

CIRCULARLY POLARIZED THERMAL RADIATION IN α -MoO₃/ β -Ga₂O₃ TWISTED LAYERS

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ABSTRACT. We numerically investigated the possibility to obtain circularly polarized infrared thermal emission from a bilayer scheme taking advantage of the strong anisotropy of low symmetry materials such as β -Ga₂O₃ and α -MoO₃. Our results show that it is possible to achieve a high degree of circular polarization over 0.85 at two typical emission frequencies related to the excitation of β -Ga₂O₃ optical phonons. Our simple but effective scheme could set the basis for a new class of lithography-free thermal sources for IR bio-sensing.

1. INTRODUCTION

Narrowband, highly directional, and polarization tunable infrared (IR) sources are required for several applications ranging from IR sensing [1,2] to biomedical diagnostics and target detecting [3,4] as well as for the realization of integrated IR photonics [5,6]. Besides the availability of the highly efficient but expensive quantum cascade laser (QCL) IR sources, a lot of effort has been recently spent to propose inexpensive alternatives by taking advantage of thermal radiation sources. Thanks to the development of micro/nano technologies [7], several approaches based on gratings [8], metamaterials [9,10], metasurfaces [11] and nanoantennae [12,13] have been studied and realized to control and tailor the typically incoherent and broadband thermal radiation from heated bodies. The mechanism at the base of this tailored emission is the excitation of resonant states i.e. surface plasmon-(SP) or phonon-(SPh) polaritons, Bloch surface waves, magnetic polaritons, to name a few. However, all these approaches rely on the use of lithographic techniques to artificially create absorption/emission resonances as well as a strong birefringent effective response. In particular, birefringence is necessary for polarization sensitive applications.

An extreme form of birefringent behavior is called hyperbolicity and it is achieved when one diagonal element of the material dielectric tensor is negative, whilst the others are positive [14]. Hyperbolic metamaterials obtained by periodic sub-wavelength patterned surfaces have been studied for enhanced thermal radiation and nanoscale heat transfer [15,16]. Recently, several van der Waals (vdW) materials have attracted considerable attention because of their natural hyperbolicity in the IR [17]. Among vdW materials, hexagonal boron nitride hBN is one of the most investigated [18]. The hBN exhibits two different Reststrahlen bands (negative values of the real part of the permittivity) along either the in-plane

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or out-of-plane directions. Nevertheless, it does not display in-plane anisotropy thus it cannot be used to distinguish orthogonal polarizations states of the electromagnetic field at normal incidence.

Among the vdW polar materials displaying in-plane anisotropy, molybdenum trioxide (α -MoO₃) has been the object of a conspicuous number of recent studies [19,20]. Its strong anisotropy, allowing in-plane and off-plane hyperbolicity, has been used to obtain mid-IR waveplates [21] and tunable [22] and broadband [23] absorption in the far-field. Concerning the excitation of surface waves, hyperbolic phonon polaritons have recently been shown in α -MoO₃ flakes from 818 cm⁻¹ to 974 cm⁻¹ [20,24]. Furthermore, the excitation of SPh polaritons waves in twisted flakes of 2D α -MoO₃ has been proposed and experimentally verified [25]. Such control of SPh polaritons in twisted flakes has interesting applications for tunable near-field radiative heat transfer [26] by adjusting the tilt angle between the receiver and the emitter. It has been shown that twisted α -MoO₃ layers can exhibit circular dichroism [27,28] and can be used to generate spin thermal radiation [29]. However, using twisted structures of the identical anisotropic materials to achieve spin thermal radiation has more restrictions on the materials. For this reason, α -MoO₃ was combined with an ideal quarter wavelength plate [29]. However, a tunable combination of two real anisotropic materials (i.e. controlling the tilt angle between them) could be adopted for practical applications and for integrated thermal sources.

Low-symmetry materials have recently emerged as possible candidates for anisotropic optical applications [30]. Among them, β -Ga₂O₃ [31,32] has been used to experimentally demonstrate the excitation of shear phonon polaritons in the IR [33,34]. Moreover, this material perfectly matches the usable wavelength range of α -MoO₃.

Here we propose a combination of two twisted layers obtained from these two strongly anisotropic materials. Our calculations, based on a complete Stokes parameters analysis [35] of emitted radiation, show that it is possible to obtain circularly polarized thermal radiation in the mid-IR with a twisted bilayer system having about two microns of total thickness. As a starting point we study the emission properties and the birefringence behavior of single layers of α -MoO₃ and β -Ga₂O₃ on gold substrates. After the choice of one of the two materials as the emitter, we add a properly tilted and sized layer of the other, acting as a quarter wave plate to obtain thermally radiated circularly polarized light.

2. RESULTS

In order to quantitatively study the degree of polarization (*DoP*) and the degree of circular polarization (*DoCP*) of the thermal radiation by our system we use a characterization approach based on the evaluation of the Stokes parameters. For reciprocal materials the Stokes parameters can be expressed in terms of the polarized emissivities as [35,36]:

$$\begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \frac{S_{0,bb}(\omega, k_{\parallel})}{2} \begin{bmatrix} \epsilon_p + \epsilon_s \\ \epsilon_p - \epsilon_s \\ \epsilon_{45^\circ} - \epsilon_{135^\circ} \\ \epsilon_R - \epsilon_L \end{bmatrix}; \quad (1)$$

where $k_{\parallel}$ is the wave vector projection onto the surface (x-y) plane, $S_{0,bb}$ is the emissivity of the black body, $\epsilon_{p,s}$ are the *p*- and *s*- polarized relative emissivities, $\epsilon_{45^\circ, 135^\circ}$ are the 45°- and 135°- polarized relative emissivities and $\epsilon_{R,L}$ are the *right*- and *left*- circular polarized relative emissivities respectively. Limiting our discussion to reciprocal media at thermal equilibrium, polarized emissivities can be evaluated by calculating the polarized absorptivities α , according to Kirchhoff's law:

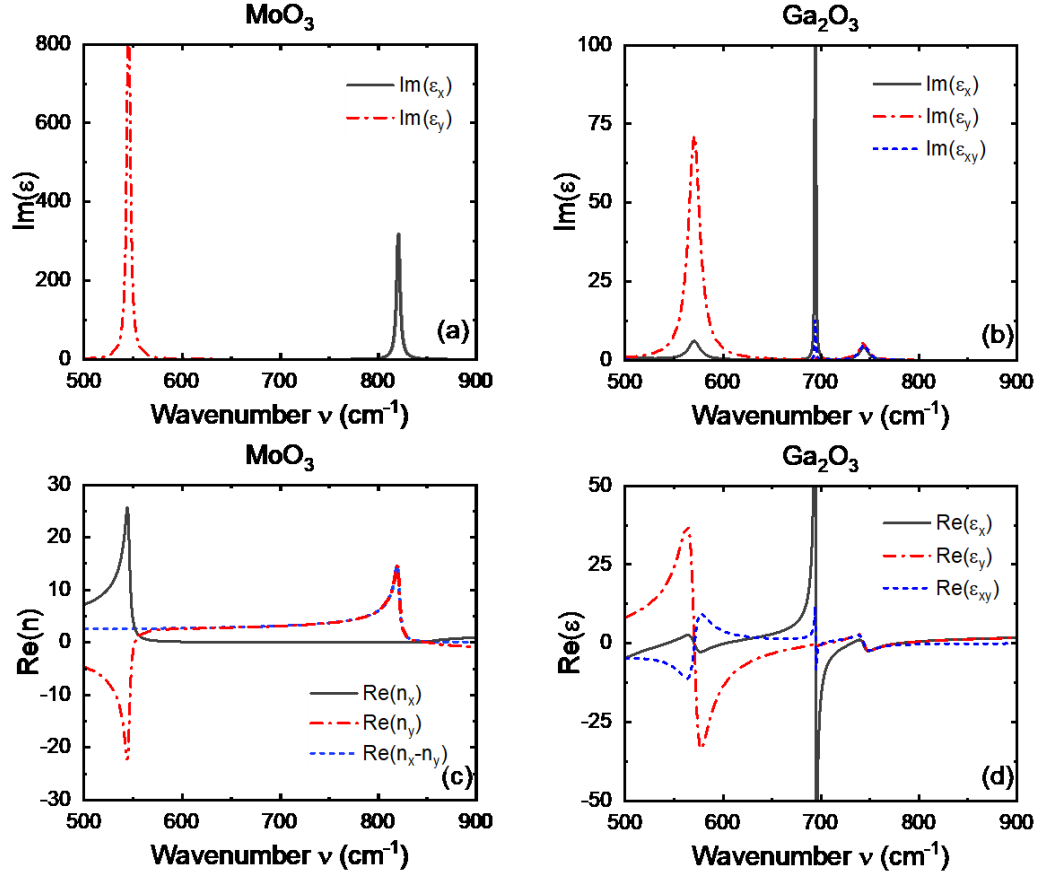


Figure 1. Imaginary part of the in-plane components (xx, xy, yy) of the dielectric permittivity tensor of (a) MoO_3 , (b) Ga_2O_3 . Real part of the refractive indices along x (n_x) and y (n_y) directions and Δn for (c) MoO_3 and (d) Ga_2O_3 .

$$\epsilon_{p,s,45^\circ,135^\circ,R,L} = \alpha_{p,s,45^\circ,135^\circ,R,L} \quad (2)$$

Polarized absorptivities have been retrieved by evaluating $1-R-T$ where R is reflectance and T is transmittance evaluated for the different polarized states of the incident light with a 4×4 transfer matrix method [37]. The DoP and $DoCP$ are then defined as:

$$DoP = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0}; \quad (3)$$

$$DoCP = \frac{|S_3|}{S_0}; \quad (4)$$

Being both DoP and $DoCP$ limited in the range $[0,1]$ and $DoCP \leq DoP$. $DoP=1$ stands for perfectly polarized light, thus $DoCP=1$ corresponds to perfectly circularly polarized light.

Our approach is based on the combination of two layers on top of gold substrate. The bottom layer acts as an emitter while the second acts as a quarter-wave plate. Both layers are required to be strongly anisotropic, however the emitting layer should provide high emission/absorption efficiency for only one polarization component while the upper one should be as transparent as possible with a strong anisotropic real part of the refractive index. Our numerical method takes into account for multiple reflections, nevertheless the optimization procedure is performed at fixed wavelength, starting with a peak in the DoP from the emitting layer. We expect that they could play a relevant role in multilayer stacks, here we only focus on the double-layer scheme. Further refinements will be taken into account in later work.

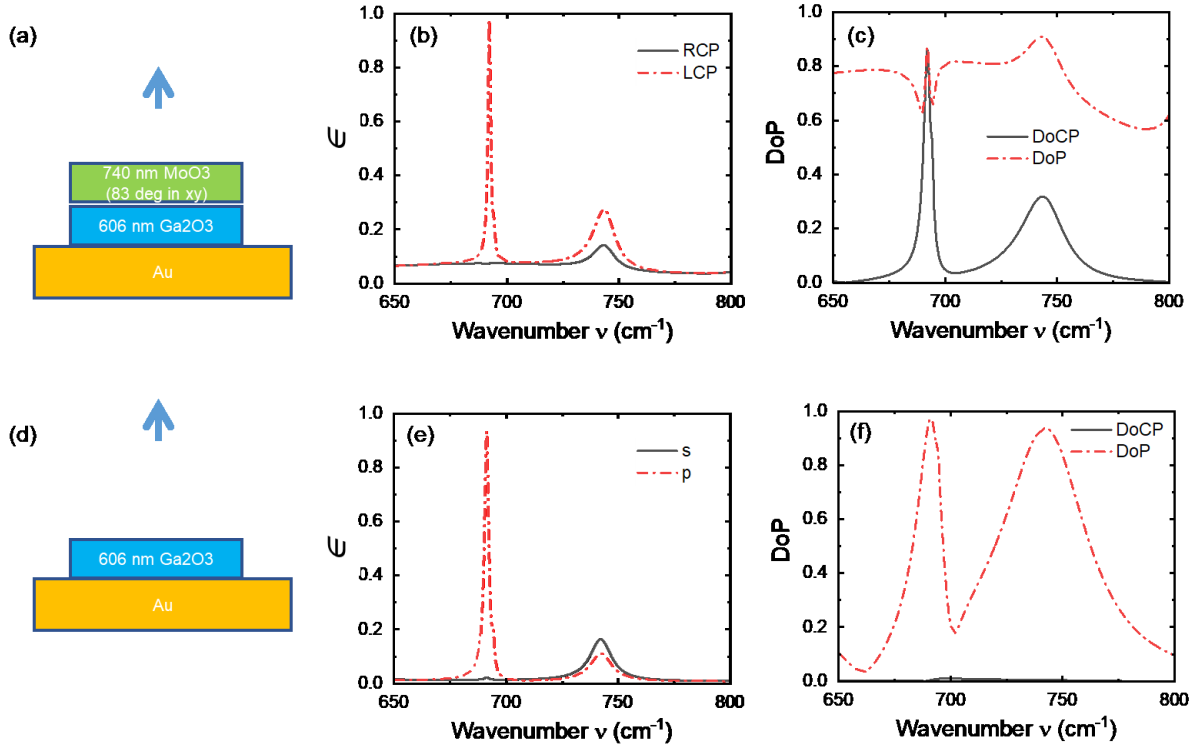


Figure 2. (a) Schematic of twisted bi-layer structure designed for $\nu=691 \text{ cm}^{-1}$, obtained by adding a α -MoO₃ layer on top of the β -Ga₂O₃ layer; (b) circularly polarized emissivity and (c) degree of polarization of the twisted bi-layer structure; (d) schematic of single β -Ga₂O₃ layer as the emission source with its (e) linearly polarized emissivity and (f) degree of polarization.

As a first step we study both α -MoO₃ and β -Ga₂O₃ optical properties in the IR with the aim to identify the frequency ranges where they can be efficiently combined. We limit our study to the thermal radiation emitted along the normal direction to the surface. Thus, if we consider the x - y plane as the surface plane we look for emission along the z -axis. We also select the orientation of the crystals so that x - and y - axis coincides with the x' - and y' - crystal axes. In this case, we are interested in the in-plane values of the dielectric permittivities. In figure 1a-b we report the values of the imaginary part of ϵ_{xx} , ϵ_{yy} and ϵ_{xy} components of the dielectric permittivity tensor of α -MoO₃ and β -Ga₂O₃ respectively. The imaginary part of the dielectric permittivity is related to absorption/emission. We note that β -Ga₂O₃ has three main bands of emission around 570 cm^{-1} , 690 cm^{-1} and 740 cm^{-1} corresponding to optical phonons excitation. Similarly, α -MoO₃ has two main emission peaks at 550 cm^{-1} and 820 cm^{-1} . As previously mentioned, in our emitter/waveplate approach we want to avoid overlaps in the emission bands in order to clearly distinguish the role of the emitter from the role of the waveplate. For this reason, we exclude from further investigation the lower frequencies (570 cm^{-1} and 550 cm^{-1}). In figure 1c-d we show the real part of the refractive index components n_x and n_y for both materials and their $\Delta n = n_x - n_y$. We note that α -MoO₃ has a wide band (from 600 cm^{-1} to 750 cm^{-1}) of high and almost constant anisotropy which makes it very suitable to be used as a wave-plate. Thus we focus our attention on the design of an efficient β -Ga₂O₃ emitting layer around 690 cm^{-1} and 740 cm^{-1} with a high DoP combined with a properly tilted and sized α -MoO₃ layer acting as wave-plate to obtain a tailored thermal source with optimized $DoCP$. We employed particle swarm optimization (PSO), a search algorithm that finds optimal solutions by iteratively trying to improve candidate solutions, and finally obtained structures with highest circular emission at specific frequencies.

The first scheme is based on the emission properties of a 606 nm thick β -Ga₂O₃ layer capped with a 740 nm α -MoO₃ layer, tilted by an angle of 83° in the x - y plane with respect to the $x'y'$ axes of the β -Ga₂O₃ layer. Results depicted in figure 2 have been obtained with the indirect method, (i.e. calculating the absorptivity and applying Kirchhoff's law) and validated by the direct method based on fluctuation electrodynamics [35]. Figure 2e shows a high peak of almost linearly polarized emission from the β -Ga₂O₃ layer on top of gold substrate (sketched in figure 2d) at $\nu=691$ cm⁻¹. The corresponding *DoP* behavior is reported in figure 2f. We note a *DoP* of about 0.97 and a zero *DoCP*. With the addition of the α -MoO₃ layer (sketched in figure 2a) we obtain *DoP*=*DoCP*=0.87 at nearly the same frequency (see figure 2c). According to figure 2b it means that the thermal emission at 691 cm⁻¹ is left circularly polarized with a *DoCP*=0.87. Multiple reflections at every interface are responsible for the slightly frequency shift of the bi-layer with respect to the emission peak of the single layer.

We finally propose a second scheme to obtain circularly polarized thermal emission at $\nu=737$ cm⁻¹. In this case we need a thicker slab of β -Ga₂O₃ to reach high polarized emissivity (figure 3e) and a value of *DoP* of 0.93 (figure 3f). Circular polarization is then achieved by adding a 324 nm thick layer of α -MoO₃ as sketched in figure 3a, rotated in the x - y plane of 114° with respect to the $x'y'$ axes of the β -Ga₂O₃ layer. We note that in this case we obtain a resulting *DoP* = *DoCP* = 0.85 (figure 3c) and the thermal emitted radiation at 737 cm⁻¹ is mostly left circularly polarized (figure 3b). In both cases, right-hand circular polarization can be obtained with the same scheme, by adjusting the tilting angle of the MoO₃ layer.

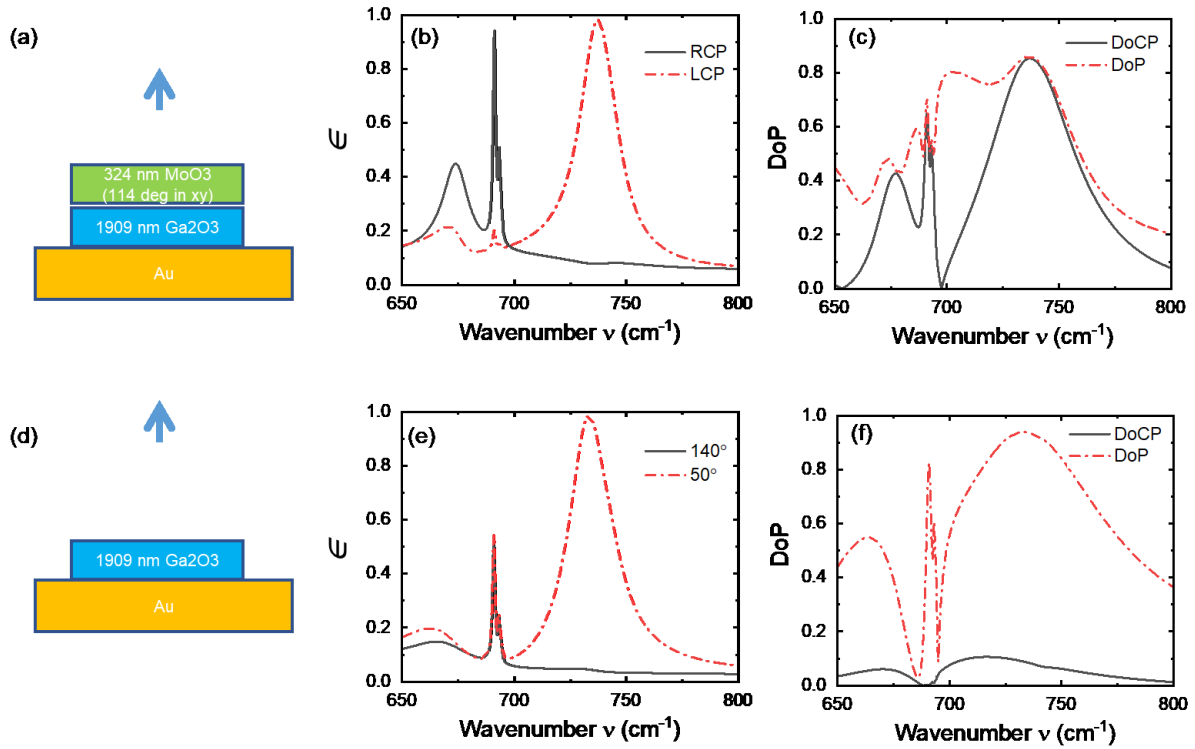


Figure 3. (a) Schematic of twisted bi-layer structure designed for $\nu=737$ cm⁻¹, obtained by adding a α -MoO₃ layer on top of the β -Ga₂O₃ layer; (b) circularly polarized emissivity and (c) degree of polarization of the twisted bi-layer structure; (d) schematic of single β -Ga₂O₃ layer as the emission source with its (e) linearly polarized emissivity and (f) degree of polarization.

3. CONCLUSION

We proposed a lithography-free method based on a double-twisted layer scheme to obtain circularly polarized thermal radiation in the mid-IR, at two specific frequencies related to β -Ga₂O₃ optical phonons excitation. The almost linearly polarized emission from the single slab is converted into circular polarization by adding a properly sized and tilted α -MoO₃ layer acting as a quarter-wavelength plate. In both cases, we take advantage of the strong natural anisotropy related to the low symmetry class of the investigated materials without the need of further processing techniques. The achieved degree of circular polarization is higher than 0.85 for the two proposed schemes. We believe that this approach could lead to the development of low-cost, integrated thermal sources of circularly polarized light for IR sensing applications.

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