Actively tuning anisotropic light–matter interaction in biaxial hyperbolic material $\alpha$-MoO$_3$ using phase change material VO$_2$ and graphene

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Anisotropic hyperbolic phonon polaritons (PhPs) in natural biaxial hyperbolic material $\alpha$-MoO$_3$ has opened up new avenues for mid-infrared nanophotonics, while active tunability of $\alpha$-MoO$_3$ PhPs is still an urgent problem necessarily to be solved. In this study, we present a theoretical demonstration of actively tuning $\alpha$-MoO$_3$ PhPs using phase change material VO$_2$ and graphene. It is observed that $\alpha$-MoO$_3$ PhPs are greatly dependent on the propagation plane angle of PhPs. The insulator-to-metal phase transition of VO$_2$ has a significant effect on the hybridization PhPs of the $\alpha$-MoO$_3$/VO$_2$ structure and allows to obtain actively tunable $\alpha$-MoO$_3$ PhPs, which is especially obvious when the propagation plane angle of PhPs is 90°. Moreover, when graphene surface plasmon sources are placed at the top or bottom of $\alpha$-MoO$_3$ in $\alpha$-MoO$_3$/VO$_2$ structure, tunable coupled hyperbolic plasmon–phonon polaritons inside its Reststrahlen bands (RBs) and surface plasmon–phonon polaritons outside its RBs can be achieved. In addition, the above-mentioned $\alpha$-MoO$_3$-based structures also lead to actively tunable anisotropic spontaneous emission (SE) enhancement. This study may be beneficial for realization of active tunability of both PhPs and SE of $\alpha$-MoO$_3$, and facilitate a deeper understanding of the mechanisms of anisotropic light–matter interaction in $\alpha$-MoO$_3$ using functional materials.

Keywords: light–matter interaction, hyperbolic material, phase change material, graphene

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1. Introduction

Hyperbolic materials (HMs) exhibit unique optical, electronic and magnetic properties, their natural hyperbolicity offers advantages over other conventional metal and dielectric materials$^{[1–6]}$. As a special class of HMs, biaxial $\alpha$-phase molybdenum trioxide ($\alpha$-MoO$_3$) has drawn much attention due to its enigmatic optoelectronic characteristics throughout the infrared regions$^{[7–9]}$. So far, $\alpha$-MoO$_3$ composing of octahedral unit cells with nonequivalent Mo–O bonds along its three principal crystalline axes (between 550 cm$^{-1}$ and 1000 cm$^{-1}$) has attracted huge attention owing to its ability to excite highly anisotropic phonon polaritons (PhPs) with in-plane hyperbolic and elliptic dispersion$^{[10]}$, which gives rise to a versatile platform for studying anisotropic light–matter interaction. Because of its biaxial anisotropy and hyperbolicity, $\alpha$-MoO$_3$ is an excellent candidate to provide more rich physical phenomena than other HMs, thus an in-depth understanding of $\alpha$-MoO$_3$ is greatly significant for many different fields involving light–matter interactions, such as near-field radiative heat transfer, super-resolution technologies, spontaneous emission (SE) and spectral absorption$^{[11–14]}$. However, the active tunability of PhPs excited by resonance between photons and $\alpha$-MoO$_3$ is still a challenge owing to their inherent crystal lattice resulting in inefficient tunability.

Stacking and combining functional materials into a heterostructure offers a flexible and promising way to couple material properties and reveal underlying physical phenomena$^{[15,16]}$. As a typical functional material, because of its electrostatic doping possibility and ability to generate higher confinement and lower losses,$^{[15,16]}$ graphene becomes an excellent candidate for the realization of tunable surface plasmon polaritons (SPPs) via applying extra gate voltage in the mid-infrared and terahertz frequency ranges$^{[17–19]}$. Meanwhile, graphene can be flexibly combined with other micro/nanostructures and HMs to obtain coupled hybridization characteristics, which provides an alternative route to realize tunable optoelectronic devices$^{[20,21]}$. Over the past few years, graphene has been applied to realize tunable hyperbolic plasmon–phonon dispersion, near-field radiative heat transfer, SE enhancement and spectral absorption$^{[22–26]}$. Except for graphene, phase-change materials (PCMs) including Ge$_2$Sb$_2$Te$_5$ (GST) and vanadium dioxide (VO$_2$) have witnessed abundant achievements toward actively tunable light–matter interaction from the perspectives of principles and applications$^{[15,16,27]}$. Compared to that from amorphous GST.
to crystalline GST at about 438 K,\[^{16}\] the phase-transition temperature of VO\(_2\) from insulator to metal is about 340 K,\[^{28}\] which is easy to achieve and has little influence on the corresponding cooperating materials. Meanwhile, the phase-transition temperature of VO\(_2\) can be tuned by doping VO\(_2\) flake with W, Ti, Mg, and Al nanoparticles forming \(M_xV_{1-x}O_2\) \((M = W, Ti, Mg, \text{ or Al})\),\[^{29-33}\] such as the transition temperature can be tailored to 295 K by setting the composition \(x\) at 1.5% in \(W_xV_{1-x}O_2\).\[^{33}\] Because its advantages over other PCMs, VO\(_2\) serves as the basis for potential applications such as temperature-adaptive radiative coating, energy storage and reconfigurable electronics.\[^{34,35}\]

Since the dispersion modes including phonons and plasmons can be tuned in layered van der Waals material systems using functional materials,\[^{15,16}\] in this study, a theoretical platform is proposed to realize anisotropic light–matter interaction in \(\alpha\)-MoO\(_3\) by using phase transition properties of VO\(_2\) and SPPs of graphene. Hybridization PhP dispersion modes of \(\alpha\)-MoO\(_3\) and coupling hybridization dispersion between \(\alpha\)-MoO\(_3\) PhPs and graphene SPPs based on VO\(_2\) substrate will be detailedly discussed, meanwhile the propagation plane angle in biaxial \(\alpha\)-MoO\(_3\) will also be considered. Furthermore, comparative analysis of actively tunable anisotropic SE enhancement in \(\alpha\)-MoO\(_3\)-based structures will also be investigated.

2. The geometry and numerical model

In order to actively tune and control PhP behavior of the biaxial HM \(\alpha\)-MoO\(_3\) along its main crystallographic directions, a VO\(_2\) layer is used as the substrate owing to its attractive dielectric properties during the temperature-tuned insulator-metal phase transition, and the complex refractive index of VO\(_2\) is refereed from Ref. 29. The above-mentioned schematic is displayed in Fig. 1(a), where a Cartesian coordinate system is established, \(x\), \(y\), and \(z\) axes represent the \([100]\), \([001]\) and \([010]\) crystal directions of the \(\alpha\)-MoO\(_3\), respectively; \(\phi\) is the propagation plane angle of PhPs with respect to the \([100]\) crystal direction, \(t\) is defined as the \(\alpha\)-MoO\(_3\) thickness. Here \(\alpha\)-MoO\(_3\) is a natural biaxial HM and its optical response is dominated by the phonon absorption, whose principal permittivity components can be expressed as the following Lorentz equation:\[^{9}\]

\[
\varepsilon_j = \varepsilon'_j \left(1 + \frac{\omega^2}{\omega^2_{\text{LO}} - \omega^2 - i\Gamma_j} \right),
\]

where \(j = x, y, z\) denotes the three crystal principal axes, corresponding to the crystalline directions \([100]\), \([001]\) and \([010]\) of \(\alpha\)-MoO\(_3\), respectively; \(\varepsilon'_j\) represents the high-frequency dielectric constant; \(\omega^2_{\text{LO}}\) and \(\omega^2_{\text{TO}}\) refer to the longitudinal and transverse optical phonon frequencies; \(\Gamma_j\) is the broadening factor of the Lorentzian line shape, and the corresponding parameters can be referred from Ref. 9.

![Fig. 1](image)

The real parts of the dielectric tensor components \(\varepsilon_j\) in \(\alpha\)-MoO\(_3\) are displayed in Fig. 1(b), three Reststrahlen bands (RBSs) exist in the mid-infrared range from 545 cm\(^{-1}\) to 1010 cm\(^{-1}\). RB-1, RB-2 and RB-3 of \(\alpha\)-MoO\(_3\) are respectively in the ranges from 545 cm\(^{-1}\) to 851 cm\(^{-1}\) (the gray region), from 820 cm\(^{-1}\) to 972 cm\(^{-1}\) (the green region) and from 958 cm\(^{-1}\) to 1010 cm\(^{-1}\) (the purple region), which originate from in-plane phonon modes along the \([001]\), \([100]\) and \([010]\) crystalline directions, respectively. It is worth noting that RB-1 and RB-2 belong to the type-II hyperbolic band, while RB-3 to the type-I hyperbolic band. In addition, the relative permittivity tensor of \(\alpha\)-MoO\(_3\) for the propagation angle \(\phi\) is obtained from \(\varepsilon(\phi) = R(\phi)\varepsilon_{\alpha\text{-MoO}_3}R^T(\phi)\), where \(R(\phi)\) is the rotation matrix for transformation of coordinate system, and the relative permittivity tensor of \(\alpha\)-MoO\(_3\) can be written as\[^{36,37}\]

\[
\varepsilon(\phi) = \begin{pmatrix}
\varepsilon_x \cos^2 \phi + \varepsilon_y \sin^2 \phi & \sin \phi \cos \phi (\varepsilon_y - \varepsilon_z) & 0 \\
\sin \phi \cos \phi (\varepsilon_x - \varepsilon_z) & \varepsilon_y \sin^2 \phi + \varepsilon_z \cos^2 \phi & 0 \\
0 & 0 & \varepsilon_z
\end{pmatrix}.
\]

Since graphene supports SPPs in the mid-infrared region, graphene layers are added at the top or bottom of \(\alpha\)-MoO\(_3\) layer to realize additional PhPs tunability of the \(\alpha\)-MoO\(_3\) and investigate coupling hybridization dispersion between \(\alpha\)-MoO\(_3\) PhPs and graphene SPPs. In this study, graphene is treated as an ultrathin dielectric layer with the thickness \(\Delta = 0.34\) nm by an effective permittivity \(\varepsilon = 1 + i\sigma_\parallel/(\omega\varepsilon_0\Delta)\),\[^{24}\] \(\varepsilon_0\) is vacuum permittivity, and its conductivity \(\sigma_\parallel\) modeled as a sum of the intraband and interband terms is given by \(\sigma_\parallel = \sigma_{\text{intra}} + \sigma_{\text{inter}}\) written as\[^{24}\]

\[
\sigma_{\text{intra}} = \frac{i}{\omega + \frac{1}{\tau_\parallel}} \frac{2\varepsilon^2/\kappa T}{\pi h^2} \ln \left[ \frac{2\cos \left( \frac{\mu_\parallel}{2k_B T} \right)}{\tau_\parallel} \right],
\]

\[
\sigma_{\text{inter}} = \frac{\varepsilon^2}{4\hbar} \int \frac{G(\hbar \omega/2)}{(\hbar \omega)^2} \left[ \frac{\varepsilon^2}{(\hbar \omega)^2} \frac{\sinh^2 \left( \frac{\hbar \omega}{2T} \right)}{\hbar^2} \right] d\xi,
\]

where \(G(\xi) = \sinh(\xi/\kappa T)/\cosh(\mu_\parallel/\kappa T) + \cosh(\xi/\kappa T); \tau_\parallel, \varepsilon, \mu_\parallel, \kappa, \mu_\text{T}, h\) and \(T\) are the relaxation scattering time, electron charge, Boltzmann constant, chemical potential, reduced Planck’s constant, and temperature, respectively.
3. Results and discussion

3.1. Actively tuning biaxial PhPs of \(\alpha\)-MoO\(_3\) based on VO\(_2\) substrate

The dispersion relations of the above-mentioned heterostructures are calculated by an intensity of the imaginary part of the Fresnel reflection coefficient \(\text{Im}(r^{pp})\)\(^{[15,16]}\), where \(r^{pp}\) is the Fresnel reflection coefficient for \(p\)-polarized wave, which is calculated using the 4 \(\times\) 4 matrix\(^{[38-40]}\)
\[
\begin{pmatrix}
r^{ss} & r^{pp} \\
r^{ps} & r^{pp}
\end{pmatrix}
\]
Meanwhile, the whole results are calculated by using the MATLAB. As shown in Fig. 2, the hybridization PhP dispersion of the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure along its main crystallographic directions at the metal- and insulator-state of VO\(_2\) are calculated. Meanwhile, since \(\alpha\)-MoO\(_3\) is a biaxial HM, the effects of the propagation plane angle \(\phi\) on the PhP dispersion modes of \(\alpha\)-MoO\(_3\) are also considered at \(\phi = 0^\circ\), \(\phi = 45^\circ\) and \(\phi = 90^\circ\), respectively. The thickness of the \(\alpha\)-MoO\(_3\) layer is 150 nm to ignore the quantum confinement effects.\(^{[9]}\) It is worth noting that the color bars of the hybridization dispersion modes are appropriately adjusted to clearly show the differences. When VO\(_2\) is in the metal state shown in Figs. 2(a)–2(c), two hybridization PhP dispersion modes of the \(\alpha\)-MoO\(_3\)/VO\(_2\)(I) structure at \(\phi = 0^\circ\) are gathering inside RB-2 and RB-3, and the dispersion modes lead to group velocity \(v_g = \text{dw} / \text{dg} > 0\)\(^{[16]}\) in RB-2 and \(v_g < 0\) in RB-3, respectively. Two PhP dispersion modes of the \(\alpha\)-MoO\(_3\)/VO\(_2\)(I) structure at \(\phi = 90^\circ\) occur at RB-1 and RB-3, the group velocity in RB-2 keeps the same sign as that at \(\phi = 0^\circ\), meanwhile the group velocity is \(v_g > 0\) in RB-1. Interestingly, PhP dispersions of the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure at \(\phi = 45^\circ\) occur simultaneously within its three RBs and just span the part frequency range of its RBs. Moreover, with the increase of the propagation plane angles from \(\phi = 0^\circ\) to \(\phi = 90^\circ\), PhP dispersions of the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure within RB-3 always exist because of the contribution of permittivity tensor along [010] crystalline directions, while PhP dispersions of the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure within RB-1 gradually widen to nearly 851 cm\(^{-1}\) and within RB-2 gradually narrow to disappear, which are attributed to the overall consideration of the rotated permittivity tensor between [100] and [001] crystalline directions.\(^{[36,37]}\) In addition, similar results can also be obtained when VO\(_2\) is in the insulator state shown in Figs. 2(d)–2(f), while the hybridization PhP dispersion modes of \(\alpha\)-MoO\(_3\) are different before and after VO\(_2\) phase transition, and the results about the adjustment resulting from bottom VO\(_2\) are considered from the whole structure. Regardless of the phase transition state of VO\(_2\), the hybridization PhP dispersion modes in \(\alpha\)-MoO\(_3\) exhibit obviously in-plane anisotropic behaviors.

![Fig. 2. Hybridization dispersions of the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure for different propagation plane angles \(\phi\) at (a)–(c) metal state and (d)–(f) insulator state of VO\(_2\). The dashed lines of different colors represent the intrinsic RBs of \(\alpha\)-MoO\(_3\).](image-url)

As an example, hybridization PhP dispersion modes of the \(\alpha\)-MoO\(_3\)/VO\(_2\)(M) and \(\alpha\)-MoO\(_3\)/VO\(_2\)(I) structures at \(\phi = 90^\circ\) are analyzed, as shown in Figs. 2(c) and 2(f). Since the insulator-to-metal transition has a profound impact on the dielectric properties of VO\(_2\), compared to that in the \(\alpha\)-MoO\(_3\)/VO\(_2\) structure, the hybridization PhP branches in the \(\alpha\)-MoO\(_3\)/VO\(_2\)(I) structure significantly approach to the light line and widen, which can be attributed to its inherent lossy enhancement of VO\(_2\) once it undergoes a phase transition from the insulator state to metal state. Furthermore, in order to clearly describe that VO\(_2\) can actively tune \(\alpha\)-MoO\(_3\) PhPs, hybridization PhP dispersions of the \(\alpha\)-MoO\(_3\)/VO\(_2\)(M)
and α-MoO₃/VO₂(I) structures varying with the wavevector components \( k_x \) and \( k_y \) at the wavenumber of 780 cm\(^{-1}\) are calculated as shown in Figs. 3(a) and 3(b). The excitation range of hybridization PhP dispersion branches reads \(-\sqrt{-\varepsilon_x/\varepsilon_y} < k_y/k_0 < \sqrt{-\varepsilon_x/\varepsilon_y}\), \(k_y/k_0 < \sqrt{-\varepsilon_x/\varepsilon_y}\), \(^{[11]}\) the corresponding dielectric function components of α-MoO₃ at the wavenumber of 780 cm\(^{-1}\) are \(\varepsilon_x = 20.7 - i0.8\) and \(\varepsilon_y = -1.95 - i0.07\). As shown in Figs. 3(a) and 3(b), the bright color within the regions up and down the origin being bounded by the two white dashed lines of \( k_y \approx \pm 3.26k_0 \) is of the hybridization PhP dispersion branches. Once VO₂ undergoes an insulator-to-metal phase transition at the wavenumber of 780 cm\(^{-1}\), the complex dielectric function of VO₂ will convert from \(\varepsilon_{\text{VO}_2(0)} = 0.72 + i0.96\) to \(\varepsilon_{\text{VO}_2(I)} = -10.91 + i103.48\), thus the dielectric function differences of VO₂ between metal state and insulator state leading to the hybridization PhP dispersion modes in the α-MoO₃/VO₂ structure are different, and its hybridization PhP dispersion branches gradually move towards the directions of a smaller vector. Furthermore, the analytical expression for the dispersion relations of the α-MoO₃/VO₂ structure under the quasistatic approximation is utilized and can be written as

\[
\frac{k_p}{k_0} = \frac{\xi}{k_0 l} \left[ \arctan \left( \frac{\varepsilon_y}{\varepsilon_x} \right) + \arctan \left( \frac{\varepsilon_{\text{VO}_2(l)}}{\varepsilon_x} \right) + l \pi \right],
\]

where \(\varepsilon_1\) and \(\varepsilon_{\text{VO}_2}\) are the permittivities of air and VO₂, \(\mathbf{t}\) is the α-MoO₃ flake thickness, \(\varepsilon_x\) is the α-MoO₃ permittivity along the [010] crystalline directions, and \(\xi = i \left( \varepsilon_y/\varepsilon_x \right) \cos^2 \phi + \varepsilon_y \sin^2 \phi \). As shown in Figs. 3(a) and 3(b), the hybridization PhP dispersion branches of the α-MoO₃/VO₂ structure with green dashed lines agree well with those using the 4 \(\times\) 4 matrix.

![Fig. 3. Hybridization dispersions of (a) α-MoO₃/VO₂(M) and (b) α-MoO₃/VO₂(I) structures varying with wavevector components \(k_x\) and \(k_y\) at the wavenumber of 780 cm\(^{-1}\).](image)

### 3.2. Coupling hybridization dispersion between α-MoO₃ PhPs and graphene SPPs

The previous study clarified that active tunability of α-MoO₃ PhPs can be achieved through the phase transition of VO₂ without changing the structural parameters. Since graphene supports SPPs in the mid-infrared and terahertz region, \(^{[18,19]}\) which provides a flexible way to investigate coupling hybridization dispersion modes in α-MoO₃, two heterostructures are proposed to realize coupling hybridization dispersion between α-MoO₃ PhPs and graphene SPPs. As a benchmark, a graphene layer is added on the top of the α-MoO₃/VO₂ structure as shown in Fig. 4(g) and its hybridization dispersion modes will be investigated. Here, \(\mu_g = 0.3\) eV is chosen and fixed to investigate coupling hybridization dispersion modes, using other graphene chemical potentials can also obtain similar conclusions, and the results are not shown here. At \(\phi = 0^\circ\) shown in Figs. 4(a) and 4(d), coupling hybridization hyperbolic plasmon-phonon polariton (HPPP) dispersion modes between α-MoO₃ PhPs and graphene SPPs occur within its RBs, \(^{[42]}\) which are not pure PhPs displayed in Figs. 2(a) and 2(d) of the α-MoO₃/VO₂ structure. Especially within RB-2, the coupling hybridization polariton branches in α-MoO₃ are broader and move closer to the light cone because of the inherently lossy of the graphene SPPs, and the group velocity obviously increases when compared with that of the α-MoO₃/VO₂ structure in Figs. 2(a) and 2(d). Compared to the hybridization dispersion in the α-MoO₃/VO₂ structure, outside its RBs, surface plasmon-phonon polaritons (SPPPs) are formed by coupling with α-MoO₃ PhPs through the dominant contribution of graphene SPPs, and possess positive group velocity keeping similar with graphene SPPs. \(^{[42]}\) Similar results can also be obtained at \(\phi = 45^\circ\) [Figs. 4(b) and 4(e)] and \(\phi = 90^\circ\) [Figs. 4(c) and 4(f)]. It is worth noting that increasing the graphene chemical potential in the graphene/α-MoO₃/VO₂ structure, coupling hybridization dispersion branches gradually shift to the direction of the light line and the characteristics of graphene SPPs in the coupled hybridization dispersion become more obvious, which can be attributed to the strong coupling between α-MoO₃ PhPs and graphene SPPs. In addition, changing the phase transition state of VO₂ can also actively tune both coupled HPPP and...
SPPP dispersion modes in the graphene/α-MoO$_3$/VO$_2$ structure without changing the structural parameters.

Based on the graphene/α-MoO$_3$/VO$_2$ structure, an additional heterostructure with the placement of a surface plasmon source on either side of the α-MoO$_3$ is considered to further investigate both HPPP and SPPP dispersion modes, as schematically shown in Fig. 5(g). Figures 5(a)–5(f) show the hybridization dispersion modes of the graphene/α-MoO$_3$/graphene/VO$_2$ structure, here keeping $\mu_1 = \mu_2 = 0.3$ eV unchanged for simplicity. Compared with that in graphene/α-MoO$_3$/VO$_2$ structure, the coupling effect is even more pronounced via the extra contribution of the graphene SPPs at the bottom of the α-MoO$_3$ layer. When compared with that of the VO$_2$ substrate in insulator state shown in Figs. 5(a)–5(c), the hybridization dispersion modes of the graphene/α-MoO$_3$/graphene/VO$_2$(M) structure shown in Figs. 5(d)–5(f) obviously move closer to the light cone and widen, especially for $\phi = 90^\circ$ displayed in Fig. 5(f), which can be attributed to the reflection reinforcement at the VO$_2$ layer once it changes from insulator state to metal state.\cite{15}

Furthermore, the hybridization dispersion modes of the graphene/α-MoO$_3$/VO$_2$ and graphene/α-MoO$_3$/graphene/VO$_2$ structures varying with the wavevector components $k_x$ and $k_y$ at the wavenumber 780 cm$^{-1}$ are also calculated in Figs. 6(a)–6(d) to illustrate the differences. Coupling hybridization HPPP dispersion modes occur within the asymptotes $k_y \approx +3.26k_x$ and $k_y \approx -3.26k_x$, and coupling hybridization SPPP dispersion modes appearing

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig4}
\caption{Hybridization dispersions of the graphene/α-MoO$_3$/VO$_2$ structure at (a)–(c) metal state and (d)–(f) insulator state of VO$_2$ for different propagation plane angles $\phi$. (g) Schematic illustration of the graphene/α-MoO$_3$/VO$_2$ structure. The dashed lines of different colors represent the intrinsic RBs of α-MoO$_3$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig5}
\caption{Hybridization dispersions of the graphene/α-MoO$_3$/graphene/VO$_2$ structure at (a)–(c) metal state and (d)–(f) insulator state of VO$_2$ for different propagation plane angles $\phi$. (g) Schematic of the graphene/α-MoO$_3$/graphene/VO$_2$ structure. The dashed lines of different colors represent the intrinsic RBs of α-MoO$_3$.}
\end{figure}
on the left and right sides of the origin are mainly derived from the contribution of graphene SPPs. When VO$_2$ in the graphene/$\alpha$-MoO$_3$/VO$_2$ structure experiences the insulator-to-metal phase transition, its hybridization HPPP dispersion branches on the top and bottom sides of the origin gradually move towards the direction of zero-wave vector. Furthermore, as another graphene layer is added at the bottom of $\alpha$-MoO$_3$ layer in the graphene/$\alpha$-MoO$_3$/VO$_2$ structure shown in Figs. 6(c)–6(d), coupling hybridization dispersion modes between $\alpha$-MoO$_3$ PhPs and graphene SPPs are stronger, and hybridization HPPP dispersion branches obviously move towards the origin.

3.3. Actively tuning anisotropic SE enhancement in $\alpha$-MoO$_3$-based structures

In this section, we present a comparative analysis of anisotropic SE enhancement in $\alpha$-MoO$_3$-based structures to illustrate the VO$_2$ phase change so as to realize actively tunable SE. As compared with that in free space, the lifetime of an excited quantum emitter being placed near a special photonic material system can be modified, which is called Purcell effect. Hybridization dispersion mode modulated interaction with graphene and $\alpha$-MoO$_3$ on VO$_2$ substrate has significant effects on the SE rate of an excited quantum emitter. When the moment of the dipole on a system is along the z-direction, its SE rate (Purcell spectrum) can be described as:

$$
\frac{\Gamma}{\Gamma_0} = 1 + \frac{3}{2k_0^2} \text{Re} \left[ \int_0^{2\pi} f_{pp}(x, k_x, k_y) \frac{2i k_x k_y}{k_z} \right],
$$

where $\Gamma$ and $\Gamma_0 = \alpha_0^2 |\beta|^2 / 3\pi\varepsilon_0 \hbar c^3$ are the SE rate of a dipole emitter near the system and in free space, $d_0$ is the distance between the dipole emitter and proposed systems, $f_{pp}(x, k_x, k_y)$ is the total Fresnel reflection coefficient of the proposed systems.\[38,39\]

Then, the SE rates of $\alpha$-MoO$_3$-based structure are investigated and discussed. The effects of the geometric parameters on the SE rates of the $\alpha$-MoO$_3$/VO$_2$ structure are firstly discussed in Fig. 7, the basic geometrical parameters are set as $d_0 = 100$ nm, $t = 150$ nm, and $\phi = 0^\circ$. As shown in Figs. 7(a) and 7(b), with the increasing $d_0$ from 80 nm to 120 nm, the SE rates of the $\alpha$-MoO$_3$/VO$_2$ structure obviously reduce, which indicates that the SE rates is inversely proportional to the distance $d_0$ \[23,45,46\]. Results in Figs. 7(c) and 7(d) show that the SE rates of the $\alpha$-MoO$_3$/VO$_2$ structure keep the same order with the increasing $\alpha$-MoO$_3$ thickness $t$ from 50 nm to 200 nm. Then, the effects of the propagation plane angle $\phi$ on the SE rates of the $\alpha$-MoO$_3$/VO$_2$ structure are also discussed in Figs. 7(e) and 7(f), the enhancement regions of SE rate mainly gather within its hyperbolic bands, with increasing the propagation plane angle $\phi$ from 0 to 90$^\circ$, the SE enhancements within RB-3 always exist, while the SE enhancements at RB-1 and RB-2 greatly rely on propagation plane angle $\phi$, which can be attributed to the contribution of $\alpha$-MoO$_3$ PhPs. The above discussion indicates the SE enhancements mainly rely on the distance $d_0$ and the propagation plane angle $\phi$. Moreover, results in Fig. 7 indicate the SE rate of the $\alpha$-MoO$_3$/VO$_2$ structure significantly change as the VO$_2$ substrate undergoes the insulator-to-metal phase transition, which provides a feasible way to actively tune the SE rates of the $\alpha$-MoO$_3$.
Since the conductivity of graphene can be modulated by applying an extra gate voltage, which is an advantage for using graphene for photonics applications. Subsequently, SE rates of the graphene/α-MoO$_3$/VO$_2$ structure are theoretically investigated, as shown in Figs. 8(a) and 8(b). The SE rates of the graphene/α-MoO$_3$/VO$_2$ structure keep similar characteristics with those in α-MoO$_3$/VO$_2$ structure, meanwhile the ratio of SE rate of the α-MoO$_3$/VO$_2$ structure with and without adding different graphene layers is introduced for evaluating the tuning performance, which is defined as

$$\text{Ratio} = \frac{\text{SE rate of the structure with graphene}}{\text{SE rate of the structure without graphene}}.$$

Figures 8(c) and 8(d) give the ratio of SE rate of the α-MoO$_3$/VO$_2$ system with and without adding graphene layers. When compared to SE rates of the α-MoO$_3$/VO$_2$ structure, SE rates with adding graphene layers obvious enhance outside the RBs and are independent of the propagation plane angle $\phi$, which can be attributed to the contribution of the SPPPs. While SE rates within the RBs are mainly caused by the strong coupling between graphene plasmons and α-MoO$_3$ phonon polaritons, its enhancement is dependent on the propagation plane angle $\phi$. In addition, SE rates of the graphene/α-MoO$_3$/graphene/VO$_2$ structure are also considered, as shown in Figs. 8(e)–8(h), except for the magnitude, its SE rates are basically the same as those in the graphene/α-MoO$_3$/VO$_2$ structure, thus the graphene layer at the bottom of the α-MoO$_3$ layer in the graphene/α-MoO$_3$/VO$_2$ structure just plays a role in adding flexibility. When comparing the proposed α-MoO$_3$-based structures, the α-MoO$_3$/VO$_2$ structure is easy to realize, while the graphene/α-MoO$_3$/VO$_2$ and graphene/α-MoO$_3$/graphene/VO$_2$ structures can provide richer physical modes.

![Fig. 7. The effects of the geometric parameters on the SE rates of the α-MoO$_3$/VO$_2$ structure. [(a), (b)] the distance $d_s$, [(c), (d)] the α-MoO$_3$ thickness $t$, [(e), (f)] the propagation plane angle $\phi$, keeping the other geometric parameters $d_l = 100 \text{ nm}$, $t = 150 \text{ nm}$, and $\phi = 0^\circ$ unchanged.]

![Fig. 8. SE rates and the ratio of SE rate of the α-MoO$_3$/VO$_2$ structure with and without adding graphene layers: (a)–(d) graphene/α-MoO$_3$/VO$_2$ structure, (e)–(h) graphene/α-MoO$_3$/graphene/VO$_2$ structure, with $d_l = 100 \text{ nm}$, $t = 150 \text{ nm}$, $\mu_g = 0.3 \text{ eV}$.]

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4. Conclusion

In summary, a theoretical platform is proposed to actively tune anisotropic light–matter interaction in biaxial HM $\alpha$-MoO$_3$ using phase change material VO$_2$ and graphene. For the $\alpha$-MoO$_3$/VO$_2$ structure, the hybridization PhP dispersion modes in $\alpha$-MoO$_3$ exhibit obvious in-plane anisotropic behaviors, and the hybridization PhPs in $\alpha$-MoO$_3$ are greatly dependent on propagation plane angle, its hybridization PhP dispersion modes are gathering inside RB-2 and RB-3 at $\phi = 0^\circ$ and RB-1 and RB-3 at $\phi = 90^\circ$, while its hybridization PhP dispersion modes simultaneously occur in all RBs and just span the part frequency range of RBs at $\phi = 45^\circ$. Meanwhile, the insulator-to-metal phase transition of VO$_2$ has a significant effect on the hybridization PhPs of $\alpha$-MoO$_3$/VO$_2$ structure, and allows to realize an active tunable $\alpha$-MoO$_3$ PhPs, especially at $\phi = 90^\circ$. Furthermore, as different graphene layers are added at the top or bottom of $\alpha$-MoO$_3$ in $\alpha$-MoO$_3$/VO$_2$ structure, actively tunable HPPPs inside its RBs and SPPPs outside the RBs are realized via the coupled function between $\alpha$-MoO$_3$ PhPs and graphene SPPs. In addition, actively tunable SE enhancement of $\alpha$-MoO$_3$ can be realized via controlling the VO$_2$ phase transition and the graphene chemical potential. The high susceptibility of PhPs of $\alpha$-MoO$_3$ opens new possibilities for potential applications in combination with strongly correlated quantum materials including PCMs and graphene.

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