Thresholdless behavior and linearity of the optically induced metallization of NbO₂

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The optical excitation of NbO₂ is known to induce a transient metallicity that partially recovers on a subpicosecond speed and only fully recovers after several nanoseconds. Here, through low-fluence pump-probe spectroscopy, the short-lived and long-lived components of NbO₂'s optically induced modulation are found to be linearly dependent on pump fluence in the low-fluence regime. Through an in-depth analysis, the short-lived and long-lived modulations are found to be caused by photocarrier-induced refractive index changes and photothermally induced changes in the lattice temperature of NbO₂, respectively, and not an insulator-metal transition.

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I. INTRODUCTION

In recent years, interest in all-optical computing devices, in particular, all-optical modulators that modulate the intensity of an optical signal by means of another optical signal, has grown [1]. Using nonlinear effects in silicon waveguides [2], photonic crystals [3,4], and graphene [5], all-optical modulators have been demonstrated; however, their reliance on weak nonlinear phenomena necessitates large device sizes that make them unattractive for most applications.

Highly correlated oxide materials, such as VO₂ and NbO₂, have been proposed as potential materials for nanoscale all-optical modulators. These materials undergo an insulator-metal transition (IMT) accompanied by a crystal phase transition (CPT) when their temperature is raised above an IMT temperature [6,7]. Upon undergoing an IMT, the optical properties of such materials also change drastically, and this has enabled many groups to demonstrate high-performance electro-optic [8,9] and thermo-optic [10] modulators with nanoscale sizes.

In recent years, it has also been shown that the IMT of VO₂ can be induced optically through excitation by a femtosecond laser pulse. Whether or not this optically induced IMT is the result of a Mott-Hubbard transition or Peierls transition has been and continues to be the focus of a large number of research articles [11–17]. Yet despite this large amount of work, the exact nature of the IMT is still not fully understood [18].

A thorough understanding of the mechanism of VO₂'s IMT is complicated by the low temperature (340 K) of the IMT [19]. To isolate the optically and thermally induced phenomena, interest has grown in another closely related material, NbO₂, whose IMT is at a much higher temperature (1081 K). In the first study of the optically induced IMT of NbO₂, NbO₂

was pumped by a femtosecond laser pulse at 800 nm and the transient change in the reflectance at 800 nm was measured [19]. The NbO₂ exhibited a transient decrease in reflectance with a recovery described by three time constants: an ultrafast fs response attributed to electron-electron scattering of photoexcited carriers, a short-lived ps response attributed to the recovery of grains of NbO₂ that undergo a purely electronic IMT, and a long-lived ns response attributed to the recovery of grains of NbO₂ that undergo both an electronic IMT and a structural CPT.

A later study of the optically induced IMT of NbO₂ studied the transient change in the terahertz conductance of NbO₂ after excitation by an 800-nm pump laser pulse [20]. The terahertz conductivity of NbO₂ was shown to exhibit a transient increase governed by the same three timescales. Nonlinearities in the long-lived response at 10 and 17.5 mJ/cm² were attributed to the IMT being induced by photocarrier excitation and photothermal excitation, respectively.

There are still several aspects of the NbO₂ pump-probe response that are not well understood. First, it is unclear whether or not the short-lived ps recovery and long-lived ns recovery are the result of an IMT, or if they are the result of photoexcited carriers in the NbO₂. In addition, it is not yet clear that NbO₂ can exhibit a short-lived modulation without the additional long-lived modulation, which is critical for NbO₂ to be applied to ultrahigh-speed all-optical modulators.

In this paper, we study the transient modulation of the transmittance and reflectance of an NbO₂ thin film at 1550 nm (C band) after excitation by a 766-nm femtosecond laser pulse. We examine the short-lived and long-lived responses in the low-fluence regime and demonstrate that both responses scale linearly with fluence with no evidence of an IMT threshold. Through an analysis of the photothermally induced temperature change, the long-lived modulation is found to be a result of the photothermal excitation of NbO₂. Additionally, an in-depth theoretical analysis that accounts for the Burstein-Moss effect, band shrinkage effect, and free carrier absorption effect shows that the short-lived modulation results from photoexcited carrier-induced changes in the optical properties of NbO₂.

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FIG. 1. Relative change in (a) transmittance and (b) reflectance of the NbO₂ to the 1550-nm probe pulse plotted against the pump-probe delay. Insets show the components of the pump-probe response.

II. RESULTS

To study the femtosecond response of NbO₂, a 60-nm NbO₂ thin film was fabricated on a 0.5-mm-thick Al₂O₃ *c*-plane (0001) substrate using pulsed laser deposition (PLD) and the transient modulation after photoexcitation was measured. Full details of the fabrication, characterization, and measurement of the NbO₂ thin film are given in the Supplemental Material [21].

The modulation in the transmittance of the NbO₂ film at 1550 nm upon excitation by the pump pulse is shown in Fig. 1(a). The response is similar to that seen when both the pump and probe pulses have a wavelength of 800 nm [19]. An initial strong decrease in the transmittance that recovers after several ps is followed by a long-lived decrease in the transmittance that recovers in several hundred ps to several ns. To better understand the dynamics of the modulation, the response was fit to a three-exponential decay curve [19],

$$\frac{\Delta T}{T} = \left[1 + \operatorname{erf}\left(\frac{t - t_0}{\tau_p}\right) \right] \left(A_1 e^{\frac{t-t_0}{\tau_1}} + A_2 e^{\frac{t-t_0}{\tau_2}} + A_3 e^{\frac{t-t_0}{\tau_3}} \right), \quad (1)$$

where t_0 is the delay between the pump and probe pulses, τ_p is related to the laser pulse width and the response time of the material, and τ_1 , τ_2 , and τ_3 are the time constants of the three decay rates. The components of the decay for the 1.29 mJ/cm² curve are shown in the inset of Fig. 1(a). There is an ultrafast decay with a τ_1 of 324 fs, a short-lived decay with a τ_2 of 3.24 ps, and a long-lived decay with a τ_3 of 543 ps. As the pump fluence increases, the exact values of the decay time constants vary, but the three timescales were consistently observed.

The reflectance was subsequently modeled with Eq. (1). The ultrafast, short-lived, and long-lived modulations are all observed; however, whereas the short-lived and long-lived modulations correspond to decreases in the transmittance at

FIG. 2. Modulation in the 1550-nm transmittance of the NbO₂ at the minimum of transmittance (blue) and at a 100-ps delay (red). Center left inset: Low-fluence region. Top inset: Ratio of the modulation at 100 ps and at the transmittance minimum.

1550 nm and the reflectance at 800 nm [19], they represent increases in the reflectance at 1550 nm.

Subsequently, the modulation in the transmittance for pump fluences between 4.6 μJ/cm² and 1.4 mJ/cm² was measured. The maximum transmittance modulation [$T_{\min}$ in Fig. 1(a)] and the modulation at 100 ps after the maximum modulation ($T_{100\text{ps}}$) are shown in Fig. 2. $T_{\min}$ includes the combined effect of all three modulations (ultrafast, short lived, and long lived), whereas $T_{100\text{ps}}$ includes only the long-lived modulation. For the entire range of fluences studied, $T_{\min}$ and $T_{100\text{ps}}$ scale linearly with pump fluence, with no evidence of any threshold. Furthermore, the ratio of $T_{\min}$ to $T_{100\text{ps}}$, as shown in the upper inset of Fig. 2, was relatively constant (only fluctuating at very low fluences where the signal-to-noise ratio was poor).

III. DISCUSSION

These results contradict the idea that the long-lived modulation is due to an IMT. Based on the Mott-Hubbard description, an IMT occurs when the concentration of excited carriers in the conduction band reaches a threshold level at which they sufficiently screen the Coulomb interaction and cause the band gap of the insulator to collapse [18]. This results in a drastic increase in the metallic reflection of the material, and therefore, a nonlinearity in the fluence versus modulation curve should be expected. For this low-fluence regime, however, no such threshold exists for fluences as low as 4.6 μJ/cm².

It can also be shown that a nonlinearity and IMT onset cannot exist below 4.6 μJ/cm². By assuming that all of the absorbed pump light contributes to the excitation of photoexcited carriers, the photoexcited carrier concentration N can be calculated as

$$N = \frac{(1 - R - T)\Phi_{\text{pump}}}{t_{\text{film}}E_{\text{ph}}}. \quad (2)$$

FIG. 3. Thermally induced modulation in NbO₂'s transmittance relative to room-temperature transmittance (black) compared to the long-lived modulation at different pump fluences (red).

Here, Φ_{pump} is the pump fluence, t_{film} is the thickness of the NbO₂ film (60 nm), E_{ph} is the photon energy of the pump (1.62 eV for a 766-nm pump), and R and T are the reflectance (27.6%) and transmittance (32.3%) of the sample at the pump wavelength. Using this equation, a pump fluence of $4.6 \mu\text{J}/\text{cm}^2$ would correspond to a photoexcited carrier density of only 1.18×10^{18} carriers/ cm^3 . This is several orders of magnitude lower than the intrinsic carrier concentration of NbO₂ just before the thermally induced IMT [22], and since a thermally induced IMT does not occur when the thermally excited carrier concentration reaches this concentration, the concentration must be insufficient to trigger an IMT.

The simplest explanation for the origin of the long-lived modulation is that it is the result of photothermally induced changes in the lattice temperature of NbO₂ and the resulting thermo-optic changes in the optical properties of NbO₂. When NbO₂ absorbs the pump light, excited electrons and holes are generated, and as they relax and recombine, absorbed energy is released. Because NbO₂ has an indirect band gap, this energy is primarily released in the form of phonons that raise the lattice temperature. The temperature change ΔT is given by

$$\Delta T = \frac{(1 - R - T)\Phi_{\text{pump}}M_u}{t_{\text{film}}\rho_{\text{NbO}_2}C_p}, \quad (3)$$

where M_u is the molar mass of NbO₂ (124.9 g/mol), ρ_{NbO_2} is its density (6.10 g/ cm^3), C_p is its heat capacity (57.35 J/mol K [23]), and Φ_{pump} , t_{film} , R , and T are the same as above. The lattice temperature change induced for the fluence range measured in Fig. 2 was first calculated according to the equation above. $T_{100\text{ps}}$ was then plotted against the induced temperature change and compared to the thermally induced modulation in Fig. 3. The thermally induced modulation was calculated by measuring the transmittance of the NbO₂ film at $1.55 \mu\text{m}$ over a temperature range of 20–60 °C and then calculating the modulation in the transmittance relative to the transmittance at 20 °C. Figure 3 shows a strong agreement between the measured thermally induced modulation and the long-lived photoinduced modulation, supporting the argument that the long-lived modulation is a result of the photothermally induced change in NbO₂'s lattice temperature. The recovery of the long-lived modulation, then, is simply due to the dissipation of the excess heat into the substrate.

The ps recovery time of the short-lived modulation suggests an origin related to photoexcited carriers in the NbO₂. When electrons and holes are excited in a semiconductor material, three main phenomena occur that work to alter the electronic band structure and optical properties of the material. They are the Burstein-Moss effect, the band-gap shrinkage effect, and the free carrier absorption effect [24].

In the Burstein-Moss effect, also known as the band filling effect, the excitation of electrons from the valence band to the conduction band of a semiconductor results in a reduction of the absorption coefficient of the semiconductor near the band-gap energy [24]. This is because excited electrons occupy states in the conduction band and leave behind holes in the valence band, preventing other electrons from absorbing photons and being excited. The change in the absorption coefficient $\Delta\alpha$ can be modeled as

$$\Delta\alpha(E) = \alpha_0(E)[f_v(E_a) - f_c(E_b) - 1], \quad (4)$$

where α_0 is the original absorption coefficient and $f_v(E_a)$ and $f_c(E_b)$ are the electron occupation probabilities at an energy E_a below the valence band maximum and an energy E_b above the conduction band minimum, respectively. The calculations of f_v and f_c are elaborated on in the Supplemental Material [21]. α_0 was determined from ellipsometric measurements of NbO₂'s refractive index. From the absorption coefficient, the change in the imaginary part of the refractive index $\Delta n''$ can be calculated using the relation $\Delta n'' = \frac{\Delta\alpha\lambda}{4\pi}$, and the real part of the refractive index can be determined from the Kramers-Kronig relation,

$$\Delta n' = \frac{c_0\hbar}{\pi q_e} \mathcal{P} \int_0^\infty \frac{\Delta\alpha(E')}{E'^2 - E^2} dE', \quad (5)$$

where c_0 is the speed of light in vacuum, $\hbar$ is the reduced Planck's constant, E is the photon energy, and q_e is the fundamental electron charge [24]. $\mathcal{P}$ denotes the Cauchy principal value of the integral.

The band-gap shrinkage effect describes the phenomena where high concentrations of electrons and holes in a semiconductor material cause a decrease in the band-gap energy due to electron-electron and hole-hole interactions [24]. The amount of band-gap shrinkage ΔE_g caused by a carrier concentration N can be approximated by an analytical equation proposed by Wolff [24–26],

$$\Delta E_g = -\frac{e}{2\pi\epsilon_0\epsilon_s} \left(\frac{3}{\pi}\right)^{\frac{1}{3}} N^{\frac{1}{3}}, \quad (6)$$

where ϵ_0 is the vacuum permittivity and ϵ_s (23.3 [22]) is the static relative permittivity of the semiconductor material. To account for the band-gap shrinkage caused by the initial carrier concentration before photoexcitation N_0 , the equation above is modified and the equation describing the additional band-gap shrinkage only becomes

$$\Delta E_g = -\frac{e}{2\pi\epsilon_0\epsilon_s} \left(\frac{3}{\pi}\right)^{\frac{1}{3}} [(N + N_0)^{\frac{1}{3}} - N_0^{\frac{1}{3}}]. \quad (7)$$

An approximation of the change in the absorption coefficient can be determined by simply shifting the measured absorption profile by an energy equal to the change in the

FIG. 4. (a) Change in the real (blue) and imaginary (red) refractive indices of NbO₂ induced by photoexcited carriers for a 1 mJ/cm² pump pulse, caused by the Burstein-Moss effect (dashed), the band-gap shrinkage effect (dotted), and the free carrier effect (dotted dashed). (b) Modulation in the transmittance (blue) and reflectance (red) resulting from the refractive index change shown in (b). Inset: Change in 1.55-μm transmittance and reflectance modulation with fluence.

band-gap energy [27], giving

$$\Delta\alpha(E) = \alpha_0(E - \Delta E_g) - \alpha_0(E). \quad (8)$$

Using the same method used for the Burstein-Moss effect, the $\Delta n'$ and $\Delta n''$ for the band-gap shrinkage effect were calculated.

The last effect of interest is the free carrier absorption effect. Excited carriers in a semiconductor act as free carriers with a corresponding Drude response that alters the semiconductor's optical properties [23]. The change in the relative permittivity due to a concentration N of photoexcited carriers is described by the Drude-Lorentz model [28],

$$\Delta\epsilon_r = -\frac{Nq_e^2}{\epsilon_0 m^* \omega^2 + i\omega\gamma}, \quad (9)$$

where m^* is the electron effective mass, γ is the damping frequency, and ω is the frequency of light in rad/s. Using a second-order Taylor expansion, the change in the complex refractive index Δn can be calculated as

$$\Delta n = \frac{\Delta\epsilon_r}{2n_0} - \frac{\Delta\epsilon_r^2}{8n_0^3}, \quad (10)$$

where n_0 is the original refractive index.

The resulting $\Delta n'$ and $\Delta n''$ for the three effects discussed above are shown in Fig. 4(a). Over the majority of the visible and near-infrared (NIR) wavelength range, the band-gap shrinkage effect dominates the photoexcited carrier-induced refractive index change. At the probe wavelength of 1.55 μm, an overall increase in both the real and imaginary parts of the refractive index is observed. Figure 4(b) shows the expected differential transmittance based on the refractive index

changes for a 1 mJ/cm² pump pulse over the visible and NIR wavelength range. Details of the calculation are given in the Supplemental Material [21]. The expected differential transmittance at 1.55 μm shown in Fig. 4(b) (−2.54%) and the measured $T_{\min}$ at 1 mJ/cm² in Fig. 2 (−2.99%) are very similar. Another important observation is that the differential reflectance becomes negative over the range of approximately 650 nm to 1 μm, which matches the decrease in the reflectance at 800 nm observed by Beebe *et al.* [19]. Despite the agreement in sign between the predicted differential reflectance at 1.55 μm and experimental measurements shown in Fig. 1, the predicted differential reflectance is clearly overestimated. This is likely due to the assumption that the band-gap shrinkage causes a uniform shift in the absorptivity curve at all wavelengths, whereas the effect of the band-gap shrinkage effect on the band structure is weaker further from the band-gap edge [29]. The agreement between the value at $T_{\min}$ and the predicted modulation based on photoexcited carrier-induced refractive index changes provides strong evidence that the latter is the origin of the short-lived modulation. It also explains why a ps time constant was observed for the recovery of the short-lived modulation, as electrons and holes are expected to recombine on a ps timescale.

To further support the above conclusion, the scaling of the predicted modulation with pump fluence was also examined [Fig. 4(b) inset]. As expected, at 1.55 μm a differential reflectance increasing with pump fluence and a differential transmittance decreasing with pump fluence is observed. The magnitude of the differential transmittance for different pump fluences is also very similar to that observed in Fig. 2; however, unlike the measured differential transmittance, the predicted differential transmittance scales sublinearly with pump fluence. Since the initial carrier concentration N_0 was relatively high, though, the deviation from linearity in the range of 0 – 1 mJ/cm² is only very slight. Furthermore, a more accurate calculation of ΔE_g would include higher-order terms [30], and this would improve the linearity.

The last feature of interest is the ultrafast modulation. Here, the ultrafast modulation at 1.55 μm was found to correspond to a decrease in both the transmittance and reflectance of NbO₂. Only an increase in the imaginary part of the refractive index with no or a minimal change in the real part of the refractive index will result in this behavior. In both prior studies of NbO₂'s photoinduced modulation, the relaxation of the ultrafast modulation was attributed to the relaxation of the photoexcited carriers from their initial nonthermal distribution to a quasithermal distribution through electron-electron and electron-phonon scattering [19,20]. In materials that show this kind of effect, the strengths of the band filling and free carrier absorption effects are modified due to the change in the effective masses of the photoexcited carriers as they relax, and this results in a change in the modulation [31]. However, from the modeling shown in Fig. 4, the band filling effect should cause a decrease in absorption, and the free carrier absorption effect a large change in the real part of NbO₂'s refractive index, and therefore neither of these effects can explain the ultrafast modulation observed. Another possibility is that the ultrafast modulation represents the increase in absorption caused by two-photon absorption of the pump and

probe pulse. This would also explain why the recovery time constant of the ultrafast modulation was limited by the laser pulse width in both this study and the one performed by Beebe *et al.* [19].

IV. CONCLUSION

The results here show that the long-lived and short-lived modulation of NbO₂ after excitation by a femtosecond laser seen at low pump fluences are not the result of a photoinduced IMT. Instead, the long-lived modulation was found to be caused by photothermally induced changes in the lattice temperature of NbO₂, and the short-lived modulation was determined to be caused by photoexcited carrier-induced changes in the refractive index of NbO₂. The ultrafast modulation is also likely to be related to two-photon absorption.

Despite the short-lived modulation not representing a purely electronic IMT, the large modulation observed is very promising for the development of nanoscale all-optical

modulators. Given the photothermal nature of the long-lived modulation, it can be significantly reduced by using a longer-wavelength pump pulse to reduce the excess above band-gap energy as well as using ultrathin and nanostructured NbO₂ to increase the rate of heat dissipation. This has the potential to enable NbO₂ based all-optical devices with speeds as high as 100s of GHz.

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