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 exciting magnetic resonance with phase transition of vanadium dioxide (VO₂). Numerical simulation shows a broad absorption peak at the wavelength of 10.9 μm when VO₂ is a metal, but it shifts to 15.1 μm 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 assisted 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 plasmon-phonon coupling is also discussed. The results here would be beneficial for active control in novel electronic, optical and thermal devices.

OCIS Codes: (160.3918) Metamaterials; (300.1030) Absorption; (310.6628) Subwavelength structures.

The unique phase transition behavior of vanadium dioxide (VO₂) [1-3] has drawn lots of attentions recently and many applications have been found. Optical properties of VO₂ changes dramatically when phase transition from dielectric to metal occurs at 68°C, which can be thermally induced by temperature control. Applications of VO₂ have been demonstrated in optical information storage [4], strain sensing [5], and lithium-ion batteries [6]. Moreover, dielectric VO₂ possesses several optical phonon modes in the infrared, which has been employed to modulate radiative heat transfer [7] in designing novel thermal devices such as thermal diodes/rectifiers [8,9] and thermal transistors [10].

Efforts have been made recently in tunable metamaterials made of phase transition VO₂. Dicken et al. [11] demonstrated a resonance-frequency-tunable metamaterial by depositing split-ring resonators on a VO₂ film. Kats et al. [12] showed ~10% resonance wavelength tunability with a plasmonic antenna array on a VO₂ film in the mid-infrared. They also demonstrated an ultra-thin tunable perfect absorber based on the VO₂ phase transition [13]. Previous designs were realized by modulating the excitation condition of surface plasmon polariton (SPP) at the interface of subwavelength plasmonic nanostructures and the supporting VO₂ film upon phase transition. However, the tunable frequency range was quite limited from the reported work.

Magnetic resonance has been studied intensively for designing selective thermal emitters [14,15] and perfect metamaterial absorbers [16]. 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 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 [17]. The induced electrical current by the resonant magnetic field is realized by the high-frequency vibration of ions in SiC, rather than free charges in metals.

Fig. 1 Proposed 1D tunable metamaterial structure with period Λ = 1.5 μm, strip width w = 1.25 μm, layer thicknesses h = 0.5 μm, d₁ = 0.3 μm, and d₂ = 1 μm. The phase transition of VO₂ can be controlled by modulating the temperature.

In this study, we present an 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. Fig. 1 depicts the proposed tunable metamaterial structure, which is made of a one-dimensional VO₂ periodic grating structure (period Λ = 1.5 μm and strip width w = 1.25 μm) on stacked MgF₂ and VO₂
film. The thicknesses of the VO$_2$ grating and thin films are $h = 0.5$ μm, $d_1 = 0.3$ μm, and $d_2 = 1$ μm. The temperature of the structure can be modulated to thermally control VO$_2$ phase transition.

When the temperature is above 68°C, VO$_2$ is an isotropic metal, whose electrical permittivity $\varepsilon_m$ can be described by a Drude model as [1]

$$
\varepsilon_m = -\varepsilon_\infty \frac{\omega_p^2}{\omega^2 + i\omega\Gamma}
$$

where $\omega$ is angular frequency, $\varepsilon_\infty = 9$ is the high-frequency constant, $\omega_p = 8000$ cm$^{-1}$ is the plasma frequency, and $\Gamma = 10000$ cm$^{-1}$ is the collision frequency. When the temperature is below 68°C, VO$_2$ becomes dielectric but with uniaxial anisotropy. Considering (200)-oriented crystal VO$_2$ with optical axis normal to the surface [1], it exhibits ordinary dielectric response denoted as $\varepsilon_0$ when incident electric field is perpendicular to optical axis, and extraordinary response $\varepsilon_\ell$ when electric field is parallel to optical axis. Both components can be described by a classical oscillator model as

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

where $\omega_j$ is the phonon vibration frequency, $\gamma_j$ 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 [1] for both ordinary ($\alpha$) and extraordinary ($\beta$) components with a total of eight phonon modes for $\varepsilon_0$ and nine modes for $\varepsilon_\ell$. In the simulation, the permittivity tensor was employed to consider the uniaxial anisotropy of dielectric VO$_2$:

$$
\bar{\varepsilon}_{\text{dielectric}} = \begin{pmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_\ell \end{pmatrix}
$$

Fig. 2(a) plots the real parts for the permittivity of metallic and dielectric VO$_2$ in the mid-infrared regime from 5 μm to 20 μm 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 [18]. 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 μm to 16.7 μm for $\varepsilon_0$ and from 17.5 μm to 18.8 μm for $\varepsilon_\ell$, respectively. As pointed out by Ref. [17], negative permittivity is required to excite phonon-mediated magnetic resonance in polar materials. The unique material properties of metallic and dielectric VO$_2$ suggest the potential in exciting magnetic resonance in 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 VO$_2$ phase transition temperature of 68°C. The optical constants of MgF$_2$ were obtained from Palik’s data [19]. A linearly polarized plane wave was incident normally onto the metamaterial structure with transverse-magnetic (TM) waves, 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 [14,15]. A numerical error less than 2% was verified with sufficiently fine mesh sizes.

As shown in Fig. 2(b), the normal absorbance in the infrared region from 5 μm to 20 μm was thus obtained by $\alpha = 1 - R - T$ based on energy balance. When the temperature is above 68°C, the VO$_2$ is at metallic phase and the metamaterial exhibits a broad absorption band peaked at the wavelength of 10.9 μm with almost 100% absorption. However, when VO$_2$ becomes dielectric at temperatures less
than 68°C, the absorption band is narrower and shifts to the peak wavelength of 15.1 μm 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 two bumps on the shoulder of absorption peak around 16.5 μm and 19 μm, which are caused by the abrupt changes of optical properties of dielectric VO₂ due to strong phonon vibration.

In fact, both absorption peaks are caused by the excitations of magnetic resonance at both phases of VO₂. 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 thus the resulting large resonance wavelength shift are due to different optical behaviors of different energy carriers that excite the magnetic resonances.

To illustrate the underlying mechanism responsible for the large absorption peak shift, 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. 3(a) and 3(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/H₀|^2 at different locations.

When VO₂ is at metallic phase, the electric field vectors inside the MgF₂ layer underneath the VO₂ strips suggest an anti-parallel current loop, along with the strong localization of magnetic field, as shown in Fig. 3(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 [14,15]. 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 μm 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. 3(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 μm. 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 [20], which is a counterpart of SPP in the infrared regime, phonon-mediated magnetic resonance [17] is the counterpart of magnetic resonance in plasmonic metamaterials made of metallic nanostructures [14-16].

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. 4. 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 = d₁ = 0.5 μm.

The spectral normal absorptance of the hybrid structure is plotted in Fig. 4. The absorption peaks when VO₂ is in either metallic or dielectric phase 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 VO$_2$, while the absorption peak with dielectric VO$_2$ becomes narrower, after the bottom VO$_2$ film was replaced by a gold substrate. The successful excitation of magnetic resonance between metallic VO$_2$ strips and the bottom gold film is easy to understand, since there exist free charges in both metals. 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 VO$_2$ film. Surprisingly, the strong absorption with dielectric VO$_2$ 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 vibration of optical phonons at high frequency at the top interface of the MgF$_2$ 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 μm.

In conclusion, we have demonstrated a wavelength-tunable metamaterial by tailoring magnetic resonance conditions with phase transition VO$_2$. The absorption peak shifts from 10.9 μm to 15.1 μm upon the VO$_2$ 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 VO$_2$ and phonon-mediated counterpart in dielectric VO$_2$, 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. The insights and understanding gained in this work will facilitate the design of novel tunable metamaterials in active control of electronic, optical, and thermal devices.

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