Dynamics and efficient conversion of excitons to trions in non-uniformly strained monolayer WS₂

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In recent years, there has been ongoing effort in achieving efficient transport of excitons in monolayer transition metal dichalcogenides subjected to highly non-uniform strain. Here we investigate the transport of excitons and trions in monolayer semiconductor WS₂ subjected to controlled non-uniform mechanical strain. An atomic force microscope (AFM)-based setup is applied to actively control and tune the strain profiles by indenting the monolayer with an AFM tip. Optical spectroscopy is used to reveal the dynamics of the excited carriers. The non-uniform strain configuration locally changes the valence and conduction bands of WS₂, giving rise to effective forces attracting excitons and trions towards the point of maximum strain underneath the AFM tip. We observe large changes in the photoluminescence spectra of WS₂ under strain, which we interpret using a drift-diffusion model. We show that the transport of neutral excitons, a process that was previously thought to be efficient in non-uniformly strained two-dimensional semiconductors and termed as ‘funnelling’, is negligible at room temperature, in contrast to previous observations. Conversely, we discover that redistribution of free carriers under non-uniform strain profiles leads to highly efficient conversion of excitons to trions. Conversion efficiency reaches up to about 100% even without electrical gating. Our results explain inconsistencies in previous experiments and pave the way towards new types of optoelectronic devices.

Two-dimensional materials from the class of transition metal dichalcogenides (TMDCs) are actively considered for applications in photonics, electronics and optoelectronics. TMDCs feature a direct bandgap in the monolayer limit, exhibit an unusual spin–valley locking and can host tunable single-photon emitters. Prototype TMDC-based electronic devices, including transistors, p–n junctions and solar photoconversion devices, have already been demonstrated. Moreover, the high Young’s modulus of TMDCs (~170 N m⁻¹ for monolayer WS₂) invites applications of these materials in flexible electronics.

The physical properties of TMDCs change under mechanical strain. In the simple case of constant uniaxial strain, the bandgap energy is reduced by ~50 meV per % (refs 15,20) and the phonon-assisted coupling is altered too. The phonon-assisted coupling is altered too. The bandgap reduction is double, ~100 meV per %, for uniform biaxial strain due to the added effect of the strain component along two axes. In conventional semiconductor materials, the advent of ‘strain engineering’, controlled strain-induced modification of the bandgap, allowed directing flows of excitons in quantum wells and led to the performance improvement of strained silicon metal–oxide–semiconductor field-effect transistors. In the same vein, strain engineering of TMDCs has been realised. The theoretical proposal of ref. 18 considered a spatially varying strain profile induced in a suspended TMDC membrane by the sharp tip of an atomic force microscope (AFM). In such a configuration, a force proportional to the bandgap energy gradient acts on a photoexcited electron–hole pair (an exciton), transporting it to the centre of the strain ‘funnel’ at the location of the tip (Fig. 1b). Two features make this setup attractive for efficient solar photoconversion. First, spatial variation of the local bandgap broadens the absorption spectrum of the TMDC. Second, excitons transported to the location of the AFM tip can be efficiently extracted and converted to electrical current, although lower photovoltage is expected due to the reduced bandgap. Experimental signatures consistent with the funnel effect have been previously observed in wrinkled few-layer MoS₂ (ref. 24) and monolayer MoS₂ nanobubbles. Nevertheless, these previous experiments did not allow for induction of predictable strain profiles, dynamic control and tunability of the strain magnitude, or quantitative analysis of the funneling efficiency. Because of that, the mechanisms governing transport and dynamics of excitons in a non-uniform strain profile have not been fully investigated.

Here, we experimentally realized the setup originally proposed by ref. 18. Highly non-uniform and in situ tunable strain profiles are induced in suspended monolayer WS₂ by the tip of an all-electrical AFM under both ambient and vacuum conditions, while optical fingerprints of funneling are collected by a high-resolution optical system. We observe large changes in the photoluminescence spectra as a function of strain. By comparing our results to a simple drift–diffusion model, we decouple the contributions of two effects: funneling of excitons and trions (charged excitons) and funneling of free charge carriers. Contrary to prior expectation, we find that funneling of excitons is a very inefficient process, with less than 4% of photoexcited excitons reaching the funnel centre even at the highest achievable strain. In contrast, funnelled free carriers are found to dominate optical spectra by binding to neutral excitons to form trions with a conversion efficiency of up to about 100%. Taken together, our results explain inconsistencies in previous experiments and open a new pathway towards ultra-efficient TMDC-based photoconversion and optoelectronic devices.

Results
To study a controllably strained TMDC, we require a suspended sample that can be approached from one side by an AFM tip while allowing optical access for excitation and detection from the other side. To produce suspended samples, we perforated holes with different radii ranging from 0.5 to 2.0 μm in a 50-nm-thick silicon nitride (Si₃N₄) using focused ion beam milling. Monolayer WS₂ was mechanically exfoliated onto a polydimethylsiloxane film and transferred on top of the holes using a dry-transfer
technique (Fig. 1c). Full details on the fabrication of the samples is given in the Supplementary Information.

To fulfil the main experimental challenge of this work—the induction of well-defined and controlled non-uniform strain profiles in a suspended monolayer of WS$_2$—we developed a novel AFM-based apparatus (Fig. 1a). A suspended monolayer is indented by an AFM tip from below, while being optically excited from above. Critically, the AFM cantilever is based on a piezo-resistive technology, allowing all-electric motion actuation and deflection readout. All-electrical operation of the cantilever allows for optical excitation/detection of WS$_2$ without the disturbing effect of laser sources normally employed in conventional AFMs. The described AFM setup is capable of topographic imaging both in tapping (Fig. 1d) and contact mode, recording force-distance curves for nanoindentation experiments, and, most importantly, applying a constant force on the suspended membrane while optical measurements are performed. The stability of the AFM under constant force application is 5–20 nN in the force magnitude, and 1–2 nm in the z-piezo displacement. Last but not least, the whole AFM system, being all-electrical and compact, can be incorporated into an optical cryostat, allowing additional measurements of the system under vacuum.

The optical part of the setup consists of a high-resolution objective (numerical aperture 0.75) and a periscope mounted on a scanning stage capable of nanometre-resolution positioning. The sample is excited by a continuous-wave laser source at 532 nm (spot size full-width at half-maximum (FWHM) 600 nm, power 30 μW unless specified otherwise) and the photoluminescence is directed to a spectrometer (Andor). Overall, our unique AFM/spectroscopy setup combines full AFM and full spectroscopic characterization capabilities. This combination is critical for the investigation of exciton transport in non-uniform strain profiles under both ambient and vacuum conditions.

We start by spatially locating a pristine WS$_2$ monolayer suspended over a hole. This is done using tapping-mode AFM imaging to avoid puncturing delicate monolayers. After locating the centre of the suspended portion of the WS$_2$ monolayer, we perform a nanoindentation experiment at that location using a commercial AFM (Supplementary Information). From this data, we extract the Young's modulus ($\mu_y$) and pre-tension ($\sigma_Y$) of the membrane, the parameters necessary for the accurate determination of the strain profile (Supplementary Information). Once preliminary characterization is complete, the sample is controllably indented by locking the proportional-integral-derivative (PID) loop at the desired force value, with the AFM tip still positioned at the centre of the membrane (determined from maximal redshift of the photoluminescence with respect to zero strain), and photoluminescence spectra versus strain are acquired.

Non-uniform strain profiles are parameterized by a single value, the maximum strain $\varepsilon_{\text{max}}$ that is reached at the middle of the membrane (Supplementary Information). We limited our investigation to $\varepsilon_{\text{max}} \approx 2.8\%$ as most measured samples ruptured when strained higher than 3%. We emphasize that this value is not the intrinsic breaking strain of TMDs, which has been measured to be higher than 10% (ref. 15). Unfortunately, in our setup, the membrane has to be kept under strain for an extended period of time for photoluminescence measurements. Unavoidable fluctuations in the PID loop and thermal drifts lead to rupturing at strain smaller than the intrinsic value. Figure 2a shows the evolution of the photoluminescence spectra of sample A as it is progressively indented. At zero strain, the photoluminescence spectrum is described by a non-symmetric Gaussian peak (Supplementary Information). As the strain is increased, this peak broadens and finally evolves at high strain into a two-peak structure: the redshifted ‘red’ peak and higher-energy ‘blue’ peak. A similar strain-dependent two-peak structure is seen in every measured sample, for example, in sample B (Fig. 2b).

The amplitudes of the two peaks are sample dependent. In sample B, for instance, the ‘red’ peak has much higher spectral weight compared with sample A.

Previous work has suggested a tempting interpretation of the two-peak structure. The ‘red’ peak could stem from emission of the excitons funnelled to the point of the highest strain, whereas the ‘blue’ peak could stem from excitons that did not reach the funnel centre, thus emitting throughout the sample as the laser excitation spot exceeds the characteristic funnelling length. A very high density of excitons is expected at the membrane’s centre in this interpretation. This should lead, in turn, to a rapid non-radiative Auger recombination of excitons, which is known to be effective in TMDs. To test the role of Auger recombination, we recorded photoluminescence spectra for sample B at a relatively low laser power of 8 nW. At this power, only a few excitons are present in the entire sample at any given time and the role of Auger recombination should be negligible. Figure 2b shows that only the ‘red’ peak remains at low power while the ‘blue’ peak vanishes. In principle, such behaviour is consistent with reduced Auger recombination and implies more efficient funnelling at low power.

Finally, we tested for the contribution of charged excitons (trions) emission in our sample, as the binding energy of charged excitons in WS$_2$ is suspiciously close to the energy separation between the ‘red’ and ‘blue’ peaks. While the photoluminescence spectra of samples A and B do not exhibit any trion contribution...
at zero strain, we later demonstrate that such contribution can arise when strain is increased. Therefore, we n-doped sample C by measuring it under vacuum. The desorption of water and nitrogen from the sample surface increases the density of free electrons, which, in turn, bind to neutral excitons to form negatively charged trions. Indeed, well-understood peaks corresponding to neutral excitons in unstrained samples disappears at high strains.

**Discussion**

To answer these questions, we analyse the drift–diffusion equations governing exciton transport in our system. For a non-uniform density of excitons \( n(r) \), the steady-state continuity condition for excitonic diffusion \( J_D = D \nabla n(r) \) and drift \( J_D = \mu n(r) \nabla u(r) \) currents yields\(^{34}\):

\[
\nabla (D \nabla n(r)) + \nabla (\mu n(r) \nabla u(r)) - \frac{n(r)}{\tau} - n^2(r) R_e + S(r) = 0
\]

(1)

Here \( D \) is the diffusion coefficient, \( \mu = \frac{e D}{k_B T} \) is the mobility (where \( k_B \) is the Boltzmann constant and \( T \) is temperature), \( R_e \) is the Auger recombination rate, \( \tau \) is the exciton lifetime and \( S(r) = \frac{\sqrt{2 \sigma}}{2 \pi \sigma^2} \psi^2 \exp(-r^2/2\sigma^2) \) is the exciton generation rate in a Gaussian illumination profile with intensity \( I_0 \) and \( \sigma = \text{FWHM}/2\sqrt{2}\ln 2 \). Unless stated otherwise, we use material constants \( D = 0.3 \text{ cm}^2\text{s}^{-1} \), \( \mu = 12 \text{ cm}^2\text{Vs}^{-1} \), \( R_e = 0.14 \text{ cm}^2\text{s}^{-1} \) and \( \tau = 1.1 \text{ ns} \).

The change of the bandgap due to the strain is assumed to be \( E_g - E_g^0 = -0.05 \times \varepsilon(r) \), where \( \varepsilon(r) \) is the strain of the strain tensor.\(^{16,22}\). A detailed analysis and simulations of equation (1) along with various calculations of \( u(r) \) are shown in the Supplementary Information. Figure 3 (blue dashed curves) shows the photoluminescence spectra obtained from the numerical solutions of equation (1) (Supplementary Information). These solutions clearly do not match the experimental data (red curves). Indeed, while the ‘red’ peak of Fig. 2 could be interpreted as corresponding to a very efficient funnelling process, the numerical solution of equation (1) exhibits funnelling efficiency (defined as the fraction of all photoexcited excitons reaching the location of the AFM tip) that never exceeds 4% (Supplementary Information).

It is instructive to develop intuitive understanding for the observed low funnelling efficiency. While the drift term in equation (1) ‘pushes’ the exciton towards the funnel centre with the force proportional to \( \nabla u(r) \), the diffusion term randomizes that motion (Fig. 1b). The average distances travelled by an exciton during its lifetime due to drift and diffusion respectively can be evaluated within a simple Drude approximation. We find that the diffusion length \( L_{\text{diff}} = \sqrt{D \tau} \approx 180 \text{ nm} \) is much larger compared with the drift length averaged over the excitation spot, \( \langle L_{\text{drift}} \rangle = \langle -\nabla u(r) \rangle \mu \tau \approx 5 \text{ nm} \). Dominating contribution of the diffusion leads, in turn, to inefficient funnelling. One could claim that we observe a rather low funnelling efficiency due to charging in the system.\(^{14}\). This is not the case here as our analysis using the drift–diffusion equation is blind to any charging effects and is also valid for a funnel that does not exhibit charging effects.

We can analyse the relative contributions of drift and diffusion in another, more quantitative way. It is easy to show that, on average, exciton current flows towards the funnel centre (drift dominates...
over diffusion) if the following condition is met (Supplementary Information):

$$k_B T \geq \frac{S(r) V\mu(r)}{V S(r)}$$  \hspace{1cm} (2)

Both the left-hand side and the right-hand side of this formula are plotted in Fig. 4b as red dotted and black solid lines, respectively. We see that the condition above is only fulfilled for the small portion of the membrane ($r < 250 \text{ nm}$), and that the diffusion term dominates the rest of the membrane, leading to inefficient funneling. We therefore posit that funneling cannot be as efficient at room temperature as predicted in ref. [18]. We note that equation (2) also suggests that higher funneling efficiency may be possible at cryogenic temperatures.

If funneling is so inefficient, what other physical mechanism is responsible for the data of Fig. 2? The hint comes from the data of sample C suggesting that the ‘red’ peak at high strain evolves from the trion peak at zero strain. To achieve the contribution of trions into our model, we use the same equation (1), but with $n(r) = n_n(r) + n_{tr}(r)$ where $n_n$ ($n_{tr}$) is the exciton (negatively charged trion) density. Although there are differences in the physical constants $D, \mu, R_0$, and $\tau$ between excitons and trions, we have found that the solution of $n(r)$ does not change substantially in a broad range of possible values (Supplementary Information); thus, we used the same values for both species.

It is easy to see that the density of trions near the device centre is expected to be strongly strain dependent. Indeed, the relative densities of neutral and charged excitons depend on the density of background electrons (doping level) $n_e(r)$ in our device. While at zero strain background carriers are uniformly distributed throughout the device, the applied non-uniform strain lowers the top of the conduction band $u(r)$. Quantitatively, assuming that the density of background electrons is described by the Boltzmann distribution, we obtain

$$n_e(r) = \frac{N_0 e^{e u(r)/k_B T}}{\int e^{e u(r)/k_B T} dr}$$ \hspace{1cm} (Supplementary Information).

Here $N_0$, which is strain dependent, represents the number of free carriers in the whole area of the strained membrane for any given time, and $\Delta u(r)$ is change of the energy of the top of the conduction band from the zero strain value (Supplementary Information). The expression above makes it clear that the electrons are effectively ‘funnelling’ towards the point of the highest strain at the centre of the membrane. As a consequence, photoexcited neutral excitons present near the membrane centre bind to free electrons forming trions. To quantitatively determine the intensity of trion emission, equation (1) is solved for $n(r)$ as before, and $n_{tr}$ and $n_n$ are determined from $n_e(r)$ using the law of mass action $n_{tr}$ (Supplementary Information).

Once the carriers densities $n_n$ and $n_{tr}$ are determined, the entire photoluminescence spectrum is calculated by the following expression:

$$\langle \text{PL} \rangle = \int_0^\infty [\text{PL}_{ex}(u_e(r)) n_{ex}(r) + \text{PL}_{tr}(u_{tr}(r)) n_{tr}(r)] dr$$ \hspace{1cm} (3)

Here $\text{PL}_{ex}$ ($\text{PL}_{tr}$) is the spectral line of the excitons (trions) and is taken from the Gaussian fits of the spectrum at zero strain. Our model does not account for changes in quantum yield as a function of strain, as this change has been shown to be small enough to be negligible in our modelling. Figure 3 shows a comparison between the model (black dashed line), using $N_0$ as a single fit parameter, and the experimental results (red line) for relevant strain and excitation intensities for all samples. The model is in much better agreement with the experimental data, especially compared with the model that does not include trion effects (thin blue dashed line). We therefore conclude that strain-related free carrier funneling, followed by conversion of neutral to negatively charged excitons, is the dominant process in our samples.

To ensure that the effect we are observing is a strain-dependent exciton-to-trion conversion, we analysed the spatial photoluminescence maps for sample A. Each spectrum was fitted with two emission line profiles. The excitone line was fitted to a Gaussian emission line, and the trion to a modified Gaussian to account for the electron recoil effect. Examples of these fits are presented in the Supplementary Information. Finally, in Fig. 5 we plotted the spatially resolved ratio of trion-to-exciton intensity. While at zero strain that ratio is uniform across the membrane, at high strain the trion peak is observed predominantly at the centre of the membrane. This is the direct evidence for large density of free carrier at the centre of the membrane and of the resulting efficient exciton-to-trion conversion.

To illustrate the mechanism responsible for the appearance of the strain-dependent trion contribution, we show in Fig. 4c the calculated spatial dependency of neutral and charge exciton densities in sample A (calculations for other samples are shown in the Supplementary Information). We see that the trion density $n_{tr}(r)$ steadily increases and becomes much larger than $n_n(r)$ towards the centre of the funnel, consistent with funneling of free carriers to that region. At the same time, in the region of the sample that predominantly contributes to the observed signal due to the Jacobian rdr (defined as the shaded area in Fig. 4a), $n_{tr}(r) > n_n(r)$. This explains comparable magnitudes of the trion and exciton peaks in sample A (Fig. 3a). In contrast, in samples B (low excitation) and C the doping level is higher. In that situation, we find (Supplementary Information) that $n_{tr}(r) \ll n_n(r)$ in the relevant area of the device, meaning that photoexcited neutral excitons are converted into trions with conversion efficiency approaching 100%.
Our findings suggest several important implications. First, strain-dependent exciton-to-trion conversion produces experimental signatures that may appear similar to that of neutral excitons funnelling, but much stronger in amplitude. This suggests that previous reports of exciton funnelling might have strongly overestimated its efficiency. Second, while we experimentally realize...
the controlled funnelling geometry of the theoretical proposal\textsuperscript{16}, the dominant process in such a device is found to be diffusion rather than drift, at least at room temperature. This means that the photoconversion mechanism proposed by ref. \textsuperscript{18} may not be feasible. Finally, and perhaps most importantly, the strain-dependent exciton-to-trion conversion may constitute another, more efficient photoconversion mechanism compared with that of ref. \textsuperscript{15}. We speculate that the energetics of energy harvesting of weakly bound trions may be advantageous to that of strongly bound neutral excitons.

To summarize, in this work we have presented a novel experimental setup that allows full dynamical control of strain amplitude and profile in optically interrogated TMDC monolayers. We revealed that even in highly strained TMDCs, the funnelling of the excited carriers is not nearly as efficient as previously thought. However, we discovered that in the presence of non-uniform strain, another process, neutral-to-charged exciton conversion, becomes dominant. It is noteworthy that this former process, while being physically very different from the latter, can produce similar experimental signatures, leading to possible misinterpretations. Finally, we note that in the future it will be especially interesting to study the role of funnelling at cryogenic temperatures, where the role of diffusion is minimized.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and material availability are available at https://doi.org/10.1038/s41566-019-0581-5.

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Data availability
The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
M.G.H. and K.I.B. planned and designed the experiment and wrote the paper. M.G.H. and J.N.K. performed the nanoindentation experiments. M.G.H., J.N.K. and M.Q. fabricated the samples. The experiments, data analysis and the theoretical model were done by M.G.H. K.G. contributed to the theoretical model.

Competing interests
The authors declare no competing interests.

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