Switchable Purcell enhancement of photoluminescence by GST film

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Abstract. In the present paper perovskite radiation enhancement on crystalline GST film compared to amorphous one has been studied. The photonic local density of states has been calculated by angular spectrum representation of the dyadic Green’s function. The Purcell factor has been calculated for perovskite luminescent on both amorphous and crystalline GST film. Almost 80% enhancement has been observed at wavelength 950 nm for system with perovskite thickness 25 nm, GST thickness 110 nm.

1. Introduction

Halide perovskites are widely studied as an emergent material for optoelectronic and photovoltaic applications. Comparing with analogs which are used today the perovskites offer ease of processing using solvent methods such as spin-coating or blade/slot-die coating. The energy gap of perovskites is altered by variation of halogen anion composition (Cl, Br and I), which allows tuning emission of the perovskite-based device over the whole visible spectrum [1].

Dielectric environment is known to influence the rate of emission centers. This effect was described by E.M. Purcell by considering a factor which is equal to a ratio of emission rate in environment normalized to emission rate of the same center in a homogeneous space [2]. The Purcell effect causes the luminescent enhancement in vicinity of metallic substrate because of induced surface plasmons [3]. However the interface between halide perovskite and metal leads to strong chemical degradation [4] that disables such devices in practice.

During the last decade phase change materials based on germanium-stibium-tellurium alloy (GST) attract a lot of attention in photonics due to non-volatile phase transition between crystalline (with high conductivity) and amorphous (low conductivity) phases induced by optical pulses [5, 6]. Thus, GST can be used as a substrate for halide perovskite which acts as switchable dielectric environment for the emission centers properties.

Here we show enhancement of perovskite photoluminescence by switching GST substrate from amorphous to crystalline phase. We calculate photonic local density of states (LDOS) by using angular spectrum representation of the dyadic Green’s function. Further we integrate Purcell factor over distance.
between emitter and GST and uncover photoluminescence enhancement for perovskite on crystalline phase compared to amorphous one.

2. Results

In the present study we consider a layered structure composed of glass substrate, GST, perovskite and air. The scheme of system is represented in Fig. 1. The perovskite emits spherical waves propagating in both hemispheres. The waves reflect at boundaries perovskite-air, perovskite-GST and GST-glass. We assume the glass and air layers are semi-infinite in the normal direction. The GST thickness ranges from 50 to 200 nm and the perovskite thickness from 20 to 70 nm. The permittivity of perovskite is set to be 5.5 [7], the permittivity of GST is taken according to the literature data [6].

![Figure 1. Schematic of the layered structure.](image)

We calculate the local density of states as imaginary part of trace of the dyadic Green’s function [3]:

$$\rho(\mathbf{r}, \omega_0) = \frac{2\omega_0}{\pi c^2} \text{Im} \{ \text{Tr}(G(\mathbf{r}, \omega_0)) \}$$  \hspace{1cm} (1)$$

where $G(\mathbf{r}, \mathbf{r}'; \omega_0)$ is a dyadic Green function, $\omega_0$ is a frequency, $\rho(\mathbf{r}, \omega_0)$ is the photonic LDOS at position $\mathbf{r}$.

By using angular spectrum representation the problem is reduced to plane waves defined by tangential wave vector projection onto the interface plane $\mathbf{k}_r$. To consider all reflections at the boundaries it is convenient to solve the problem for individual plane waves by the approach based on transfer matrix [8]. After that we integrate plane waves over all $\mathbf{k}_r$ and obtain Green’s function for layered structure. The Purcell factor is obtained by dividing the calculated photonic LSOD to one for sample with no GST layer. Figure 2 demonstrates the value of Purcell factor of perovskite on the crystalline (red curve) and amorphous (black curve) GST film versus wavelength for structure with GST layer thickness of 90 nm, perovskite layer thickness of 25 nm. The emitter is localized at a distance 45 nm off the GST layer. The Purcell factor corresponding to amorphous GST has the maximum at wavelength 525 nm while the maximum of one for the crystalline phase is shifted 15 nm to shorter wavelengths. Note that the specific shape of the curve depends on the position of the emitter.
Figure 2. Purcell factor for perovskite on crystalline (red curve) and amorphous (black curve) GST layer. Perovskite layer thickness is 25 nm and GST layer thickness is 90 nm. The emitter is localized at the distance 45 nm from the interface with GST.

To evaluate the luminescence enhancement for the crystalline GST compared to amorphous one we integrate the local density of states over emitter position in the perovskite layer. Due to GST bandgap less than perovskite bandgap the near-surface charge carries to the GST layer and recombine in nonradiative processes. Therefore, we assume that the emission occurs due to recombination of excitons in the perovskite at a distance greater than 10 nm from the interface with GST. Figure 3 represents the results of factor Purcell calculations for perovskite thickness 25 nm and GST thickness 110 nm. For such configuration, the maximal value of Purcell factors ratio observed at the wavelength 950 nm and equal to 1.79.

Figure 3. Purcell factor for perovskite on GST film in crystalline (red curve) and amorphous (black curve) phase and its ratio (blue curve). Perovskite thickness is 25 nm and GST thickness is 110 nm.
3. Summary
To summarize, we have revealed the switchable Purcell enhancement of perovskite photoluminescence by GST films. The Purcell factor values for perovskite radiation on amorphous and crystalline GST film have been calculated. Almost 80% value of enhancement has been obtained for the structure with GST thickness 110 nm, perovskite thickness 25 nm at the wavelength 950 nm.

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References
[1] Protesescu L et al 2015 Nano Lett. 15 3692
[2] Purcell E 1946 Phys. Rev. 69 37
[3] Novotny L and Hecht B 2012 Principles of nano-optics (Cambridge: Cambridge University Press)
[4] Behrouznejad F et al. 2016 J. Mater. Chem. A. 4 13488
[5] Wuttig M and Yamada N 2007 Nat. Mater. 6 824
[6] Makarov S et al. 2017 Laser Photonics Rev. 11 1700108
[7] Yan W et al. 2020 Opt. Express 28 15706
[8] Rybin M et al. 2016 Sci. Rep. 6 1