

## RESEARCH ARTICLE

# High-Sensitivity Visualization of Ultrafast Carrier Diffusion by Wide-Field Holographic Microscopy

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Ultrafast transient microscopy is a key tool to study the photophysical properties of materials in space and time, but current implementations are limited to  $\approx 1\text{-}\mu\text{m}$  fields of view, offering no statistical information for heterogeneous samples. Recently, we demonstrated wide-field transient imaging based on multiplexed off-axis holography. Here, we perform ultrafast microscopy in parallel around a hundred diffraction-limited excitation spots over a  $\approx 60\text{-}\mu\text{m}$  field of view, which not only automatically samples the photophysical heterogeneity of the sample over a large area but can also be used to obtain a 10-fold increase in signal-to-noise ratio by computing an average spot. We apply our microscope to study the carrier diffusion processes in methylammonium lead bromide perovskites. We observe strong diffusion due to the presence of hot carriers during the first picosecond and slower diffusion afterward. We also describe how many-body kinetics can be misleadingly interpreted as strong diffusion at high excitation densities, while at weak excitation, real diffusion is observed. Therefore, the vast increase in sensitivity offered by this technique benefits the study of carrier transport not only by reducing data acquisition times but also by enabling the measurement of the much smaller signals generated at low carrier densities.

## Introduction

Characterization of charge carrier transport processes in inorganic, organic, and hybrid semiconductors is the key for the development of next-generation photovoltaic devices [1]. Carrier diffusion constants and lengths can be measured with various experimental techniques, such as photoluminescence (PL) quenching [2,3], Hall effect [4], ultrafast terahertz spectroscopy [5,6], transient grating [7,8], and microwave photoconductivity [9,10]. However, most of these approaches rely on observables indirectly related with the diffusion combined with a model, resulting in large discrepancies among their results [11]. On the other hand, time-resolved PL and transient absorption (TA) microscopy provide a direct way to image exciton and carrier diffusion in real space and real time. While PL has temporal resolution limited to the picosecond time scale and is sensitive to bright excited states only, TA microscopy enables the observation of ultrafast diffusion phenomena down to the femtosecond time scale and is also sensitive to nonemitting states.

State-of-the-art TA microscopes used to study carrier diffusion in organic and inorganic materials all rely on photoexcitation of a single diffraction-limited spot and probe the response of the sample around that spot either with a wide-field or with a scanning probe configuration. Since only a spatially confined area of the sample is excited, the dynamic spread of the TA signal around this spot is indicative of diffusion of the photo-generated carriers. However, because of the limited frame rates of multipixel camera sensors, most TA microscopes use confocal designs, exploiting single-pixel detectors and fast raster scanning [12–17]. Wide-field cameras have been used only with low-repetition-rate ( $< 1$  kHz) laser pulses [18] or by compromising shot-noise limited detection and acquiring images at a frame rate much lower than the laser repetition rate [19–24]. This limits the sample area that one can measure within reasonable times to a few squared micrometers and prevents one from capturing large-scale heterogeneities of the sample in a single image. To determine the average properties of the material, it is therefore often necessary to average data from measurements sequentially performed on different parts of the sample

**Citation:** Hörmann M, Visentin F, Zanetta A, Osmond J, Grancini G, van Hulst NF, Liebel M, Cerullo G, Camargo FVA. High-Sensitivity Visualization of Ultrafast Carrier Diffusion by Wide-Field Holographic Microscopy. *Ultrafast Sci.* 2023;3:Article 0032. <https://doi.org/10.34133/ultrafastscience.0032>

Submitted 2 January 2023

Accepted 30 May 2023

Published 21 June 2023

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[16,17,22,24], substantially increasing data acquisition times and leaving open the question of how representative of the entire sample the few measured spots are.

Recently, we have introduced an ultrafast holographic transient (UHT) microscope, which overcomes the camera frame-rate limitation by exploiting off-axis holography to decouple the signal demodulation frequency from the frame rate of the camera and, thus, enabling shot-noise limited, large field-of-view transient microscopy [25]. Here, we exploit the wide field of view provided by the UHT microscope to photoexcite the sample in parallel with a hundred diffraction-limited spots, yielding diffusion data around all of them in a single measurement. This approach presents 2 major advantages: (a) The carrier diffusion properties can be characterized for hundreds of spots simultaneously covering a large area, experimentally assessing the effect of spatial heterogeneity on the photophysics of the sample; (b) when only the average behavior is desired, the signals from different spots can be averaged to increase the signal-to-noise ratio (SNR).

## Materials and Methods

### Experimental design

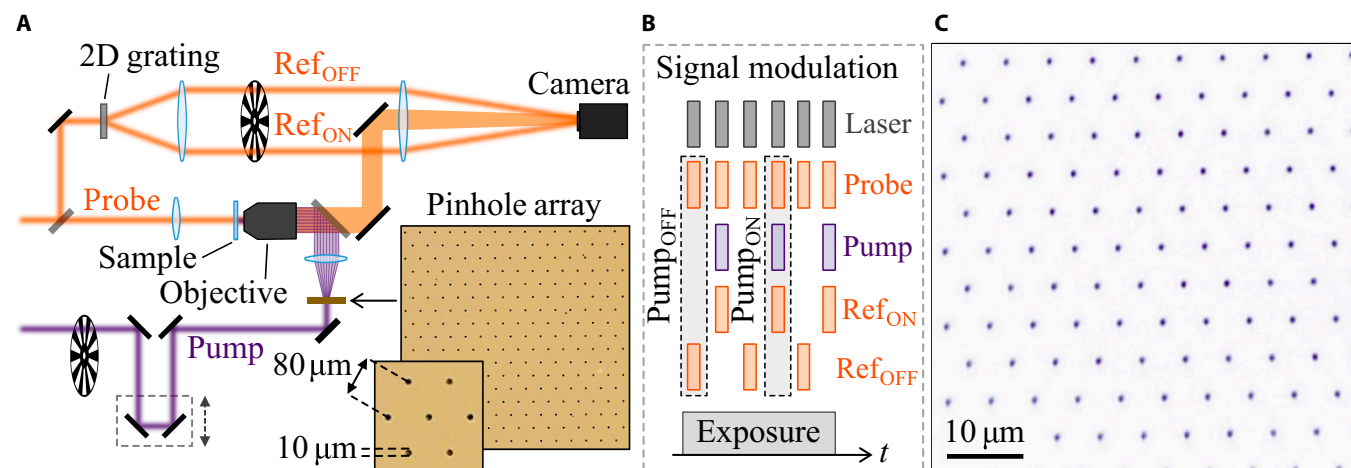
Figure 1A shows a sketch of the experimental setup, the holographic aspects of which have been described in detail before [25,26]. Briefly, it consists of a wide-field TA microscope in which the probe pulse is made to interfere on the camera with 2 reference pulses in a diagonally off-axis multiplexed holography configuration [27]. The references are modulated synchronously with the pump according to the scheme of Fig. 1B: One reference is present only when the pump illuminates the sample, while the other is present only when the pump is blocked. In the spatial 2-dimensional (2D) Fourier transform of the image, the interference terms are displaced from zero momentum (DC), a direct result of the angle between the wave vectors of the reference and image waves (see Fig. S1). The interference term for each wave can be cropped and shifted to zero, after which an inverse Fourier transform reconstructs the amplitude and phase of the original image. Since each reference is associated with either the pumped or the unpumped sample, a pixel-by-pixel subtraction of these images corresponds to the TA image of the whole field of view. The simultaneous

acquisition of pumped and unpumped sample images allows to decouple modulation frequency and camera frame rate, with only the latter being limited by the camera's technology. Thus, the UHT microscope acts as an all-optical wide-field digital lock-in camera, with the advantage of allowing a direct demodulation of the entire 2D image in a single exposure. As a consequence of the high modulation frequency, the resulting TA images show shot-noise limited signal fluctuations [25]. Further, the holographic nature of the technique recovers both amplitude and phase of the signal, which allows postacquisition digital processing of the image, such as computational focusing through the angular spectrum method [28] and hence recording out-of-focus TA dynamics of the sample [26].

We create the aforementioned excitation pattern by propagating the pump beam through an array of small apertures of 10  $\mu\text{m}$  in diameter spread in a hexagonal lattice of a characteristic length of 80  $\mu\text{m}$  on a 200-nm-thick gold foil (inset of Fig. 1A). A relay imaging system composed of the objective lens and an additional lens images the pinhole array into the sample plane and further allows freely adjusting the excitation spot size. Since each hole acts as a small aperture, diffraction effects must be considered, which, in the case of a periodic grating, manifest themselves in the form of the Talbot effect (see the Supplementary Materials for details) [29–32]. In this work, a demagnification of 13.89 is chosen to obtain  $\approx 100$  Gaussian spots with 756-nm full width at half maximum (FWHM) diameter spread across the 60- $\mu\text{m}$  field of view (Fig. 1C).

### Setup description

Spectrally tunable pulses are generated by a home-built non-collinear optical parametric amplifier, pumped by a 2-kHz, 100-fs amplified Ti:Sapphire laser system (Libra 2, Coherent), as described previously [33]. The 400-nm pump pulse is directly obtained by frequency doubling the laser output and modulated at 1 kHz with a mechanical chopper (MC2000B, Thorlabs). A 90:10 beam splitter generates the probe and reference beams from the noncollinear optical parametric amplifier output. The probe is focused onto the sample with a  $f = 200\text{-mm}$  achromatic lens (AC-254-200-A, Thorlabs) to cover the whole field of view. A microscope objective (numerical aperture = 0.8; UPLXAPO20X, Olympus) images the sample onto a complementary metal-oxide semiconductor camera (Basler ace acA720-520um, Basler AG)



**Fig. 1.** UHT experimental setup. (A) Schematic of the experimental setup. Inset: Image of the pinhole array. (B) Signal modulation. (C) Image of the pump illumination pattern obtained using a mirror at the sample position.

with a nominal magnification of  $\times 67$ . Holograms (540 pixels  $\times$  540 pixels) are recorded at 166 fps with an exposure time of 5.6 ms. To allow holographic imaging with femtosecond pulses [34], the reference beam is relay-imaged onto the same camera, and the 2 reference pulses are obtained by placing a 2D diffraction grating (MS-561-600-Y-A, HOLO/OR) in the conjugate image plane to allow interference over the entire camera chip. Two of the first diffraction orders are spatially selected and are modulated at 1 kHz with a second mechanical chopper to achieve the modulation scheme illustrated in Fig. 1B. The time delay between probe and reference pulses is adjusted by maximizing the interference fringe contrast with a mechanical delay line. The variable time delays between pump and probe pulses are obtained by means of a computer-controlled translation stage (M-531.PD1, Physik Instrumente).

### Sample preparation

MAPbBr<sub>3</sub> hybrid perovskite samples were analysed in form of thin films deposited on glass substrates by solution-based spin coating process. The glass substrates (2 cm  $\times$  2 cm) were sequentially cleaned in soap water (Hellmanex III, Hellma), acetone and 2-propanol by ultrasonication for 15 min. Then, a 10-min O<sub>2</sub> plasma cleaning treatment was done to improve their wettability. The MAPbBr<sub>3</sub> solution was prepared by mixing MABr (methylammonium bromide, Greatcell) and PbBr<sub>2</sub> (lead bromide, 99.99%, TCI America) powders in a 1:1 stoichiometric ratio. The powders were then dissolved in a 4:1 DMF:DMSO vol/vol solvent mixture [DMF (*N,N'*-dimethylformamide), 99.8%, Sigma-Aldrich; DMSO (dimethyl sulfoxide), 99.8%, anhydrous, Sigma-Aldrich] and stirred at room temperature for 2 h until complete dissolution. The so-formed solution was then deposited on top of the glass substrates by spin coating at 5,000 rpm for 35 s with an acceleration of about 1,500 rpm/s.

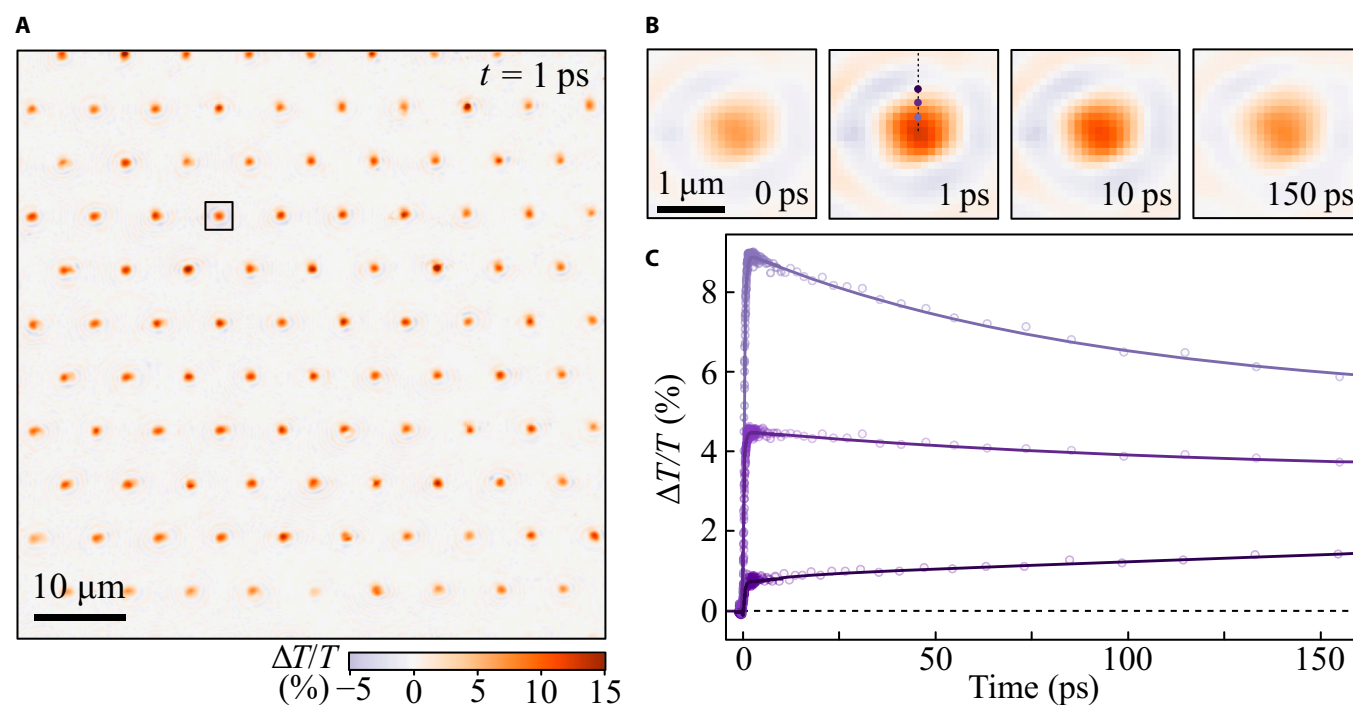
After 6 s from the starting of the process, 150  $\mu$ l of chlorobenzene (99.8%, anhydrous, Sigma-Aldrich) was dripped on the spinning substrates as solvent quenching step, to favor the crystallization of the polycrystalline perovskite thin film. Finally, the films were annealed for 30 min at 100  $^{\circ}$ C.

### Pinhole array fabrication

AZ 5214 E photoresist was spin-coated on a previously cleaned microscope glass substrate and baked for 90 s at 100  $^{\circ}$ C. The design of the hexagonal lattice of Fig. 1A (inset) was exposed with a maskless aligner (MLA 150, Heidelberg Instruments) at a fluence of 100 mJ/cm<sup>2</sup> with an exposure wavelength of 375 nm, developed in AZ 726 MIF for 1 min, and rinsed in deionized water. Subsequently, a 2-nm Ti adhesion layer and 200-nm Au film were deposited on the substrate by thermal evaporation (Lab 18 Thin Film Deposition System, Kurt J. Lesker Co.). The result was left for lift-off in acetone for one night and rinsed in 2-propanol.

### Results and Discussion

To validate the concept, we perform wide-field TA microscopy with  $\approx 150$ -fs temporal resolution on thin films of the methylammonium lead bromide (MAPbBr<sub>3</sub>) perovskites, photo-excited above the bandgap at  $\lambda = 400$  nm and probed at their band edge at  $\lambda = 525$  nm with  $\Delta\lambda_{\text{FWHM}} = 7$ -nm probe pulse bandwidth (Fig. S3). Because of the low exciton binding energy of this material, we are essentially probing free charge carriers [35,36]. Figure 2A shows a typical TA image obtained with this technique, which is equivalent to performing a hundred of confocal TA microscopy experiments in parallel. For each spot, we can study the temporal evolution of the TA image (Fig. 2B): The positive differential transmission ( $\Delta T/T$ ) comes from the



**Fig. 2.** Wide-field TA microscopy of MAPbBr<sub>3</sub> thin films. (A) TA image of a thin film of MAPbBr<sub>3</sub> probed at 525 nm at 1-ps delay. (B) Temporal evolution of a representative spot indicated by the square in (A). (C) TA kinetics of 3 pixels corresponding to different distances from the center of the excited spot. The full lines are the result of a multiexponential fit to help visualize the data.



photobleaching of the band edge transition, and the expansion of the spot size at longer times is due to diffusion of charge carriers. Carrier diffusion can be also observed in the TA kinetics for each pixel (Fig. 2C), showing increasing signals at longer times far from the spot's center.

To extract the charge carrier diffusion properties, we perform a Gaussian fit of the spot in the TA image and extract the diffusion coefficient  $D$  from the mean square deviation (MSD) as [11,14,19,20]:

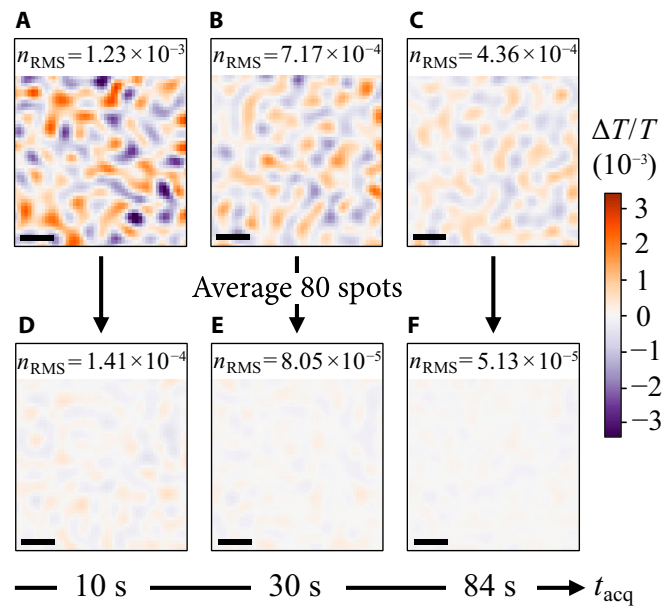
$$\text{MSD} \equiv \sigma^2(t) - \sigma^2(0) = 2Dt \quad (1)$$

where  $\sigma^2(t)$  is the variance of the Gaussian distribution at time  $t$ . To analyze the data, we isolate  $5\text{-}\mu\text{m} \times 5\text{-}\mu\text{m}$  areas around each spot, and 2 different approaches are possible: (a) first, fit the spots individually and then compute the average variance; or (b) first, calculate the average spot and then fit it to extract its variance. With the first approach, we obtain both the average variance and the variance for each spot, allowing us to visualize how the diffusion properties are distributed across the photoexcited spots. On the other hand, averaging the spots allows us to significantly improve the SNR by a factor of the square root of the number of spots, rendering smaller signals detectable with a significant reduction of measurement times, which is convenient for the 2-kHz repetition rate of the laser system used.

To demonstrate the SNR improvement by averaging multiple spots, we analyze the noise at a negative delay time, where the signal is zero and only noise is present, and compare the fluctuations of single spots with the average spot at the same delay. Figure 3A to C shows maps around a single spot at a negative delay as the total acquisition time for the full image is increased from 10 to 84 s, while Fig. 3D to F shows the corresponding maps if the average of 80 spots is taken. By averaging 80 spots, we increase the SNR by a factor of  $\sqrt{80}$ , which is reflected in Fig. 3D showing less noise than Fig. 3C despite the acquisition time being reduced by almost one order of magnitude.

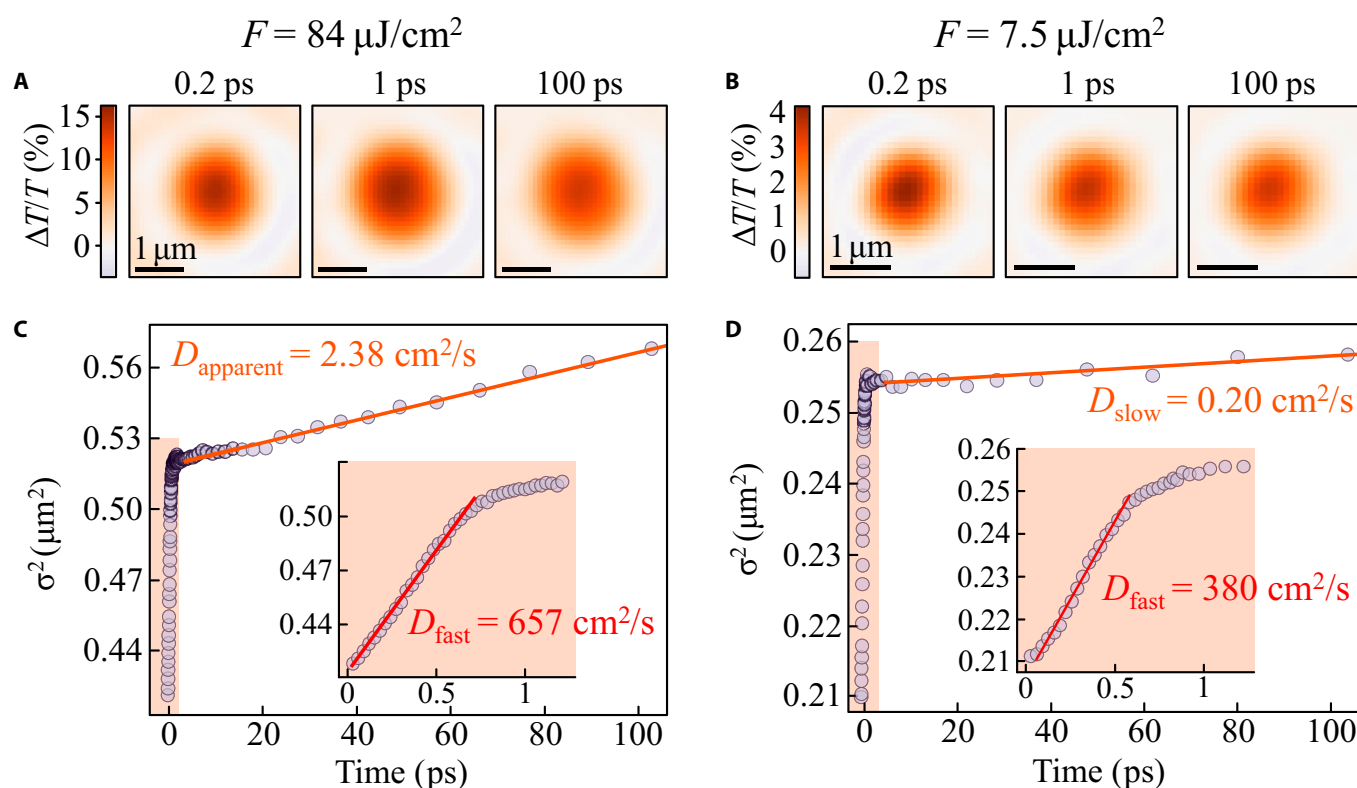
Having validated our technique, we apply it to TA microscopy experiments on a polycrystalline thin film of MAPbBr<sub>3</sub>, whose diffusion properties have been described in detail before [37–41]. The experiments are performed at room temperature for 2 different excitation fluence regimes, namely,  $F = 84$  and  $7.5 \mu\text{J}/\text{cm}^2$ , corresponding to carrier densities of  $2.75 \times 10^{18}$  and  $2.45 \times 10^{17} \text{ cm}^{-3}$ , respectively (see the Supplementary Materials for fluence characterization and carrier density estimation). The spots are first averaged, and the average spot is fitted with a 2D Gaussian function to extract a diffusion coefficient as described previously.

Figure 4A and B shows the time evolution of the average spot for both fluences. While the low-fluence transient spot size in Fig. 4B is comparable to that obtained by directly imaging the excitation spot at the sample position (Fig. 1C), the high-fluence images show much larger spots, reflecting a saturation of the TA signal near the peak due to the high photon flux. Indeed, a power increase by a factor of 11 results in a signal increase by only a factor of  $\sim 4$  (Fig. 4A and B), showing that the high fluence is well inside the saturation regime. Figure 4C and D shows the variance of the charge carrier distribution obtained by fitting the average spot with the 2D Gaussian function. Both fluences show a rapid increase in the variance in the first picosecond, due to hot carrier diffusion with a coefficient of the order of hundreds of square centimeters per second, followed by a slower diffusion of cold carriers, as already observed in similar samples [14]. The same diffusion behavior has also been observed for hot electrons in gold thin films [42]. The fast diffusion, which



**Fig. 3.** Noise reduction by averaging multiple spots for different acquisition times. (A to C) Root mean square noise  $n_{\text{RMS}}$  around a 50-pixel  $\times$  50-pixel area at a negative pump–probe delay. The acquisition time refers to the total time from several individual frames with 5.6-ms integration each. (D to F) Noise of the average of 80 such regions at negative pump–probe delay. Scale bars,  $1 \mu\text{m}$ . The SNR improvement as a function of the data acquisition time is summarized in Fig. S7.

occurs in a quasi-ballistic regime, correlates perfectly with the cooling time of hot carriers and was shown to be absent after excitation without excess energy [14]. Moreover, ballistic ultrafast, sub-100-fs, diffusion has also been reported, which may lead to a larger distribution even around time zero (see Fig. S8) [21,22]. Carrier–carrier interactions dominate the photophysical behavior in such ultrashort time scales, so experiments with higher temporal resolution are required to address it [43]. The diffusion coefficient at later times is computed using Eq. 1 by fitting the points from 5 ps, where effects of the rapid initial expansion are no longer present. This yields  $D = 2.38 \text{ cm}^2/\text{s}$  for  $F = 84 \mu\text{J}/\text{cm}^2$  and  $D = 0.20 \text{ cm}^2/\text{s}$  for  $F = 7.5 \mu\text{J}/\text{cm}^2$ . Previously reported carrier diffusion coefficients of MAPbBr<sub>3</sub> fall in the range of  $D = 0.16$  to  $1.5 \text{ cm}^2/\text{s}$ , measured with time-resolved PL microscopy [37], transient grating spectroscopy [38,39], transient reflectivity spectroscopy [40], and microscopy [41], with the variability attributed to differences in defect density and grain size between the samples [11,41]. The diffusion coefficient that we obtain with low-fluence excitation measurement falls within the lower boundaries of this range, while the one obtained at high fluences, which we name  $D_{\text{apparent}}$ , significantly exceeds reported values, suggesting that many-body interactions are responsible for the difference and more complex models have to be introduced to extract the true value of  $D$  under such conditions [44]. Indeed, moderately high fluences are associated to a substantial amount of Auger recombination (see Fig. S6) [45,46]. The nonlinear dependence of the Auger process on the density of excitons leads to a faster  $\Delta T/T$  decay for the central part of the spot, in which the excitation density is higher because of the Gaussian spatial profile of the pump pulse, resulting in apparent broadening of the distribution, as already reported in other works [18,24]. Since typical excitation densities used in TA microscopy are much larger than those associated with natural sunlight illumination, care must be taken to obtain the diffusion



**Fig. 4.** MAPbBr<sub>3</sub> average diffusion properties. (A and C)  $F = 84 \mu\text{J}/\text{cm}^2$ . (B and D)  $F = 7.5 \mu\text{J}/\text{cm}^2$ . (A and B) Temporal evolution of the average spot over 100-ps time scale. Scale bars, 1  $\mu\text{m}$ . (C and D) Variance of the average spot versus time and extracted diffusion coefficient. Inset: Zoom of the first picosecond.  $D_{\text{fast}}$  describes the fast diffusion of hot carriers that cool down to a value  $D_{\text{slow}}$  in few picoseconds, while the value  $D_{\text{apparent}}$  observed in (C) is the result of many-body interactions occurring at high excitation densities that cannot be captured by a linear model considering only carrier diffusion.

coefficient in a fluence regime below the threshold for many-body effects. Averaging the spots as presented here is a powerful option in this sense, since we have demonstrated the capability of our wide-field TA microscope to reach low levels of noise with short measurement times, even with a low-repetition-rate (2 kHz) laser.

## Conclusion

In summary, we have presented a new ultrafast microscopy technique that simultaneously acquires transient signals of 100 diffraction limited spots covering a sample area of  $\approx 60 \mu\text{m} \times 60 \mu\text{m}$ . This expansion of the spatial sampling by 2 orders of magnitude compared to current techniques is enabled by the all-optical lock-in camera concept, where off-axis holography with 2 reference waves modulated synchronously with the pump is used to encode  $\text{pump}_{\text{ON}}$  and  $\text{pump}_{\text{OFF}}$  images in  $k$ -space [25]. This key step decouples the camera frame rate from the signal demodulation rate, allowing shot-noise limited detection of the transient signals even when using camera frame rates much lower than the laser repetition rate, which was the limiting factor preventing wide fields of view to be obtained in ultrafast TA microscopy. We demonstrate how to profit from this large field of view using a pinhole array to create a structured photoexcitation pattern at the sample position that consists of around 100 diffraction limited spots. As a result, transient imaging at and around all these spots can be obtained in a single measurement. The data can be analyzed spot by spot to obtain statistics, or the individual spots can be averaged before the analysis. The latter option is particularly attractive

in the context of studying the photophysics of new photovoltaic materials, as it offers a route to obtain high-quality data of low signals, allowing lower excitation carrier densities to be used to avoid spurious many-body kinetics [47–49] that are irrelevant under natural sunlight excitation. Finally, we note that the 20 $\times$  objective used in this work has a numerical aperture of 0.8 with a field number of 26.5. Hence, it is capable of imaging a 1.3-mm field of view if a top-end camera with a sufficient number of pixels is used, which would enable us to achieve more than  $10^4$  diffraction-limited excitation spots in a single measurement. Work in this direction is currently in progress.

## Acknowledgments

**Funding:** M.H. and G.C. acknowledge financial support from the Marie Skłodowska-Curie project 812992—“MUSIQ”. F.V., M.L., G.C., and N.F.v.H. acknowledge funding from HORIZON-EIC-2021-PATHFINDEROPEN-01 project “TROPHY” (grant agreement no. 101047137). G.G. and A.Z. acknowledge the “HY-NANO” project that has received funding from the European Research Council (ERC) Starting Grant 2018 under the European Union’s Horizon 2020 research and innovation program (grant agreement no. 802862) and support from the Ministero dell’Università e della Ricerca (MUR) and the University of Pavia through the program “Dipartimenti di Eccellenza 2023–2027”. M.L. and N.F.v.H. acknowledge support through the RTI2018-099957-J-I00 and PGC2018-096875-B-I00 projects funded by MCIN/AEI/10.13039/501100011033/FEDER, the MICINN “Severo Ochoa” program for Centers of Excellence in R&D CEX2019-000910-S, Fundació Privada Cellex, Fundació

Privada Mir-Puig, and the Generalitat de Catalunya through the CERCA program. N.F.v. H. acknowledges the financial support by the European Commission (ERC advanced grant 670949-LightNet). **Competing interests:** The authors declare that they have no competing interests.

## Data Availability

The data reported in this article are freely available upon request to the authors.

## Supplementary Materials

Ultrafast holographic transient microscopy  
Optical characterization of MAPbBr<sub>3</sub> thin films

Fluence characterization

Carrier densities

Ultrafast ballistic diffusion

SNR improvement by averaging multiple spots

Talbot effect

Fig. S1. Holographic recording and image processing.

Fig. S2. Optical microscope image of a thin film of MAPbBr<sub>3</sub>.

Fig. S3. Static absorption spectrum of a thin film of MAPbBr<sub>3</sub>, pump, and probe spectra.

Fig. S4. TA spectroscopy of a thin film of MAPbBr<sub>3</sub>.

Fig. S5. 2D Gaussian fit of the average pump spot of Fig. 1C.

Fig. S6. Fluence-dependent TA kinetics of a thin film of MAPbBr<sub>3</sub>.

Fig. S7. SNR improvement by averaging 80 spots versus acquisition time.

Fig. S8. Ultrafast ballistic diffusion.

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