show that these hybrid heterostructures exhibit very large, *volatile*, and *nonvolatile* photoinduced modulations of both the amplitude and the critical temperature of the MIT. Moreover, there are interesting similarities and differences between the two VOx-based heterostructures. Such significant enhancement of the photosensitivity to light in Mott insulators provides an advanced material for basic research studies and a promising platform for optoelectronic applications.

We fabricated 10 ± 1 nm thick epitaxial V_2O_3 film on top of r-cut sapphire substrates using rf magnetron sputtering from a V_2O_3

target, in an 8 mTorr high-purity argon (>99.999%) atmosphere. The substrate temperature during deposition is 720 °C, and the sample is cooled at a rate of 80 °C/min after growth. A 10 \pm 1 nm thick epitaxial VO₂ film was grown by reactive sputtering on top of an TiO₂ substrate (oriented along the (001) plane). A 4 mTorr argon/oxygen mix (8% O₂) was used during deposition, and the substrate was kept at 600 °C during the growth and later cooled down at a rate of 12 °C/min. CdS 80 ± 1 nm thick film was grown on top of the VOx with rf magnetron sputtering from a CdS target, in a 2 mTorr pure argon atmosphere at 150 °C. Electrical transport properties of a CdS film grown directly on Al₂O₃ are shown in the supplementary material. In each CdS/VO_x bilayer, two Au (40 nm) electrodes were patterned on top of the CdS/ VO_x heterostructures films. XRD measurements were done in a Rigaku SmartLab system at room temperature. Single-phase growth is confirmed by XRD, epitaxially along the (012) direction for V₂O₃, textured along $\langle 002 \rangle$ for VO₂, and hexagonal phase direction H $\langle 002 \rangle$ for CdS. In all samples, negligible changes in the crystal structure were observed upon exposing them to light (see supplementary material). Transport measurements were carried out on a Montana C2 S50 Cryocooler and TTPX Lakeshore cryogenic probe station, using a Keithley 6221 current source and a Keithley 2182A nanovoltmeter. A white LED (Thorlabs model MCWHLP1) was used for the photoconductivity measurements.

Figure 2 shows the hysteretic resistance of the samples as a function of temperature in the dark, due to the first order MIT of over five orders of magnitude in CdS/V₂O₃ and three orders of magnitude in CdS/VO₂. This temperature-dependent behavior is identical to that of bare VO₂ and V₂O₃, shown in Fig. 3, confirming that growing CdS over the vanadium oxide does not modify their properties. Figure 2 shows the effect of illumination of these samples with varying light power densities. As the light power density is increased, the MIT temperature is reduced and the resistance of the insulating ground state decreases. The effects are most notable in the CdS/V₂O₃ bilayer, where the MIT is almost completely suppressed for a light power density of 731 mW/cm². This results in a resistance drop of more than 6 orders of magnitude compared to the case with no light. The light effects are smaller, though still significant, for VO2 with TMIT decreasing by \sim 15 K when illuminated with a power of 731 mW/cm², meaning a drop of about two orders of magnitude in the resistance at 308 K.

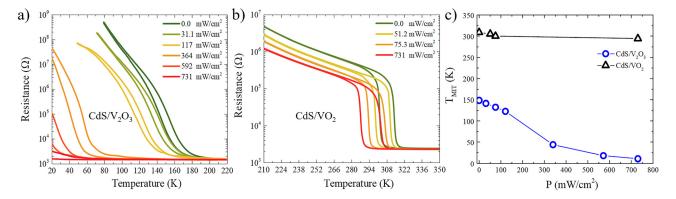


FIG. 2. Light-induced modification of the metal-insulator transition in CdS/vanadium oxide heterostructures. Electrical transport measurements of the resistance as a function of temperature from the bilayers (a) CdS/V₂O₃ and (b) CdS/VO₂. (c) Values of metal-insulating transition temperatures (T_{MIT}) plotted as a function of the power density of the light source.

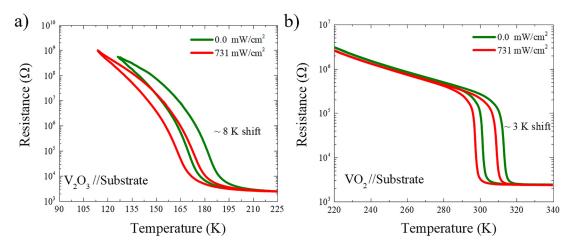


FIG. 3. Light-induced modulation of the metal-insulator transition in bare vanadium oxide films. Electrical transport measurements of the resistance as a function of the temperature in bare vanadium oxide thin film without CdS: (a) V_2O_3 (10 \pm 1 nm) and (b) VO_2 (10 \pm 1 nm). The green curve corresponds to resistance vs temperature without light and the red curve with light. In both cases, a small shift in the onset of the transition is labeled.

In the absence of CdS, the effect of light directly on the vanadium oxide is much smaller, as shown in Fig. 3. This implies that the CdS plays a crucial role in our observations and that sample heating does not play a role. To investigate further the origin of this effect, we repeated the photodoping experiments with thicker VO_X films (100 \pm 2 nm), which exhibit no changes in the $T_{\rm MIT}$ (see the supplementary material). This points to a proximity effect localized at the CdS/VO_X interface. To further corroborate this, we fabricated samples in which a 10–30 nm thick, insulating Al₂O₃ layer was sandwiched between the vanadium oxide and the CdS (see the supplementary material), so that any proximity effect would be suppressed. No MIT modification was observed in this case.

There are additional major differences between the response of the two CdS/vanadium oxide hybrid heterostructures. Figure 4 shows the resistance vs temperature (R vs T) for the two types of heterostructures, measured with and without light, as a function of time. The green curves show the R vs T before illuminating the samples, the red curves during illumination, and the black dashed curves show the state after turning the light off. The V_2O_3 bilayer presents a *volatile* modulation of the MIT [Fig. 4(a)], i.e., the R vs T recovers its original shape immediately after the light is switched off. This contrasts to the *nonvolatile* modulation in the VO_2 . The R vs T does not recover its original shape, and the MIT remains suppressed for hours after the light has been turned off. This nonvolatile change, however, is not permanent. The blue curve shows that after 8 h at room temperature, the original MIT returns to its initial behavior. More information can be found in the supplementary material. Importantly, this demonstrates that the photodoping process does not introduce permanent damage into the sample.

A qualitative explanation of these effects arises by considering the charge carriers in the CdS and VO_X as well as the fact that Mott MITs are highly sensitive to the carrier concentration.³⁶ It is well

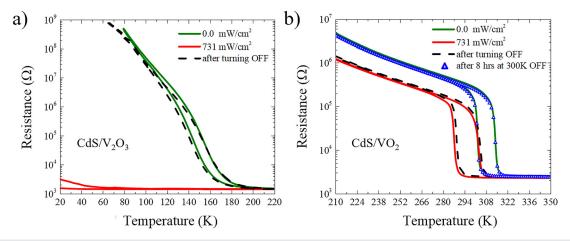


FIG. 4. Time dependence of the light-induced modulation. (a) Volatile changes in the transition temperature of the CdS/V $_2$ O $_3$ heterostructure. (b) Nonvolatile changes observed in the CdS/VO $_2$ sample. In both cases, the dash lines show the resistance vs temperature immediately after turning off the light. In VO $_2$ case, the blue curve(triangles) shows the recovery of the original state after 8 h at room temperature.

documented that conductivity of CdS in the photo-excited state is through electron carriers. Ton the other hand, while in V_2O_3 , the electrical conduction is by holes and that of the VO_2 is by electrons. The critical carrier concentration needed to undergo a Mott MIT can be affected by adding more photoelectrically created electrons from the CdS, which leak into the VO_X . If the work function of the CdS falls between that of V_2O_3 and VO_2 , the observed differences in the effect would have a natural explanation. This would explain qualitatively three important facts discovered in our experiments: (1) the effect is an interface effect, (2) the light completely disrupts the MIT in V_2O_3 and has a minor effect on the VO_2 , and (3) the effect is volatile in V_2O_3 and nonvolatile in VO_2 .

Alternatively, the differences observed may suggest that the underlying mechanism for the MIT modulation in the various CdS/VOx samples is different. The nonvolatile modulation observed in VO2 could be explained by light-induced chemical reactions. On the other hand, CdS is also one of the most prominent photocatalysts.^{34,40} Irradiation with photons of energy larger than the bandgap may create electron-hole pairs, which would produce reduction/oxidation reactions at the CdS/VOX interface. This would reduce the VO2 into a different Magnéli phase. 41 After the light is switched off, those chemical changes would persist and only slowly reverse over time. The volatile V₂O₃ case is different and would be compatible with a modulation of the MIT caused by photodoping. Electrons generated by light in the CdS would transfer across the interface into the ultrathin (hole conducting) V₂O₃ layer, doping it and destabilizing the Mott insulating phase. Future work will be performed to identify which of these speculative scenarios lead to the effect we observe.

In conclusion, we have demonstrated large, light-induced modulation of the MIT in CdS/VO_X heterostructures. For CdS/ V₂O₃, the T_{MIT} modulation is as large as 140 K, and the giant resistance reduction is as high as six orders of magnitude. For CdS/ VO₂, the modulation is much smaller, although the resistance can be reduced by two orders of magnitude in the middle of the transition. A single study showed that light irradiation can be used to modify the insulating resistivity of VO2, 30 with minimal effect on the transition temperature. The modification we observe is volatile in V₂O₃ bilayers and nonvolatile in VO₂ bilayers. We show that these effects are caused by the intimate contact at the CdS/vanadium oxide interface. A simple qualitative explanation suggests that these effects are caused by the differences in the electronic carriers in CdS (electrons), V₂O₃ (holes), and VO₂ (electrons) and their work functions. An alternative possible explanation for these effects involves photodoping in V₂O₃, ⁴² and CdS-mediated photocatalysis in VO₂. 43,44 Further experimental work is under way to elucidate the precise mechanism. The volatile and nonvolatile behavior in two closely related material systems opens up the possibility for the use of these systems for different neuromorphic applications such as synaptors and neuristors.

In a broader sense, our results show a very promising approach toward the development of hybrid functionalities in materials using the possible transfer of electronic responses in a properly engineered heterostructure. This may have further applications as functional materials useful in other optoelectronic applications or systems where a different functionality can affect each other when incorporated into hybrid heterostructures.

See the supplementary material related to the structural characterization of the analyzed heterostructures.

We thank R. C. Dynes, A. Hoffmann, J. A. Schuller, and Y. Takamura for useful conversations. We thank Francisco Schuller for supplying the Au for the electrodes. This collaborative work was supported as part of the "Quantum Materials for Energy Efficient Neuromorphic Computing" (Q-MEEN-C), an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Basic Energy Sciences under the Award No. DE-SC0019273. A.R.-C. thanks the economic support of the mobility research program Salvador de Madariaga from Spanish Ministry of Science.

I.K.S. and H.N. conceived the idea. H.N. and J.d.V. designed the experiment. H.N. and Y.K. fabricated the samples. H.N. performed the transport measurements with assistance from J.d.V., N.M.V., A.R.C., P.L., and E.Q. The x-rays diffraction measurements were performed by H.N., C.A., M.H.L., and I.Z.; I.Z., O.S., and A.F. carried out the x-ray diffraction analysis. H.N., A.F., and I.K.S. wrote the manuscript. All authors participated in the discussion of the results and corrected multiple iterations of the manuscript.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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