

Krūvinikų difuzijos ir rekombinacijos režimai iš garų fazės nusodintuose švino halogenidų perovskituose

Regimes of carrier diffusion and recombination in vapor deposited lead-halide perovskites

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Metal halide perovskites are attractive materials for realization of cheap and effective solar cells, thin film transistors, and light emitters [1]. Carrier diffusion at high excitations, however, is poorly addressed in perovskites, even though it governs the diffusion length and determines the efficiency of a photonic device. To fully understand the diffusion length dependence on carrier density, we performed direct and independent measurements of carrier diffusivity and recombination rate in several methylammonium lead-halide perovskite layers by applying the light-induced transient grating technique. We demonstrate the existence of two distinct carrier diffusion regimes within the density range of $10^{18} - 10^{20} \text{ cm}^{-3}$.

The carrier diffusivity increased with photoexcitation in all samples at densities above 10^{18} cm^{-3} due to two different physical mechanisms. In iodide and bromide layers, where diffusion coefficient was high even at low densities ($0.7 \text{ cm}^2/\text{s}$), the increase in diffusivity was moderate and caused by carrier degeneracy. In contrast, the mixed and chloride layers with a small low-density diffusivity ($0.01 - 0.04 \text{ cm}^2/\text{s}$) exhibited almost linear increase of D due to saturation of carrier traps and consequent carrier de-localization. On the other hand, the carrier lifetime decreased with excitation due to band-to-band radiative recombination and density-dependent nonradiative process. Assuming that latter is Auger recombination and accounting for the saturation of recombination coefficients due to state filling, we determined the non-degenerate values of bimolecular recombination coefficient $(1-7) \times 10^{-10} \text{ cm}^3/\text{s}$ and Auger recombination coefficient $(1.4-4.5) \times 10^{-28} \text{ cm}^6/\text{s}$. Finally, we demonstrate that two scenarios are possible where diffusion length either increases or drops with excitation, which is determined by compositional and structural quality of material, point defect density, and character of carrier diffusion.

From the application point of view, these results show that different strategies have to be used for performance optimization of various perovskites. In the mixed and chloride perovskites, there is a room for improvement of diffusion length via optimization of crystalline and compositional quality. In particular,

chlorine related point defects have to be eliminated in order to enhance the lifetime of carriers. On the other hand, the diffusion length in high quality iodide and bromide perovskites is limited by fundamental processes of nonlinear recombination at high carrier densities; therefore, operation at lower drive currents is preferable where maximum diffusion length is required.

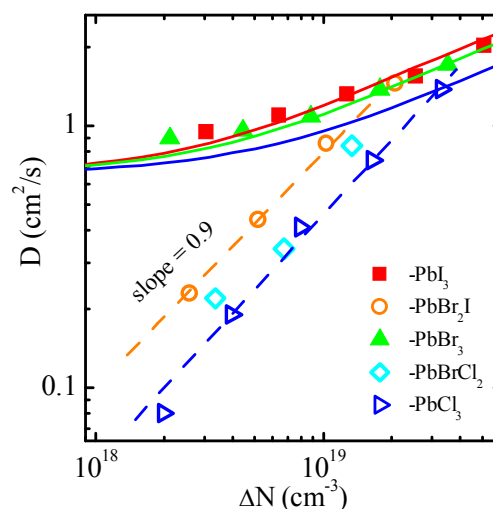


Fig. 1. Diffusion coefficient D determined from the light-induced transient grating transients as a function of photoexcited carrier density ΔN in different perovskite layers.

Keywords: lead-halide perovskites, carrier diffusion, light induced transient grating, time-resolved photoluminescence, carrier recombination.

References

- [1] Z. Xiao, R.A. Kerner, L. Zhao, N.L. Tran, K.M. Lee, T.-W. Koh, G.D. Scholes, B.P. Rand, Efficient perovskite light-emitting diodes featuring nanometre-sized crystallites, *Nat. Photonics*. (2017).