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Perovskite-GST hybrid platform for optical memristors with complex optical response

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Abstract. This study explores the optical properties of a perovskite-GST hybrid platform, revealing a remarkable, up to twofold, increasing of photoluminescence intensity of halide perovskite thin film by switching the phase of GST substrate from amorphous to crystalline. The underlying mechanisms responsible for this strong modulation in photoluminescence intensity are related to interaction between the halide perovskite and the GST substrate, which we are investigating through comprehensive morphological and optical characterization techniques, highlighting the different reflectance properties of the crystalline and amorphous phases of GST. We consider this hybrid platform as a promising architecture for a neuromorphic system, leveraging the volatile properties of halide perovskites with the non-volatile characteristics of GST. This architecture aims to mimic the functionalities of the human brain, including the behavior of neurons and synapses, thus opening new avenues for the development of energy-efficient and highly adaptive computing systems. Our findings contribute to a deeper understanding of hybrid perovskite-GST platform and its potential in future technological applications in neuromorphic systems.

Keywords: halide perovskite, phase-change materials, volatile memory, nonvolatile memory

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Гибридная платформа перовскит-GST для оптических мемристоров со сложным оптическим откликом

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Аннотация. Данное исследование изучает оптические свойства гибридной платформы на основе перовскита и GST, демонстрируя увеличение интенсивности фотолюминесценции тонких пленок галогенидного перовскита до двух раз при переключении фазы GST с аморфной на кристаллическую. В этой работе мы фокусируемся на исследовании возможных механизмов такой модуляции и ее потенциальных применений для нейроморфных систем.

Ключевые слова: галогенидный перовскит, материалы с фазовым переходом, волатильная память, неволатильная память

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Introduction

Nowadays, artificial intelligence (AI) and machine learning are making significant progress as they can be applied to various aspects of our lives. While AI software continuously improves through machine learning techniques, it is also important to develop the technical basis of AI. Commonly, advanced AI systems use bio-inspired neuromorphic architectures that operate based on the principles of the human brain, utilizing volatile neurons and non-volatile synapses [1]. To increase the operational speed and energy efficiency of such systems, the scientific community is actively exploring novel architectures and materials suitable for this application. Hybrid platforms with halide perovskite, such as silicon-perovskite, already demonstrate high potential in optoelectronic applications, including Si-perovskite tandem solar cells and photodetectors [2, 3]. Here, we study the properties of halide perovskite in combination with germanium-antimony-tellurium (GST) phase-change alloy as it is a promising platform for neuromorphic applications. This combination possesses the energy-independent (non-volatile) properties of GST [4] and the energy-dependent (volatile) characteristic of halide perovskite [5], effectively imitating the functionality of the human brain.

Materials and Methods

$CsPbBr_3$ solution was synthesized using stoichiometric mixtures of 0.3 mmol CsBr (>99.999%) and 0.3 mmol $PbBr_2$ (> 99.9%) in dimethyl sulfoxide. The solution was prepared and stored in an inert atmosphere of nitrogen-filled glove box. Halide perovskite thin films were deposited via spin-coating method using a one-step spin-cycle with 3000 rpm to obtain a film with thickness of 80–100 nm. Amorphous GST (a-GST) films were deposited on the glass substrate by a magnetron sputtering. Switching from a-GST to crystalline GST (c-GST) phase was made by local heating of one half of the substrate on the hotplate at 200°C, which allows us to obtain substrate with a-GST and c-GST areas. Reflectance (R) and photoluminescence (PL) spectra were measured using a QE pro Ocean optic spectrometer connected to the Axio Imager.A2m (Carl Zeiss SMT) optical microscope with optical fiber. For PL measurements, optical pumping was performed using a 350 nm wavelength UV lamp. Optical pumping for time-resolved photoluminescence (TRPL) measurements was achieved using a TEMA femtosecond pulse laser with a wavelength of 350 nm, frequency of 10 kHz, and a laser fluence of 10 nJ/cm^2. Atomic force microscopy (AFM) was made on AIST-NT SmartSPM 1000.

Results and Discussion

In this work we studied the optical properties of perovskite-GST platform by comparing the R and PL spectra of $CsPbBr_3$ halide perovskite deposited on a-GST and c-GST thin film.

Fig. 1, *a* presents a photo of a 100 nm GST thin film deposited on a glass substrate. The different crystal structures of the a-GST and c-GST phases result in different optical properties. In the transmission mode, where the light source is positioned behind the sample, c-GST appears as a darker region compared to the lighter a-GST. The morphology of the thin film of $CsPbBr_3$ perovskite deposited on top of both the a-GST and c-GST phases was analyzed using AFM. The results, shown in Fig. 1, *b*, indicate no significant differences, suggesting that the GST phase does not affect the formation of the perovskite thin film. Fig.1, *c* presents an optical microscope image in reflection mode of the perovskite thin film on the GST layer, clearly divided into two regions: lighter area associated with c-GST and darker area with a-GST.

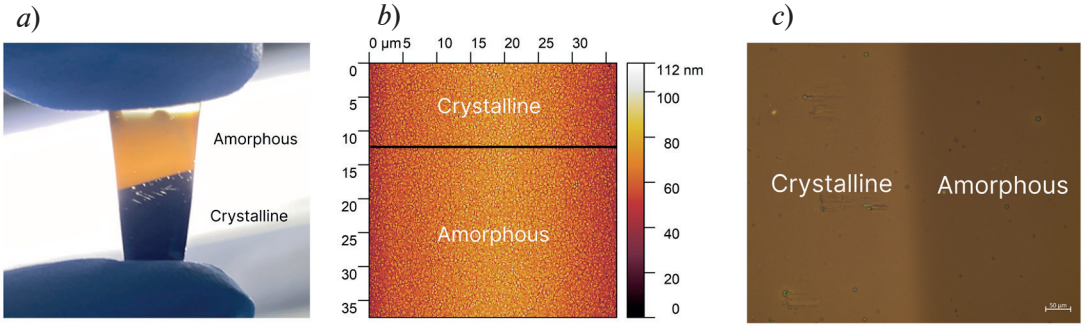

Fig. 1. Photo of 100 nm GST thin film in both phases of the glass substrate (*a*). AFM image of $CsPbBr_3$ film on top of the a-GST and c-GST (*b*). Image made by optical microscope of $CsPbBr_3$ thin film on the border of a-GST and c-GST (*c*)

In addition to morphology study and optical imaging, we measured the R spectra for both GST phases. The c-GST, with a higher refractive index due to highly ordered crystal structure, demonstrated increased reflection, as illustrated in Fig. 2, *a*. This trend remains consistent even after the deposition of the $CsPbBr_3$ thin film on top of the GST layer, as shown in Fig. 2, *b*. Notably, the reflection peak observed near 525 nm in Fig. 2, *b* corresponds to the bandgap energy of the perovskite layer, which is supported by absorption spectra measurements also presented in Fig. 2, *b*. Furthermore, the photoluminescence (PL) peak of the $CsPbBr_3$ thin film, depicted in Fig. 2, *c*, is located at the same wavelength of 525 nm, indicating that it represents band-edge photoluminescence.

Furthermore, the reflection difference of the two GST phases can influence the PL intensity of $CsPbBr_3$ thin film, as demonstrated in Fig. 2, *c*. The PL intensity of perovskite film deposited on top of the c-GST is twice higher compared to the same film deposited on a-GST. The strong PL modulation can be attributed to several factors. The primary factor is the difference in reflectance, which likely has the most significant impact. Additionally, two other important factors influence the PL intensity of halide perovskite. The first factor is Purcell effect which involves a modification of the local density of photonic states due to the GST impact. The second factor is the difference in defect density at the perovskite-GST interface between the crystalline and amorphous phases. Carriers can migrate to and localize at the defects present at this interface. Regarding the last two factors, the crystalline state is more favorable for emission enhancement [6].

To demonstrate the influence of different defect densities in the two phases, we conducted TRPL measurements on $CsPbBr_3$ perovskite film deposited on both a-GST and c-GST, as illustrated in Fig. 2, *d*. To extract the non-radiative lifetime (τ_{nr}) from the decay data plot, the ABC-model fitting was employed [7]. In this analysis, the C coefficient, which corresponds to Auger recombination, was set to zero due to the low pump fluence of 10 nJ/cm^2. The equation (1) governing the dependence of charge carrier density (n) on time (t) is presented below:

$$\frac{n(t)}{n_0} = \frac{\exp\left(-\frac{t}{\tau_{nr}}\right)}{\left(1+\tau_{nr}\left(\frac{n_0}{\tau_r}\right)\left(1-\exp\left(-\frac{t}{\tau_{nr}}\right)\right)\right)} + D, \quad (1)$$

Here, n represents the charge carrier density, t is a time, τ_{nr} denotes the trap-assisted lifetime, τ_r is a radiative lifetime, n_0 is the initial charge carrier density, and D is a constant. Based on the fitting results τ_{nr} for $CsPbBr_3$ film on c-GST was found to be 14.58±0.2 ns, while for the film on a-GST, it was 13.95±0.16 ns. This small difference (approximately 4-5%) in τ_{nr} indicates a minor variation in defect density. Consequently, the possible impact of defect density at the perovskite-GST interface – between the crystalline and amorphous phases – on photoluminescence modulation is several times lower than that of the Purcell effect [8] and an order of magnitude lower compared to the reflectance factor. Therefore, this influence can be considered negligible.

The PL enhancement attributed to the Purcell effect is approximately 15–20% [6, 8]. However, when we account for the reflectance factor, we observe an enhancement of around 100%. This demonstrates that reflectance is a primary factor in PL modulation.

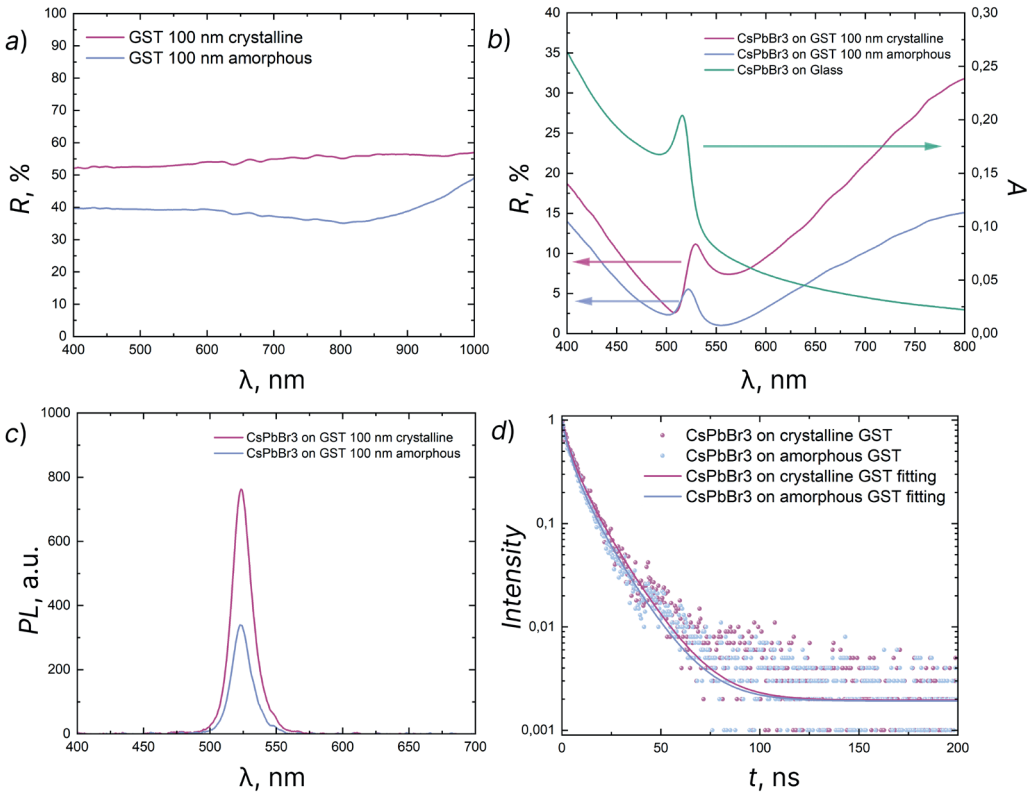

Fig. 2. R spectra of a-GST and c-GST thin film on the glass substrate (*a*). R spectra of $CsPbBr_3$ thin film on top of the a-GST and c-GST alongside with absorption spectra of $CsPbBr_3$ thin film (*b*). PL spectra of $CsPbBr_3$ thin film on top of the a-GST and c-GST layer. (*d*) TRPL measurement of $CsPbBr_3$ thin film on crystalline and amorphous GST and fitting with ABC-model (*c*)

Conclusion

Our research highlights the potential of tuning the PL signal of perovskite thin film through the phase switching of GST substrate for possible application in photoluminescence computing. Our findings indicate that the difference in reflectance between a-GST and c-GST is the primary factor influencing the modulation of the PL signal. This platform could significantly enhance the development of all-optical memristors, utilizing direct laser writing for GST phase switching, resulting in a distinct PL response from halide perovskite thin films. Furthermore, we propose that optoelectronic devices employing electrical field-induced GST phase transitions (or hybrid optical-electrical) might also be a promising direction for future research.

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