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Letter

$AgScP_2S_6$ van der Waals Layered Crystal: A Material with a Unique Combination of Extreme Nonlinear Optical Properties

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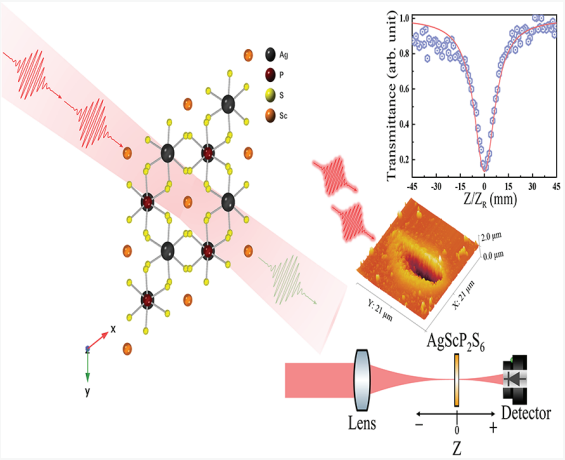
ABSTRACT: Research in two-dimensional layered materials (2DLMs) has exploded over the past several years for a variety of applications in photonics and optoelectronics. The 2D nature of these materials allows for a very local electronic probe of material as well as flexible integration with other functional components. Herein, using the femtosecond Z-scan technique, we report a giant two photon absorption (TPA) process and its saturation in the van der Waals gapped silver scandium thiophosphate ($AgScP_2S_6$) crystal. We have found a TPA coefficient of the order of 10^4 cm/GW which is orders of magnitude larger compared to many existing semiconductors and nonlinear crystals. Furthermore, we found a TPA cross-section of 10^3 GM and characterized the optical limiting (OL) response (0.2 mJ/cm^2) and the multipulse laser damage threshold (1.09 ± 0.19 J/cm^2). The combination of giant TPA, extremely low OL, and very high damage threshold suggests that this material could be extremely useful in applications like optical limiters or switches.



In all aspects of photonics devices, e.g., pulsed lasers, optical switches, optical modulators, and photodetectors, the nonlinear optical (NLO) materials play a critical role.[1−4] Current NLO devices for nonlinear nanophotonic applications such as quantum nanophotonics and on-chip nanophotonics are based on traditional semiconductors like β-barium borate and lithium niobate, which exhibit a bulk 3D crystalline setting.[5,6] However, these traditional NLO materials are technically limited due to their low NLO response and available fabrication and integration technologies.[7−9] Therefore, it is imperative to discover new materials with large NLO responses and harness their electronic and optical properties that can be device-integrated with new functionalities and high performance for future optoelectronics and nanophotonics. Two dimensional layered materials (2DLMs), due their exceptional physical properties, are the topic of intense research for a wide variety of applications in electronics, photonics, and optoelectronics. Increasing research efforts toward NLO properties of 2DLMs have been performed in the recent past, including materials such as graphene and transition metal dichalcogenides (TMDCs). The strong optical nonlinearity and ultrafast response of these materials showed signs of unique NLO features like ultrafast saturable absorption, second/third harmonic generation, and optical limiting.[10,11] Success of 2DLMs in nonlinear optics can be counted on from the fact that these materials possess broad and tunable absorption, large optical and thermal damage threshold, ultrafast recovery time, and high mechanical and thermal stability.[12] Graphene and its derivatives have been extensively used for making optical limiting material due to its ultrabroadband response.[13] Two photon absorption (TPA) saturation is a third order nonlinear process involving the participation of two coherent photons where an electron is promoted to an excited energy level, the energy gap of which corresponds to combined energies of two photons. This process has been experimentally demonstrated in monolayer and few layer WS_2, MoS_2, and $MoSe_2$ with large TPA coefficients.[14,15] NLO responses at 1064 and 532 nm from two-dimensional hexagonal boron nitride (h-BN) nanosheets and their hybrid gel glasses were investigated with TPA coefficients in the range of 3.84−5.24 cm/GW (at 532 nm) and 4.62−7.66 cm/GW (at 1064 nm), thus making this system an efficient optical limiter in the visible to near-infrared region.[16]

Another class of exotic 2D materials are known as layered quaternary metal thio/selenophosphates and have a general

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