Nonlinear self-action of ultrashort guided exciton–polariton pulses in dielectric slab coupled to 2D semiconductor

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Abstract

Recently reported large values of exciton–polariton nonlinearity of transition metal dichalcogenide (TMD) monolayers coupled to optically resonant structures approach the values characteristic for GaAs-based systems in the regime of strong light-matter coupling. Contrary to the latter, TMD-based polaritonic devices remain operational at ambient conditions and therefore have greater potential for practical nanophotonic applications. Here, we present the study of the nonlinear properties of Ta₂O₅ slab waveguide coupled to a WSe₂ monolayer. We confirm that the hybridization between the waveguide mode and the exciton resonance in WSe₂ gives rise to the formation of guided exciton–polaritons with Rabi splitting of 36 meV. By measuring transmission of ultrashort optical pulses through this TMD-based polaritonic waveguide, we demonstrate the strong nonlinear dependence of the output spectrum on the input pulse energy. We develop a theoretical model that shows agreement with the experimental results and gives insights into the dominating microscopic processes which determine the nonlinear pulse self-action: Coulomb exciton–exciton interaction and scattering to an incoherent excitonic reservoir. Based on the numerical simulation of nonlinear phenomena in our polariton system, we conclude that it may support a quasi-stationary solitonic regime of pulse propagation at intermediate pump energies. Our results provide an important step for the development of nonlinear on-chip polaritonic devices based on 2D semiconductors.

1. Introduction

Two-dimensional transition metal dichalcogenides (TMDs) are actively used as alternative materials to III–V semiconductors [1–6] to realize the strong light-matter coupling regime between excitons and photons [7]. Contrary to their bulk counterparts, monolayer TMDs are direct band-gap semiconductors possessing strong excitonic response in the visible-to-near-IR spectral range. The Wannier–Mott excitons in these materials are characterized by large binding energies (up to 200–500 meV for neutral excitons) and large exciton oscillator strengths [8–10]. This makes possible the formation of hybrid light-matter quasiparticles—exciton–polaritons at cryogenic [11–16] and room temperatures [17–21] in TMD-based systems with excitons resonantly coupled to a spatially confined photonic mode.

The peculiar features of exciton–polaritons are due to their hybrid nature [22, 23]. They inherit an extra small effective mass and a coherence length on
the micrometer scale from the photonic component [24]. At the same time, the excitonic component gives rise to strong polariton–polariton interactions, which lead to a high nonlinear optical response [25]. These notable properties make systems based on exciton–polaritons excellent candidates for the practical implementation of active all-optical on-chip photonic devices.

Recently, the nonlinear optical response has been investigated in TMD-based polariton systems where atomically-thin semiconductors were integrated with photonic structures such as open cavity [14], dielectric Bragg stacks supporting Bloch surface waves [13], and photonic crystal slabs supporting either leaky modes [17] or bound states in continuum [15]. Furthermore, the enhanced nonlinear response of polaritons in TMD-based structures has been shown for Rydberg excitons [26, 27], trions [14] and indirect excitons in Moiré-type structures [28]. The nonlinear polaritonic response typically manifests itself as a power dependent shift of polariton branches due to either blueshift of the exciton energy arising from the Coulomb interaction [28] or the nonlinear quenching of Rabi splitting due to the phase space-filling effects [14]. Both mechanisms come into play with the increase of polariton density.

The study of the polariton nonlinearities in TMD-based systems is still under heavy both theoretical [28, 29] and experimental [13–15, 26, 30] investigations. The reported values of exciton–polariton nonlinearity—1–3 $\mu$eV$\cdot$µm$^{-2}$ [14, 15] and trion-polariton nonlinearity—37 $\mu$eV$\cdot$µm$^{-2}$ [14] at cryogenic temperature in MoSe$_2$-based systems are not much lower than values of the strength of the exciton–polaritons interaction per one quantum well, extracted from the measurements in GaAs-based systems—1–100 $\mu$eV$\cdot$µm$^{-2}$ [25, 31–35]. This makes it possible to demonstrate in TMD-based polariton systems various nonlinear effects that are well-known in devices based on classical semiconductors, such as polariton nonlinearities of neutral exciton [14, 15] and charged excitonic complexes [14, 26], nonlinear polariton parametric emission [36], bosonic condensation and lasing in hybrid TMD-GaAs systems [30, 37]. Most of the presented polariton systems represent a TMD monolayer integrated with various microcavities. In turn, TMD-based polariton waveguides [13] may open the way for demonstrating some nonlinear dynamic effects such as solitons [38, 39] and self-phase modulation [38], which were studied in polaritonic waveguides based on III-V semiconductors.

In this work, we comprehensively study the nonlinear evolution of intense femtosecond polaritonic pulses propagating along a single-mode planar waveguide integrated with a WSe$_2$ monolayer. We experimentally confirm the strong light-matter coupling between photonic mode of the waveguide and excitonic resonance in TMD. Further, we perform power-dependent measurements revealing complex nonlinear reshaping of transmitted pulse spectrum depending on the energy of an input pulse. To explain our experimental findings, we then develop a theoretical model that takes into account a polariton nonlinearity driven by Coulomb exciton–exciton interaction and scattering of coherent excitons into an incoherent reservoir, demonstrating satisfactory agreement with the experimental results. Finally, based on the model we analyze the spatio-temporal evolution of the ultrashort polariton pulses realized in the experiment. In particular, we confirm that the spectral broadening of the output signal experimentally observed at intermediate pump levels may correspond to the realization of quasi-stationary solitonic regime of pulse propagation.

2. Results

2.1. Experiment

The studied sample, as depicted in figure 1(a), consists of an unpatterned Ta$_2$O$_5$ slab waveguide coupled to WSe$_2$ monolayer surrounded by two coupler/de-coupler gratings. In the first stage, the gratings were etched in a 90 nm-thick Ta$_2$O$_5$ layer deposited on SiO$_2$/Si (1 µm/525 µm) substrate.

Further on, a mechanically exfoliated 50 µm-long monolayer of WSe$_2$ was transferred on the surface of the slab waveguide between the gratings (see section 5). Such a configuration allows us to study nonlinear processes that occur during the propagation of laser pulses through the structure. Figure 1(b) shows the measured lower polariton dispersion with the avoided crossing between uncoupled photon and exciton modes characterized by a Rabi splitting of $\hbar\Omega_0 \approx 36 \pm 1$ meV.

In the experiment, the sample was placed into a closed-cycle helium cryostat integrated with a setup for the Fourier plane imaging allowing to perform the angle-resolved spectroscopy measurements at 7 K. The sample was excited by laser pulses with a spectral linewidth of 19 meV corresponding to a $\sim$130 fs temporal full width at half maximum (FWHM). The pulses are coupled at an optimized angle through the input grating, propagate 100 µm along the waveguide, and decouple through the output grating. It is important to note that the effective length of the interaction between the excitons and the waveguide mode is $\approx$50 µm, which is defined by the size of the area covered by a WSe$_2$ monolayer along the propagation direction.

Figures 2(a) and (b) show the evolution of output pulse spectra after the propagation through the waveguide with the increase of the pulse energy. The data were acquired for two negative detunings of the central frequency of the input pulse from the exciton resonance: $\delta_1 = -11$ meV and $\delta_2 = -41$ meV. At the
Figure 1. (a) Sketch of a Ta$_2$O$_5$ slab waveguide integrated with a WSe$_2$ monolayer. Periodic gratings are used to couple and decouple femtosecond pulses. (b) Angle- and frequency-resolved photoluminescence measured from the area of the decoupler covered by a TMD monolayer. Solid lines represent exciton ($X_0$) and photon ($C_{ph}$) modes. Dashed line shows the lower polariton branch dispersion obtained from the fitting of the data with coupled oscillator model. (c) Scheme showing the several processes accounted for in the model: exciton–photon coupling, scattering into the exciton reservoir, photon emission.

Figure 2. (a), (b) Transmitted pulse spectra measured for the input pulse energy increasing from the bottom to the top and normalized to their respective peak values. The estimated input pulse energies are specified above the respective curves. The input pulse spectrum is shown at the bottom of each panel. Its shape is assumed to match the transmitted pulse spectrum under high-energy excitation (the details are given in the text). The dashed vertical line ($X_0$) denotes the frequency of the uncoupled exciton resonance. Presented data correspond to negative detunings of the input pulse central frequency of (a) $-11$ meV and (b) $-41$ meV from the exciton resonance. (c), (d) Results of the theoretical modelling, corresponding to the upper panels. Depending on the detuning and power of the input pulse, the output spectrum demonstrates complex behaviour of both its linewidth and spectral position. Black circles represent the position of the centre of mass of the pulse, defined as the first moment of the spectral power distribution for each spectrum shown.
Figure 3. Power dependencies of the laser pulse (a), (b) total transmittance and (c), (d) peak position calculated as a first moment for two negative detunings of (a), (c) $\delta_1 = -11$ meV and (b), (d) $\delta_2 = -41$ meV. Experimental and simulated results are presented by the circles and solid lines, respectively. In panels (a) and (b), the three characteristic regions are clearly visible: low-energy linear (constant transmittance) regime (I), intermediate-energy nonlinear (growing transmittance) region (II), high-energy nearly linear (constant transmittance) mode. In the latter case, the exciton resonance becomes strongly blueshifted such that the majority of the pulse radiation energy propagates without polaritonic absorption.

Small detuning and low pulse energies, the measured transmitted pulse spectra exhibit a distinct asymmetric shape. This asymmetry arises from the proximity of the laser pulse central frequency to the exciton resonance, causing efficient absorption in the high-energy spectral range. With the increase of the pulse power, the shape of the output spectrum for $\delta_1 = -11$ meV detuning becomes more symmetric, and for both series of the measurements, we observe the blueshift–redshift crossover of the power-dependent peak position. To complement these experimental findings, we conducted numerical simulations based on the theoretical model described below, as depicted in figures 2(c) and (d).

Note that at high excitation power, both in the experiment and the simulations, the normalized output pulse spectrum becomes nearly independent of the pump power. This is due to the excitonic blueshift, which leads to the suppression of both the effective nonlinearity and the absorption. It is important to emphasize that this happens because the blueshift of the exciton resonance frequency decreases the excitonic fraction in the polariton at fixed frequency. This decreases the effective nonlinearity and the losses experienced by the polaritons but does not necessarily mean the transition to a weak coupling regime. Meanwhile at high powers, the output pulse spectrum almost corresponds to the spectrum of the input pulse coupled into the system (shown with black curves in figure 2).

To quantify the output laser pulse after its propagation through the waveguide in each measurement, we calculate the area under the spectral curve normalized to the incident power and the first moment of the spectral power distribution in order to determine the overall pulse transmittance (figures 3(a) and (b)) and the output pulse central frequency (figures 3(c) and (d)), respectively. Notably, figures 3(c) and (d) clearly show a blueshift–redshift crossover of the power-dependent peak position. As the excitation power increases, we observe a pronounced blueshift of the peak position during the middle power regime (Region II), followed by a redshift at high powers (Region III). Transmittance and the output pulse central frequency exhibit strongly nonlinear dependencies on the incident pulse energy,
which are defined by the microscopic mechanisms of the nonlinear response in the system. Below we provide a theoretical model describing these mechanisms.

2.2. Model

To describe the evolution of laser pulses in the transmittance spectra after their propagation through the waveguide with increasing pulse energy, we used a theoretical model that takes into account the strong exciton–photon coupling, exciton–exciton repulsion within the pulse and the presence of an incoherent exciton reservoir. In particular, we assume that photons can be absorbed creating coherent excitons, which can in turn annihilate emitting coherent photons. This phenomenon leads to the hybridization of the photons and excitons and the formation of a polariton gap in the dispersion of the elementary excitations. The observed nonlinear effects in such systems primarily arise from exciton–exciton interactions, which lead to a shift in the exciton resonance frequency.

To provide a satisfactory description of the experimental data, it is crucial to consider the formation of an incoherent excitonic reservoir, where coherent excitons generated by the optical pulse can undergo scattering. These incoherent excitons lie far outside the light cone, and thus cannot emit coherent photons. However, the reservoir density strongly affects the blueshift of the excitonic line, thus acting as a non-instantaneous nonlinearity.

In our modelling we use the following set of the equations describing the coherent exciton–photon wavepacket interacting with incoherent reservoir of excitons [40]:

\[
\left( \frac{\partial}{\partial t} + v_g \frac{\partial}{\partial x} + \gamma_{ph} \right) A = i \frac{\Omega_g}{2} |\psi|^2 + f(t,x),
\]

\[
\frac{\partial |\psi|^2}{\partial t} = - \left( \gamma_e + \frac{\mu}{2} \right) |\psi|^2 + i \alpha (|\psi|^2 + \rho) + i \frac{\Omega_g}{2} A,
\]

\[
\frac{\partial \rho}{\partial t} = - \Gamma \rho + \mu |\psi|^2,
\]

Here \( \hbar \Omega_g = 36 \text{ meV} \) and \( v_g = 45 \mu \text{m ps}^{-1} \) are Rabi splitting and photon group velocity estimated from the measured polariton dispersion (figure 1(b)). The loss rate of excitons due to scattering to the exciton reservoir \( \hbar \gamma_e = 3 \text{ meV} \) and all other exciton losses \( \hbar \gamma_e = 9 \text{ meV} \) are chosen to get the best agreement between the theory and the experiment (figure 3). We used the typical value of the nonlinear parameter \( \alpha \) for TMD monolayers from literature \( \hbar \alpha = 1 \mu \text{eV} \mu \text{m}^2 \) [14, 15].

The guided photons are described by the slow varying amplitude \( A \) of the fundamental mode of the waveguide with group velocity \( v_g \). The physical meaning of \( A \) and \( \psi \) is that the photon and coherent exciton densities are expressed as \( |A|^2 \) and \( |\psi|^2 \), respectively. We neglect the background waveguide and material dispersions (the frequency dependencies of the mode group index and material refractive index, respectively) because they are much smaller compared to the dispersion caused by the coupling of the photons with the excitons in the vicinity of the exciton resonance (see figure 1(b)). For the same reason we neglect the bare Kerr nonlinearity of an empty waveguide. The effective linear photonic losses (Ohmic, scattering on the imperfections, etc) are accounted for by the dissipation rate \( \gamma_{ph} \). The interaction strength between the photons and the excitons is characterized by the Rabi frequency \( \Omega_g \). A similar model describing excitons and photons in mean field approximation is described in [41] and the references therein. The system is excited by a coherent optical pulse which is transferred to the photon guided mode by the coupler described by the driving force \( f(x,t) \) in the right-hand side of the equation (1a).

To reproduce the experimental results, we introduce also a reservoir of incoherent excitons which is characterized by the incoherent exciton density \( \rho \). The coherent excitons lose their coherence with the rate defined by \( \mu \) and this populates the reservoir of the incoherent excitons. We assume that the incoherent excitons decay with the rate \( \Gamma \), which is supposed to be much less compared to \( \gamma_e \). Thus the equation (1c) is actually the rate equation accounting for the transition from the coherent to incoherent excitons and for the decay of the incoherent excitons. We note that the process of decoherence contributes to the decrease of the coherent exciton density and thus the effective losses for the coherent excitons are the sum of their annihilation rate \( \gamma_e \) and the decoherence rate \( \gamma_e \Gamma \). In the polariton context, the model with the incoherent exciton reservoir is used, for instance, in [42].

We neglect the photonic losses (as we work in the waveguide regime) and reservoir lifetime (as reservoir excitons are dark and long living), \( \hbar \gamma_{ph} = 0, \hbar \Gamma = 0 \). We would like to mention that in reality there are processes that make \( \gamma_{ph} \) and \( \Gamma \) to be finite. However, these values are so small that their influence on the polariton propagation is negligible at the propagation distances in our study. We acknowledge that for longer propagation distances these losses can become of importance.

The nonlinear parameter \( \alpha \) is the proportionality coefficient defining the exciton resonant frequency shift \( \Delta \omega \) through the total density of the excitons \( \Delta \omega = \alpha (|\psi|^2 + \rho) \). Let us remark here that the nonlinearity introduced in our model is conservative in the sense that absorption in exciton or in photon field does not depend on the field intensity. However, in terms of a polariton mode, the effective propagation length is a function of the polariton intensity. The explanation is that the effective losses experienced by polaritons depend on Hopfield coefficients.
These coefficients in their turn depend on the resonant frequency of the excitons and thus depend on the polariton intensity. So the conservative nonlinearity affects the structure of the polariton mode and by this changes the effective propagation length of polaritons.

The pump $f$ is an amplitude of the incident light multiplied by the transmission coefficient connecting the intensity of the incoming light to the intensity of the light coupled to the guided mode. We fitted this coefficient by comparing the simulated and experimental data. Careful optimization of the model parameters confirmed that in our system, the Coulomb-type nonlinearity leading to the exciton resonance blueshift (defined by $\alpha$) dominates over another possible source of nonlinearity, namely the quenching of Rabi splitting originating from phase space filling effects. This conclusion agrees well with previously reported both theoretical [43] and experimental [15] studies of the nonlinear response of exciton–polaritons in TMD-based systems.

3. Discussion

The theoretical model presented above provides good semi-quantitative description of the experimental data (see figures 2 and 3).

Let us first analyze the dependence of pulse transmittance on the input pulse energy shown in figures 3(a) and (b). For both detunings, one can distinguish three characteristic regions. Under both low (Region I) and high (Region III) input pulse powers, the system behaves linearly, i.e. transmittance is nearly energy-independent. In Region I, the nonlinearity is small, while in Region III, only a small portion of the pulse energy is absorbed before the strong blueshift of the exciton resonance, therefore the overall transmitted pulse energy remains almost unaffected. Therefore, we can use the high-energy spectrum as a reference while maximizing light coupling efficiency in the experiment. For intermediate input powers (Region II), the system reveals a strongly nonlinear response: the transmittance increases by several orders of magnitude, the central frequency of the output pulse manifests complex non-monotonic behaviour, and pronounced spectrum broadening occurs compared to the low- and high-energy regimes (see figures 2(a) and (c)).

First, we note that at the low input pulse energy, the output spectrum of the pulse is significantly redshifted compared to the spectrum of the input pulses (black curves in figure 2). The explanation for this is that the contribution of the exciton subsystem to the effective losses is overwhelmingly higher compared to its photonic counterpart. This means that the polaritons experience the highest losses at the exciton resonance frequency. The frequency of the input pulse is lower than the exciton resonance and thus the blue part of the pulse spectrum gets absorbed much stronger than the red part. This explains why the center of the output spectrum is red-shifted in respect to the center of the input one.

At intermediate pump energies, the nonlinearity comes into play via two mechanisms. First, it blueshifts the exciton resonance with the increase of input pulse energy. This reduces the difference between the absorption rates for different frequencies, and so one can expect that the redshift of the output pulse becomes lower under higher pump powers. As a result, with the increase of the input pulse energy the central frequency of the output spectrum should undergo a blueshift.

The second nonlinearity-driven mechanism in the system is a parametric process that results in the spectral broadening of the pulse directly observed at the intermediate pump powers in the experiment (figure 2(a) and (c)). Let us consider the pulse dynamics assuming that frequency-dependent polaritonic losses are not affected by the nonlinear effects (exciton blueshift is disabled), but the spectral broadening takes place. The parametric process increases the width of the spectrum. However, high frequencies experience much stronger absorption than the lower frequencies. This means that the high-frequency wing of the output spectrum does not grow much—these frequencies are absorbed. In contrast, the lower frequencies, including the ones generated by the parametric process, survive, and the spectrum extends into lower frequencies. As a result, the central frequency of the pulse should redshift. Therefore, if the process of spectrum broadening is efficient and dominates over the nonlinear modification of the effective losses, with the increase of the input pulse energy the central frequency of the output pulse undergoes redshift.

If we perform a numerical simulation with account for these two competing nonlinear mechanisms with no reservoir enabled (see supplementary material, figure S1 for details), we see that the behaviour of the central frequency is not monotonic. Only at high powers does the exciton blueshift sufficiently reduces the losses for higher frequencies to counteract the initial redshift. Meanwhile, in the experiment, no redshift is observed at intermediate pump pulse energies (see figures 2(a) and (b), Region II).

In order to explain the experimentally observed behaviour of the central frequency of the output pulse spectrum we can surmise that the parametric process in the experiment is less efficient compared to the model without a reservoir. Therefore, the model should be modified to suppress the dependence of the efficiency of the new frequency generation on the polariton density. This can be done by introducing a non-instantaneous nonlinearity, assuming
Figure 4. (a) Simulated evolution of temporal profile of the pulse propagating along the waveguide for the input pulse energies of 11 and 258 pJ and detuning of $-11$ meV. In the low-energy regime (11 pJ), the dispersion-driven temporal pulse elongation is observed. On the contrary, for the input pulse energy of 258 pJ, the pulse becomes about 30% shorter and remains stable during the propagation over several tens of microns, which is a signature of a quasi-solitonic behaviour. Simulated (b) duration, (c) peak position, and (d) spectral width of the pulse depending on propagation distance and input pulse energy. Pulse duration and spectral width are determined as a second moment $\mu_2$ of the spectral and temporal power distributions, respectively. For the incident pulse spectrum of a Gaussian shape, the second moment is equal to the variance $\sigma^2$ and related to the full width at half maximum as $\text{FWHM} = 2\sqrt{2\mu_2} \ln 2$.

that the coherent excitons can get scattered into the incoherent ones. The incoherent excitons do not take part in the parametric process spreading the spectrum but still contribute to the exciton blueshift and thus nonlinear blueshift of polaritonic losses. The use of this model (equation (1)) allows us to obtain good semi-quantitative agreement with the experiment. It is worth mentioning that there may be other physical mechanisms (for example, generation of electron–hole plasma) that could lead to a similar effect. However, in the studied system the best agreement with the experiment was obtained for the model accounting for the reservoir, i.e. generation of the incoherent excitons.

Let us now mention the possible reasons for the observed discrepancy between the experiment and the simulations. Note that in the system under study, losses depend on frequency so sensitively that the attenuation varies by at least three orders of magnitude for the given propagation distance within the spectral width of the initial pulse. Consequently, to accurately reproduce the attenuation curve quantitatively, we need to know the spectrum of the initial pulse with the precision much exceeding experimentally achievable values.

The experimental data on the power dependent reshaping of the spectrum of the output signal allows us to reconstruct the spatio-temporal dynamics of the pulse. Figure 4 shows the simulation results for the pulse propagation in the polariton waveguide. Under low input energies (Region I), the pulse undergoes polaritonic absorption leading to the redshift of its spectrum and strong temporal broadening (see also green dashed curve in panel (a)). For high input pulse energies (Region III), pulse parameters remain nearly unchanged due to the strong exciton blueshift, which weakens the nonlinear processes. The intermediate regime (Region II) is illustrated in figure 4(a). While propagating, the pulse first gets compressed, and then its duration (figure 4(b))
and spectral width (figure 4(d)) remain nearly constant for approximately 30 microns. Such behaviour may indicate a quasi-stationary solitonic regime of pulse propagation, which is realized in the studied system.

4. Conclusion

We have shown that strong light-matter coupling in a TMD-based polaritonic waveguide gives rise to pronounced nonlinear effects in the propagation of ultrashort optical pulses. We observe power dependent suppression of the losses at large pulse energies and blueshift–redshift crossover of the pulse central frequency. These features are well-reproduced by a theoretical model which accounts for the excitonic reservoir and Coulomb interaction between the excitons. The experimentally observed spectral broadening at intermediate pulse powers is a manifestation of a new frequency generation process. Our modelling shows that it is accompanied by the temporal shortening of the pulse. These results form the basis for further experiments confirming that such a polaritonic system supports quasi-stationary regimes of soliton propagation.

5. Methods

5.1. Sample fabrication

We fabricated two samples in which a monolayer was placed between the coupler and the decoupler. All nonlinear measurements presented in the main text were made using Sample A, where the monolayer had a length of 50 µm along the pulse propagation direction (figure 5(a)). However, when a monolayer does not cover a grating that plays the decoupler role, it is impossible to measure the polaritonic dispersion directly. In such a case, only the photonic mode can be observed during light propagation and outcoupling through an uncovered waveguide area where photons cannot interact with excitons. Therefore, we fabricated a second device (Sample B), where we transferred a monolayer to cover part of the grating and part of the waveguide (figure 5(b)). This made it possible to measure the value of Rabi splitting in a polaritonic system based on WSe$_2$ integrated with a slab waveguide (figure 1(b)). The main difference between the samples is the distance between the gratings etched in the waveguide, which was 100 µm for Sample A and 50 µm for Sample B.

5.2. Optical measurements

The optical measurements were conducted in our custom-built experimental setup using back focal (Fourier) plane imaging, enabling direct extraction of dispersion diagrams in frequency-momentum space. Laser pulses and white light, polarized along the grating that corresponds to the transverse-electric field mode, were focused onto the input grating coupler with an objective lens. The output grating decoupled the light from the waveguide, which was collected by the same microscope objective (Mitutoyo Objective 50x/0.65) and spatially filtered, allowing only the radiation from the output grating to be detected. To obtain the angle-resolved signal, the Fourier plane of the objective was projected onto the entrance slit of an imaging spectrometer (Princeton SP 2550), and the light was detected by a CCD detector (PyLoN 400BR eXcelon). The sample was mounted in an ultra-low-vibration closed-cycle helium cryostat (Advanced Research Systems, DMX-20-OM) and maintained at 7 K.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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