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Article

Third-Order Nonlinear Optical Properties and Saturation of Two-Photon Absorption in Lead-Free Double Perovskite Nanocrystals under Femtosecond Excitation

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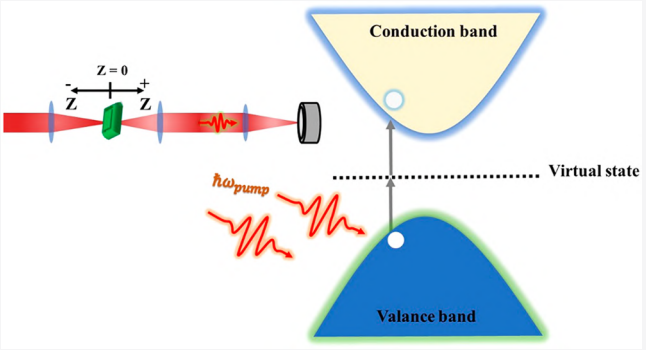
ABSTRACT: Lead halide perovskites have been widely explored in the field of photovoltaics, light-emitting diodes, and lasers due to their outstanding linear and nonlinear optical (NLO) properties. But, the presence of lead toxicity and low chemical stability remain serious concerns. Lead-free double perovskite with excellent optical properties and chemical stability could be an alternative. However, proper examination of the NLO properties of such a material is crucial to identify their utility for future nonlinear device applications. Herein, we have made use of femtosecond (fs) Z-scan technique to explore the NLO properties of $Cs_2AgIn_{0.9}Bi_{0.1}Cl_6$ nanocrystals (NCs). Our measurements suggest that under nonresonant fs excitation, perovskite NCs exhibit strong two-photon absorption (TPA). The observed saturation of TPA at high light intensities has been explained by a customized model. Furthermore, we have demonstrated a change in the nonlinear refractive index of the NCs under varying input intensities. The strong TPA absorption of lead-free double perovskite NCs could be used for Kerr nonlinearity-based nonlinear applications such as optical shutters for picosecond lasers.



KEYWORDS: *Z-scan, TPA saturation, double perovskite, nonlinear optics, nonlinear refractive index*

Strong interaction of intense light with matter comes under the class of nonlinear optics, which becomes extremely important for modern technologies. The nonlinear optical properties of a specific material play an important role in revealing light−matter interactions and ultrafast dynamics.[1,2] Formation of new secondary optical fields and variation of phase and frequency instigated by polarization made the NLO effect a keystone for the manipulation of photons in advanced technologies such as optical computation, information processing and storage, and telecommunication.[3−6] The NLO effect has been identified in a series of materials including transition metal dichalcogenides (TMDCs),[7,8] graphene,[9,10] hexagonal boron nitride (h-BN),[11] and metal organic frameworks (MOFs).[12,13] The materials possessing optical nonlinearities find applications in optical switches, optical data storage,[14,15] and lasers.[16] Nowadays, switching of optical signals in optical communication is achieved through optical-to-electronic-to-optical (OEO) transmutation components. Operation of such photonic devices is based on the instantaneous Kerr effect, which occurs when bound electronic charges of material are virtually excited by photons having energies less than the bandgap resulting in change in refractive index of the material.[17] The Kerr effect in thin films is generally weak, and in that case direct single or multiphoton absorption process can bring the change in refractive index that is significantly larger than the change due to Kerr effect. In contrast, the linear electro-optic or Pockels effect (a second order process) where the change in refractive index varies linearly with the electric field (of laser beam) finds application in electro-optic modulators. However, due to intrinsic centrosymmetric structures, traditional perovskites ($CsPbX_3$ or $MAPbX_3$) are not useful for applications based on the Pockels effect.[18] A recent report shows that germanium based perovskite ($CsGeI_3$) exhibits an electro-optic coefficient which is better than that of $LiNbO_3$.[19] It is worth mentioning that the optical Kerr effect is present in all centrosymmetric media, but the strength of this effect is weaker than the linear electro-optic effect. Furthermore, carrier induced third-order nonlinearities depend on carrier diffusion length and recombination of carriers in a material.

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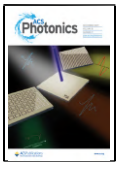

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