Ultrafast synergistic excitation for in-situ computing

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Ultrafast synergistic excitation for in-situ computing

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Abstract
Nonlinear optical phenomena (NLOPs) in two-dimensional (2D) materials can be envisioned for neuromorphic functions at the device and related system level. But it has been attended rarely that transition among multi-energy states as one origin of NLOPs directly used for neuromorphic functions, which is assisted to understand nature of device-level nonlinear optical neuromorphic performance. Here we introduced a pump-probe-control technology to reveal multi-energy-state transition in multilayer molybdenum disulfide, enhancing nonlinear signals by transitions from two-photon absorption to synergistic excited states absorption and enabling an in-situ computing concept within an array of pure 2D flakes. Optical weighted average calculation and artificial neural network were realized without the fabrication of complex extrinsic structures, while preserving the femtosecond speed and femto-Joule power consumption, revealing the feasibility of pump-probe-control technology for nonlinear neuromorphic functions.
Introduction

Exploring nonlinear optical phenomena (NLOPs) is fundamental to advanced photonic data sensing, communicating, and computing technologies in the era of artificial intelligence\textsuperscript{1-3}. Especially, the strong Coulomb interactions confined in the atom-thin plane endow two-dimensional (2D) materials with a plethora of NLOPs\textsuperscript{3}, including saturable absorption\textsuperscript{4}, stimulated emission\textsuperscript{5}, and high harmonic generation\textsuperscript{6,7}. Therefore, 2D materials are expected for remarkable functions, such as nonlinear modulators\textsuperscript{6}, sensors\textsuperscript{8}, converters\textsuperscript{9}, and synapses\textsuperscript{10},\textsuperscript{11}, for which the intrinsic tunability, ultrafast, power-efficient, and massive parallelism nature are advantageous to keep pace with the unprecedentedly increasing amount of data\textsuperscript{5-7,10,12}. The nonlinear dynamics in 2D materials have been intensively investigated through pump-probe technology, however, the limited signal-to-noise ratio is a congenital restriction for neuromorphic functions due to the short interaction length along the out-of-plane direction\textsuperscript{3,6,7,13}. Previous studies have proposed electronic and all-optical strategies to enhance the nonlinear signals in single flakes\textsuperscript{3,6,7,13,14}. For electronic strategies, electric fields dynamically modulate the nonlinear signals by changing the oscillating strength of excitons or breaking the crystal inversion symmetry\textsuperscript{6,14}. For all-optical methods, the adjacent media integrated with 2D materials determine the nonlinear signals\textsuperscript{3,7,13,15}. For example, coherent plasmonic excitations launched in nanocavities and metamaterials modulate the nonlinear signals by different geometric structures\textsuperscript{7,13}. The remote dipole-dipole interactions induced by quantum dots control the nonlinear signals by the thicknesses of the quantum dots and the distances to the 2D film\textsuperscript{3}. Although future neuromorphic functions based on pump-probe technology can be envisioned, implementing these strategies to fabricate an integrated array may face the contradiction between performance and fabrication complexity. Integrating gating electrodes into an array is feasible, but the degraded response speed and higher power consumption are unavoidable in electronic systems\textsuperscript{6,14}. Present all-optical methods can preserve high performance, but precisely integrating an array with various geometric structures may complicate the fabrication procedure\textsuperscript{7,13}. Enhancing the nonlinear signals based on the intrinsic properties of 2D materials opens new a way to resolve this contradiction. Based on the special distribution of density of states (DOS) in TMDs\textsuperscript{4,15-18}, synergistic transitions between different energy states in TMDs\textsuperscript{9,16} inspired us to introduce a new control pulse in the pump-probe setup to enhance nonlinear signals\textsuperscript{4}. The ultrashort laser pulses at the femtosecond level can preserve the fast speed and low energy consumption, which may also avoid the fabrication of complex extrinsic structures at the same time.

In this work, transient excitation and relaxation under the pump, probe, and control pulses were analyzed in multilayer molybdenum disulfide (MoS\textsubscript{2}), for which the intensity showed a strong correlation with the pulse time delay and a linear dependence with the pulse power. The pump-probe process without the control pulse revealed two-photon absorption (TPA) near the K points of the Brillouin zone\textsuperscript{19}. The transient intensity enhanced by the control pulse was dominated by synergistic excited states absorption (SESA) shifted to momentum space between the K and \Gamma points\textsuperscript{16}. The final excitation states were located near the high-energy nested conduction bands (HENCBs), where the higher DOS accounted for the enhancement\textsuperscript{16}. More interestingly, the intensity enhancement gradually quenched in thicker samples, related to the Coulomb screening-induced generation of ionized electron-hole pairs rather than excitons\textsuperscript{16,20}. 
The thermalization of ionized electron-hole pairs accounted for the prolonged relaxation\(^{16, 21}\). Based on the ultrafast excitation and relaxation, we can propose an in-situ computing concept, in which the nonlinear signals are generated, processed, and computed within a 2D array with different flake thicknesses. The weighted average calculation (WAC) was realized by directly encoding the thickness as weights. The pulse spot size determines the equivalent unit size, showing the potential for a higher integration density. In addition, the 2D array was feasible for an artificial neural network (ANN) by encoding time delay as weight\(^{11}\), with an operating speed at femtosecond level and power consumption of few femto-Joule per spike, revealing that the pump-probe-control technology can make an array of 2D flakes practical for nonlinear optical neuromorphic functions.

**Results**

The experimental setup is shown in Fig. 1a, in which the pump pulse was at 1040 nm with a width of 220 fs, and the probe pulse was at 902 nm with a width of 100 fs. The probe pulse was frequency doubled to generate the control pulse at 451 nm. Two time-delay lines were used to modulate the pump (control) time delay, defined as the excitation time difference of the pump (control) pulse relative to the probe pulse. The three pulses with matched Gaussian distribution were focused on exfoliated multilayer MoS\(_2\) samples (Fig. 1a, bottom left inset). We modulated the intensity of the pump beam at 2.2 MHz and detected the resulting intensity modulation of the probe beam by a photodiode at the same frequency with a lock-in amplifier. More experimental details are included in methods\(^4\). Pump-probe pulses with adjusted pump time delay stimulated Sample A to evaluate the control pulse’s contribution, in which control time delay was zero (Fig. 1a, bottom right inset). The pump and probe power were \(P_{\text{Pump}} = P_{\text{Probe}} = 2\) mW, and the control power was \(P_{\text{Ctl}} = 5.5\) mW. The mapping results in Figs. 1b-1g showed relatively low intensity for the selected pump time delays when the control pulse was off (mapping results for all pump time delays in Extended Data Fig. 1). At the same pump time delays, the control pulse induced strong signal enhancement in Figs. 1h-1m (mapping results for all pump time delays in Extended Data Fig. 2), indicating a different mechanism from saturable absorption\(^4, 22\). The intensity distributions across the sample as marked by the dashed lines in Fig. 1b and 1h are summarized in Fig. 1n, showing stronger pump time delay-dependence under the control pulse excitation. In more detail, intensity excitation and relaxation dynamics were analyzed for different regions (marked in Fig. 1d and 1j). All regions shared similar intensity evolutions with the maximum intensity located at the pump time delay of \(~0.015\) ps when the control pulse was off (Fig. 1o, Extended Data Fig. 3a). While stronger intensity variations occurred across different regions, with the intensity peak shifted to a larger pump time delay of \(~0.04833\) ps, when the control pulse was on (Fig. 1p, Extended Data Fig. 3b). This delay shift indicates all regions share the same excitation path, as will be discussed later. Based on Fig. 1o and 1p, the calculated average intensity gain \(IG = \frac{I_{\text{control on}}}{I_{\text{control off}}}\) (Fig. 1q, Extended Data Fig. 3c) and the normalized intensity difference \(\Delta I = \frac{I_{\text{normalized control on}}}{I_{\text{normalized control off}}}\) (Extended Data Fig. 3d) characterized the relaxation dynamics. The maximum gain and intensity difference were shifted to \(~0.18167\) ps, due to the extended relaxation time under the control pulse excitation.
The intensity variations for different regions were attributed to the thickness correlation, therefore atomic force microscopy (AFM) measurement was conducted for sample A (Extended Data Fig. 4), and the thickness distributions for regions marked in Fig. 1d and 1j were summarized in Fig. 2a. Without the control pulse, the intensity showed weak thickness-dependence, as shown in Fig. 2b and 2c for the pump time delay of 0.08167 ps and 0.18167 ps, respectively (more results in Extended Data Fig. 5). At the same pump time delay, obvious thickness-dependence emerged with the control pulse included, as shown in Fig. 2d and 2e (More results in Extended Data Fig. 6). When the thickness was below ~70 nm, the intensity decreased monotonically with the increase of thickness. For thicker locations over ~70 nm, the intensity enhancement was quenched, indicating the screening of the control pulse. This thickness dependence can be observed from the intensity gain distributions in Extended Data Fig. 7.

Fixing the pump and probe power at $P_{\text{Pump}} = P_{\text{Probe}} = 4$ mW and scanning the pump time delay, a stronger intensity dynamic was observed when the control power increased from 0 mW to 8 mW in Sample B (Fig. 2f, Extended Data Fig. 8), indicating that the intensity enhancement was mainly attributed to the control pulse. Similar intensity evolution under the control powers of 7 mW and 8 mW may stem from carrier saturation. The non-shifted intensity peak and identical relaxation lifetime reveal that the control power did not alter the final states of the carriers. At zero pump time delay, the mapping results under the control power of 0 mW and 4 mW are shown in Fig. 2g, in which the marked region was used to calculate the average intensity in Fig. 2f. Detailed mapping results under all pump time delays are shown in Extended Data Fig. 9 and 10.

Under zero pump and control time delays, the pump-probe-control pulses excited sample C to reveal the pulse power dependence, for which only one pulse power was monotonically increased from 0 mW to 7 mW, while the other two pulse powers were fixed at 2 mW. Before reaching the saturated condition, the linear relationship between pulse power and intensity was observed in Fig. 2h, calculated from the marked regions in Fig. 2i (mapping results for all power conditions in Extended Data Fig. 11-13). The intensity followed $I \propto P_{\text{Pump}} \times P_{\text{Probe}} \times P_{\text{Ctl}}$. Since the three pulses exhibited synergistic effects on the intensity dynamics, we infer that the intensity enhancement under the control pulse may be accompanied by excitation and relaxation time shifts. Fixing the pump, probe, and control powers at 2 mW, and adjusting the control time delay, the excitation started at an earlier negative control time delay of $\sim -0.53367$ ps, and the relaxation was greatly prolonged to over 4 ps, compared with the case of no control pulse (Fig. 2j), which could be feasible for applications in the time domain. This prolonged relaxation was also revealed from the pump-probe mapping (pump-probe-control mapping) results under pump time delay scanning (control time delay scanning) in Extended Data Fig. 14 (Extended Data Fig. 15).

Qualitatively shown in Fig. 3a, the A and B exciton near the band edge are residing at the K points of the Brillouin zone. The C peak is determined by transitions between K and $\Gamma$ points, distributed in a circular region around the $\Gamma$ point where the conduction band (CB) and valence band (VB) are nearly parallel. Such special band alignment generates larger joint DOS for much higher absorption near the C peak, and stronger many-body effects can occur under external excitation. To theoretically analyze carrier transitions, the A and B excitons are
treated as the same state by neglecting the spin-orbit coupling. Therefore, the synergistic excitation mainly involves five specific bands in the momentum space: the VB maximum and CB minimum near the K points, the parallel VB and CB between the K and Γ points, and the HENCBs near the Γ point. Because the pump and probe photon energies were below the bandgap, the pump-probe signal with no control pulse is attributed to TPA, matching the linear correlation with pulse powers. The excited states are localized near the K points, with the generated electron-hole bound state dominated by the Coulomb attraction (Fig. 3b)\textsuperscript{23}. The relatively low intensity is related to the limited DOS, and this excited state could decay fast by phonon-assisted recombination\textsuperscript{20}. With the increase of thickness, the limited changes in DOS (Fig. 3d) account for similar intensity dynamics in Fig. 1o.

The control pulse with photon energy near the C peak effectively promotes transitions in the parallel bands. Since the intensity monotonically increases with pulse powers (Fig. 2h), the stimulated emission is excluded. The enhanced intensity is attributed to SESA, with excited states in the HENCBs, where the nested CBs could provide much higher DOS (Fig. 3c)\textsuperscript{16}. Fractional carriers radiatively relax to the edge states near the K points, leading to a shifted intensity peak in Fig. 1p. However, the increased DOS in thicker samples can also appreciably screen the Coulomb interactions\textsuperscript{20}, leading to obvious intensity gain differences. The screening effect is due to a high density of excited hot carriers localized in the HENCBs, which effectively reduce the electronic bandgap and exciton binding energy\textsuperscript{20}. The reduced exciton binding energy lowers the exciton oscillation strength, therefore the excited hot carriers are more likely to generate ionized electron-hole pairs rather than excitons, which finally relax through dominant intraband scattering (Fig. 3e)\textsuperscript{20, 21}. The greatly prolonged relaxation lifetime under control pulse delay could be explained by the thermalization of ionized electron-hole pairs to the lattice temperature\textsuperscript{20, 21}. Therefore, the control pulse shows no influence on the intensity gain when the screening-induced carrier ionization dominates.

An in-situ computing concept was evaluated in a pure array of 2D flakes. An application of optical WAC is schematically shown in Fig. 4a, which uses the control power as the input signal. Based on the linear correlation between intensity and control power, the calculated derivative showed a linear correlation with flake thickness \( \frac{dI}{dP_{Ctl}} \propto h_t \), making it feasible to directly encode the thickness as weight \( h_t \propto w \) (Fig. 4a, left inset, Extended Data Note 1). By exciting different flakes, the collected signals are summed to acquire the WAC output (Fig. 4a, right inset). Experimentally, the pump and probe powers were 2 mW, and the pump and control time delays were zero. The correlations between control power and intensity are shown in Fig. 4b for 6 MoS\textsubscript{2} units, and the \( \frac{dI}{dP_{Ctl}} \) values are mapped into normalized weights Fig. 4c (Extended Data Fig. 16). After setting the control power, the WAC results for 3 units were acquired through optical means simply in Fig. 4d.

Based on the excitation and relaxation for different thicknesses, an ANN was established by adjusting the control time delay as weight. The optical ANN is schematically shown in Fig. 4e. The pump time delay was zero, and the control time delay was marked as the signal delay \( \Delta \tau \) (Fig. 4e, inset). Under the pump, probe, and control power of 2 mW, we mapped the intensity into a normalized weight (\( I \propto w \)), which led to a logarithmic relationship between weight and...
signal delay because the intensity relaxation is generally fitted with exponential equations (Fig. 4f). The ANN was applied for pattern recognition in simulation, with the SoftMax as the activation function and the cross-entropy as the cost function (detailed ANN setup is discussed in Extended Data Note 2). The specific patterns for letters H, U, S, and T in the training dataset are shown in Fig. 4g, encoded by the control power for each pixel (full training dataset given in Extended Data Fig. 17). The initial signal delays were set to zero, and after training a single-layer ANN with 36 synapses in 30 epochs, the signal delays were updated to modulate the output intensity (Fig. 4h, Extended Data Fig. 18). The corresponding initial and final weights are shown in (Fig. 4i, Extended Data Fig. 19). The average intensity of each epoch after updating the signal delay was simulated in Fig. 4j-4m for the H, U, S, and T patterns, respectively. By comparing the pattern labels and the predictions, the accuracy reached 100%, and the cost decreased to 1.697 (Fig. 4n). Detailed predictions for the training and testing dataset are summarized in Extended Data Fig. 20 and 21, respectively, proving the feasibility for neuromorphic functions based on the pump-probe-control technology.

Discussion and conclusion

The pulse power and pulse time delay enrich the synergistic transitions in multilayer MoS₂, and this pump-probe-control strategy is reproducible in more samples (Extended Data Fig. 22-25). Generally, the control pulse changes the carrier transition mechanism from TPA to SESA, but special phenomena in MoS₂ should be noted. The intensity enhancement was quenched at the crystal edge within ~1 μm range for some samples, even though the edge region and the adjacent bulk region shared the same thickness (Extended Data Fig. 26, Note 3). Although the intensity enhancement discussed above is a bulk rather than an edge effect, the physical symmetry-broken at the crystal edge may block carrier diffusion and assist Coulomb screening. Besides, wrinkles can form during sample preparation, which generally showed an enhanced intensity compared with adjacent flat regions regardless of the control pulse, attributed to the localized strain-induced carrier funneling. However, some wrinkles showed intensity reduction with the control pulse off, and intensity enhancement with the control pulse on (Extended Data Fig. 27, Note 4). The switching from anti-funneling- to funneling-like dynamics indicates that the crystal straining may exhibit non-monotonic effects on carrier transition, which is possibly related to the wrinkle’s geometric parameters. Therefore, a deeper understanding of pump-probe-control experiments may enrich new computing mechanisms for this field.

In-situ computing was discussed based on the above mechanism. For optical WAC applications, encoding the thicknesses into weights is easier for 2D materials, compared with 3D bulk materials. The unit size is determined by the laser spot size, which can be narrowed down to achieve a higher integration density. For ANNs based on this technology, the spike speed is at hundreds of femtoseconds, and the power consumption is at several femto-Joule, showing the potential to develop ultrafast and power-efficient neuromorphic systems with simplified fabrication. Besides, the excited states in 2D materials can be transferred into electronic signals in-situ, assisting the matching between optical and electronic modules for further chip-scale integration. Although the thickness-controllable growth of 2D film is still a severe challenge, recent progress in robotic assembly opens a realistic way to fabricate an array
of 2D flakes\textsuperscript{27}. Except for multilayer MoS\textsubscript{2}, the intrinsic nonlinear responses in many 2D materials are advantageous\textsuperscript{22}, and it deserves more efforts to develop new applications based on pump-probe-control excitation. Moreover, encoding the pulse power and time delay can be feasible for optical communications, and an intensity modulator is discussed in Extended Data Fig. 28 and Note 5. Therefore, the pump-probe-control technology may also enlighten compact integration of optical communication and neuromorphic computing systems.

References


Data availability
The data on which the figures are constructed and from which the conclusions are drawn are available in an online open dataset provided by the authors. All findings of this study are archived on Zenodo.

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Author contributions

Competing interests
The authors declare no competing interests.

Supplementary information
Methods
Extended Data Note 1–5
Extended Data Fig. 1–28
Extended Data Movie 1–6
**Fig. 1 | Signal modulation with control pulse.**

- **a.** Schematic of the pump-probe-control system.
- **The abbreviation of each component:** HWP, half-wave plate; PBS, polarization beam splitter; BBOC, beta-barium borate crystal; AOM, acousto-optical modulator; TD, time-delay line; H, pinhole; QWP, quarter-wave plate; BPF, band-pass filter; PM-SM-F, polarization-maintaining single-mode fiber; DM, dichroic mirror; SL, scanning lens; TL, tube lens; OB, objective; OC, oil condenser; BPF, band-pass filter; PD, photodiode. Bottom left inset, schematic of multilayer MoS$_2$ excited by the pump-probe-control pulses. Bottom right inset, the pulse setup with scanning pump time delay and zero control pulse delay.
- **b-m.** The imaging results of sample A with the control pulse off (**b-g**) and the control pulse on (**h-m**), under different pump time delays. Scale bars, 10 μm.
- **n.** Intensity distribution across the same line (red line in (b) and blue line in (h)) under different pump time delays.
- **o-p.** The average intensity evolution with scanning pump time delay for eight selected regions (marked in (d) and (j)), when the control pulse was off (**o**) and on (**p**).
- **q.** The average intensity gain with scanning pump time delay for the eight selected regions.
Fig. 2 | Thickness, power, and pulse time delay dependence. 

a. The thickness distribution for the selected regions in Fig. 1d and 1j.

b-e. The thickness-intensity correlation at the pump time delay of 0.08167 ps (b, d) and 0.18167 ps (c, e), with the control pulse off (b-c) and the control pulse on (d-e). The right panels show the intensity distribution in each region.

f. The normalized intensity evolution with turned pump time delay under different control powers in sample B. The shaded area shows the error. The inset shows the schematic pulse setup.

g. The intensity mapping results in sample B under the control power of 0 mW and 4 mW. The intensity data in (f) was acquired in the marked region. Scale bars, 5 μm.

h. The linear dependence of intensity with pump, probe, or control power in sample C. The solid line shows the linear fitted results, and the shaded area shows the error.

i. The intensity mapping results in sample C under different power conditions. The intensity data in (h) was acquired in the marked region. Scale bars, 10 μm.

j. The prolonged intensity decay with the control pulse on. The inset shows the schematic pulse setup.
**Fig. 3** Excitation mechanism with/without control pulse. 

a. The schematic band structure near the bandgap. Inset, the schematic absorption characters of MoS$_2$.

b. The intensity dynamic with the control pulse off is dominated by TPA at the K points.

c. The intensity dynamic with the control pulse on is dominated by SESA between the K and Γ points.

d-e. The thickness-correlated excitation and relaxation with the control pulse off (d) and the control pulse on (e).
Fig. 4 | In-situ optical neuromorphic computing based on pump-probe-control technology.

a. The schematic of the optical WAC system. Left inset, the mechanism of encoding thickness into weight. Right inset, the procedure of WAC. b. The control power-intensity correlations for 6 cells with different thickness. c. The derivative of intensity with respect to the control power is linearly encoded as weight. d. The 3-inputs WAC results. e. The schematic of the optical ANN system. Left inset, the computing mechanism of turning the control time delay. f. The linear correlation between transient intensity and weight, and the logarithmic correlation between signal delay and weight. g. The 3×3 patterns of H, U, S, and T in the training dataset. h. The initial and final delays for each synapse. i. The initial and final weight for each synapse. j-m. The simulated average intensity for label H (j), U (k), S (l), and T (m) in each epoch. n. The simulated accuracy and cost in 30 epochs.
Supplementary Files

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