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## Next-Generation Ultrafast Photoluminescence Spectroscopy: Integration of Transient Grating Optical Gate and Advanced Femtosecond Laser Technology

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1 Next-Generation Ultrafast Photoluminescence  
2 Spectroscopy: Integration of Transient Grating  
3 Optical Gate and Advanced Femtosecond Laser  
4 Technology

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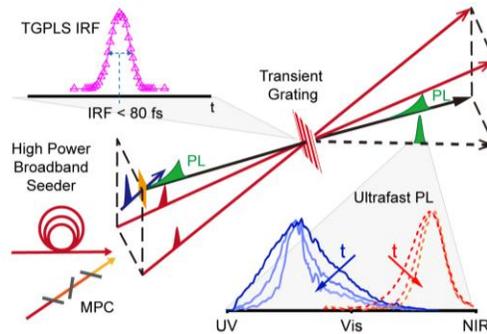
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1 **Abstract:**

2 We demonstrated a high-performance ultrafast broadband time-resolved photoluminescence  
3 (TRPL) system based on the transient grating photoluminescence spectroscopy (TGPLS)  
4 technique. The core of the system is a Kerr effect-induced transient grating (TG) optical gate  
5 driven by high repetition rate ultrashort laser pulses at 1030 nm with micro-Joule pulse energy.  
6 Satisfying the demands of spectroscopy applications, the setup achieves high sensitivity, rapid  
7 data acquisition, ultrafast time resolution, and a wide spectral window from ultraviolet to near-  
8 infrared. The time resolution can be further improved to achieve <80 fs instrument response  
9 function by employing multiple pulse compression (MPC) technique to temporally compress the  
10 driving pulses. This work presents a new benchmark for ultrafast TRPL.

11

12 **TOC GRAPHICS**



13

14

15 **KEYWORDS** Ultrafast Spectroscopy; Ultrafast Photoluminescence; Photochemistry;  
16 Photophysics; Ultrafast Laser; Nonlinear Pulse Compression; Optoelectronic Materials.

17

1 Optical time-resolved spectroscopies serve as pivotal tools in unraveling the intricacies of  
2 photophysical and photochemical mechanisms by capturing spectral evolution over time. Time-  
3 resolved photoluminescence spectroscopy (TRPL),<sup>1-3</sup> in particular, stands out as a fundamental  
4 and broadly utilized method for delving into photo-induced dynamics. By specifically targeting  
5 emissive species, TRPL offers detailed insights with straightforward data interpretation, making it  
6 invaluable for understanding complex photophysical processes.

7 Recent breakthroughs in ultrafast laser technology have significantly enhanced TRPL's  
8 capabilities. Utilizing femtosecond laser pulses for excitation, alongside nonlinear optical gates as  
9 ultrafast optical shutters, this approach allows researchers to probe photoexcitation processes down  
10 to sub-picosecond timescales.<sup>4-6</sup> This temporal window is critical for capturing the fleeting  
11 moments of photoexcitation dynamics, where many crucial processes unfold.

12 Fluorescence up-conversion spectroscopy (FLUPS),<sup>1,7</sup> a traditional ultrafast TRPL method that  
13 employs sum-frequency generation for gating, exemplifies these advancements. FLUPS offers  
14 exceptional temporal resolution and signal-to-noise ratio (S/N), but it is subject to the narrow  
15 detection bandwidth per measurement dictated by phase-matching conditions.

16 However, the time-dependent spectral evolution captured by TRPL is immensely informative. It  
17 reveals not just population kinetics but also sheds light on various phenomena such as the  
18 identification of excited states,<sup>8</sup> energy transfer mechanism,<sup>9</sup> photo-induced structural changes,<sup>5,10</sup>  
19 and carrier dynamics,<sup>11,12</sup> including cooling and thermalization.<sup>13-15</sup> From a technological  
20 standpoint, addressing the challenges of achieving high time resolution, broad detection  
21 bandwidth, efficient throughput, and superior data quality remains at the forefront of TRPL  
22 research. Enhancing these aspects is pivotal for pushing the boundaries of what can be understood  
23 about the transient states and dynamics of photoexcited materials.

1 The performance of a pulse fluorometry system can be evaluated by considering its signal-to-  
2 noise ratio. Disregarding the noise and quantum efficiency of the detector and the efficiency of  
3 optics in the system, we can have the equation:

$$4 \quad S/N(t, \lambda) = \frac{P(t, \lambda) * Q_G(\lambda) * Q_S(\lambda) * R * T}{[(P(t, \lambda) * Q_G(\lambda) * Q_S(\lambda) + B * Q_S(\lambda)) * R * T]^{0.5}} \quad (1)$$

6  
7 where  $P(t, \lambda)$  is the photoluminescence (PL) photon number in the detection time window,  $Q_G(\lambda)$ ,  
8  $Q_S(\lambda)$  are the time-resolved gate efficiency and camera detection efficiency, respectively,  $B$  is the  
9 signal background,  $R$  is the repetition rate of the pulsed laser system, and  $T$  is the integration time  
10 of the detector. The inherent challenge in ultrafast TRPL spectroscopy stems from the inherently  
11 low  $P(t, \lambda)$  within the ultrafast time window. The optimization of the S/N through gating methods  
12 and the choice of light sources is key to obtaining high-quality TRPL data.

13 Currently, second and third-order nonlinear gates are utilized in broadband ultrafast PL  
14 spectroscopy. Broadband upconversion,<sup>16–18</sup> characterized by its low background and wide  
15 detection bandwidth that spans from 420 to 750 nm, faces limitations on the selection of gate  
16 wavelength and nonlinear crystals due to stringent phase-matching conditions. The PL-seeded  
17 noncollinear optical parametric amplifier<sup>19–21</sup> provides a detection bandwidth of up to 150 nm in  
18 the visible range. However, the interaction between quantum noise and the pump introduces  
19 significant noise to the amplified signals, which further impairs the sensitivity of the system.<sup>22</sup> The  
20 third-order Kerr effect<sup>4,23–26</sup> offers an even wider detection bandwidth and greater flexibility in the  
21 choice of gate medium and laser wavelength.<sup>4,24,27</sup> However, the conventional optical Kerr gate is  
22 limited by the imperfection of polarizer pairs, which suffers from a strong signal background  
23 caused by leakage of long-lived fluorescence.

1 In addition to the gate methods, the selection of ultrafast light sources is also crucial for the  
2 performance of TRPL. Equation 1 indicates that the S/N of TRPL measurement can be  
3 dramatically improved by employing a high repetition rate laser source. Unlike pump-probe-based  
4 ultrafast spectroscopy methods, TRPL can adapt to high repetition rate lasers without the  
5 constraints of camera readout rates and the complexity of synchronization electronics. This  
6 adaptability has been exemplified using an optical Kerr gate paired with a high repetition rate Ti-  
7 sapphire regenerative amplifier.<sup>4</sup> However, the performance of such systems is often constrained  
8 by the intrinsic background noise of the optical Kerr gate and reduced gate efficiency,  
9 consequently yielding a lower S/N due to the low pulse energy typical of high repetition rate Ti-  
10 sapphire amplifiers.

11 In this study, we present significant advancements in ultrafast PL spectroscopy, including  
12 enhanced sensitivity, accelerated data acquisition, improved ultrafast time resolution, and  
13 expanded spectral detection range. These improvements were achieved simultaneously, marking  
14 a considerable leap forward in the field. Our strategy integrates an advanced ultrafast PL gating  
15 technique with cutting-edge femtosecond light sources, focusing on the application of transient  
16 grating PL spectroscopy (TGPLS).<sup>5</sup> Utilizing a transient grating as the optical gate, TGPLS meets  
17 the contemporary demands for TRPL, including sub-picosecond time resolution and broadband  
18 detection, making it an exceptionally potent tool for investigating photophysical phenomena such  
19 as ultrafast energy transfer,<sup>28</sup> exciton relaxation,<sup>29,30</sup> carrier cooling,<sup>31,32</sup> and singlet exciton  
20 fission.<sup>33-35</sup> Furthermore, the low background noise characteristic of TGPLS, combined with high  
21 repetition rate lasers, provides an optimal setting for high S/N detection, alongside fast data  
22 acquisition.

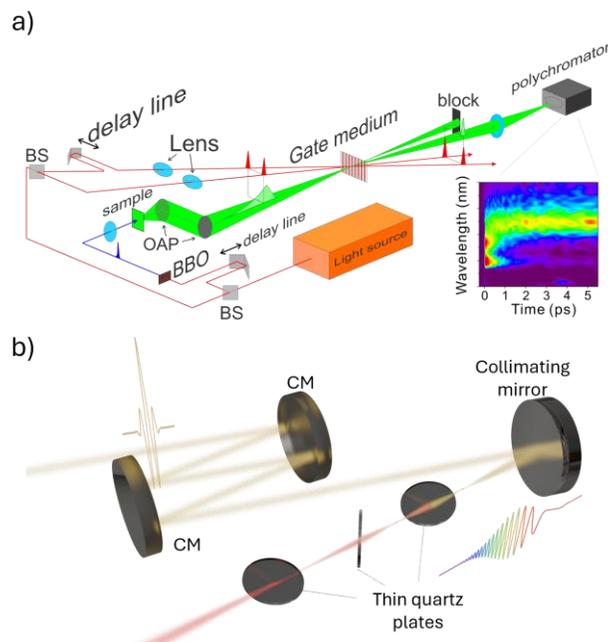
1 Driving the nonlinear optical gate effectively necessitates a laser amplifier capable of delivering  
2 high pulse energy (greater than 10  $\mu$ J). Achieving both high pulse energy and high repetition rates  
3 in standard ultrafast Ti-Sapphire systems is challenging due to the high thermal load inherent from  
4 the  $\sim$ 34% quantum defect<sup>36</sup> and the requirement for expensive cryogenic cooling.<sup>37,38</sup>  
5 Additionally, the lack of efficient diode pumps further restricts the output power to several Watts.<sup>39</sup>  
6 In contrast, diode-pumped Ytterbium ultrafast lasers with  $\sim$ 5% quantum defect can deliver  
7 hundreds of times higher average power<sup>40,41</sup> and have found extensive use in medical and industrial  
8 contexts.<sup>42,43</sup> However, the pulse widths of these sources typically exceed 200 fs or even in the  
9 picosecond regime, strongly limiting the attainable time resolution and detection bandwidth for  
10 spectroscopic applications. In response, we introduce TGPLS systems integrated with advanced  
11 Ytterbium femtosecond amplifiers and nonlinear pulse compressor to address these problems.

12 TGPLS utilizes an ultrafast transient grating as a nonlinear optical gate, providing background-  
13 free and broadband photoluminescence detection. The configuration of the TGPLS system is  
14 depicted in Figure 1a. The layout of Ti-sapphire laser-based TGPLS can be easily adapted to Yb  
15 lasers, owing to the versatility provided by third-order nonlinear interactions of transient grating.  
16 Specifically, the output from the Yb laser serves for both gate and excitation beams. For gate beam  
17 generation, we utilize a 50/50 beamsplitter to split the fundamental light into two horizontally  
18 polarized beams, which are focused and converged within a gate medium to form a transient  
19 grating. To prevent thermal effects and potential damage from the high-power femtosecond laser,  
20 all mirrors directing the gate beams are dielectric mirrors optimized for 1030 nm. For excitation  
21 beam generation, the 515 nm and 343 nm wavelengths are produced through second and third-  
22 harmonic generation in BBO crystals. The intensity of the excitation is controlled using a tunable  
23 attenuator, comprising a thin-film polarizer and a half-wave plate for 1030 nm. Moreover, a

1 suitable short-pass filter is placed after the BBO crystal to eliminate any residual of the  
2 fundamental laser. The excitation beam is then directed/focused onto the sample, and the PL  
3 signals emitted by the sample are captured using an off-axis parabolic mirror (OAP, Edmund  
4 Optics #87-406) and subsequently directed into the gate medium by another OAP (Newport  
5 50332AL). The spatial arrangement of the PL and gate beams adheres to a boxcar geometry,  
6 characterized by a crossing angle of approximately 7 degrees between the two gate beams. These  
7 gated PL signals are then analyzed by a spectrograph (Princeton Instruments SP2150), coupled  
8 with either a deep-cooled, back-side illuminated camera (Finger Lake MicroLine with Hamamatsu  
9 S10140-1108 sensor) or an intensified CCD camera (Princeton Instruments PIMAX 3). To  
10 mitigate interference from the pump beams (343 nm or 515 nm) and the central gate pulses at 1030  
11 nm, appropriate long-pass and short-pass filters (Semrock FF01-790/SP-25 or Thorlabs  
12 FESH0950) are employed.

13 Our study employed two advanced femtosecond amplifier sources to drive the TGPLS system.  
14 The first source, a short-pulse Yb fiber amplifier (Tangerine SP, Amplitude System) is capable of  
15 delivering 200  $\mu$ J pulse energy at up to 175 kHz repetition rate with a central wavelength of 1030  
16 nm. The spectral output, boasting a 60 nm bandwidth at 1030 nm, underwent pulse shaping before  
17 serving as the seed for a fiber chirped pulse amplifier, culminating in short pulse operation<sup>44</sup>. This  
18 resulted in the generation of sub-120 fs pulse width centered at 1030 nm, verified through  
19 frequency-resolved optical gating. The second femtosecond laser source incorporated was a solid-  
20 state Yb:KGW laser (Pharos, Light Conversion) with a pulse duration of 190 fs. To further enhance  
21 the time resolution, we employed nonlinear pulse compression techniques for the first time,  
22 specifically multiple plate compression (MPC).<sup>45,46</sup> This innovative approach enables more precise  
23 investigations into ultrafast photophysical processes.

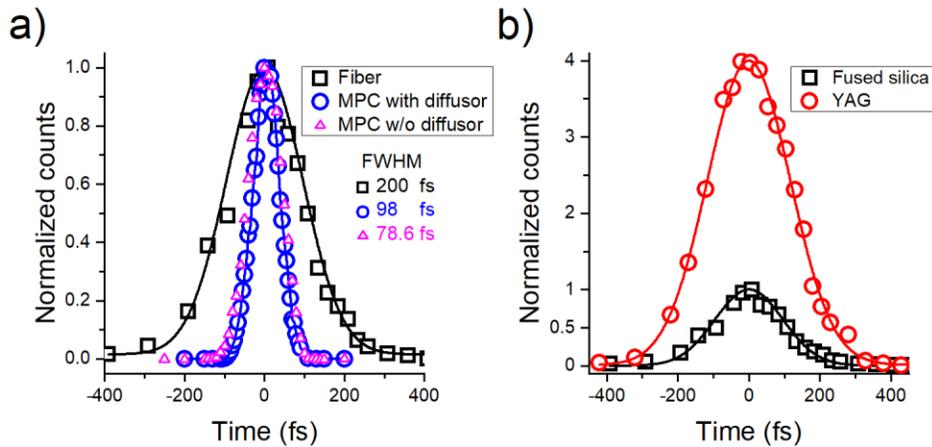
1 MPC (Figure 1b) employs multiple thin plates to temporally compress the ultrafast laser pulse  
2 with high conversion efficiency<sup>45,46</sup>, functioning as a high-brightness, ultrashort, and stable light  
3 source suitable for ultrafast spectroscopy. In our MPC setup, we initially focused the fundamental  
4 laser beam onto a 200- $\mu\text{m}$ -thick quartz plate using a concave mirror with a focal length of 50 cm,  
5 and we positioned three identical quartz plates successively beyond the self-focusing region  
6 produced by the preceding plate. To minimize surface reflection losses, all quartz plates are  
7 oriented at Brewster's angle. We introduced an iris after the plates to selectively block the outer  
8 rim of the beam caused by nonlinear propagation to further enhance the beam homogeneity. The  
9 beam is then collimated by another concave mirror with 40 cm focal length, followed by two  
10 chirped mirrors (CM39 and PC1611, Ultrafast Innovation) for dispersion compensation. The  
11 compressed pulse width is determined using a homemade second harmonic generation frequency-  
12 resolved optical grating (SHG-FROG)<sup>47</sup> technique (Figure S1). The algorithm successfully  
13 retrieves a trace (Figure S1b) and a spectrum (Figure S1c, blue line) that closely resemble the  
14 actual measurements (Figure S1a and Figure S1c, grey pattern), confirming the integrity of  
15 retrieval. The broadband spectrum and nearly constant phase (Figure S1c, blue solid line and red  
16 dashed line, respectively) result in a pulse duration of 48.8 fs (Figure S1d, blue line), approaching  
17 the transform-limited value of 44.6 fs (Figure S1d, grey pattern). The final output pulse energy is  
18 160  $\mu\text{J}$ , reflecting a high throughput of 80%.



1  
 2 **Figure 1.** (a) Scheme of TGPLS. BS, beamsplitter; OAP, off-axis parabolic mirror. (b) Conceptual  
 3 diagram of MPC. CM: Chirped mirror.

4 To evaluate the time resolution of the TGPLS system, we measured the instrument response  
 5 function (IRF)<sup>48</sup> by examining the spatially incoherent light at 515 nm generated by the pump and  
 6 a 1 mm thick fused silica diffusor, which mimics the PL produced in samples from a point light  
 7 source. Figure 2a illustrates the time-resolved gated signals and their fittings for the 515 nm light  
 8 captured by our TGPLS setup. Utilizing the Yb fiber amplifier and a 1 mm fused silica window as  
 9 the gating medium resulted in a full width at half maximum (FWHM) of the laser scattering (Figure  
 10 2a, black squares) of approximately 200 fs, comparable with results from a Ti-sapphire amplifier  
 11 with 100 fs pulse width.<sup>5</sup> Similar measurements were conducted on the MPC-based TGPLS  
 12 (Figure S2), yielding a curve with 98 fs FWHM (Figure 2a, blue circles). Notably, performing the  
 13 measurement without a diffusor yields a signal with 78.6 fs FWHM (Figure 2a, magenta triangles),  
 14 indicating that sub-80 fs temporal resolution is achievable with appropriated pulse chirp pre-

1 compensation. Given that the IRF is predominantly constrained by the laser pulse duration, the  
2 synergistic employment of MPC with industrial-grade laser systems presents a straightforward and  
3 effective approach to bridging the technological gap for high-power, sub-80 fs pulse duration laser  
4 systems in spectroscopic applications.



5  
6 **Figure 2.** (a) Normalized IRF signals (symbols) and fittings (lines) of TGPLS systems with 1 mm  
7 thick fused silica as gate medium and driven by Yb fiber laser (black) and MPC. Blue and magenta  
8 symbols represent IRF signals with and without a diffuser in the MPC configuration, respectively.  
9 (b) Normalized IRF signals (symbols) and fittings (lines) of TGPLS systems driven by Yb fiber  
10 laser with a 1 mm fused silica window (black) and a 1 mm YAG crystal (red) as gate medium,  
11 respectively.

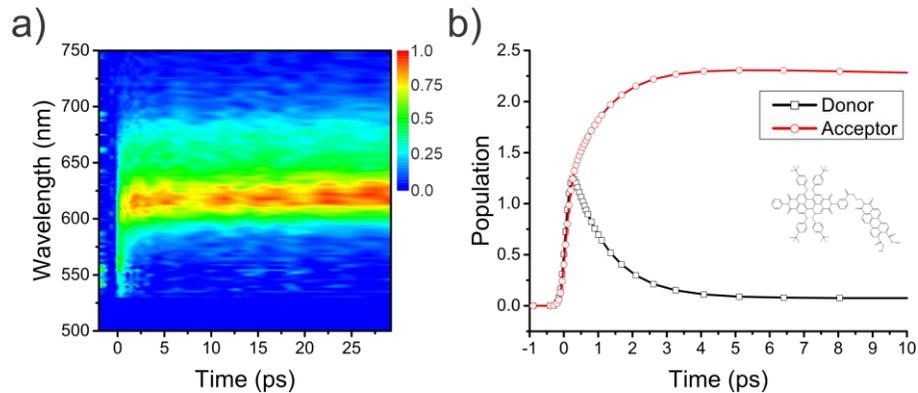
12 In comparison with Ti-sapphire systems, Yb lasers emit photons with lower energy, which  
13 reduces background noise stemming from multiphoton excitations within the gate medium. This  
14 advantage permits stronger gate pulse energy for higher gate efficiency, particularly when using a  
15 high nonlinearity Kerr gate medium with a smaller energy bandgap than fused silica. For example,  
16 utilizing a 1 mm thick undoped Yttrium aluminum garnet (YAG) crystal (Figure 2b, red circles)  
17 as the gate medium gives a peak value of the gated laser scattering signal approximately fourfold

1 compared to that achieved with fused silica (Figure 2b, black squares) at the cost of degraded  
2 temporal resolution (increased from 200 fs to 250 fs). Gate mediums such as CS<sub>2</sub> and Benzene<sup>4</sup>  
3 have the potential to further enhance gate efficiency, albeit at the potential cost of degraded time  
4 resolution.

5 To evaluate the capabilities of our Yb laser-driven TGPLS system, we initially investigated the  
6 ultrafast PL dynamics of a perylene multiple chromophore array tetraphenoxy diimide (dimer,  
7 depicted in Figure 3b inset).<sup>9</sup> This dimer was engineered to exhibit high (>99%) intra-molecular  
8 energy transfer efficiency, leading to a rapid decay of donor PL within sub-picosecond to  
9 picosecond timescales. Figure 3a presents the TGPL spectrogram of the dimer dissolved in  
10 chloroform which is measured in a 1 mm optical path cuvette and excited at 515 nm with 1 nJ per  
11 pulse. The data processing procedure included dark current subtraction, wavelength, and intensity  
12 calibration, while no smoothing techniques were applied. The spectra distinctly reveal the ultrafast  
13 spectral evolutions, where the swift PL decay within the 550 to 570 nm range indicates the donor's  
14 photoluminescence. By applying global fitting,<sup>9</sup> we can extract the PL kinetics for both the donor  
15 and acceptor. The observed ~1 ps decay time of the donor (Figure 3b, black line) and the  
16 corresponding rise in the acceptor's PL (Figure 3b, red line) align with previously reported findings,  
17 affirming the high intra-molecular energy transfer efficiency of the dimer and showcasing the  
18 precision of our TGPLS system in capturing ultrafast PL dynamics.

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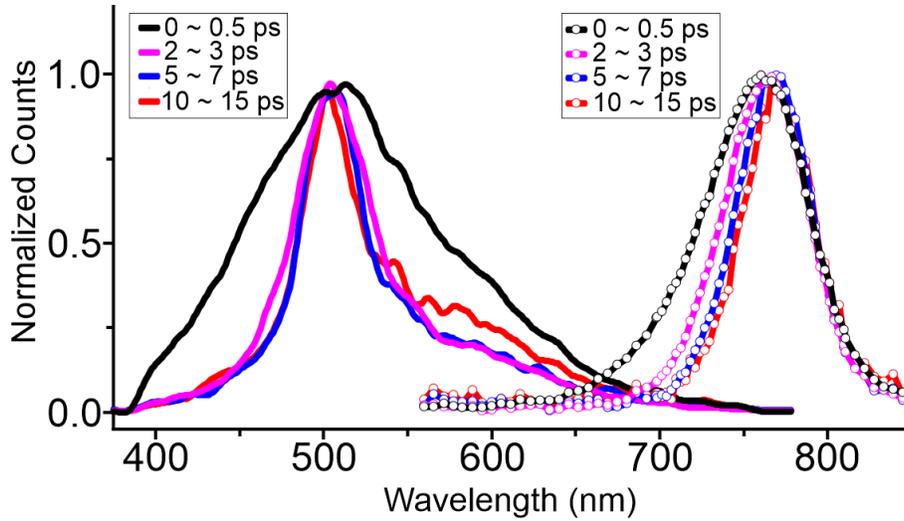


3

4 **Figure 3.** (a) TGPL spectrogram of dimer. (b) The species kinetics of the donor and acceptor. The  
5 inset depicts the molecular structure of the dimer is depicted.

6 To illustrate the broadband detection capabilities in our TGPLS system, we measured the  
7 broadband PL in perovskites. These materials have excellent optical and electronic properties for  
8 photonic applications and are distinguished by their wide band gap tunability and high emission  
9 quantum efficiency.<sup>49-51</sup> Accordingly, the broadband detection capability of our ultrafast PL setup  
10 serves as an excellent tool for investigating photophysical processes in perovskites. Figure 4 (solid  
11 lines) illustrates the transient PL of mixed-halide perovskite,<sup>52</sup> excited at 343 nm (obtained by  
12 third-harmonic generation of the 1030 nm fundamental pulse), spanning a spectral range from 385  
13 nm to 750 nm. These results confirm the effective capture of transient PL in the ultraviolet-visible  
14 (UV-visible) region using the 1030 nm transient grating gate. It is worth noting that the spectral  
15 bandwidth limitations in the UV range are primarily attributed to the optics, grating-based  
16 spectrometer, and CCD camera. Therefore, employing a prism spectrometer and UV-specific  
17 optics could potentially expand the detection bandwidth further into the UV spectrum.

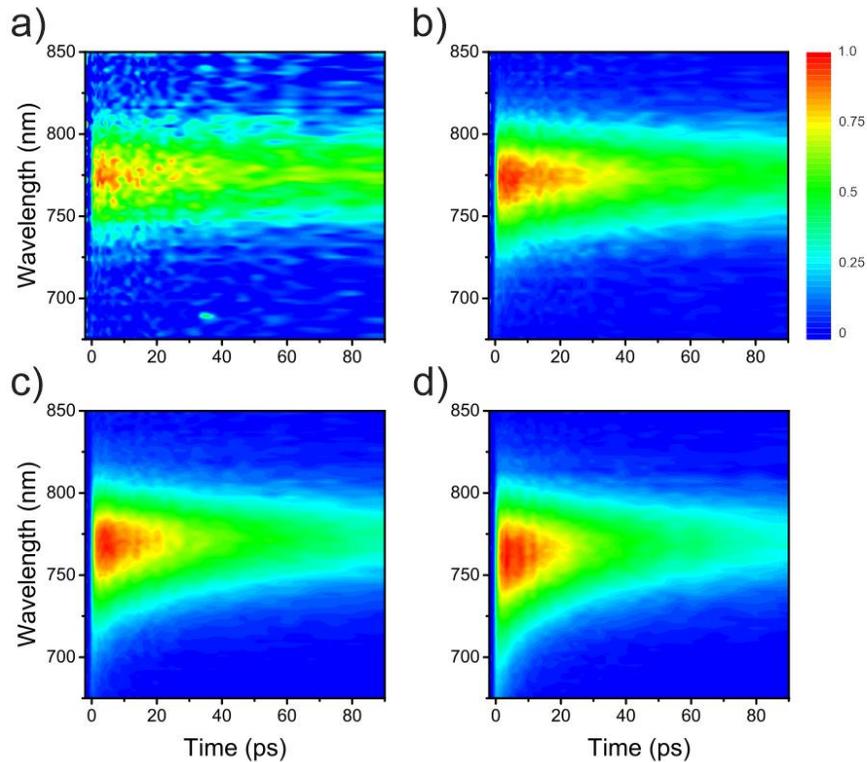
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1  
 2 **Figure 4.** TGPL spectra at selected times of mixed-halide perovskite samples excited at 343 nm  
 3 (solid lines) and  $\text{CH}_3\text{NH}_3\text{PbI}_3$  thin film excited at 515 nm (dotted lines).

4 A notable advantage of employing a 1030 nm fundamental pulse as the gate beam is the extended  
 5 spectral window into the near-infrared (NIR) region, which is particularly relevant for advanced  
 6 materials used in photovoltaic applications<sup>53–55</sup> and light-emitting devices for biomedical  
 7 purposes.<sup>56,57</sup> However, a significant challenge in TRPL with nonlinear optical gates arises from  
 8 the overlap between the NIR emission wavelength and the intense fundamental wavelength of the  
 9 Ti-sapphire laser around 800 nm. To highlight the advantageous NIR detection capability of our  
 10 Yb-based TGPLS system, we explored the ultrafast broadband PL in NIR region from  
 11  $\text{CH}_3\text{NH}_3\text{PbI}_3$  thin-film samples under 515 nm excitation. Figure 4 (dotted lines) and Figure 5  
 12 present the TGPL spectra and spectrogram of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  thin films, revealing spectral evolution  
 13 on sub-picosecond to picosecond timescales under various excitation pulse energies. The initial  
 14 broad PL bandwidth and the subsequent fading on the blue wavelength part reflect hot carriers  
 15 generated from the above bandgap excitation and carrier relaxation from higher excited state to  
 16 the band edge, respectively. As the excitation fluence increases, hot carriers are generated at higher

1 excitation states, which are reflected by the bluer tail shortly after excitation (Figure 5). The high  
2 efficiency of our system facilitated data collection under different experimental conditions. For  
3 example, a 2D spectrogram derived from an average of 4 scans (70 time steps per scan) can be  
4 acquired within 15 minutes. Moreover, our TGPLS system features advanced background  
5 suppression, effectively addressing the background issues in TRPL experiments posed by  
6 materials with high PL quantum yield and extended PL lifetime ( $>10$  ns). The empirical results  
7 demonstrated a virtually neglectable background (1.1% of the maximum signal) at negative time  
8 delays using a backside illumination CCD camera during a 2-second exposure (200,000 shots),  
9 and the low background noise facilitates clear observation of signals under mild pumping condition,  
10 which helps avoid unwanted nonlinear effects from intense excitation, including exciton-exciton  
11 annihilation<sup>58-60</sup> and Auger recombination<sup>61-63</sup>.



12

1 **Figure 5.** The spectrograms of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  thin films excited at 515 nm with excitation fluence  
2 of (a)  $1 \mu\text{J}/\text{cm}^2$ , (b)  $3 \mu\text{J}/\text{cm}^2$ , (c)  $6 \mu\text{J}/\text{cm}^2$ , and (d)  $15 \mu\text{J}/\text{cm}^2$ .

3 Our work presents a high-performance ultrafast broadband PL spectroscopy system that  
4 leverages state-of-the-art ultrafast laser sources and TGPLS. The system achieves sub-80 fs  
5 temporal resolution and a UV-NIR probe window, as demonstrated through IRF and perovskite  
6 measurements. The PL data can provide crucial insights into photophysics and photochemistry,  
7 including processes like carrier cooling and relaxation, as well as energy and charge transfer  
8 mechanisms. This information is invaluable for investigating electronic and electron-phonon  
9 coupling dynamics, which are vital for semiconductor optoelectronic applications, fundamental  
10 material research and advanced material design. The enhanced sensitivity due to high gate  
11 efficiency, low background noise, and high repetition rate of our system enables experiments  
12 conducted under sub-nJ excitation pulse energy without incurring nonlinear artifacts from samples.  
13 Additionally, TGPLS offers considerable design flexibility for optimization across diverse  
14 samples and applications. This platform is poised to significantly enhance the exploration of  
15 photoexcitation dynamics in numerous fields, marking a substantial advancement in the  
16 capabilities of TRPL spectroscopy.

17

## 18 **ASSOCIATED CONTENT**

### 19 **Supporting Information**

20 The Supporting Information is available free of charge at

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22 [journal/download.aspx?id=171156&guid=1004fb81-2aab-4b5e-9d27-6c7d854cb11b&scheme=1](https://www2.cloud.editorialmanager.com/lpor-journal/download.aspx?id=171156&guid=1004fb81-2aab-4b5e-9d27-6c7d854cb11b&scheme=1)

1 FROG measurement results and setup diagram for TGPL setup based on MPC: ([PDF](#))

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16

## 17 **Author Contribution**

18 S.-D.Y. and K.C. proposed the concept. M. H, J. M. H., S.-D.Y., and K.C. supervised the work.  
19 C.-Y.L., Z.-C.J., I. W., B.-H. C., C.-H. L., and K.C. established the spectroscopy setup, carried  
20 out and analyzed the spectroscopy measurement. L.L., Y.Z., B.-H. C., and C.-H. L. supported the  
21 ultrafast laser sources. L.-S. C. provided the perovskite samples. C.-Y.L., Z.-C.J., and K.C. wrote  
22 the manuscript. All authors discussed and revised the manuscript.  
23

## 24 **Notes**

25 The authors declare no competing financial interests.

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