Conference paper
UDC: 53.093
DOI: https://doi.org/10.18721/JPM.191.126

The role of charge carrier diffusion in halide perovskite luminophores with memory for optical computing

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Abstract. The optical elements that can combine memory and signal modulation have become a topic of interest in the field of neuromorphic computing systems. In particular, the optical analogue of a memristor called memlumor exhibits promising features in which the change of the output luminescence depends not only on the excitation light signal but also on the state of the material. Metal halide perovskite luminophores are considered to be suitable for memlumor implementation as they exhibit modulation of photoluminescence due to the interaction of structure defects with the environment and previous interactions, thereby possessing memory. Luminescence in perovskite materials is described via Shockley–Reed–Hall model which takes into account charge carriers dynamics in the structure. Additionally, the diffusion of charge carriers also plays a key role and highly depends on memlumor's size. This paper explores the perovskite memlumor's functionality based on their characteristic size to identify optimal parameters and suitable designs for future optical neuromorphic computing architectures.

Keywords: neuromorphic systems, memlumors, metal halide perovskites, photoluminescence, charge carrier diffusion, quantum yield, time-resolved photoluminescence, Shockley–Read–Hall model, optical computing systems

Funding: The work was supported by the Russian Science Foundation (project 25-29-00520, https://rscf.ru/project/25−29−00520/).

Citation: Ekgardt A.A., Marunchenko A.A., Sapozhnikova E.V., Verkhogliadov G.A., Pushkarev A.P., The role of charge carrier diffusion in halide perovskite luminophores with memory for optical computing, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 19 (1.1) (2026) 156−161. DOI: https://doi.org/10.18721/JPM.191.126

Конференционная статья
УДК: 53.093
DOI: https://doi.org/10.18721/JPM.191.126

Роль диффузии носителей заряда в галогенидных перовскитных люминофорах с памятью для оптических вычислений

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Аннотация. Оптические элементы, сочетающие функции памяти и модуляции сигнала, вызывают значительный интерес в области нейроморфных вычислительных систем. В частности, оптический аналог мемристора – мемлюмор. Он обладает

перспективными свойствами: выходной сигнал люминесценции зависит не только от возбуждающего светового сигнала, но и от текущего состояния материала. Металлогалогенидные перовскитные люминофоры подходят для реализации мемлюморов, поскольку демонстрируют модуляцию фотолюминесценции за счет взаимодействия структурных дефектов с окружающей средой, учитывая при этом историю предыдущих воздействий, что обеспечивает эффект памяти. Люминесценцию в перовскитах описывает модель Шокли–Рида–Холла, учитывающая динамику носителей заряда. Кроме того, диффузия носителей заряда играет ключевую роль и сильно зависит от размеров мемлюмора. В статье исследуется функциональность перовскитных мемлюморов в зависимости от их характерных размеров для определения оптимальных параметров и подходящих элементов для будущих оптических нейроморфных вычислительных архитектур.

Ключевые слова: нейроморфные системы, мемлюморы, металлогалогенидные перовскиты, фотолюминесценция, диффузия носителей заряда, квантовый выход, время-разрешенная фотолюминесценция, модель Шокли–Рида–Холла, оптические вычислительные системы

Финансирование: Работа выполнена при поддержке Российского фонда науки (проект 25-29-00520, https://rscf.ru/project/25-29-00520/).

Ссылка при цитировании: Экгардт А.А., Марунченко А.А., Сапожникова Е.В., Верхоглядов Г.А., Пушкарев А.П. Роль диффузии носителей заряда в галогенидных перовскитных люминофорах с памятью для оптических вычислений // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2026. Т. 19. № 1.1. С. 156–161. DOI: https://doi.org/10.18721/JPM.191.126

Introduction

The von Neumann architecture, which is used in the vast majority of most modern computers, faces significant limitations in performance and energy efficiency, especially when applied to artificial intelligence and large-scale data processing tasks [1]. An alternative approach, such as optical neuromorphic computing architectures inspired by biological neural networks, are attracting increasing attention. A key component in such systems is the memlumor [2], a device that combines memory functionality with photoluminescence, enabling optical neuromorphic computing. Metal halide perovskites considered to be a promising material for memlumors due to their high charge carrier mobility, high photoluminescent quantum yield and simple synthesis methods [3]. Memory effects in perovskite materials occur on time scales ranging from milliseconds to minutes due to their photochemical activity under the influence of light. Furthermore, the thickness and lateral dimensions of the elements are important for maximizing photoluminescence and for the memlumor's operation itself, as in perovskites the carrier diffusion length exceeds several microns [4]. When excitation light is focused on an array of small elements linked together or a large bulk element of the material, the carriers quickly spread throughout the sample, which modulates the carrier concentration on the plane and thus inevitably influences the formation of the luminescence signal [5]. The focus of this work is to estimate the influence of memlumor's size on its functionality. For this purpose, two types of experiments are carried out: an experiment with wide uniform illumination and multiple perovskite $CsPbBr_3$ crystals of different effective sizes (from 100 nm to 1000 nm), as well as time-resolved luminescence at different pulse repetition rates and perovskite $MAPbBr_3$ films of different thicknesses (from 70 nm to 450 nm).

Materials and Methods

To produce $CsPbBr_3$ microcrystals the initial solution was synthesized using mixtures of 0.3 mmol CsBr and 0.3 mmol $PbBr_2$ in dimethyl sulfoxide. The solution was spin-coated onto a glass substrate at 6000 rpm spin cycle, which was later heated up to 70 °C in a Petri dish filled with 180 ml of isopropanol. The 0.32 ml of solution was then drop-cast onto a substrate and remained heated for 10 minutes in order to facilitate the crystal growth.

A series of $MAPbBr_3$ solutions (0.4 M, 0.6 M, 0.8 M, 1 M, 1.5 M) were prepared and spin-coated onto a glass substrate at 3000 rpm spin cycle for 40 seconds in order to create polycrystalline perovskite films with various thickness (70 nm, 170 nm, 200 nm, 240 nm and 450 nm).

Scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used to analyze the structure's size, providing precise determination of linear dimensions and morphology. Photoluminescent images of crystals were obtained on an Axio Imager.A2m (Carl Zeiss SMT) optical microscope. Image processing was performed using the ImageJ and Gwyddion software packages, and numerical modeling of charge carrier dynamics was carried out using a Shockley–Read–Hall model with diffusion, allowing for comparison between experimental and theoretical results. The time-resolved photoluminescence (TRPL) measurements were performed using a TEMA femtosecond pulse laser with a wavelength of 350 nm and frequency of 10–200 kHz and 100 nJ/cm^2 fluence.

Results and Discussion

Images of the $CsPbBr_3$ crystal were obtained on a photoluminescent microscope and PL of each crystal was estimated according to pixel intensity. SEM was used to measure the exact size of the crystals and evaluate their shape (Fig. 1, *a*). The study of photoluminescent properties of CsPbBr3 microcrystals depending on their linear size (in the range of 100–1000 nm) showed that the integral value of photoluminescence remains almost unchanged (Fig. 1, *b*), while the quantum yield decreases inversely proportional to the square of the size (Fig. 1, *c*). As the linear size increases, the computational density also decreases inversely proportional to the square of the size (Fig. 1, *d*).

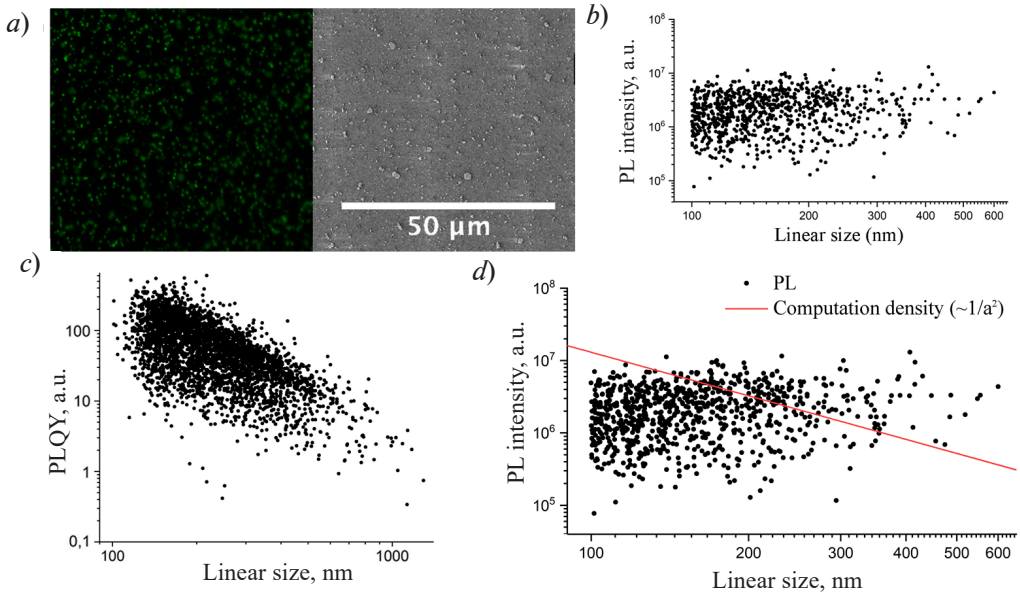

Fig. 1. Fluorescent (left) and SEM (right) images of the same area of $CsPbBr_3$ crystals (*a*); dependence of the photoluminescence intensity on the linear size of the crystal (*b*); dependence of the photoluminescence quantum yield on the linear size of the crystal (*c*); dependence of the photoluminescence intensity and computation density on the linear size of the crystal (*d*)

For $MAPbBr_3$ films with thicknesses of 70–450 nm, the charge carrier dynamics was evaluated using TRPL method that allows measuring the lifetime of nonradiative and radiative processes in structure and estimate charge carrier density. The resulting TRPL kinetics (Fig. 2, *a*) exhibit two components of lifetime: a fast one (Fig. 2, *b*) (on the order of nanoseconds), associated with diffusion, and a slow one (in the range from 10 to 100 nanoseconds).

The description of luminescence in $MAPbBr_3$ films is carried out using the Shockley–Reed–Hall model that also consider the charge carrier diffusion and memlumor formalism (1).

$$\begin{cases} \dfrac{d}{dt}\big(n(t)\big) = g(t) - k_r n(t)\, p(t) - k_t\big(N_t - n_t(t)\big) n(t) + D_n \cdot \dfrac{\partial^2 n}{\partial z^2}, \\ \dfrac{d}{dt} n_t(t) = k_t\big(N_t - n_t(t)\big) n(t) - k_n n_t(t)\, p(t), \\ \dfrac{d}{dt}\, p(t) = g(t) - k_r n(t)\, p(t) - k_n n_t(t)\, p(t) + D_p \cdot \dfrac{\partial^2 p}{\partial z^2}, \\ PL = k_r n(t)\, p(t) - PLQY\big(\vec{X}(t), I(t), t\big) \cdot I(t), \end{cases} \quad (1)$$

where $g(t)$ is the photogeneration rate of charge carriers, $I(t)$ is the pumping intensity (input signal), k_r is the constant of the rate of radiative recombination, N_t is the concentration of electron traps (defects), k_t is the constant rate of electron capture, k_n is the constant rate of trap release as a result of nonradiative recombination of trapped electrons with free holes, D_n and D_p are the diffusion coefficients for electrons and holes, respectively. This model considers the processes of photoexcitation ($g(t)$), photoluminescence (radiative recombination) ($k_r n(t)p(t)$), electron trap capture ($k_t(N_t - n_t(t))n(t)$) and trap release during nonradiative recombination ($k_n n_t(t)p(t)$).

Numerical modeling using Shockley–Read–Hall theory with added charge carrier diffusion and memlumor formalism confirmed that the fast component is associated with diffusion as it becomes more prominent with the increase of film thickness. (Fig. 2, *c*, *d*).

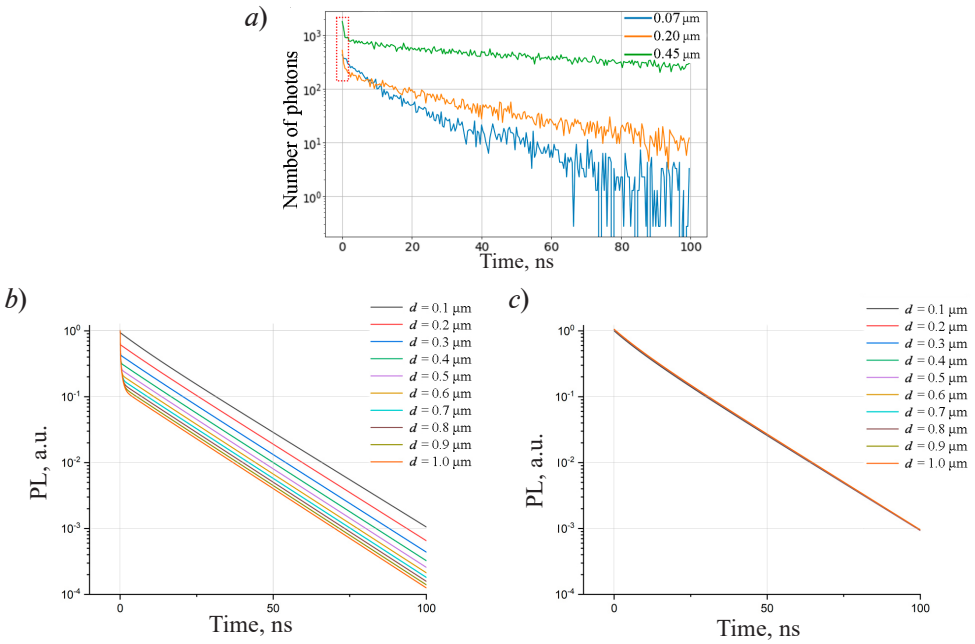

Fig. 2. TRPL kinetics for $MAPBBr_3$ films with thicknesses of 0.07 μm, 0.20 μm, 0.45 μm at a pump frequency of 30 kHz. TRPL experimental result (red frame is the fast component of luminescence attenuation associated with diffusion) (*a*); TRPL numerical modeling, obtained according to a diffusion-adjusted model (*b*); model without diffusion for a material with a thickness from 0.1 μm to 1 μm (*c*)

To further evaluate the role of charge carrier diffusion in memlumors based on metal halide perovskite films, it is necessary to look at the functionality of memlumors, namely, the modulation of the luminescence quantum yield depending on the pulse repetition rate of pump signal. As the film's thickness increases, the quantum yield also increases, while the luminescence peak decreases (Fig. 3, *a*, *b*, *c*). Due to the release time of traps (in order of tens to hundreds of microseconds) they do not have time to release themselves in the interval between pulses. As a result, more free holes remain, and, accordingly, photoluminescence also increases. After integrating the area under the decay curves for samples of different thicknesses curves for the modulation of the quantum yield were obtained (Fig. 3, *d*). Thus, it is revealed that the modulation is maximized for a 70 nm thick sample and minimized for a 450 nm one, suggesting that diffusion negatively

affects modulation. It can be seen from the TRPL kinetics that the role of diffusion increases with the increasing thickness, however, it is difficult to separate its effect on the quantum yield from the effect of the defect factor, which requires further quantitative studies.

The results indicate a preference for using independent elements with minimized diffusion to create high-density memlumor arrays, comparable in density to modern transistor circuits (up to 10^{10} cm^{-2}).

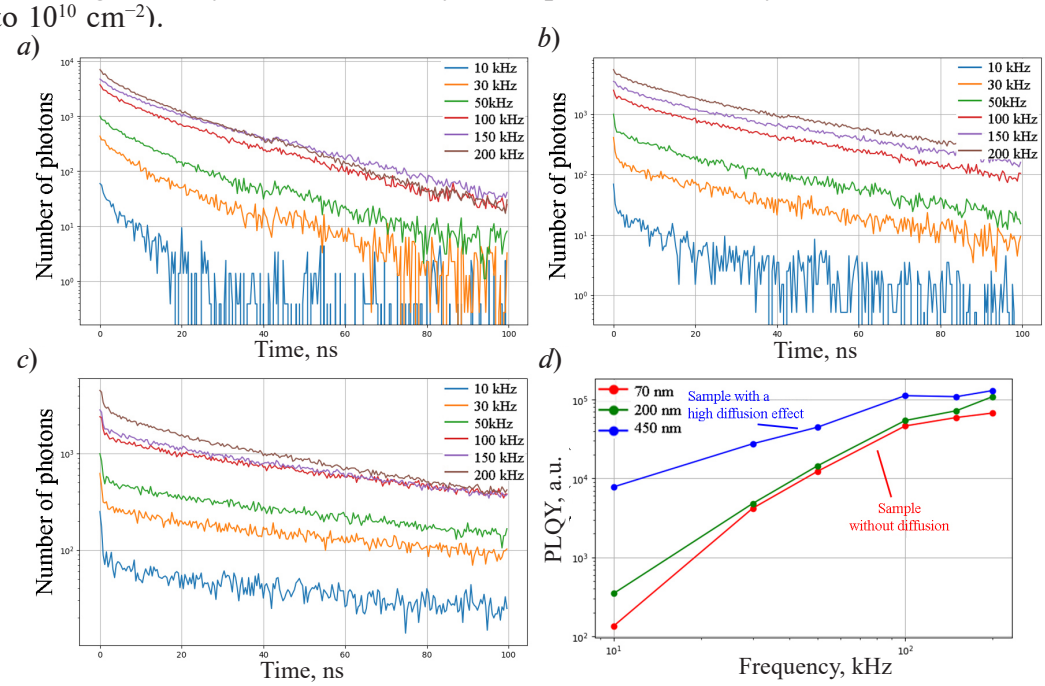

Fig. 3. TRPL kinetics measured at various repetition rates (10–200 kHz) for films with a thickness of 70 nm (*a*); 200 nm (*b*); 450 nm (*c*); dependence of the photoluminescence quantum yield on the pulse repetition rate for films with thicknesses of 70, 200 and 450 nm (*d*)

Conclusion

This work demonstrates the potential of metal halide perovskites for developing energy-efficient optical neuromorphic systems based on memlumors. Experimental and theoretical studies confirmed the influence of charge carrier diffusion on luminescent properties and highlighted the importance of minimizing diffusion for increasing element density. The results indicate the preference for using independent elements with minimal diffusion to create high-density memlumor arrays. The obtained data can be used to design new architectures for optical computing systems and high-density memlumor arrays with high performance and energy efficiency.

Acknowledgments

The work was supported by the Russian Science Foundation (project 25-29-00520, https://rscf.ru/project/25-29-00520/).

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Received 15.12.2025. Approved after reviewing 26.02.2026. Accepted 26.02.2026.