Active terahertz metamaterials based on the phase transition of VO₂ thin films

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Abstract

Vanadium dioxide (VO₂) thin films were prepared on single crystal sapphire substrates by pulsed laser deposition. VO₂ films exhibited a significant resistivity drop ($>10^4 \Omega\text{-cm}$) and large optical transmittance change ($> 60 \%$) in the near-infrared region across their semiconductor-to-metal transition. Hybrid metamaterial devices designed for the THz frequency regime were fabricated by combining double split-ring resonators (SRRs) with phase changing VO₂ films. By changing the conductivity of VO₂ via temperature, the behavior of the SRR gap was adjusted from capacitive to resistive in order to modulate the THz beam transmission at their resonance frequencies. A modulation efficiency greater than 50 % was achieved at the magnetic resonance frequencies (0.3 THz and 0.7 THz) in these hybrid SRR-VO₂ metamaterial devices.

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1. Introduction

Metamaterials, artificially designed/engineered electromagnetic composite structures, have attracted great interest from the scientific community due to their potential applications in super-lenses [1], optical cloaking [2,3], and structures with a negative index of refraction [4]. Typical metamaterial structures consist of arrays of metal patterns in a dielectric medium with dimensions much smaller than the operating wavelengths for a given portion of the electromagnetic spectrum, including microwave, THz, and optical frequencies. Metamaterials operating in the THz regime have attracted great interest due to their potential applications in security monitoring (THz imaging), non-destructive testing, wireless communications, and materials characterization [5-8]. A common design of THz metamaterials is based on a periodic array of subwavelength scale split-ring resonators (SRRs) due to their ability to achieve magnetic resonances through their inductor-capacitor (LC) resonance [9]. The key parameters to determine the resonant frequency are the choice of SRR materials and the dimensions of the SRRs. It is of considerable interest to develop tunable metamaterials by adjusting the capacitance in the resonator gap to modulate the resonant frequency, amplitude, or phase of the resonance for achieving active metamaterial devices, such as filters, modulators and sensors [8,10]. One promising approach is to fabricate a hybrid structure comprising of a metamaterial structure incorporating functional materials like VO₂.

VO₂ has been realized as an excellent tunable functional material because of its significant change in optical and electrical properties resulting from a structural phase change from a semiconducting monoclinic phase (low-temperature) to a metallic tetragonal phase (high-temperature) at ~68 °C [11]. This semiconductor-to-metal transition (SMT) takes place when a thermal, electrical, magnetic or optical stimulus is applied and it can occur at ultrafast timescales (~100 femtosecond) [12-14]. Recently, considerable efforts have focused on fabricating

frequency tunable active metamaterials exhibiting narrow-band operation for a given portion of the electromagnetic spectrum, including microwave, THz, and optical frequencies, by tuning the electrical and magnetic modes of SRRs using the phase changing properties of VO₂ [15-17]. In this paper, we demonstrate hybrid metamaterial devices designed for multi-band THz frequencies by fabricating double SRR arrays on VO₂ films by a conventional lift off process. We then investigate the resonance behavior using terahertz time-domain spectroscopy (THz-TDS) and compare the measured resonances with that predicted by full wave electromagnetic simulations. The results of this work show that a modulation efficiency of over 50 % can be achieved at the resonant frequency of these SRR-VO₂ hybrid metamaterial devices.

2. Experimental

2.1. VO₂ film growth

VO₂ thin films were deposited on c-axis, single crystal sapphire substrates by pulsed laser deposition (PLD). Details of the deposition conditions of VO₂ thin film have been described elsewhere [18]. Briefly, a KrF excimer laser (Lambda Physik LPX 305, $\lambda = 248$ nm, pulse duration = 30 ns) was operated at a pulse rated of 10 Hz and was focused onto a rotating VO₂ target (ACI Alloys) with a fluence of 1.5 J/cm². Films were grown at 550 °C and 2.5 Pa of oxygen gas. The samples were cooled down to room temperature at the same oxygen pressure. The resulting film thickness was 100 nm. The electrical switching properties of the VO₂ films were characterized at temperatures between 25 and 100 °C by a four-probe resistivity method. The optical transmittance spectra were measured in air using a UV-visible-near IR spectrophotometer [JASCO, V-670]. X-ray diffraction (XRD) [Rigaku rotating anode X-ray generator with Cu K α radiation] was used to characterize the crystal structure of the films.

2.2. Hybrid metamaterial devices fabrication

Two types of hybrid metamaterial devices were fabricated for this work: one was built on planar VO₂ films (referred to “SRR-planar VO₂ device”) and the other was built on patterned VO₂ films (referred to “SRR-patterned VO₂ device”). Periodic arrays of double SRR metamaterials were fabricated on both planar and patterned 100 nm thick VO₂ films previously deposited on sapphire substrates by a conventional lift-off process. First, the negative photoresist (NR7-3000PY) was deposited over the VO₂ films by spin coating and then exposed by a UV light through a mask to generate the SRR structures. After developing the SRR patterns, a 200 nm Au film with a 20 nm Ti adhesion layer were deposited by e-beam evaporation and followed by lift-off in acetone. Figure 1(a) shows a schematic illustration of the unit cell with dual-resonance SRR metamaterial patterns, which consists of two square split-rings located oppositely with dimensions of $l = 50 \mu\text{m}$, $w = 6 \mu\text{m}$, $t = 3 \mu\text{m}$, $g = 4 \mu\text{m}$ and the lattice period of $P = 100 \mu\text{m}$. Figure 1(c) shows an optical micrograph belonging to some of the SRR arrays on VO₂. For the SRR-patterned VO₂ devices, small VO₂ pads ($6 \mu\text{m} \times 8 \mu\text{m}$) were deposited first on sapphire substrates by lift-off and then the 200 nm thick Au SRR arrays were deposited on top of VO₂ pads (see Figs. 1d, 1e, and 1f).

2.3. Device characterization and simulation

The devices were characterized using a terahertz time domain spectrometer (THz-TDS). The spectrometer consisted of a femtosecond laser (Pharos, Light Conversion Ltd.), photoconductive antennas (Ekspla), delay line, and associated data acquisition. The oscillator in the femtosecond laser was used to excite the photoconductive antennas and had an operating wavelength centered at 1030 nm with a pulse width of ~80 fs and a repetition rate of 75 MHz. The samples were mounted on a two inch diameter heated stage with a circular aperture (8 mm

diameter). The heated stage was first centered between the THz emitter and detector and aligned to ensure that the stage did not introduce experimental artifacts such as diffraction or resonance from the aperture. A schematic diagram of the THz-TDS system is shown in figure 2. The frequency dependent transmission was determined by the ratio of the Fourier-transformed amplitude spectra of the sample to that of the heated stage with no sample mounted. Thus, the measured transmission spectra included the Fabry-perot resonance effect occurring from the Al_2O_3 substrate (0.5mm thick). Full-wave electromagnetic simulations were carried out using HFSS from ANSOFT. The Au SRRs and VO_2 thin films were modeled using the finite conductivity boundary condition, while the periodicity of the structure was simulated using periodic boundary conditions and Floquet ports.

3. Results and discussion

VO_2 films used in this research were synthesized by PLD and were first optimized to achieve stoichiometric VO_2 films with the largest change in electrical conductivity and optical transmission in the near infrared (IR) region ($\sim 2.5 \mu\text{m}$). XRD results verified that the deposited films only exhibited the VO_2 monoclinic phase with no signs of impurity phases as demonstrated by the absence of other peaks [18]. At room temperature the electrical resistivity of VO_2 film was semiconducting with a large resistivity of $30 \Omega\text{-cm}$. When the temperature increased above the phase transition temperature to $\sim 90^\circ\text{C}$, the resistivity dropped by more than 4 orders of magnitude ($\sim 6 \times 10^{-4} \Omega\text{-cm}$), as seen in Fig. 3(a). At room temperature, the optical transmittance in the near IR region was the highest ($\sim 66\%$) and upon heating the sample to 90°C the transmittance reached a minimum ($\sim 7\%$), indicating good optical switching properties in the near IR [Fig. 3(b)].

Figure 4 shows the frequency-dependent transmission spectra of a hybrid SRR-planar VO₂ device with two different phases (semiconducting and metallic) of VO₂ film. THz-TDS measurements were performed by placing the sample plane normal to the electromagnetic radiation with the incident E-field polarized either perpendicular or parallel to the SRR gaps in order to excite different modes in the resonators. For the E-field perpendicular to the SRR gap, two transmission dips were observed at 0.33 THz and 0.69 THz, which correspond to the magnetic resonances of the outer and inner rings, respectively. These magnetic resonances are obtained due to resonant currents circulating around the SRR rings leading to charge accumulation at the gap, even though both parallel and perpendicular orientations are magnetically inactive as the magnetic field is in the plane of the SRR (thus unable to excite resonant circulating currents around SRR rings) [19]. Similar magnetic resonance effects caused by the electric field excitation have been observed in previous reports [20]. When the E-field is oriented parallel to the SRR gap [Fig. 4(b)], only a single resonance dip is observed at 0.8 THz. This transmission dip is related to the dipole resonance due to a pair of linear currents in the SRR side lengths parallel to the incident E-field. It is clear that the dipole resonance at 0.8 THz shifts to lower frequency compared to the simulated resonance at 0.83 THz. The sample was subsequently heated to ~ 90 °C, which is above the semiconductor-metal transition temperature (~68 °C). At this temperature VO₂ became metallic, shorting out all the SRR structures, and thus exhibiting very low transmission over the entire frequency range.

In order to achieve modulation only at the desired frequencies, small VO₂ pads (6 μm x 8 μm) were used on the ring gaps, which are slightly larger than the gap area (4 μm x 6 μm). [See Fig.1 (d-f)]. In this design the quality factor (Q) of the resonators was greatly increased due to the reduced quantity of VO₂ and associated dissipation loss compared to the previous design (SRRs on planar VO₂). Accordingly, upon heating the sample above the transition temperature to ~90

°C, a strong modulation effect was achieved at the magnetic and electric resonances. Figure 5 shows the frequency-dependent transmission spectra of a hybrid SRR-patterned VO₂ device at two different temperatures (25 and 90 °C) under an applied E-field perpendicular to the SRR gap. At 25 °C, two transmission dips were observed at 0.35 THz and 0.72 THz, which correspond to the magnetic resonances of the outer and inner rings, respectively. These magnetic resonances are obtained due to resonant currents circulating around the SRR rings and charge accumulation at the gap [19]. Notably, the linewidth of these resonances is narrower than those observed in the SRR-planar VO₂ device (see Fig.4a) due to the higher quality factor. At ~90 °C, however, these magnetic resonant dips disappear and only a broad and weak dip (dipole resonance) was observed between 0.84 and 0.94 THz. This weak resonance can be improved by modifying the geometry of the gap in order to reduce its resistance. As the temperature was raised above 70 °C, the VO₂ pads became metallic and thus the gap of the SRR turned conductive, resulting in closed rings (i.e., $g=0$). The transmission of these closed rings revealed only a single broad range stop band, which closely overlapped the resonance of the SRR array in parallel orientation (data is not shown). The simulated transmission spectra are also shown in Figs. 5(a) and 5(b), which agree very well with the experimental results at both temperatures.

In order to understand the nature of resonances in the SRR structures built on both planar and patterned VO₂ films, the simulated electric field distribution of both devices at their resonant frequencies is shown in Figure 6. It is clear that the electric field intensity concentrates on the gap of the outer SRR [Figs. 6(a) and 6(c)] and on the gap of the inner SRR [Figs. 6(b) and 6(d)] at its magnetic resonant frequencies. At 25 °C, although both planar and patterned devices showed similar resonator behavior as seen in Fig. 6, the magnetic resonant peaks in the patterned VO₂ device were much sharper due to the reduced quantity of VO₂ (see Fig.4a) and associated parasitic losses compared to the SRR structures on the planar VO₂ film (see Fig.5a). Upon

heating the samples to 90 °C, the patterned VO₂ devices showed a strong modulation effect at the resonant frequencies, while the planar VO₂ devices showed no modulation effect at their resonant frequencies. Thus, the patterned VO₂ device is recommended to modulate THz waves.

Figure 7(a) shows the comparison of transmission spectra of a hybrid SRR-patterned VO₂ device measured at 25 and 90 °C, indicating a clear transmission change between two temperatures at the observed resonances. As can be seen in Fig. 7(b), the modulation efficiency, defined as $(T_{\max} - T_{\min}) / T_{\max}$, was calculated to be ~60 % and 54 % at 0.35 THz and 0.72 THz, respectively with a bandwidth of ~30 GHz (FWHM) due to the high Q of the magnetic resonances, while the wider resonance in the frequency range from 0.84 THz to 0.94 THz showed a modulation efficiency of only 22 %. It is clear that upon transition of the VO₂, the low temperature magnetic resonances observed at 0.35 and 0.72 THz are removed by closing the gaps in the rings, resulting in the observed broad dipole resonance of the SRRs at 0.84 - 0.94 THz. This broad resonance can be used for broadband data transmission in communication systems even though the modulation efficiency is slightly low (20 - 40 %). Finally, it should be pointed out, that the low efficiency in this resonance can be improved by increasing the VO₂ film thickness used in the gap or by modifying the gap geometry because thicker VO₂ films (but less than its skin depth, ~1000 nm) can provide less Ohmic losses between VO₂ and Au SRR in the gap, and the Q factor can be improved by decreasing gap spacing or increasing the gap width of the SRRs. Furthermore, in this work, although we have measured the devices at only two different temperatures, the electrically or optically driven SMT in VO₂ can be used to fabricate more practical hybrid SRR-VO₂ devices that can be tuned actively in resonance frequency and amplitude.

4. Conclusion

In this work, high quality VO₂ films were prepared by pulsed laser deposition exhibiting a significant resistivity drop ($>10^4 \Omega\text{-cm}$) and large optical transmittance change ($> 60 \%$) in the near-infrared region across their semiconductor-to-metal transition. Hybrid metamaterial devices designed for the THz frequency regime were fabricated by building double split-ring resonator arrays on patterned VO₂ film by a conventional lift off process. By changing the conductivity of VO₂ via temperature, the conductance in the SRR gap was adjusted to modulate their resonance frequencies and associated THz transmission. The experimental measurements agreed very well with the simulation results for these SRR metamaterials based on both semiconducting and metallic VO₂. Finally, a resonance modulation efficiency of $>50 \%$ was achieved in these SRR-VO₂ hybrid metamaterial devices.

Acknowledgments

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Figure Captions

Figure 1. Hybrid SRR metamaterials based on planar $\text{VO}_2/\text{Al}_2\text{O}_3$ [(a) – (c)] and patterned $\text{VO}_2/\text{Al}_2\text{O}_3$ [(d) – (f)]. (a) and (d) Schematics of SRR-planar VO_2 and SRR-patterned VO_2 devices, respectively. (b) and (e) Diagrams of SRR unit cell: the dimensions of the SRR are $l = 50 \mu\text{m}$, $w = 6 \mu\text{m}$, $t = 3 \mu\text{m}$, $g = 4 \mu\text{m}$, and $P = 100 \mu\text{m}$. (c) and (f) Optical microscope images of array of SRR structures on planar VO_2 and patterned VO_2 , respectively.

Figure 2. Schematic illustration of terahertz-time domain spectroscopy (THz-TDS) setup.

Figure 3. (a) Temperature-dependence of the electrical resistivity of a VO_2 thin film deposited on sapphire substrate. (b) Optical transmittance spectra of a VO_2 thin film measured at 25 °C and 90 °C. The film thickness was $\sim 100 \text{ nm}$.

Figure 4. Measured transmission spectra of hybrid SRR metamaterials based on planar $\text{VO}_2/\text{Al}_2\text{O}_3$ for the electric field polarization (a) perpendicular and (b) parallel to the SRR gap at two different temperatures (25°C and 90 °C). Simulation results (dashed lines) are also shown for both orientations.

Figure 5. Measured transmission spectra of hybrid SRR metamaterials based on patterned $\text{VO}_2/\text{Al}_2\text{O}_3$ at 25°C (a) and 90 °C (b). The E-field was applied perpendicular to the SRR gap. Simulation results (dashed line) are also shown in (a) and (b).

Figure 6. Simulated electric field distribution of hybrid SRR metamaterials based on planar VO₂/Al₂O₃ [(a) and (b)] and patterned VO₂/Al₂O₃ [(c) and (d)] at (a) 0.33 THz, (b) at 0.69 THz, (c) 0.35 THz and (d) 0.72 THz. The electric field was applied perpendicular to the SRR gap for both devices. The color scale bar was normalized by the maximum intensity value to be a range from 1 to 0.

Figure 7. (a) Comparison of measured transmission spectra of hybrid SRR- patterned VO₂ device at 25°C and 90 °C. (b) Transmission modulation efficiency, defined as $(T_{\max}-T_{\min})/T_{\max}$, obtained from (a). The vertical dashed lines in (a) and (b) indicate the magnetic resonances.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7