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Wavelength-tunable infrared metamaterial by tailoring magnetic resonance condition with VO₂ phase transition

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In this work, we report the design of a wavelength-tunable infrared metamaterial by tailoring magnetic resonance condition with the phase transition of vanadium dioxide (VO₂). Numerical simulation based on the finite-difference time-domain method shows a broad absorption peak at the wavelength of 10.9 μ m when VO₂ is a metal, but it shifts to 15.1 μ m when VO₂ changes to dielectric phase below its phase transition temperature of 68 °C. The large tunability of 38.5% in the resonance wavelength stems from the different excitation conditions of magnetic resonance mediated by plasmon in metallic VO₂ but optical phonons in dielectric VO₂. The physical mechanism is elucidated with the aid of electromagnetic field distribution at the resonance wavelengths. A hybrid magnetic resonance mode due to the plasmon-phonon coupling is also discussed. The results here would be beneficial for active control of thermal radiation in novel electronic, optical, and thermal devices. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4896525]

I. INTRODUCTION

The unique phase transition behavior of vanadium dioxide $(VO_2)^{1-3}$ has drawn lots of attentions recently and many applications have been found. Optical properties of VO_2 change dramatically when the phase transition between dielectric and metal occurs at 68 °C, which can be thermally induced with temperature control. Applications of VO_2 have been demonstrated in optical information storage,⁴ strain sensing,⁵ and lithium-ion batteries.⁶ Moreover, dielectric VO_2 possesses several optical phonon modes in the infrared, which have been employed to modulate radiative heat transfer⁷ in designing novel thermal devices such as vacuum thermal diodes/rectifiers^{8,9} and thermal transistors.¹⁰

Progresses have been made recently in designing tunable metamaterials made of phase transition VO₂. Dicken *et al.* demonstrated a frequency-tunable metamaterial by depositing split-ring resonators on a VO₂ film.¹¹ Kats *et al.* showed ~10% resonance wavelength tunability with a plasmonic antenna array on a VO₂ film in the mid-infrared.¹² They also demonstrated an ultra-thin tunable perfect absorber based on the VO₂ phase transition.¹³ More recently, a switchable thermal antenna with periodically patterned VO₂ has been proposed.¹⁴ Designs reported in Refs. 12 and 14 were realized by modulating the excitation condition of surface plasmon polariton (SPP) at the interface between subwavelength plasmonic nanostructures and the supporting VO₂ film upon phase transition.

Magnetic resonance has been studied intensively for designing selective thermal emitters^{15,16} and perfect metamaterial absorbers.¹⁷ Magnetic resonance occurs when external electromagnetic wave couples with magnetic resonance excited inside the metamaterial structures typically in a metal-insulator-metal configuration, resulting in strong absorption or emission at the selected resonance frequency. Note that, magnetic resonance can be also excited in the mid-infrared regime with polar materials, mediated by optical phonons, rather than plasmon in metals. An infrared selective emitter made of SiC has been demonstrated by exciting magnetic resonance within its phonon absorption band.¹⁸ The electrical current induced by the resonant magnetic field is realized by the high-frequency vibration of ions in SiC, rather than free charges in metals.

In this study, we present a tunable infrared metamaterial by exciting magnetic resonance at different conditions with either metallic or dielectric VO₂, leading to highly tunable resonant wavelength upon the phase transition of VO₂. Figure 1 depicts the proposed tunable metamaterial structure, which is made of a one-dimensional VO₂ periodic grating structure (period $\Lambda = 1.5 \,\mu\text{m}$ and strip width $w = 1.25 \,\mu\text{m}$) on stacked MgF₂ and VO₂ layers. The thicknesses of the VO₂ grating and thin films are $h = 0.5 \,\mu\text{m}$, $d_1 = 0.3 \,\mu\text{m}$, and $d_2 = 1 \,\mu\text{m}$, respectively. In practice, the VO₂ grating could be formed by the



FIG. 1. Proposed 1D tunable structure with period $\Lambda = 1.5 \ \mu m$, strip width $w = 1.25 \ \mu m$, layer thicknesses $h = 0.5 \ \mu m$, $d_1 = 0.3 \ \mu m$, and $d_2 = 1 \ \mu m$. The phase transition of VO₂ can be controlled by modulating the temperature.

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strain mismatch method from a VO_2 film on a flexible substrate,^{19,20} while the temperature of the structure can be modulated to thermally control VO_2 phase transition.

II. NUMERICAL METHODS

When the temperature is above $68 \,^{\circ}$ C, VO₂ is an isotropic metal, whose electrical permittivity ε_m can be described by a Drude model as¹

$$\varepsilon_{\rm m} = -\varepsilon_{\infty} \frac{\omega_p^2}{\omega^2 + i\omega\Gamma},\tag{1}$$

where ω is angular frequency, $\varepsilon_{\infty} = 9$ is the high-frequency constant, $\omega_p = 8000 \text{ cm}^{-1}$ is the plasma frequency, and $\Gamma = 10\,000 \text{ cm}^{-1}$ is the collision frequency. When the temperature is below 68 °C, VO₂ becomes dielectric but with uniaxial anisotropy. Considering (200)-oriented crystal VO₂ with optical axis normal to the surface,¹ it exhibits ordinary dielectric response denoted as ε_O when incident electric field is perpendicular to optical axis, and extraordinary response ε_E when electric field is parallel to optical axis. Both components can be described by a classical oscillator model as

$$\varepsilon(\omega) = \varepsilon_{\infty} + \sum_{j=1}^{N} \frac{S_j \omega_j^2}{\omega_j^2 - i\gamma_j \omega - \omega^2},$$
 (2)

where ω_j is the phonon vibration frequency, γ_j is the scattering rate, S_j represents the oscillation strength, and j is the phonon mode index. The values for each parameter can be found from Ref. 1 for both ordinary ($\varepsilon_{\rm O}$) and extraordinary ($\varepsilon_{\rm E}$) components with a total of eight phonon modes for $\varepsilon_{\rm O}$ and nine modes for $\varepsilon_{\rm E}$. In the simulation, the permittivity tensor was employed to consider the uniaxial anisotropy of dielectric VO₂,

$$\overline{\overline{\varepsilon}}_{\text{dielectric}} = \begin{pmatrix} \varepsilon_{\text{O}} & 0 & 0\\ 0 & \varepsilon_{\text{O}} & 0\\ 0 & 0 & \varepsilon_{\text{E}} \end{pmatrix}.$$
 (3)



FIG. 2. Real parts of permittivity of VO_2 at either metallic or dielectric phase, which exhibits both ordinary and extraordinary dielectric response.

Figure 2 plots the real parts for the permittivity of metallic and dielectric VO₂ in the mid-infrared regime from 5 μ m to 20 μ m in wavelength. The metallic phase exhibits a negative real part of permittivity, which is crucial to excite plasmonic resonances as found in most noble metals.²¹ On the other hand, there exist several phonon modes in both ordinary and extraordinary components of the dielectric phase. As a result, negative real part of permittivity exists in the wavelengths from 12.4 μ m to 16.7 μ m for ε_0 and from 17.5 μ m to 18.8 μ m for ε_E , respectively. As pointed out by Ref. 18, negative permittivity is required to excite phononmediated magnetic resonance in polar materials. The unique material properties of metallic and dielectric VO₂ suggest the potential in exciting magnetic resonance at both phases.

The finite-difference time-domain method (Lumerical Solutions, Inc.) was used to numerically calculate the spectral reflectance *R* and transmittance *T* of the proposed tunable metamaterial above and below the VO₂ phase transition temperature of 68 °C. The optical constants of MgF₂ were obtained from Palik's data.²² A linearly polarized plane wave was incident normally onto the metamaterial structure with transverse-magnetic (TM) incidence, in which magnetic field is along the grating groove direction. Note that, magnetic resonance can be excited only at TM polarization in 1D grating based metamaterials.^{15,16} A numerical error less than 2% was verified with sufficiently fine mesh sizes. The spectral normal absorptance in the infrared region was thus obtained by $\alpha = 1-R-T$ based on energy balance.

III. RESULTS AND DISCUSSIONS

A. Tunable resonance absorption with VO₂ phase transition

As shown in Fig. 3, when the temperature is above 68 °C, the VO₂ is at metallic phase and the metamaterial exhibits a broad absorption band peaked at the wavelength of 10.9 μ m with almost 100% absorption. However, when VO₂ becomes dielectric at temperatures less than 68 °C, the absorption band is narrower and shifts to the peak



FIG. 3. Simulated normal absorptance of proposed tunable absorber in the mid-infrared upon VO_2 phase transition, showing a relative 38.5% shift of resonant absorption peak wavelength.

wavelength of 15.1 μ m with maximum absorptance of 0.97, resulting in a relative 38.5% peak wavelength shift upon the VO₂ phase transition from metal to dielectric induced thermally. Note that there exist three bumps on the shoulder of absorption peak around 13 μ m, 16.5 μ m, and 19 μ m, which are caused by the abrupt change in the optical properties of dielectric VO₂ associated with several phonon absorption modes at these wavelengths.

In fact, both absorption peaks are caused by the excitations of magnetic resonance at both phases of VO_2 . But the fundamental difference is that, one is assisted by free charges or plasmon in metallic phase, while the other is mediated by optical phonons in its dielectric phase. The different resonance conditions and thereby the resulting large resonance wavelength shift are due to different optical behaviors of different energy carriers that excite the magnetic resonances.

B. Electromagnetic field distribution at magnetic resonance

To illustrate the underlying mechanism responsible for the large absorption peaks, electromagnetic field distributions at the cross section of the metamaterial structure were plotted at the resonance wavelengths with metallic and dielectric phases of VO₂, as shown in Figs. 4(a) and 4(b), respectively. The arrows indicate the strength and direction of the electric field vectors, while the contour shows magnetic field strength normalized to the incidence as $|H/H_0|^2$ at different locations.



FIG. 4. Electromagnetic field distribution at resonance peak wavelengths when VO_2 is at (a) metallic or (b) dielectric phase. The field patterns show the exact behavior of magnetic resonance with both phases of VO_2 , but assisted by plasmon in metallic VO_2 and mediated by optical phonons in dielectric VO_2 , respectively.

When VO₂ is at metallic phase, the electric field vectors inside the MgF₂ layer underneath the VO₂ strips indicate an anti-parallel current loop, along with the strong localization of magnetic field, as shown in Fig. 4(a). The localized energy is more than five times higher than the incidence. This is the exact behavior of magnetic resonance that has been intensively studied in similar 1D grating based metamaterials.^{15,16} Due to the oscillating movement of free charges in metallic VO₂, the sandwiched MgF₂ layer serves as a capacitor, while top metallic VO₂ strip and the bottom metallic VO₂ film function as inductors, forming a resonant alternating-current circuit. When the magnetic resonance occurs, the external electromagnetic energy at the resonant wavelength of 10.9 μ m is coupled with the oscillating plasmon, resulting in almost 100% absorption inside the metamaterial structure.

When VO₂ becomes dielectric with the temperature below 68 °C, the electromagnetic field shown in Fig. 4(b) presents a similar behavior of magnetic resonance with an induced anti-parallel electric current loop and confined magnetic field inside the MgF₂ layer but at a different resonance wavelength of 15.1 μ m. The localized energy strength is about five times to the incidence. Note that, this resonant wavelength is within the phonon absorption band of the ordinary component of dielectric VO₂, in which negative permittivity exists. When optical phonons vibrate at high frequency, the fast movements of bound charges or ions form oscillating electric currents and an inductor-capacitor resonant circuit, resulting in the excitation of magnetic resonance. Since the energy carrier changes from free electrons to optical phonons upon the phase transition of VO₂ from metal to dielectric, a large shift in resonance wavelengths occurs. It should be noted that, similar to the surface phonon polariton with polar materials,²³ which is a counterpart of SPP in the infrared regime, phonon-mediated magnetic resonance¹⁸ is the counterpart of magnetic resonance in plasmonic metamaterials made of metallic nanostructures.^{15–17}

C. Hybrid magnetic resonance due to the phonon-plasmon coupling

Finally, we would like to show that a hybrid magnetic resonance mode could also occur by the phonon-plasmon coupling from a modified tunable metamaterial by replacing the bottom VO₂ layer with a gold film, as shown in the inset of Fig. 5. The period and strip width of the top VO₂ grating are kept unchanged, while the thicknesses of the grating and the MgF₂ spacer layer are $h = d_1 = 0.5 \mu m$.

The spectral normal absorptance of the hybrid structure at TM waves is plotted in Fig. 5. When VO₂ is in either metallic or dielectric phase, the absorption peaks remain almost at the same resonance wavelengths, suggesting that magnetic resonance can still be excited in both phases of VO₂. However, maximum absorptance drops slightly to 0.85 for the peak with metallic VO₂, while the absorption peak with dielectric VO₂ becomes narrower, after the bottom VO₂ film was replaced by a gold substrate. The successful excitation of magnetic resonance between metallic VO₂ strips and the bottom gold film is easy to understand, as there exist free charges in both metals. The peak absorptance drops due to



FIG. 5. Normal absorptance of the tunable structure with gold substrate instead of the bottom VO_2 layer. The absorption peaks still exist because of a hybrid magnetic resonance mode due to phonon-plasmon coupling between top dielectric VO_2 and bottom gold.

the plasmonic coupling between metallic VO₂ and Au, whose strength is weaker compared to that between two identical materials with matching plasmonic properties. On the other hand, it would be expected that it would fail to excite phonon-mediated magnetic resonance due to the removal of the bottom VO₂ film. Surprisingly, the strong absorption with dielectric VO₂ could still occur. This can be understood by the excitation of a hybrid magnetic resonance mode due to the strong coupling between optical phonons in dielectric VO_2 and plasmon in the bottom gold substrate. The high-frequency vibration of optical phonons at the top interface of the MgF₂ spacer along with the movement of plasmon at the bottom interface could still form a close-loop inductor-capacitor circuit, which successfully excites magnetic resonance at the wavelength of 14.8 μ m. Note that the absorption peak becomes narrower because Au has less intrinsic loss compared with the VO₂ substrate.

IV. CONCLUSIONS

In summary, we have numerically demonstrated a wavelength-tunable metamaterial by tailoring magnetic resonance conditions with phase transition of VO₂. The absorption peak shifts from $10.9 \,\mu\text{m}$ to $15.1 \,\mu\text{m}$ upon the VO₂ phase transition from metal to dielectric, resulting in a relative 38.5% shift in the peak wavelength. The underlying physical mechanisms lie in the plasmon-assisted magnetic resonance in metallic VO₂ and phonon-mediated counterpart in dielectric VO₂, which leads to different resonance

wavelengths. A hybrid magnetic resonance mode due to phonon-plasmon coupling was also discussed when replacing the bottom VO_2 layer with a gold film, which could simplify the metamaterial design in practice. The wavelength-tunable metamaterial absorber or emitter could find applications in tunable infrared detectors and coherent thermal emitters. The insights and understanding gained in this work will facilitate the design of novel tunable metamaterials for active control of thermal radiation in electronic, optical, and thermal devices.

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