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Nonlinear optical properties of benzylamine lead(II) bromide perovskite microdisks in femtosecond regime

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ABSTRACT

Organometal halide perovskites, an emerging class of direct bandgap semiconductors, are attractive candidates for many optoelectronic device applications. Herein, we have reported the nonlinear optical (NLO) properties of layered benzylamine lead(II) bromide perovskite microdisks (MDs) having a lateral dimension of a few micrometers and an average thickness of 35 nm, featuring narrow deep blue emission using the Z-scan technique. The NLO behavior switches over from saturable absorption to reverse saturable absorption under femtosecond laser pulse excitation. Our NLO studies have demonstrated tunable nonlinear behavior, which is attributed to the interplay between single and two-photon absorption by the carriers in the conduction band. Perovskite MDs exhibit an optical limiting behavior originating from the two-photon absorption mechanism.

Discovery of optical harmonics by Franken *et al.* in 1961 has attracted research interest in the field of nonlinear optics.[1] Nonlinearity is typically observed at very high light intensities, which can be achieved using lasers. In semiconductor materials, absorption of light results in the interaction process in which electrons of the interacting material resonate with the electric field of the incoming optical signal inducing a polarization

$$P = \varepsilon_0\left(\chi^{(1)}E(\omega) + \chi^{(2)}E^2(\omega) + \chi^{(3)}E^3(\omega) + \cdots\right). \quad (1)$$

Here, ε_0 is the dielectric permittivity of vacuum, coefficients χ^n are the nth order susceptibilities of the medium (the presence of such a term is generally referred to as the nth order nonlinearity), and E is the electric field of frequency ω associated with the optical excitation signal.[2] Up to moderate light intensity, only the first term corresponding to the linear optical response dominates, leading to phenomena like refraction, scattering, and linear absorption. However, at higher field intensity, other terms in Eq. (1) become prominent introducing a nonlinear response in the materials. Materials having a superior nonlinear optical (NLO) response are widely used in photonic devices such as optical limiters, mode-lockers, and Q-switchers.[3]

Currently used semiconductor saturation absorber mirrors have drawbacks such as the high fabrication cost and limited operational wavelength range. Therefore, the quest for new materials having superior NLO properties is continuing for the development of viable photonic devices. A variety of organic,[4] inorganic,[5] and quantum dot[6] materials have been investigated, and some of them find applications in the field of nonlinear optics. Saturable absorption (SA) behavior has been recently observed in emerging materials like 2D transition metal dichalcogenides (TMDCs),[7] graphene,[8] topological insulators,[9] and black phosphorous.[10] Organometal halide perovskites, on the other hand, have attracted many applications over conventional semiconductor materials. These materials are the emerging class of direct bandgap semiconductors, which have shown great application potential in optoelectronics devices due to their outstanding characteristics such as high color tunability from the visible to near-infrared region, high color purity with reduced FWHM (of the order of ~20 nm), high photoluminescence quantum yield, high carrier mobility, and ease of synthesis.[11–13] Recent reports have demonstrated that both organometallic halide perovskite[2,12] and inorganic perovskite QDs[14] could be potential materials for nonlinear device applications.[15] But it is imperative that the