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Dipolar-induced enhancement of parametric effects in polariton waveguides

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Exciton-polaritons are hybrid light-matter excitations arising from the non-perturbative coupling of a photonic mode and an excitonic resonance. Behaving as interacting photons, they show optical third-order nonlinearities providing effects such as optical parametric oscillation or amplification. It has been suggested that polariton-polariton interactions can be greatly enhanced by inducing aligned electric dipoles in their excitonic part. However direct evidence of a true particle-particle interaction, such as superfluidity or parametric scattering is still missing. In this work, we demonstrate that dipolar interactions can be used to enhance parametric effects such as self-phase modulation in waveguide polaritons. By quantifying these optical nonlinearities we provide a reliable experimental measurement of the direct dipolar enhancement of polariton-polariton interactions.

Adding high nonlinearities to compact, integrated optical circuits could unlock the development of new, fully photonic devices such as ultra-low-power active switches and transistors [1], parametric amplifiers or oscillators and supercontinuum sources [2]. Nonlinearities could even push forward the limit of integrated optics in quantum applications, i.e. universal quantum logic gates [3, 4]. Exciton-polaritons have demonstrated to be good candidates to supply nonlinearities to integrated optical platforms. They emerge, in inorganic semiconductors, from the strong coupling between a photonic mode and an electronic excitation (exciton) in Quantum Wells (QWs). The nonlinearities in such systems, originating from **short-range exchange** interactions in the excitonic component, have been widely studied in the mesoscopic regime, allowing the demonstration of effects such as optical parametric oscillation, [5], bistability [6], superfluidity [7, 8], pattern formation [9] or quantum vorticity [10]. Despite polariton-polariton interactions providing higher nonlinearities than standard nonlinear materials [2], they failed to bring such system fully in the quantum regime [11]. For example, in the recently observed polariton blockade, a consequence of the polaritonic interactions at the two-particles level [12, 13], only a weak violation of the classical regime has been observed, due to the low interaction over dissipation ratio. These results have strongly motivated the quest for the enhancement of the nonlinearities in polariton based systems.

Recently, it has been given evidence that increasing the lengths of aligned exciton dipoles brings an enhancement of the polaritonic interactions in both vertical microcavities [14], and waveguides (WGs) [15, 16], due to dipolar long range interactions between excitons. This

very promising phenomenon could be exploited for many nonlinear optical effects or for the design of logic gates working at the two-particles regime [17]. However, up to now, a **direct** evidence that dipolar nonlinearities can actually **truly** work to enhance **polariton interaction, for instance via resonant** nonlinear parametric effects, is still missing, with only **indirect, non-resonant indirect** proofs that have been provided, as is the overall blueshift of the exciton resonance [14–16] **or the polariton condensation threshold** [18]. Nevertheless, the overall blueshift is often due to a concurrence of different factors such as phase space filling, charge screening, reduction of the Rabi coupling and presence of dark excitonic states which can lead to puzzling results. The WG geometry, a system in which the photonic mode is confined into a 2-D slab by total internal reflection [15, 19], seems particularly promising for the study and development of optoelectronic devices [20], thanks to features such as faster propagating particles, a geometry that favors the integration of polaritonic optical circuits, high quality factors of the photonic mode and less demanding growth requirements. Most importantly, the WG platform allows the application of an external electric field perpendicular to the propagation direction able to control the polariton-polariton interaction strength through the manipulation of the exciton dipole length [16, 21].

In this letter, we demonstrate that dipolar polariton-polariton interaction do enhance parametric effects well beyond those derived by standard particle-particle exchange interaction. This is done by measuring the self phase modulation (SPM) of a resonant pulsed wavepacket propagating in the same kind of sample studied in [15, 16, 21]. Moreover, we use this effect to make a precise and unequivocal assessment of the

strength of the polariton dipolar interaction. A short pulse propagating inside a polariton WG undergoes a parametric effect [2] known as self-phase modulation (SPM). SPM is a well-known third order nonlinear effect [22] seen in optical systems as caused by the modification of the refractive index due to the propagation of the pulse itself and resulting in a spectral broadening and, for higher powers, structuring of the propagating pulse. However, the same effect can be seen, for strongly interacting particles, as a spontaneous parametric scattering, a four wave mixing process, that leads to depletion of the packet at the highest density point, in favour of a spread of particles along the propagation dispersion. Here we measure the SPM-induced pulse spectral modification in an AlGaAs/GaAs WG-QW system, directly showing the enhancement of the parametric effect by dipolar polariton-polariton interactions, introduced by means of a perpendicular electric field. The measured data are accurately fitted by a Gross-Pitaevskii (GP) model allowing extracting the real dipolar enhancement of polariton-polariton interactions. Remarkably, we measure onto the parametric effect a 5-fold enhancement of excitonic interactions due to the change in the dipole length.

The WG structure is grown on an n^+ -doped GaAs substrate, on top of which a 500 nm cladding of $Al_{0.8}Ga_{0.2}As$ was previously deposited. The structure consists of 12 pairs of 20 nm thick GaAs QW separated by 20 nm of $Al_{0.4}Ga_{0.6}As$ barriers [15, 16], resulting in a photonic mode with a quality factor of $\sim 10^4$. We process the slab WG through a dry etching technique [23] in order to fabricate a $1\ \mu\text{m}$ wide, $200\ \mu\text{m}$ long wire, confining polaritons in one additional direction and further reducing the modal volume. With this technique we also fabricate focusing gratings [24] or tapers to efficiently inject and extract the laser pulse into the small-volume waveguide. A schematic model of the WG system, comprising the input/output tapered gratings, is displayed in Fig. 1a. Finally, the slab surface is covered with a 50 nm layer of Indium Tin Oxide (ITO), that together with the doped substrate on the opposite side, enables the application of an external electric field in the direction perpendicular to the slab plane [16] (see SI for additional details on sample etching and processing). Figure 1b shows the energy dispersion of the zero-order polaritonic modes in absence of external fields.

As reported in previous works [15, 25], the geometry and polarization of the guided electromagnetic modes yield a Rabi splitting around 3 times higher for the Transverse Electric (TE) than for the Transverse Magnetic (TM) mode. The value of the Rabi splitting is $\Omega = 13.9\ \text{meV}$ for the TE mode and $\Omega = 5.8\ \text{meV}$ for the TM mode. Due to the greater exciton-TE mode coupling, all the measurements are performed on this

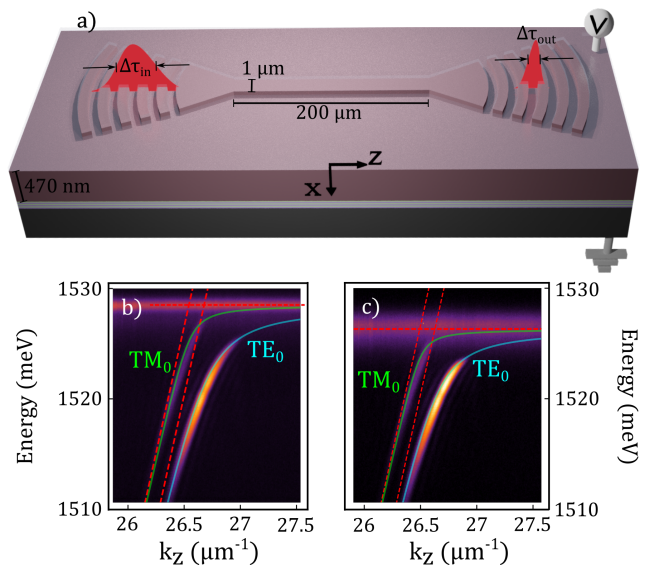


FIG. 1. a) A slab waveguide is patterned through an etching process to create a wire $1\ \mu\text{m}$ wide with input and output focusing gratings. The incident laser pulse resonantly injects population into the system, and the output signal, modified by the nonlinearities resulting from polaritonic interactions, is collected and analyzed. The doped substrate together with the ITO capping layer enable the application of an electric field along the growth direction. b) Angle and energy-resolved PL emission showing polariton dispersion in the WG system, without applied electric field. c) As in b) for an applied electric field $E = 11.9\ \text{kV/cm}$, resulting in the Stark shift of the excitonic energy. Continuous lines in b) and c) are calculated polariton dispersion.

mode. When an electric field is applied to the structure [15] the spatial profile of the potential energy of the QW is modified. The induced spatial separation between electrons and holes results in a finite dipole in a photocreated electron-hole pair [26, 27]. As a consequence, the exciton energy shifts and the Rabi splitting slightly diminishes, as it is showed in Fig. S2. The range of fields explored in this work corresponds to the regime characterized by a quadratic Stark energy shift [26], in which exciton dissociation due to the applied field can be neglected. By using a variational approach [27], we extract an induced dipole length around $4\ \text{nm}$ for Fig. 1c in agreement with previous works [14–16]. The laser pulse is focused on the input taper, that injects the light into the TE_0 mode of the micro-wire. After propagating along its $200\ \mu\text{m}$ it is extracted by a second taper, see sketch in Fig. 1a. The light is then collected and analyzed by a spectrometer, see SI for further details. During the pulse propagation, the time-dependent pulse intensity modifies the refractive index of the medium, due to $\chi^{(3)}$ nonlinearities provided by polariton-polariton interactions, resulting in the SPM effect. Figure 1a shows that the time-dependent modification of the refractive index can be intuitively understood as an

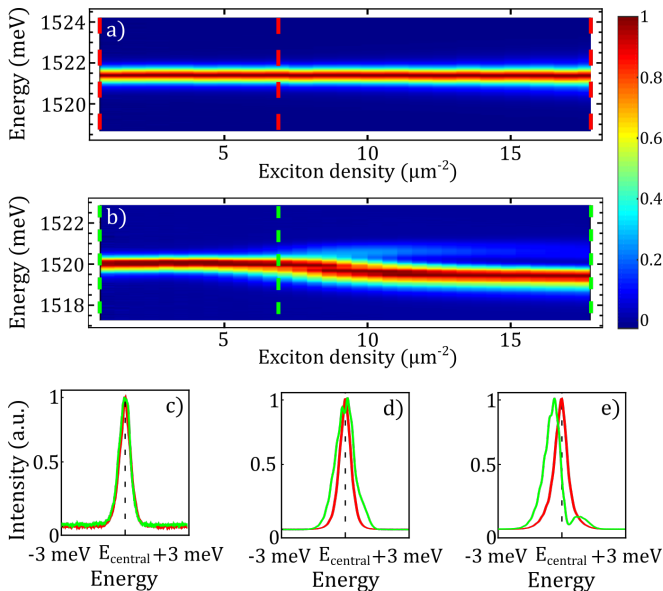


FIG. 2. **a)** Normalized output pulse spectra as a function of the exciton densities. **b)** Same as panel a) but for an applied transverse electric field of 11.9 kV/cm ; nonlinear effects in **b)** are visible at exciton densities lower than in **a)**, a signature of the dipolar enhancement of polariton - polariton interactions. **c-e)** Pulse broadening and structuring for three different exciton densities indicated by vertical lines on the 2D maps of panels **a)** and **b)**; the red spectra correspond to the case in which no electric field acts on the system, while green spectra correspond to an electric field value of 11.9 kV/cm ; spectra are energy shifted to compare shape and FWHM.

induced temporal compression of the propagating pulse and hence the observed SPM spectral broadening. This effect can also be viewed as a particle-particle scattering mechanism: two particles injected with the same energy and wavevector can interact and scatter towards two different states fulfilling the energy and momentum conservation conditions [2, 22].

Figures 2a-b show 2D representations of the pulse spectra obtained after resonant nonlinear propagation through the WG. The x axis represents the calculated exciton density, which is proportional to the excitation power (see description of the theoretical model below and a discussion of the relationship between input power and exciton density in the SI). We note that the absolute value of g , and thus the absolute value of the density, are not measured in this experiment, and instead we observe their scaling as a function of input power. Figure 2a shows the propagation without applied electric field and Fig. 2b the propagation for $E = 11.9 \text{ kV/cm}$. In both cases the laser energy is tuned to be at excitonic fraction $X^2 = 0.5$. Both figures show a density-dependent spectral broadening and structuring of the propagating pulse [2, 22]. Remarkably, the pulse broadening in presence of an applied field in Fig. 2b is

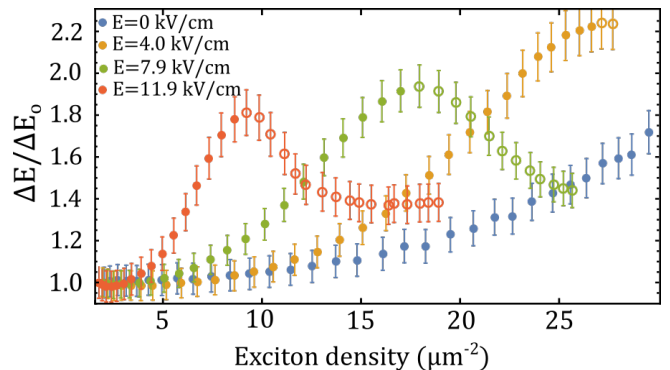


FIG. 3. FWHM of the spectra from a gaussian fit as a function of the output power for four different values of the electric field. The output pulse width is normalized to the incident width. The empty circles correspond to the cases in which the output spectrum has several peaks, and represent the linewidths of the most intense lobe.

larger and occurs at lower exciton densities than in the case without applied electric field in Fig. 2a. Moreover, in presence of the applied electric field, the output pulse shows a more pronounced structure. At higher excitation powers, the high-intensity part (the central peak) of the propagating pulse is depleted by the particle-particle scattering resulting in a more complex structure of the output pulse [22]. This is a signature of the nonlinear pulse propagation and can be understood in terms of a peak depletion due to the particle-particle scattering. Figure 2 also shows an asymmetric broadening, with the red part of the propagating pulse being more intense than the blue part. This is due to both the shape of the polariton dispersion and the increased absorption on the blue side due to the excitonic tail. We note that for $E = 11.9 \text{ kV/cm}$, the output intensities are roughly half the output intensities without any applied electric field. We assign this discrepancy to a slight increase of nonradiative losses in presence of the electric field (see also discussion below). Figures 2c-e show spectra of the output pulse at zero field (red lines) and at $E = 11.9 \text{ kV/cm}$ (green lines) at exciton densities equal to 1, 7 and 19 excitons per μm^2 , see corresponding vertical dashed lines in figures 2a and b. These figures show that at low exciton density, see Fig. 2c, the nonlinearity does not affect the pulse shape and the two spectra have identical linewidths. When the exciton density is increased, Fig. 2d, the applied electric field causes a larger broadening of the output pulse. At even higher exciton densities, Fig. 2e, the pulse propagating under the action of the applied electric field shows a pronounced structure with the presence of two lobes, while the pulse propagating without applied electric field keeps a single peak shape.

The effect of the electric field on the SPM-induced

broadening is also clearly visible in Fig. 3, showing the FWHM of the output pulse as a function of the exciton densities for different applied electric fields (keeping the same excitonic fraction $X^2 = 0.5$ for each field). Filled points refer to the range of exciton densities in which the output pulse shows a single broadened peak. Empty circles refer to the range of exciton densities in which the output pulse show a structured peak (in this case, each point represents the FWHM of the more intense lobe). As the applied electric field increases, the pulse broadening at a given polariton density increases, showing a clear dipolar enhancement of the parametric effects. Moreover, pulse breaking, i.e. the transition from filled points to empty circles, takes place at lower excitonic density when the applied electric field is increased, highlighting the dipolar enhancement of polariton-polariton scattering.

To obtain a quantitative description of the observed dipolar enhancement of the polariton-polariton interactions, we solve the coupled equations [2] 1a and 1b describing the system dynamics in the GP formalism:

$$\left[i \frac{\partial}{\partial t} + i\gamma_p + \nu_g \left(i \frac{\partial}{\partial z} + \frac{1}{2k_z} \frac{\partial^2}{\partial x^2} \right) \right] A = \left(\frac{\Omega}{2} \right) \Psi, \quad (1a)$$

$$\left[i \frac{\partial}{\partial t} + i\gamma_e - g|\Psi|^2 \right] \Psi = \left(\frac{\Omega}{2} \right) A \quad (1b)$$

where A and Ψ are the photon and exciton wavefunction amplitudes, respectively. ν_g is the ~~particles~~ photons group velocity obtained from the theoretically fitted dispersion and k_z the corresponding linear momentum. γ_p and γ_e are the decay rates of the coherent photonic and excitonic states. The evolution of the initial (~ 2 ps or ~ 145 μeV) pulse is calculated by solving the equations 1 obtaining a theoretical output spectrum for each input power. A nonlinear least squares routine is used to determine g , as well as the detuning and duration of the pump pulse, minimizing the difference between the measured and simulated matrices of power vs. spectrum. Fig. 4 shows the measured (left column) and calculated (right column) spectra as function of the input power for each of the four applied electric fields, with an excellent agreement between experimental data and simulations. It is worth noting that here we are focusing on the dipolar enhancement of polariton-polariton interactions. In particular, we have fixed the value of $g = 6$ $\mu\text{eV} \mu\text{m}^2$ at zero applied electric field [28] and found the corresponding exciton density per QW. Based on the obtained exciton density and on the measured output intensities, it is possible to retrieve the exciton density/output intensity calibration. We calibrate based on the peak value of $|\Psi|^2$ observed at the end of the waveguide in the simulation. We have then used this calibration to determine the exciton densities for the entire data set (see SI and in

particular Fig. S34 for the details). Figure 4i shows that the enhancement of the dipolar interaction with the field causes g to increase by a factor ≈ 5 under the strongest electric field applied in the experiment. To show the consistency of our approach we have reported values of g obtained on two different sets of data measured at excitonic fractions $X^2 = 0.5$ and $X^2 = 0.6$.

Remarkably, while we find a significant enhancement of polariton-polariton interactions due to the applied electric field, this enhancement is at least an order of magnitude smaller than the one reported on a sample having the same design [16] by measuring non-resonant blueshift of the polariton resonance. We believe, as mentioned above, that this discrepancy is essentially due to the fact that the polariton blueshift is only an indirect measurement of the particle-particle interaction that could be caused by many different effects. As a matter of fact, we have also performed non-resonant experiments under ultra-fast pumping, observing a wide range of possible blueshifts at a given excitation power. The cause of such behaviour is due to multiple factors not all related to the dipole enhancement and possibly dependent on extrinsic parameters. Non-resonant excitation could be responsible for the creation of trapped electric carriers that, under the action of the applied electric field and depending on the quality of the electric contacts, can induce a screening phenomenon. Such a screening may reduce the Stark effect and result in an apparent blueshift. In contrast, in this work we measure polariton-polariton interactions through a different effect, the SPM, which cannot be enhanced by other effects including screening. We stress also that the use of an ultrafast ps exciting pulse is important to reduce other spurious causes such as the creation of a long living, “dark” exciton reservoir [28], that contribute to the overall density without being detected. Particularly in the WG platform, the presence of long-lived excitonic reservoir can alter the measurement of the interaction strength by more than two orders of magnitude [29, 30]. To avoid these possible spurious effects, we resonantly inject the population with a pulsed laser of 80 MHz repetition rate and ~ 2 ps (~ 145 μeV) pulse duration (spectral width). Our results are in qualitative agreement with the enhancement reported under resonant reflectivity in the case of permanent electric dipoles stemming from indirect excitons in double QWs [14].

In conclusion, we demonstrated that parametric effects in polaritonic systems can be enhanced by polarizing their excitonic component thanks to an applied electric field. By measuring the self-phase modulation of a propagating waveguide packet, we find a 5-fold enhancement of polariton-polariton interactions. However, in case of systems with very weak nonlinearities, such a dipolar effect, which is independent of the intrinsic polariton exchange interaction, could even lead to enhancements order of magnitude higher. For instance, we believe that

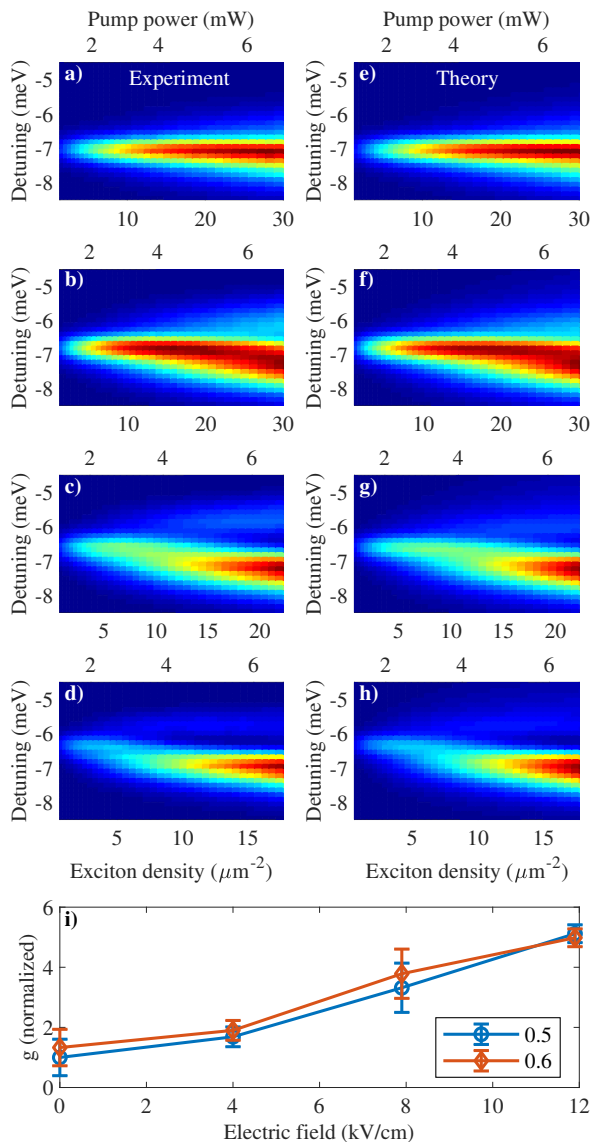


FIG. 4. a-d) Measured density maps of the output spectra as a function of estimated exciton density at the end of the waveguide for field strengths of 0 kV/cm , 4 kV/cm , 7.9 kV/cm , and 11.9 kV/cm , respectively, and placed on a constantly spaced photon energy grid for comparison to simulations, for an excitonic fraction of 0.5. e-h) Corresponding results of numerical solutions of the Gross-Pitaevskii equation for guided exciton-polaritons after nonlinear least squares fitting to determine the value of the interaction constant g . See the text for the calibration of the $|\Psi|^2$ axis. i) Electric field dependence of the fitted value of the parameter g , for two different excitonic fractions, 0.5 and 0.6, normalized to the value at zero field and fraction of 0.5.

this enhancement method could be extremely effective in TMD-based polariton systems. These results are essential to assess the relevance of dipolar polariton waveguides for future integrated and quantum optics application [31, 32] and to guide the development of a new generation of WG polaritons supporting stronger

nonlinearities, even at room temperature.

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