Ultra-strong Kerr optical nonlinearity in BiOBr nanoflakes

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ABSTRACT

We characterize the third-order optical nonlinearity of BiOBr nanoflakes via Z-Scan technique. Strong two-photo absorption (TPA) as well as high Kerr nonlinearity are observed at both 800 nm and 1550 nm, with a high TPA coefficient on the order of 10^{-7} m/W and a high Kerr coefficient on the order of 10^{-14} m²/W being achieved.

Keywords: two-photo absorption, Kerr nonlinearity, 2D materials, BiOBr.

1. INTRODUCTION

As the fundamental blocking of nonlinear photonic devices, advanced optical materials with superior optical nonlinearity have been widely explored and investigated [1-6]. Among them, two-dimensional (2D) layered materials, featuring remarkable nonlinear optical properties such as ultrafast broadband optical response [2], ultrahigh Kerr optical nonlinearity [3-6], and strong nonlinear absorption [2-5], have attracted significant interests in recent years. Bismuth oxyhalides, i.e., BiOX (X = Cl, Br, I), which consists [Bi₂O₂]²⁺ slabs interleaved by double halogen atoms with weak van der Waals interaction among adjacent slabs of halogen, have been explored as a new group of advanced layered optical materials [7]. The self-built internal static electric fields generated between different elements layers leads to an effective separation of photoinduced electro-hole pairs, which enables BiOX with prominent photocatalytic properties [7, 8] as well third-order optical performances [9, 10].

In this paper, we characterize the third-order nonlinear optical response of BiOBr nanoflakes at both 800 nm and 1550 nm via Z-scan technique. Experiment results show that BiOBr nanoflakes exhibit strong TPA and high Kerr nonlinearity in these wavelengths, with a high TPA coefficient on the order of 10^{-7} m/W and a high Kerr coefficient on the order of 10^{-14} m²/W being achieved. Moreover, it is also interesting to found that the Kerr coefficient n_2 translates from negative to positive when the wavelength is changed from 800 nm to 1550 nm. Our results confirm the strong potential of BiOBr as an advanced nonlinear optical material for the implementation of high-performance nonlinear photonic devices.

2. MATERIAL PREPARATION AND Z-SCAN SETUP

The BiOBr samples used in our experiment were mechanically exfoliated from the bulk crystals onto a glass substrate using adhesive tapes. Fig. 1(a) shows the representative morphology image and thickness profile of the BiOBr nanoflakes characterized by atomic force microscopy (AFM). The measured thickness of the BiOBr nanoflakes was ~140 nm. The Raman spectra of BiOBr nanoflake is presented in Fig. 1(b). The incident laser wavelength was 532 nm. The two phonon modes of A_{1g} (~113.2 cm⁻¹) and E_g (~160.5 cm⁻¹) are consistent with previous report [11], thus demonstrating the high quality of our prepared BiOBr nanoflakes.

Figure 1. (a) AFM image and (b) Raman spectral of BiOBr nanoflake. (c) Schematic illustration of the Z-scan system.

The third-order optical nonlinearity of the prepared BiOBr nanoflakes was measured by the Z-scan setup shown in Fig. 1(c). Ti:sapphire femtosecond pulsed lasers (Coherent, Chameleon) centered at 800 nm and 1550 nm were used to excite the samples. The repetition rate and pulse duration were ~80 MHz and ~140 fs, respectively. A half-wave plate combined with a linear polarizer was employed as an attenuator to control the input power. The laser beam was focused by an objective (magnification 10×, numerical aperture 0.25) to achieve a low beam waist of ~1.6 μm (Rayleigh range: ~20 μm). A high-definition CCD imaging system was used to locate the target material and direct the transmitted laser beam on it. Two photodiodes were employed to collect the power data of

the open- (OA) and closed-aperture (CA) measurements [12]. A Labview program was developed to synchronize the movement of the motorized stage and data collection.

3. EXPERIMENT RESULTS AND DISCUSSION

Figures 2(a) and (b) present the Z-scan results at 800 nm. Typical reverse saturation absorption (RSA) is clearly observed for the OA curve in Fig. 2(a). Since the photo energy (~ 1.55 eV) of the excitation laser is much smaller than the bandgap of BiOBr (~ 2.73 eV for the bulk and ~ 2.82 eV for the monolayer) [8], the observed RSA can be mainly attributed to the TPA of the BiOBr nanoflakes. By fitting the OA curve (solid line in Fig. 2(a)), we obtain a fit TPA coefficient β as high as $\sim 2.625 \times 10^{-7}$ m/W, much higher than graphene [3, 13]. The Kerr nonlinearity of BiOBr was characterized via CA measurement. The peak-valley CA configuration in Fig. 2(b) is a reflection of the self-defocusing effect in BiOBr nanoflakes, which corresponds to a negative Kerr coefficient n_2 . A large fit n_2 of -1.711×10^{-14} m²/W is achieved, which is close to the reported value of graphene [3, 14].

Figure 2. Z-scan results of BiOBr nanoflakes: (a) OA and (b) CA curves at 800 nm. (c) OA and (b) CA curves at 1550 nm.

The optical nonlinear responses of BiOBr at telecommunication band (1550 nm) was also investigated, as shown in Figs. 2(c) and (d). Prominent RSA behavior can be observed in the OA measurement, which indicates the strong TPA at this wavelength. However, a positive n_2 is obtained by fitting the valley-peak CA configuration in Fig. 2 (d), which is opposite to the n_2 at 800 nm. This can be attributed to the dispersion of Kerr nonlinearity associated with TPA – positive n_2 is obtained when the excitation photo energy (at 1550 nm) is below the TPA edge (half-bandgap) while it becomes negative when the photo energy (at 800 nm) is between the one-photon absorption and TPA edges [15]. Moreover, a fit n_2 as high as $\sim 3.887 \times 10^{-14}$ m²/W is obtained, which is almost three orders of magnitude larger than single crystalline silicon [1]. This highlights the high potential of BiOBr for nonlinear optical functions at telecommunication band. In Table 1, we compare the measured β and n_2 of BiOBr [16] with some other typical 2D layered materials, which confirms the superior third-order optical nonlinearity of BiOBr nanoflakes as an advanced nonlinear optical material for implementing nonlinear photonic devices, similar to other 2D materials we recently measured [17]. These results reflect the stability of the prepared BiOBr nanoflakes and confirm their strong potential as a promising nonlinear optical material for high-performance hybrid integrated photonic devices [18-19] similar to other 2D materials such as graphene oxide [20-23] and PdSe₂, potentially for both classical [24] and quantum [25-28] applications. Finally, as for Si-Ge [29-31] PdSe₂ may also offer interesting possibilities for 2nd order nonlinear effects courtesy of its complex anisotropic nonlinearity.

Table 1. Comparison of β and n_2 of various 2D layered materials

Material	Laser parameter	Thickness (nm)	β (m/W)	n_2 (m ² /W)	Reference
Graphene	1550 nm, 100 fs	5-7 layers	9×10^{-8}	-8×10^{-14}	[3]
WS ₂	1040 nm, 340 fs	57.9	$(1.81 \pm 0.08) \times 10^{-8}$	$(-3.36 \pm 0.27) \times 10^{-16}$	[4]
BP	800 nm, 100 fs	30-60	$(4.5 \pm 0.2) \times 10^{-10}$	$(6.8 \pm 0.2) \times 10^{-13}$	[5]
BiOBr	800 nm, 140 fs	140	2.625×10^{-7}	-1.711×10^{-14}	This work
BiOBr	1550 nm, 140 fs	150	1.781×10^{-7}	3.887×10^{-14}	This work

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