Transport of charge carriers is at the heart of current nanoelectronics. In conventional materials, electronic transport can be controlled by applying electric fields. Atomically thin semiconductors, however, are governed by excitons, which are neutral electron-hole pairs and as such cannot be controlled by electrical fields. Recently, strain engineering has been introduced to manipulate exciton propagation. Strain-induced energy gradients give rise to exciton funneling up to a micrometer range. Here, we combine spatiotemporal photoluminescence measurements with microscopic theory to track the way of excitons in time, space and energy. We find that excitons surprisingly move away from high-strain regions. This anti-funneling behavior can be ascribed to dark excitons which possess an opposite strain-induced energy variation compared to bright excitons. Our findings open new possibilities to control transport in exciton-dominated materials. Overall, our work represents a major advance in understanding exciton transport that is crucial for technological applications of atomically thin materials.
The mechanical exfoliation of atomically thin nanomaterials has initiated a new research field. Since then, in particular transition metal dichalcogenides (TMDs) have been intensively studied. The family of these truly two-dimensional materials exhibits a strong Coulomb interaction giving rise to the formation of excitons, which are Coulomb-bound electron-hole pairs. Having binding energies of a few hundreds of meV, they dominate the optoelectronic response of TMDs even at room temperature. Recent studies have shown that besides the regular bright excitons accessible in optical spectra, dark excitonic states play a significant role for understanding the optical response and non-equilibrium dynamics in TMDs. Here, the Coulomb-bound electrons and holes either exhibit an opposite spin or are located at different valleys (K, K’, Λ) in momentum space. The required spin-flip or momentum-transfer cannot be provided by photons, making these states dark and difficult to access in optical spectra. Very recently, momentum-dark KA excitons (with the hole localized at the Λ valley) could be visualized in angle-resolved photoemission spectroscopy. Being atomically thin, flexible, and exhibiting strong absorption and ultrafast non-equilibrium dynamics, TMDs have been considered as promising candidates for next-generation optoelectronic applications including light-emitting, detecting, and harvesting devices. Optical excitation, relaxation dynamics and transport of carriers represent the crucial many-particle mechanisms governing the efficiency of such devices. The optical response and non-equilibrium dynamics of excitons in TMDs have been well studied. Exciton transport, however, has still remained elusive. In conventional materials, external electrical fields are used to control the transport of optically excited carriers. However, deeply-bound excitons as neutral particles (consisting of negatively charged electrons and positively charged holes) are only weakly affected by in-plane electrical fields, e.g., via Stark shifts and exciton dissociation. Only in the case of spatially separated interlayer excitons in van der Waals heterostructures, an electrical control of exciton transport has been demonstrated via the energetic tuning allowed by the dipole moment. Thus, strain engineering has been introduced to manipulate the propagation of excitons in atomically thin semiconductors and could be exploited to boost the energy conversion efficiency in photovoltaics devices. TMDs are remarkably sensitive to strain resulting in a large tunability of their energy landscape. In particular, strain was shown to significantly shift exciton resonances. As a result, strain-induced energy gradients allow us to control and manipulate exciton propagation. Excitons have been observed to funnel up to a micrometer range towards spatial regions with the strongest strain gradient, the energy is minimal and a microscopic many-particle theory to investigate exciton funneling in presence of spatially inhomogeneous strain gradients in the exemplary case of a WS$_2$ monolayer. Surprisingly, we find an inverse exciton funneling (anti-funneling) toward spatial regions with low strain (Fig. 1) — opposite to the conventional funneling behavior observed so far. Shedding light on the many-particle processes behind the funneling process, we explain this counter-intuitive behavior. In presence of strain, lattice distortions induce valley-dependent variations of energies, i.e., bright and momentum-dark excitons shift in different directions (Fig. 1c). In particular, the energy of momentum-dark KA excitons increases with strain, and thus this type of excitons funnels opposite to the strain gradient toward spatial regions with minimal strain (Fig. 1e). Our joint experiment-theory study represents a major

**Fig. 1** Dark and bright exciton funneling in an atomically thin semiconductor. a Creation of an inhomogeneous strain profile in a WS$_2$ monolayer via transfer on an array of polymer micropillars. The lowest graph shows the patterned substrate, the middle one the sample topography with the WS$_2$ monolayer, and the top one the resulting strain profile in the experiment. b Illustration of funneling of bright excitons and anti-funneling of dark excitons in the strained material. Dark excitons can be activated by phonons and/or defects making them visible in optical spectra. c Strain-induced shift of dark $X_{\Lambda\Lambda}$ and bright $X_{KK}$ exciton resonances. d,e Opposite shifts of $X_{\Lambda\Lambda}$ and $X_{KK}$ give rise to reverse spatial energy gradients resulting in an unconventional anti-funneling of dark $X_{\Lambda\Lambda}$ excitons away from regions with maximum strain.
**Effects of strain gradient on exciton funneling in WS$_2$ monolayers**

In this study, we investigate the impact of strain gradients on exciton funneling in atomically thin semiconductors, including WS$_2$ monolayers. By inducing inhomogeneous strain through micropillar arrays, we observe a surprising anti-funneling behavior in which excitons drift towards regions of lower strain, opposite to the conventional funneling observed in homogeneous materials.

**Results**

**Spatiotemporal exciton funneling.** We study spatiotemporal exciton dynamics in WS$_2$ monolayers with picosecond time resolution. We pump the monolayer with ultrashort laser pulses ($\approx 200$ fs) at 2.1 eV. An objective lens focuses the laser onto a fixed sample position. By scanning a second objective lens in transmission geometry, we image the PL distribution onto a streak camera. In this way, we achieve a temporal and two-dimensional spatial resolution of the PL (see Methods for details on the experimental setup and measurement routine). From this data, we can observe ultrafast PL movies tracking exciton motion within the strain profile created by micropillars (see the movies in the SI).

**Dark-exciton anti-funneling in WS$_2$.** To explain this intriguing and unexpected anti-funneling behavior, we complement our experiments with microscopic calculations of the spatiotemporal dynamics of excitons. Here, we exploit a generalized drift-diffusion equation for exciton densities $N_i(r,t)$ yielding

$$
\dot{N}_i(r,t) = \nabla \cdot D_i(r) \nabla N_i(r,t) + \frac{1}{k_B T} \nabla \cdot \nabla \nabla \nabla (N_i(r,t) \varepsilon(r))
$$

Here, we explicitly take into account the entire exciton landscape, where the valley index $v$ includes the energetically lowest $s$ states of bright and momentum-dark excitons.

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**Fig. 2** Exciton anti-funneling. a–c Spatiotemporally resolved photoluminescence from an inhomogeneously strained WS$_2$ monolayer upon pulsed excitation at a delay time of 0 ns (orange) and after 1.8 ns (purple). The blue colormap depicts the measured strain gradient induced by the micropillars. d–f Cuts along the $x$ direction (horizontal white dashed line) in figures a–c. The dashed blue line shows the strain distribution along the $x$ direction. We find that excitons do not propagate in the absence of a strain gradient (spot A). Surprisingly, they funnel towards lower-strain regions (opposite to conventional funneling), when the excitation spot is subject to a considerably large strain gradient (spots B and C). The supplementary material includes PL movies further illustrating this surprising anti-funneling behavior.

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**Advance for the microscopic understanding of exciton funneling in atomically thin semiconductors including the entire landscape of bright and dark excitons.**

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$$

---

**Results**

**Spatiotemporal exciton funneling.** We investigate spatiotemporal exciton dynamics in a strongly spatially dependent strain profile. To create such a strain gradient, we use the concept of a nanopatterned surface, which has previously been exploited to create single-photon emitters in TMD monolayers.

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$$

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**Discussion**

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**Conclusion**

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**Data availability**

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**References**

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**Supplementary information**

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**Author contributions**

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**Competing interests**

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**Clusters of highly cited papers**

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**References**

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Fig. 3 Exciton anti-funneling: experiment-theory comparison. a,b Spatial strain profiles at the spots B and C (illustrated in Fig. 2b,c). c,d Experiment: Spatiotemporal evolution of the PL signal imaged with the streak camera after an optical excitation at the spots B and C. e,f Theory: Temporally and spatially resolved dynamics of momentum-dark KΛ excitons obtained by solving the drift-diffusion dynamics [cf. Eq. (1)] assuming the same conditions as in the experiment. g Spatially dependent strain profile (black line, extracted from the experiment, cf. Fig. 2e) and theoretically predicted diffusion coefficient (green line) and h energy of bright KK and dark KΛ excitons. The position of the investigated spots B and C are denoted by thin vertical lines. Explanation for the anti-funneling behavior observed in the experiment (Fig. 2).

Previous studies have shown that the interaction with high-dipole molecules or phonons can brighten dark exciton states and make them visible and even dominate the low-temperature PL of tungsten-based TMDs. Through funneling, dark KΛ excitons can reach spatial regions, where the spectral separation of dark and bright states becomes small, significantly enhancing the phonon-driven intervalley scattering. As a direct consequence, a large amount of originally dark KΛ excitons scatters into bright KK states and becomes visible in PL spectra (cf. Figs. 1b and SI).

Additional defect-induced activation mechanisms could further boost the effect. More details on the theoretical approach (including the total decay rate $\gamma$ and intervalley thermalization $N_{th}^{K\Lambda}$ appearing in Eq. (1) as well as funneling-induced activation of dark excitons) can be found in the Supplementary Information.

To further illustrate the anti-funneling behavior, we show the spatiotemporal evolution of the experimentally measured PL signal in Fig. 3c, d. We focus on spatial cuts along the x axis (cf. the dashed white line in Fig. 2b, c) for optical excitation close to the maximum strain gradient (corresponding to spot B and C). For a better orientation with respect to the funneling direction, we also plot the spatially dependent strain profile at these spots (cf. Fig. 3a, b). Opposite to the expected behavior, we see a clear leftward propagation away from regions with maximal strain, i.e., an anti-funneling of excitons. The propagation is more pronounced in the spot C due to the presence of a higher strain gradient. A quantitative analysis shows that excitons propagate for a few hundreds of nm within the first nanoseconds. This is sensitive to the strain profile, i.e., at spots with a larger strain gradient, the drift efficiency is higher (cf. the Supplementary Material). The anti-funneling length is in the same range as the conventional exciton propagation observed so far in literature.

The PL at room temperature is usually associated with bright excitons. However, these states cannot explain the observed anti-funneling behavior, since they propagate toward regions of high strain, where their energy is minimal (cf. Fig. 1c). In contrast, the energy of momentum-dark KΛ excitons decreases with strain and is thus the driving force for the observed anti-funneling behavior. To illustrate this, we show in Fig. 3e, f the spatially and temporally resolved occupation $N_{K\Lambda}$ of momentum-dark KΛ excitons in direct comparison to the PL measurements in Fig. 3c, d. The theoretical data is obtained by numerically evaluating the drift-diffusion Eq. (1) exactly at the conditions of the experiment, i.e., assuming the same spatially inhomogeneous strain profile shown in Fig. 2e, f. In both experiment and theory, we find a clear exciton motion to the left of the excitation spot, i.e., towards spatial regions with a smaller strain gradient (cf. the strain profile in Fig. 3a, b). This indicates that the surprising anti-funneling observed in the PL can indeed be directly ascribed to the propagation of dark excitons. This can only occur at excitation spots, where momentum-dark KΛ excitons are energetically lower or close to the bright KK states (Fig. 3h) and thus exhibit a considerable occupation. We find a more pronounced funneling after optical excitation at location C closer to the maximum of the strain gradient, since here the spatial variation of strain is steeper, resulting in a stronger drift force $-\nabla E_{K\Lambda}(r)$. In addition, we observe both in experiment and theory that interestingly the funneling is initially faster and then it slows down approaching a stationary value for the propagation. This effect is well reproduced by the theoretically predicted evolution of dark-
Our results demonstrate that strain engineering is an efficient tool to manipulate the propagation of neutral excitons. In particular, the interplay of dark and bright states offers new possibilities for tuning the optical response, dynamics, and transport, which are key processes for the realization of novel ultrathin and flexible optoelectronic devices.

Methods

Theoretical methods. The strain-dependent exciton energies entering Eq. (1) are obtained starting from the unstrained single-particle dispersion relation, adding strain-induced shifts and solving on microscopic footing the Wannier equation:

$$\frac{\hbar^2 k^2}{2m} \Psi_r(k) - \sum_q W_q \Psi_r(k + q) = E_r^0 \Psi_r(k),$$

(2)

where $m_r$ is the reduced exciton mass in exciton valley $r$. Here, $E_r^0$ corresponds to the exciton binding energy, which depends weakly on strain via $m_r$, cf. Table in Supplementary Information for the list of all appearing parameters. Furthermore, $\Psi_r(k)$ describes the excitonic wave function in momentum space, while $W_q$ is the Coulomb interaction obtained via a modified form of the potential for charges in a thin film of thickness $d$ surrounded by a dielectric environment. Taking into account anisotropic dielectric tensors and solving the Poisson equation with the boundary conditions described above yields $W_q = V_q \delta_{q,\mathbf{G}}$ with the 2D-Fourier transformed Coulomb potential $V_q$ and a non-local screening.

The exciton energy reads $E_r^0 = E_r^{\text{hBN}} - E_r^0$, where $E_r^{\text{hBN}}$ is the minimal energy separation between conduction and valence band in valley $r$. This quantity depends crucially on strain, see Table in Supplementary Information. Due to large spectral separations, we focus on the energetically lowest 1s states of the 6 most relevant exciton valleys (KK, KK, KA, KA, FK, FK). These states exhibit a center-of-mass momentum $\mathbf{Q}$ such that the state in valley $r$ and momentum $\mathbf{Q}$ has an energy $E_r^\mathbf{Q} = E_r^0 + \hbar^2 \mathbf{Q}^2/(2M_r)$ with $M_r$ being the total exciton mass. These states depend strongly on strain mostly via $E_r$, hence they are space-dependent in the case of a spatially-inhomogeneous strain profile, i.e., $E_r^\mathbf{Q} \neq E_r^0(G)$. As explained in the main text, this induces (i) a spatially-dependent diffusion coefficient $D(\mathbf{r})$ that has been microscopically evaluated in Ref. 26 (cf. Supplementary Information for more details) and (ii) a drift force $-\mathbf{v}(E_r)$ with a generalization of the electrical mobility provided by $D(\mathbf{k})\alpha$ according to the Einstein relation. It is well known that trions can also be observed in the presence of space-dependent strain profiles25, however, we focus here on the intrinsic undoped regime. The impact of spin-dark states has also been neglected, since they are expected to only lead to quantitative changes due to their similar character as KK excitons considered here.

Discussion

By combining spatiotemporal photoluminescence experiments with quantum-mechanics many-particle modeling we provide deep microscopic insights into the transport of excitons in atomically thin semiconductors. Creating a strain gradient via nanopatterned surfaces and investigating exciton propagation via spatiotemporal photoluminescence, we find an anti-funneling behavior in WS2 monolayers. Complementing our experiments with microscopic theory, we attribute the anti-funneling to dark KA excitons, which possess an opposite strain-induced energy variation compared to bright excitons and thus propagate in the opposite direction. To further prove that momentum-dark KA excitons are the origin of the inverse funneling observed in our experiments, we have performed similar measurements on a molybdenum diselenide (MoSe2) monolayer, cf. Fig. 4 and Supplementary Video S2. Contrary to the case of WS2, we find a conventional exciton funneling, i.e., the PL signal moves from low- to high-strain regions (cf. orange and purple area indicating initial and final distributions, respectively). This observation is in full agreement with our theoretical predictions, since in MoSe2 dark KA excitons are located above the bright states. Therefore, dark states are weakly occupied and are expected to have only a minor impact on exciton funneling (cf. Supplementary Information). Remarkably, a quantitative analysis of the funneling behavior shows an efficient propagation of excitons exceeding 1 μm within 0.8 ns (cf. Supplementary Information), which is beyond the reported values of some hundreds of nanometers in previous studies performed on WSe2.20,21.
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Author contributions
R.R., S.B., R.P.C., and E.M. developed the theoretical model for exciton funneling. R.S., I.N., J.K., J.A.P., R.S., S.M.d.V., and R.B. devised and performed the spatiotemporal PL measurements. All authors contributed to the writing of the manuscript.

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