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Near-infrared tailored thermal emission from wafer-scale continuous-film resonators

Alexander S. Roberts,1,∗ Manohar Chirumamilla,2 Kasper Thilsing-Hansen,1 Kjeld Pedersen,2 and Sergey I. Bozhevolnyi1

1University of Southern Denmark, Institute of Technology & Innovation, Niels Bohrs Alle 1, 5230 Odense, Denmark
2University of Aalborg, Department of Physics and Nanotechnology, Skjernvej 4, 9220 Aalborg, Denmark
3University of Southern Denmark, Mads Clausens Institute, Alsion 2, 6400 Sønderborg, Denmark
∗asro@iti.sdu.dk

Abstract: We experimentally investigate the near-infrared emission from simple-to-fabricate, continuous-film Fabry-Perot-type resonators, consisting only of unstructured dielectric and metallic films. We show that the proposed configuration is suitable for realization of narrowband emitters, tunable in ranges from mid- to near-infrared, and demonstrate emission centered at the wavelength of 1.7 µm, which corresponds to the band gap energy of GaSb-based photodetectors. The emission is measured at 748 K and follows well the emissivity as predicted from reflection measurements and Kirchhoff’s reciprocity. The considered emitter configuration is spectrally highly tunable and, consisting of only few unstructured layers, is amenable to wafer-scale fabrication at low cost by use of standard deposition procedures.

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The fields of actual and potential application of thermal radiation emission and detection are conceivable system, allowing for sensing, spectroscopic analysis or energy transport as such. To all ordinary matter, it provides an energy transfer and loss mechanism available in most meta-materials as selective thermal emitters [12–14].

1. Introduction

Progress in the tailoring of thermal emission spectra in the infrared by both nano-structured surfaces and thin-film stacks serving as gratings [1–3], photonic crystals [4–8] or resonators [9–11], has been widely reported in recent years. As thermal emission is a process inherent to all ordinary matter, it provides an energy transfer and loss mechanism available in most conceivable system, allowing for sensing, spectroscopic analysis or energy transport as such. The fields of actual and potential application of thermal radiation emission and detection are therefore correspondingly diverse, spanning from industrial and botanical control [12–14] to...
Fig. 1. (a) Working principle of the continuous resonator. Freely propagating waves can couple to the resonator through the optically thin top layer. Inside the resonator, photonic modes are confined in the vertical direction, acquiring reflection and propagation phases, $\phi_{r,\text{top}}$, $\phi_{r,\text{bottom}}$, and $\phi_{\text{prop}}$, respectively, as indicated in the right part of (a), creating a resonance when the round-trip phases equal multiples of $2\pi$. (b) Scanning electron micrograph (side view) of a continuous-film Fabry-Perot emitter detached and elevated above its substrate. Note that the 12 nm top gold layer is not well-resolved. (c) The structure of (b), showing the amorphous SiO$_2$-layer (blue), the optically thick bottom gold layer and the optically thin top gold layer (yellow). Not shown are adhesion-promoting layers of 3 nm Ti at the SiO$_2$-gold interfaces.

thermophotovoltaic energy conversion [15–17], amongst others. While nanostructuring of surfaces constitutes a very powerful tool for the tailoring of optical properties, it often requires expensive and slow fabrication techniques, hence calling for simpler structures with which to tailor the optical properties of surfaces. In this work, we present tailored thermal emission from an unstructured few-layer resonator, that is both easily fabricated and suited for the controlled tailoring of thermal near- and mid-infrared narrowband emission, see Fig. 1. The emitter conceptually consists of two gold films of different thicknesses, which are spaced by an amorphous silicon dioxide layer. The thicknesses of the gold films are such that the bottom film is fully reflecting while the top film is semitransparent and so ensures that the resonator modes couple to freely propagating waves. We design the emissivity to coincide with the band gap energy of GaSb, a low band gap semiconductor material often used for thermophotovoltaic (TPV) conversion. Its band gap energy is 0.7 eV for an unstrained material, corresponding to a vacuum wavelength of roughly 1.7 $\mu$m.

The structure was recently investigated by Yan [18], at a resonance wavelength of 1 $\mu$m, and by Zhao et al. [19] in the visible, with regard to its cold optical properties and was found to exhibit sharp resonances that are tunable in resonant wavelength, through variation of the spacer thickness. The line width and maximum absorption are determined primarily by the thickness of the top gold layer. A conceptually closely related structure, where a Bragg reflector acts as the top reflector, can be found in [20]. Previously, Wang et al. have measured thermal emission from a very similar structure, however at energies too small for photovoltaic conversion [21]. Here, we investigate the thermal emission from this structure at 1.72 $\mu$m - that is at wavelengths with direct relevance in thermophotovoltaic conversion. As the emitter requires no layer-structuring and, moreover, can be fabricated in a variety metals and dielectrics, it has the potential for cost-effective production scaling of thermal emitters for large-scale illumination of photovoltaic cells in TPV applications.

2. Thermal emission and Kirchhoff’s reciprocity

In thermodynamic equilibrium, the thermal emission of a totally absorbing and opaque object, a blackbody, is described by the radiation law first derived by Planck [22], that relates wavelength and temperature of a blackbody to its spectral radiance, $I$, which quantifies the emitted power

\[ I(\lambda, T) = \frac{2\pi h c^2}{\lambda^5} \left( \frac{1}{e^{h c / \lambda k T} - 1} \right) \]

where $h$ is Planck’s constant, $c$ the speed of light, $k$ the Boltzmann constant, and $T$ the temperature in Kelvin. Kirchhoff’s law states that the spectral radiance at a given wavelength is equal to the emissivity of the material at that wavelength.
per unit area, wavelength, and solid angle. A blackbody is an idealized object; the emission of any real object is further described by their emissive power or emissivity, $\varepsilon_{\lambda}(T)$, which quantifies the spectral radiance of an object, relative to that of a blackbody. The spectral radiance of an object with emissivity $\varepsilon_{\lambda}(T)$, can thus be expressed as

$$I(\lambda, T) = \varepsilon_{\lambda}(T)B(\lambda, T) = \varepsilon_{\lambda}(T) \cdot \frac{2hc^2}{\lambda^5} \cdot \frac{1}{e^{\frac{hc}{kBT}} - 1},$$  

(1)

where $\lambda$ is the vacuum wavelength, $T$ is the temperature of the object, $h$ is Planck’s constant, $c$ is the vacuum speed of light and $k_B$ is Boltzmann’s constant. The wavelength of maximum spectral radiance of the blackbody spectrum $\lambda_{\text{max}}$, is given by Wien’s displacement law, which can be expressed in the following reciprocal relationship between $T$ and $\lambda_{\text{max}}$:

$$\lambda_{\text{max}}(T) = \frac{b}{T} = 2898 \, \mu mK/T,$$

(2)

where $b$ is Wien’s displacement constant. Optimizing the efficiency of the emitter, defined as the power emitted around the design wavelength divided by the total power emitted by a blackbody at the same temperature, therefore, requires heating the sample to a temperature where $\lambda_{\text{max}}$ and the resonance wavelength become comparable [23].

Due to the broad spectrum of the blackbody radiation, efficient technological utilization of thermally generated emission is often challenging, apart from the rather obvious case of radiative cooling. However, for many other applications, it is desirable to emit or detect radiation only within a particular frequency band, determined by the characteristic energies of the processes involved. As the blackbody spectrum depends only on temperature and is inherently broadband, the emissivity must be tightly controlled to achieve narrowband thermal emission. Through Kirchhoff’s reciprocity, which equates emissivity and absorptivity, $\alpha_{\lambda} = \varepsilon_{\lambda} = 1 - R_{\lambda} - T_{\lambda}$, thereby relating absorptivity $\alpha_{\lambda}$ and emissivity $\varepsilon_{\lambda}$ to transmissivity $T_{\lambda}$ and reflectivity $R_{\lambda}$ [24,25]. The relation $\varepsilon_{\lambda} = 1 - R_{\lambda}$ is readily derived for non-transmitting objects (Temperature dependence is suppressed for readability). Consequently, a narrowband absorbing surface functions as a thermal narrowband emitter with a relatively high degree of temporal coherence, when heated to sufficient temperatures.

3. Fabrication and tunability of spectral properties

The emitter is fabricated on a polished silicon (p-type, c-Si (100)) substrate by electron-beam evaporation of 90 nm of gold at an evaporation rate of 0.3 Å/s, subsequent radio-frequency sputtering of the spacer layer consisting of amorphous SiO$_2$ at a rate of 0.5 Å/s and finally by electron-beam evaporation of a 12 nm gold top layer at 0.3 Å/s. Adhesion-promoting titanium layers of 3 nm thickness are evaporated between all layers at 0.3 Å/s. All deposition steps are performed in a continuously evacuated chamber at pressures of $10^{-5}$ mbar or less. The thickness of the stacked layers is characterized independently by spectroscopic ellipsometry. Figure 1(a) shows a schematic and a scanning electron micrograph of the resulting continuous-film Fabry-Perot resonator. In contrast to the optically thick bottom layer of gold acting as a reflector, the top gold layer functions as a semi-transparent mirror by choosing its thickness to be comparable to the skin depth of light. The partially reflecting film allows for coupling of the lossy resonator modes to freely propagating waves, leading to absorption at wavelengths near resonance. The spectral position of the resonance is determined by the condition that the round-trip phase equals integer multiples of $2\pi$ at resonance:

$$\phi_{r, \text{bottom}} + \phi_{r, \text{top}} + 2 \cdot \phi_{\text{prop}} = 2m\pi,$$

(3)

for integer $m$. Appropriate geometric parameters are found by reflectivity measurements, see Fig. 2. As a result of Eq. (3), the resonance frequency can be tuned over most of the infrared
Fig. 2. Schematic of the setup used for emission and reflection measurements. The sample is heated in a temperature-controlled microscope stage (HC). For the measurement of blackbody reference spectra, the heated sample cavity (HC) is replaced with a reference blackbody cavity with W1 mounted to the cavity opening. Reflection of sample and silver mirror reference is measured outside of the HC to avoid reflections from window W1, while the frequency shift is monitored inside HC. The numerical aperture (NA) of illumination is controlled with irises AS and FS, while in emission the NA and the area from which light is picked up, are controlled by irises CI and OI.

The depth of the resonance is determined mainly by the thickness of the top gold layer which, to a lesser extent, also modifies the resonance position, as the reflection phase $\phi_{\text{r, top}}$ depends on the thickness of the top layer. Moreover, the structure is flexible in terms of material choices, allowing for use in a much broader range of both temperatures and wavelengths than presented here.

### 4. Thermal emission measurements

Thermal emission is measured by heating the sample in a microscope heating stage (Linkam TS-1000) while picking up the generated thermal radiation through a long-working distance objective which is fibre-coupled to a near-infrared spectrometer (OceanOptics, NirSpec), see Fig. 2 for details. For the calculation of the emissivity, it is necessary to divide the thermal emission spectrum by the blackbody spectrum at the same temperature, which simultaneously eliminates the setup dispersion from the resulting emissivity spectrum. The blackbody in use is a high absorptivity cavity-type (Electro-Optical Industries, with a surface extinction specified at 0.97 – 0.99 at wavelengths from 500 nm to 20 $\mu$m), and spectra are taken after allowing thermal stabilization for at least 30 min. Temperatures are ramped with 0.5 K/s under both heating and cooling. A quartz window identical to W1 (Fig. 2) is mounted to the reference cavity to guarantee identical optical paths and dispersion. Reflection measurements are referenced against a silver mirror with a reflectivity of at least 97.5 % in the wavelength range investigated (Thorlabs, PF10-03-P01). Frequency shifts occurring during heating are evaluated by measuring the reflection spectra at each temperature point (also comprising thermal emission that unrelated to the reflected light), from which we subtract the generated emission. This procedure allows for the tracking of the shift in resonance frequency, as shown Fig. 4, inset. It can be seen that
Fig. 3. (a) Dependence of the reflectivity on the top gold layer thickness, for a spacer thickness of 472 nm. (b) Dependence of the reflectivity on spacer layer thickness for a top layer thickness of 12 nm. It is obvious that the emitter offers a high degree of tunability. Variation of the spacer thickness influences mainly on the position of the resonance and, to a lesser extent, also the position of the resonance through modification of the reflection phase $\phi_{r, \text{top}}$.

the observed reflectivity wavelength shift corresponds exactly to the wavelength shift observed between peaks in room temperature reflectivity and heated emissivity. It should be noted that ambient thermal emission that is reflected by the sample into the setup is not taken into account; it is negligible at the temperatures investigated here, as is clear from Eq. (1).

Figure 4 shows emission spectra taken at 673, 723 and 748 K from an emitter fabricated with a resonance at 1.72 $\mu$m at room temperature. As seen in Fig. 4(a), the emission rises rapidly with increasing temperature, which, assuming temperature-independent optical properties, is wholly attributable to the change of the blackbody spectrum. At 723 K and 748 K the emission from the emitter exhibits a clear peak at the resonance, where the blackbody emission, in contrast, continues to increase. The resulting emissivity $\varepsilon_\lambda$ is shown in Fig. 4(b). Clearly, the emissivity closely matches the absorption, approximated as the measured extinction, $1 - R_\lambda$. The observed emissivity peak is somewhat broader than the extinction peak and slightly lower in maximum emissivity. Both broadening and redshift might be explained by material temperature dependent behavior, since it is known the dielectric function $\varepsilon$ of gold (and other metals) is temperature dependent with the imaginary part of $\varepsilon$ increasing with increasing temperature, while at the same time, the reflectivity decreases [26–29], leading to a lower Q-factor and broader peak. Furthermore, thermal expansion might explain the red-shift. However, experimental factors could also play a role such as illumination conditions and emission measurement conditions not being perfectly matched (Fig. 2 illustrates such a situation where neither NA or illuminated sample area are matched) or scattering caused by surface roughness which can cause extinction in reflection measurements that is not associated with absorption, but with light being scattered out of the observed solid angle. Consequently, to minimize extinction in reflection measurements arising from scattering, it is necessary to pick up light with the largest NA possible. Thus, the extinction is not exclusively dominated by absorption as assumed by $\varepsilon_\lambda = \alpha_\lambda \approx 1 - R_\lambda$. The inset in Fig. 4 shows the temperature dependence of the resonance to be very low, amounting to a maximum shift of the resonance of less than 7 nm at 748 K, when compared to room tem-
Fig. 4. Thermal emission measurements. (a) Emission from sample (solid) and blackbody (dashed) at temperatures 673, 723 and 748 K, respectively. It should be noted that intensity measurements incorporate the setup dispersion, leading to an apparent peak at 2 µm. (b) Emissivity calculated from spectra in (a) and extinction measurements (estimated as 1-R) showing an excellent match, in agreement with Kirchhoff’s reciprocity. Inset: The total frequency shift of the resonance is roughly 6 nm and has a low degree of hysteresis.

5. Thermal stability

Degradation of the structure plays no role for short-term heating procedures at 748 K but starts to set in at even slightly higher temperatures. This degradation cannot be expected from inspection of material properties, with the bulk melting point of all materials being at least several hundred degrees Celsius higher than the temperatures investigated in this context. However, the melting point of finite materials with dimensions on the order of the grain sizes or impure materials, is known to be influenced by melting point depression [30–32], which arises both due to diffusion of impurities and due to the presence of surface lattice-sites. Both effects induce local variations in the lattice energy, causing melting to set in at lower temperatures than for the corresponding bulk material. Furthermore, the phase change occurs gradually over a range of temperatures that corresponds directly to the spread in lattice energies. As a result, temperature instability of the sample sets in well below the bulk melting point of Au. The degradation of the sample under excessive heating is shown in Fig. 5, where samples heated to 823 and 923 K are seen. The temperatures are ramped as in the emission measurements (0.5 K/s), while samples are annealed for 60 minutes. The extent of the damage caused by heating varies heavily, depending on the homogeneity of the layers. For a relatively smooth sample, only the thin top layer is damaged at 823 K (Fig. 5(d)), in marked contrast to samples with considerable surface roughness, in which case all layers are damaged, (Fig. 5(b)). It should be noted that for a combination of sufficiently high temperatures and a high degree of roughness, no equilibrium or metastable state is reached. The effect of further annealing is then to drive further changes to
Fig. 5. Scanning electron micrographs of heated samples after cooling. (a)-(b) and (c)-(e) show different sample batches, respectively, highlighting the influence of fabrication imperfections. (a) Sample A before heating. Several imperfections can be seen on the sample surface. (b) Sample A heated to 823 K, showing a complicated multitude of phase changes, cracks in the spacer layer and wrinkling of the bottom gold layer. (c), (d) Heated emitter of a fabrication batch with improved surface roughness. (c) The number of imperfections in the unheated sample are visibly reduced compared to (a). (d) Identical to c, heated to 823 K. This sample shows degradation of the top layer only, which has transformed into distinct particles. (e) Identical sample to (c), heated to 923 K, where the SiO$_2$-layer starts to fracture.

structure - this is the case in Fig. 5(b), while samples similar to Figs. 5(c)-5(e) mostly show little to no obvious structural changes after initial degradation has occurred. We anticipate that the presented structure will be further optimized for the TPV use at temperatures given by Wien’s Law, by using combinations of refractory materials [33]. This would also eliminate the need for adhesion layers and thus decrease the potential for inter-diffusion and the entailing changes in optical properties, such as increased losses [34, 35], and melting point depression. For use in the mid-infrared, however, no further modifications are necessary; an inspection of Wien’s law (Eq. (2)) reveals that $\alpha_{\text{max}}$ lies at a wavelength of $\lambda_{\text{max}} \approx 3.9 \ \mu$m at a working temperature of 748 K.

6. Conclusion

In conclusion, we have demonstrated tunable thermal narrowband emission from a simple, unstructured few-layer structure, exhibiting a resonance at the band gap energy of GaSb. The spectral properties can be modified in terms width, depth and resonance frequency through straightforward variation of fabrication parameters. The emitter is suitable for scaling to large-area fabrication and transferable to other combinations of materials, as the emitter is unstructured and relies only on standard film deposition procedures. Given the thermal stability of the sample, highly efficient emission can be achieved at wavelength of 3.9 $\mu$m, or longer, and owing to the simple physical principles of the resonance, there are large degrees of freedom concerning material combinations and geometry, which can be chosen to suit specific applications.
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