

# Enhanced broadband non-degenerate two-photon absorption in Ga-doped ZnO for ultrafast all-optical switching: supplementary material

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## 1. Full width of half maximum at different wavelengths

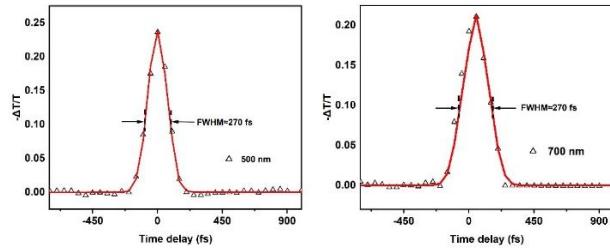


Fig. S1. Full width of half maximum at wavelengths of 500nm and 700nm.

## 2. Schematic diagram of an all-optical switching

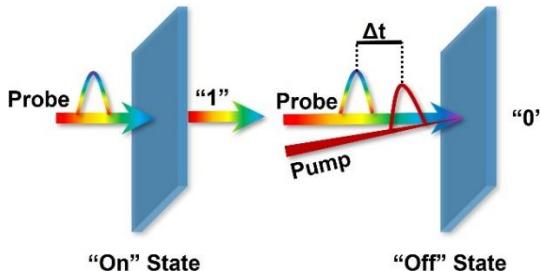


Fig. S2. All-Optical Switching ON/OFF State Schematics.

## 3. Theoretical fitting using different models

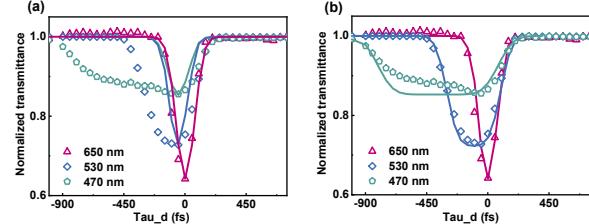


Fig. S3. (a) Normalized transmittance at different probe wavelengths under 650 nm excitation, the solid lines represent the theoretical fitting without considering GVM; (b) Normalized transmittance under the same probe and excitation wavelength, the solid lines represent the theoretical fitting with the walk-off model.

## 4. Effect of thickness and pulse width on walk-off parameters

The walk-off parameter  $\rho$  is given by:

$$\rho = \frac{L}{\omega_p \cdot c} \left[ n - \lambda \frac{dn}{d\lambda} - \left( n_p - \lambda_p \frac{dn}{d\lambda_p} \right) \right] = \frac{L}{\omega_p \cdot c} \Delta n_g$$

$$C_{ts} = \frac{\tau}{L} \left( \tau = \frac{\omega_p}{1.6651} \right) \quad (1)$$

where  $L$  represents the sample thickness,  $\omega_p$  denotes the pump pulse width,  $c$  is the speed of light and  $n(n_p)$  refers to the linear refractive index of the material at the probe wavelength (pump wavelength), the symbol  $\lambda$  ( $\lambda_p$ ) indicates the probe wavelength (pump wavelength). The threshold  $C_{ts}$  represents the threshold value for walk-off, when this value is less than 2.30 ps/mm, the TAS obtained from the experiment needs further correction to achieve an accurate TPA spectrum for GZO

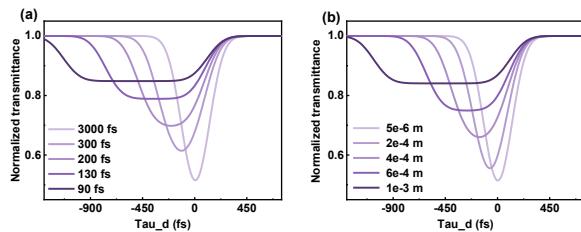


Fig. S4. (a)  $L = 0.5\text{mm}$ , the impact of different pulse widths on walk-off; (b) The pump pulse width is 190 fs, and the impact of different sample thickness on walk-off.

Table S1. The pump wavelength of 650 nm, the corrected TAS coefficient at different detection wavelengths

$\omega_p$ (ps)	0.2	0.4	0.6	1.0	1.5
$L$ (mm)	0.053	0.105	0.157	0.261	0.392
$\rho$	0.506	0.507	0.507	0.506	0.507
$C_{ts}$ (ps/mm)	2.303	2.302	2.301	2.304	2.305

6. Nondegenerate dispersion relations in different material

Table S3. Non-degenerate TPA coefficients of GZO, ZnS, GaAs and ZnSe (ZnS ZnSe and GaAs are from references[1], [2]and[3], respectively).

$\beta_{ND}(\omega_1; \omega_2)$ (cm/GW)							
$\omega_2/\omega_1$	$\beta_{ND}$ (GZO)	$\omega_2/\omega_1$	$\beta_{ND}$ (ZnS)	$\omega_2/\omega_1$	$\beta_{ND}$ (GaAs)	$\omega_2/\omega_1$	$\beta_{ND}$ (ZnSe)
1.06	1.62	1.40	0.07	1.10	~5.4	1.00	~2.0
1.15	4.30	1.45	0.33	1.20	~5.5	1.45	~3.1
1.29	10.08	1.50	0.90	1.30	~6.5	1.65	~4.0
1.47	16.97	1.55	1.51	1.42	~7.3	1.85	~4.5
1.56	17.33	1.60	2.00	1.55	~7.9	2.25	~5.0
1.60	17.58	1.67	2.88	1.72	~9.3	2.50	~6.0

## References

1. S. Chen, M.-L. Zheng, X.-Z. Dong, Z.-S. Zhao, and X.-M. Duan, "Nondegenerate two-photon absorption in a zinc blende-type ZnS single crystal using the femtosecond pump-probe technique," J. Opt. Soc. Am. B: Opt. Phys. 30(12),3117-3122 (2013).
2. L. Krauss-Kodytek, W. R. Hannes, T. Meier, C. Ruppert, and M. Betz, "Nondegenerate two-photon absorption in ZnSe: Experiment and theory," Phys. Rev. B 104(8),085201 (2021).
3. L. Krauss-Kodytek, C. Ruppert, and M. Betz, "Nearinfrared non-degenerate two-photon absorption coefficients of bulk GaAs and Si," Opt Express 29(21), 34522-34530 (2021).

## 5. The corrected TPA coefficients with 750 nm excitation wavelengths

Table S2. The  $\beta_{ND}$  at different detection wavelengths under 750 nm excitation wavelength

Probe Wavelength (nm)	$\rho$	$\beta_{ND}$ ( $10^{-11}\text{m/W}$ )	$\beta'_{ND}$ ( $10^{-11}\text{m/W}$ )
470	16.0	2.80	17.58
480	14.0	3.20	17.33
510	10.0	4.40	16.97
580	4.0	6.20	10.08
650	0.1	4.30	4.30