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Deterministic placement and effective-mass pinning of topological polariton bound states in the continuum

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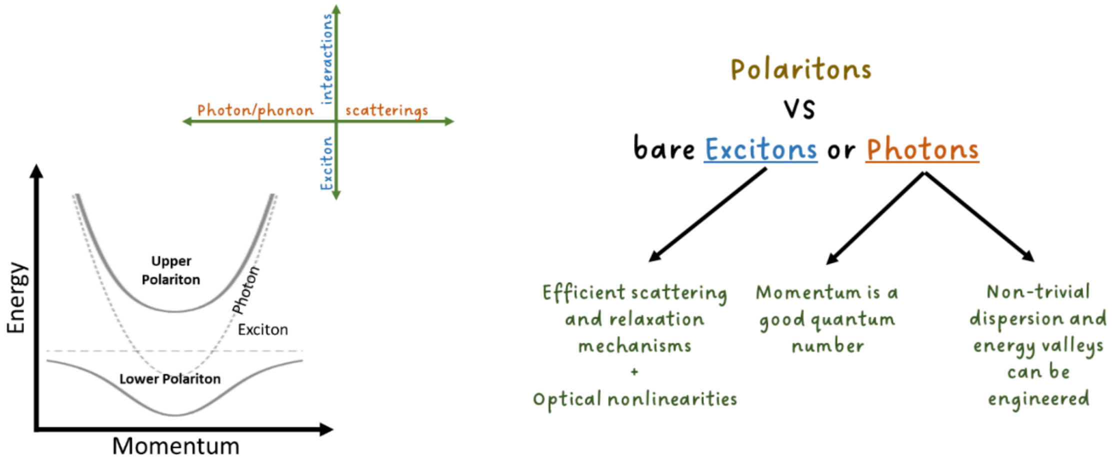
Abstract. The exciton-polaritons derived from the light-matter interaction of an optical bound state in the continuum (BIC) with the strong excitonic resonance in a transition metal dichalcogenide (TMD) monolayer can inherit ultra-long radiative lifetimes and significant nonlinearities up to room temperature. Yet such realization can be challenging with conventional approaches to the photonic cavity design, typically due to poorly-resolved Rabi splittings at room temperature and an unstable energy positioning of the BIC state. We show and experimentally validate a strategy to dramatically improve the state-of-the-art on both points, by embedding a tungsten disulfide (WS_2) monolayer deep within a Bloch-surface-wave stack, where the photonic mode is moulded by a 1D photonic crystal with a compound periodicity. In particular, we introduce a deterministic placement principle to the design of the PhC, allowing to stabilize the energy positioning of a topologically-protected BIC polariton eigenstate, with an effective mass which we can robustly pre-assign at choice as either positive or negative. This is in stark contrast to typical waveguide realizations of polariton BICs: only negative polariton effective masses can be commonly achieved, while sudden jumps to a weaker-interacting positive-effective-mass BIC are at the same time possible upon small perturbations, in fact hijacking the advantage from a topological protection when present.

1. Introduction

1.1 Exciton-polaritons

Exciton–polaritons are bosonic quasi-particles arising from the strong coupling between excitons and confined photons, acquiring hybrid properties inherited from their bare components. Namely, from confined photons they inherit a great flexibility in moulding an energy-momentum dispersion. This can typically be achieved by inducing, with nanolithographic techniques, patterns in the dielectric permittivities. The resulting photonic structures commonly go under the name of Photonic Crystals (PhCs) [1]. One of the most interesting features which could be induced are stationary points and especially isolated energy valleys in the dispersion, where bosonic population can easier build-up and lead to spectacular effects as a Bose-Einstein condensation, for instance.

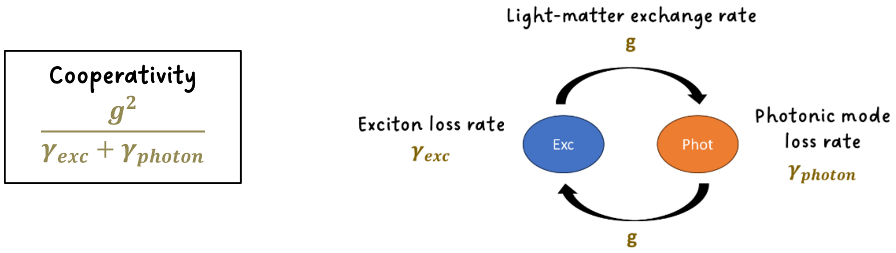


(Fig.1)
Polaritons mix the characteristics of both excitons and photons, resulting in nontrivial dispersions and powerful scattering mechanisms in both energy and momentum, resulting in a phenomenology much richer than the one available to either the bare exciton or photon.

Nevertheless, despite the possibility to arbitrarily mould the energy-momentum dispersion, the physical processes which can be induced among photons are mostly limited to scatterings of momenta, without the possibility for coherent exchanges of energy. On the other hand, from excitons the polaritons can inherit strong dipolar interactions, which indeed allow them to easily scatter at the same time energy and momentum (see Fig.1). So polaritonic platforms allow not only to engineer peculiar energy-momentum dispersions, but also to have a rich phenomenology happening on such dispersions, for instance polariton parametric scattering processes, bistability phenomena and eventually also ultralow-threshold lasing phenomena and Bose-Einstein condensation [2,3].

1.2 Cooperativity: a characterizing figure of merit for the strong light-matter interaction regime

Nevertheless one of the critical requirements in order to access the rich polariton phenomenology is having access to the most possible exciton interactions, but at the same time getting rid of the exciton incoherent losses. This point gets dramatically critical if room temperature operation is also required, as the exciton nonradiative linewidth typically grows wider. It is thus essential to inherit the most possible excitonic interactions at finite and possibly large detunings from the exciton itself. This translates into increasing the light-matter energy exchange rate (observable as a "Rabi splitting" in the dispersion) while also preserving a thin polariton linewidth, a trade-off which can be captured by a proper figure of merit which we shall call, as commonly done, "cooperativity" (see Fig.2):

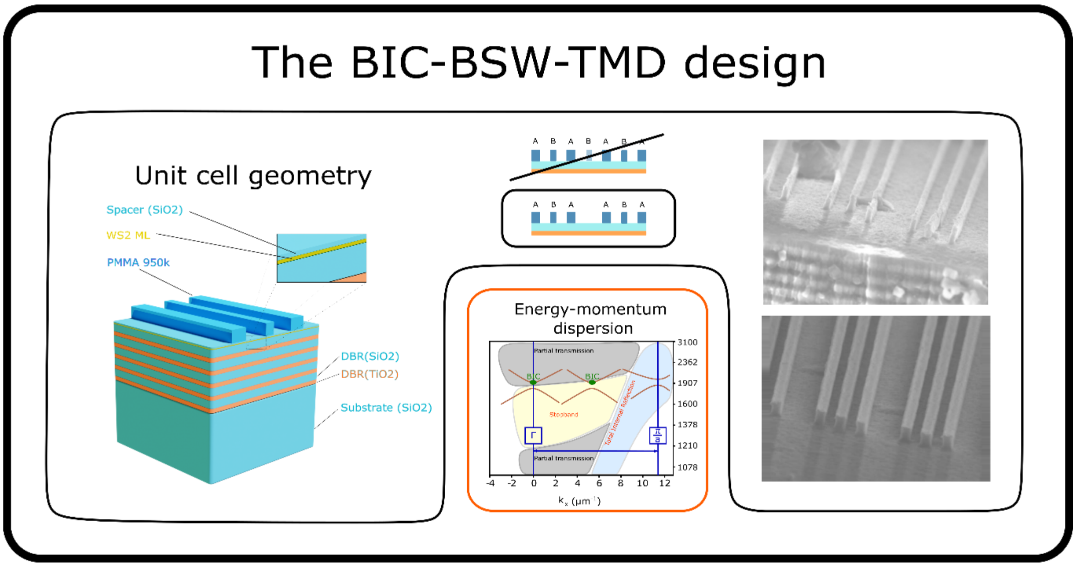


(Fig.2)

Polaritons are not only the result of a large light-matter coupling constant g, but to observe their typical phenomenology a reversible character is needed for the energy exchange between the exciton and the photon. Individual loss rates of the exciton and photon thus play a critical role as well as the coupling constant g. Their interplay in determining how strong the polariton phenomenology will be is captured by a key figure of merit commonly called "cooperativity".

Photonic BICs are a powerful tool to obtain significant improvements in the polariton cooperativity by dramatically reducing the photon loss rate. In fact, in III-V materials and at cryogenic temperatures, polariton Bose-Einstein condensation has recently been demonstrated at remarkably low thresholds, by leveraging exactly on a BIC [3].

2 The BIC-BSW-TMD platform



(Fig.3)

Our design consists of a glass dielectric mirror, on top of which we encapsulate a TMD monolayer and subsequently pattern a nontrivial superwavelength PhC of PMMA, featuring a periodic subtraction of pillars. This results in a polariton dispersion accessible from air at k~0 and with a topologically protected BIC. In virtue of the nontrivial PhC architecture, the energy position and effective mass of the BIC is deterministically fixed by the parity in the number of pillars within each grating PhC subunit. Images partially adapted from [4].

2.1 Enhancing light-matter interaction up to room temperature operation by BSWs and TMDs

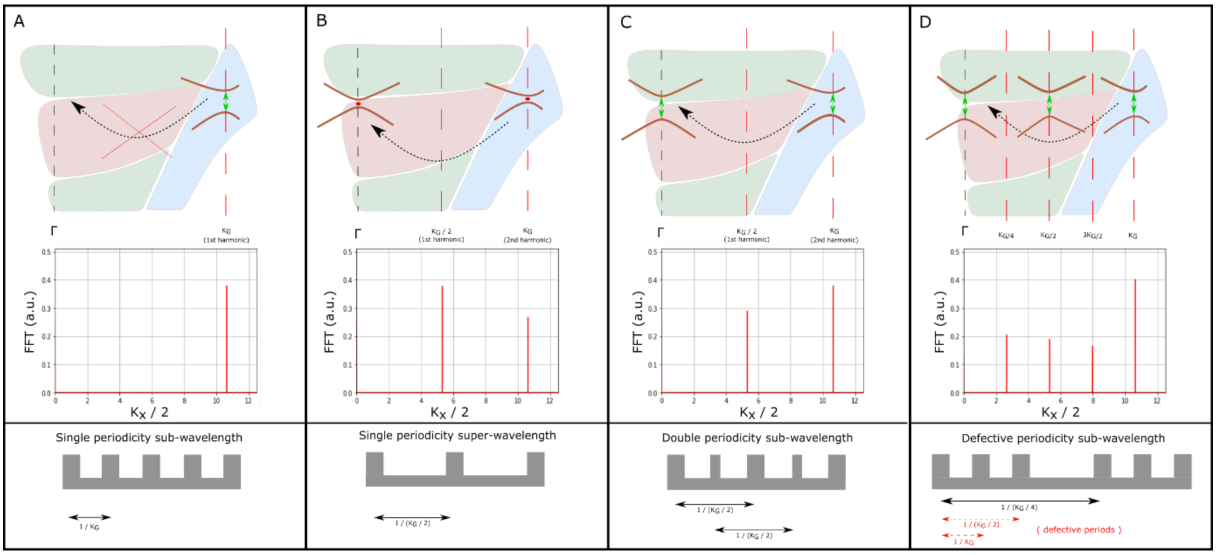
Here and more extensively in **[4]** we show that at least a couple of critical targets can be effectively added to the results in [3]; a first point would be the possibility to obtain strongly coupled exciton-polariton systems robust to room temperature operation. In work **[4]** we are indeed able to meet this first point by switching from III-V materials to TMD monolayers and jointly using a Bloch Surface Wave

(BSW) [5,6] as photon confinement mechanism. In fact, on one hand the neutral excitons in TMD monolayers exhibit a strong and thin resonance up to room temperature, due to their large oscillator strength and high binding energy. The reduced dielectric screening from the monolayer confinement additionally results in high exciton-exciton nonlinear interactions, which can be inherited by exciton-polaritons [7].

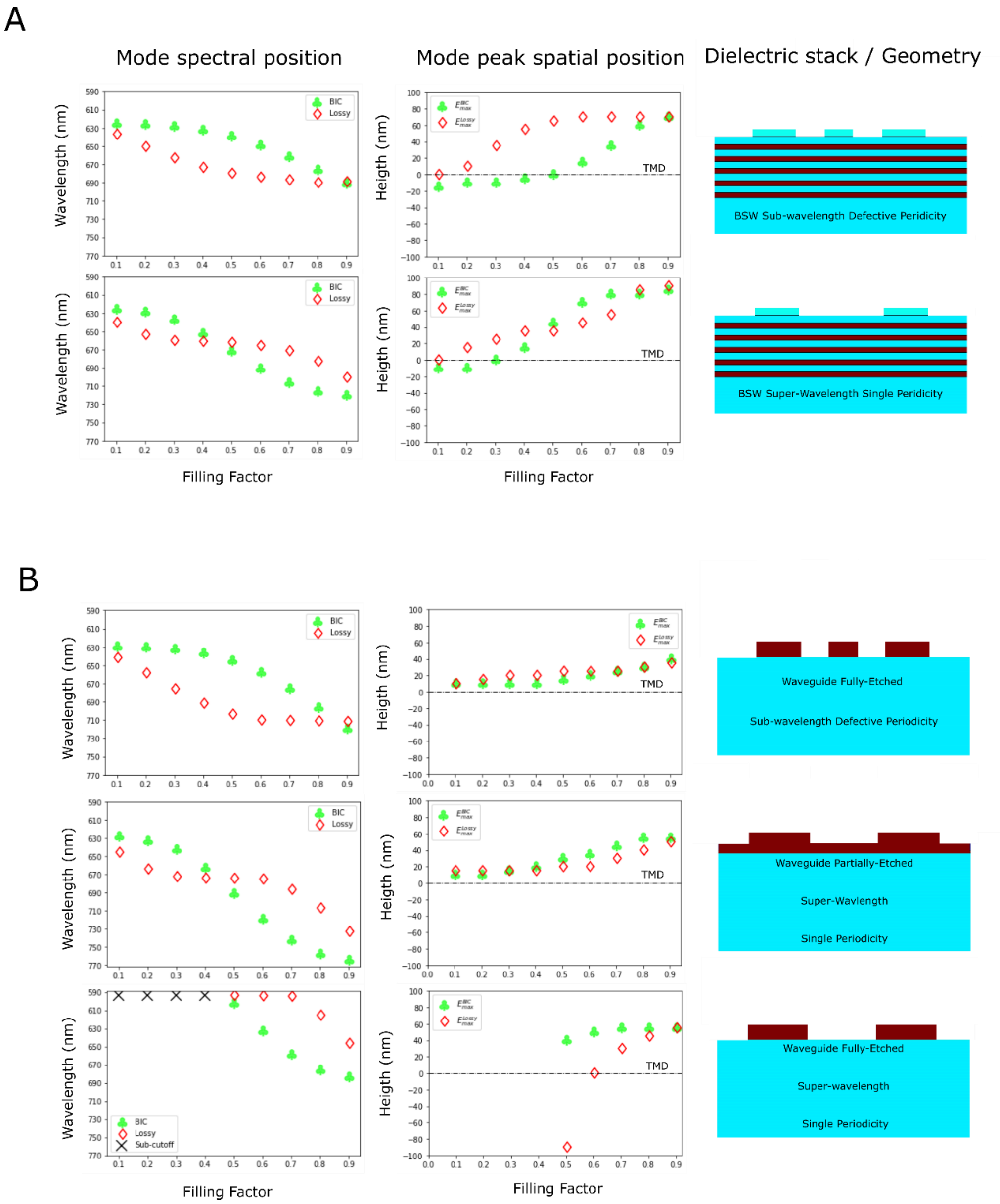
On the other hand, BSWs allow for small modal volumes while at the same time having their maximum field enhancement on a free surface, where a 2D material can be optimally coupled. This puts them in stark contrast to the (typically) two-times larger modal volumes of microcavity modes, and also to the limited surface-field enhancement of waveguide modes.

Nevertheless, a stationary point in the energy-momentum dispersion is often a desirable feature in the context of polariton physics, as it allows to easier build up population and induce nonlinear effects, as we have already mentioned. A limitation of BSWs against microcavities is that they are indeed missing such stationary points in their dispersion. This problem, as commonly done for waveguides, can be overcome by a superwavelength PhC, which can be induced on the top of our BSW-TMD-hosting stack by a nanopatterned resist layer (see Fig.3); this results in a branching of the polariton dispersion into two Bloch-like modes accessible at the Gamma-point, where two opposite effective-mass stationary points arise, separated by a wide photonic band gap (see Fig.4).

In Fig.5 we further show that a BSW system features distinctive advantages with respect to waveguide modes, beyond the competitive modal volume and the much better surface enhancements: in fact, when coupled to 2D materials on the surface, properly designed PhC-BSWs (panels C and D) are able to open large energy gaps between the two Bloch modes and at the same time have a strongly selective field enhancement for one of the two modes. The latter fact is relevant, as it can prevent the second mode from acting as an energy sink for polariton population at non zero momenta.



(Fig.4)
Fourier analysis of different grating designs and resulting energy-momentum dispersions of the corresponding Bloch eigenmodes. *KG* is the momentum crossing the BSW original eigenmode beyond the light-cone; the weight of this momentum in the Fourier spectrum of the grating will determine the width of the bandgap between the upper and lower branches; the weight of *KG*/2 will instead determine the effectiveness of the folding from *KG* back to the Γ point (i.e., the origin in reciprocal space). We show here several possible situations: (A) sub-wavelength periodicity with a strong fundamental harmonic at *KG,* opening a wide bandgap and which does not lead to any folding at Γ; (B) Super-wavelength single periodicity, resulting in a strong fundamental harmonic at *KG/2* and a weaker 2nd-order harmonic beyond the light-cone, for which the folded BSW dispersion is strongly visible at Γ, but the bandgap opening is small. In contrast with these regular solutions, it is possible to introduce grating solutions deterministically allowing to obtain an upper-branch antisymmetric mode: the alternate deformation of the grating posts (C) or the subtraction of one pillar per unit cell (D) result in similar Fourier spectra, with both a strong harmonic opening the gap (*KG*) and a robust harmonic (*KG/*2) transposing the bandstructure to Γ. Image readapted from the supplementary information of [4].



(Fig.5)

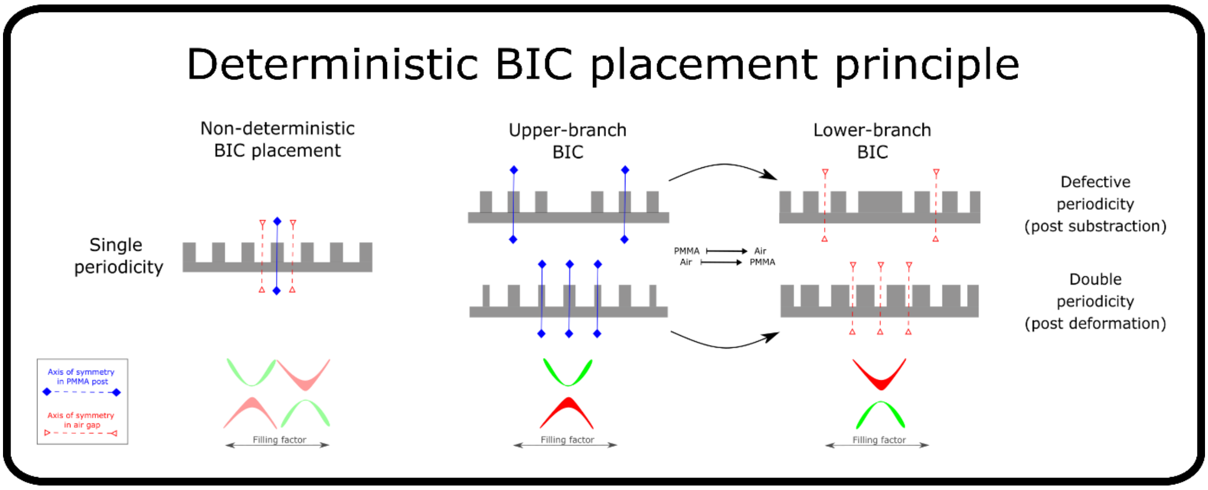
Comparison of different design solutions employing BSW modes (Fig.4A) versus Waveguide modes (Fig.4B). The column "Mode spectral position" gives a measure of the photonic bandgap appearing in the dispersion at k = 0; the column "Mode peak spatial separation" refers to the position, averaged across each unit cell, of the electric field peak for the symmetric (red) and antisymmetric (green) Bloch modes of the grating. A large "eye" is clearly opened for both the energy gap and the spatial gap, only for the BSW system, while waveguide-mode-based systems can never contemporarily open an energy gap and a spatial field enhancement gap. Image readapted from the supplementary information of [4].

2.2 Enhancing room temperature cooperativity by a polariton BIC

Provided that the point-group-symmetry of our PhC unit cell is at least C_{2v} (i.e. a 180deg rotational symmetry), a ultrahigh-Q state, namely a BIC, will always be sustained at the Gamma-point in either one of the two Bloch branches (see Fig.3 dispersion inset and Fig.4 B/C/D), protected by a mechanism relying on pure symmetry (see Sakoda book[8] for a thorough discussion). In particular - keeping our discussion focused on TE-like modes - one can argue that antisymmetric eigenstates with respect to their in-plane E-field distribution within the unit cell, won't be able to transfer power to the plane waves in vacuum; the intuitive reason is that equal portions of the near field will respectively be in relative phase and counterphase to the plane waves propagating normally to the stack plane. The opposite will happen for the symmetric modes, which will instead feature an enhanced ability to match the outer radiation, because of their homogeneous relative phase in the near field at each given plane parallel to the surface of the device. The peculiar symmetry protection mechanism featured by antisymmetric modes – i.e. the BIC modes - can also be proofed to be of "topological character", when analysed in terms of the farfield projection from such modes [9].

2.3 Deterministic energy placement of BICs

A key point of this discussion now arises, as one could ask what are the practical consequences of this so called "topological character": upon small changes of the dielectric indexes or of pattern dimensions, the topological nature of the farfield polarization pattern from a BIC in a regular grating can be proofed to robustly preserve the very existence of the BIC itself at the Gamma points in the dispersion; this is a well known mechanism, often mentioned in literature as "topological protection" [9]. Yet such protection mechanism doesn't fix in any way whether the BIC will have a positive or negative effective mass, i.e. whether it will be hosted as a local energy maximum within a lower-energy Bloch mode or as a local minimum in an upper-energy Bloch mode. It is in fact the case that, contrary to the preservation of their very existence at the Gamma point, the energy positioning of topological BICs can suddenly jump between the two Bloch branches upon small perturbations of the dielectric landscape. This is something of critical importance in the context of polariton physics, as it means that the detuning or the effective mass can discontinuously change either due to fabrication inaccuracies or upon changing pumping power through a nonlinear modulation of the refractive indexes. The solution which we present here – we shall call it a "deterministic placement principle", as illustrated in Fig.5 – allows to overcome this key issue, assuring the topological protected nature in a BIC, while at the same time deterministically assigning its energy position and the sign of its effective mass.



(Fig.5)
The presence of a large number of symmetry axes falling both in full parts and air trenches of a regular and single periodicity PhC pattern induce an instability in the energy positioning and effective mass of BIC, even when these are protected by topological mechanisms. This problem can be robustly avoided by reducing the symmetry axes in the PhC unit cell and deterministically associating them to either the high index parts of the grating or to the low index air trenches. Doing so is possible while at the same time retaining the same point-group symmetry of the unit cell and preserving a topologically-protected BIC. Image readapted from the supplementary information of [4].

In order to understand the principle, we shall recall again that due to the existence of 180deg rotational symmetry axes in regular grating structures (see Fig.5 leftmost, for instance), at the Gamma point the Bloch eigenmodes arising from the periodic modulation will always be also eigenstates of the symmetry operation; in other words, their fields will be either even or odd with respect to the symmetry axes of the structure. It turns out that the very identification of a photonic eigenmode as a BIC is entirely related to its symmetry, for the reasons we discussed in section 2.2; specifically, in case of the TE modes which we consider here (the only ones which can couple the TMD 1L neutral exciton), the BIC will correspond to the state with the in-plane E-field sign odd around the symmetry axes of the structure. One should then recall that a confined photonic eigenmode energy is related to the average refractive indexes of the region it is sustained within: eigenmodes sustained in regions with a higher average refractive index will have lower relative energy than eigenmodes in regions of lower refractive index. So we now need to match these facts into one reasoning flow:

(1) BIC states can be deterministically identified with their field symmetries.
(2) BIC TE-mode fields will always have a node falling on the symmetry axes of the structure.
(3) A BIC TE-mode, due to its nodes on the symmetry axes, will probe a refractive index which will underweight the material refractive index at the position of such symmetry axes.

If one could add, by construction of the PhC geometry, a 4th statement, namely

(4?) The PhC geometry is constructed such that symmetry axes will selectively fall only in full pillars (higher refractive index) or in air trenches (lower refractive index) of the pattern.

Then, according to whether the symmetry axes are chosen to only fall in the middle of full pillars (air trenches), we would have built a full syllogism:

(S) "The BIC TE-mode will deterministically underweight the higher (lower) refractive index", i.e. "The BIC TE-mode will deterministically fall in the lower (higher) energy branch"

We introduced point (4) as an hypothesis for the syllogism to work. But, as we show in Fig.5 (second and third columns rightmost), it is indeed possible to realize a real structure satisfying point (4), and it is possible to do so in at least a couple of different ways:

(1) A regular and selective removal of pillars, which will deterministically result in a high-energy (positive effective mass) BIC when the remaining grating subunits will count an odd number of pillars; on the reverse, a low-energy (negative effective mass) BIC will be sustained when the grating subunits will feature an even number of pillars.
(2) An alternate post deformation, such that the resulting superwavelenght unit cell features one single symmetry axis falling in the middle of a pillar or in the middle of a trench either. The former will result in a high-energy (positive effective mass) BIC, the latter in low-energy (negative effective mass) BIC.

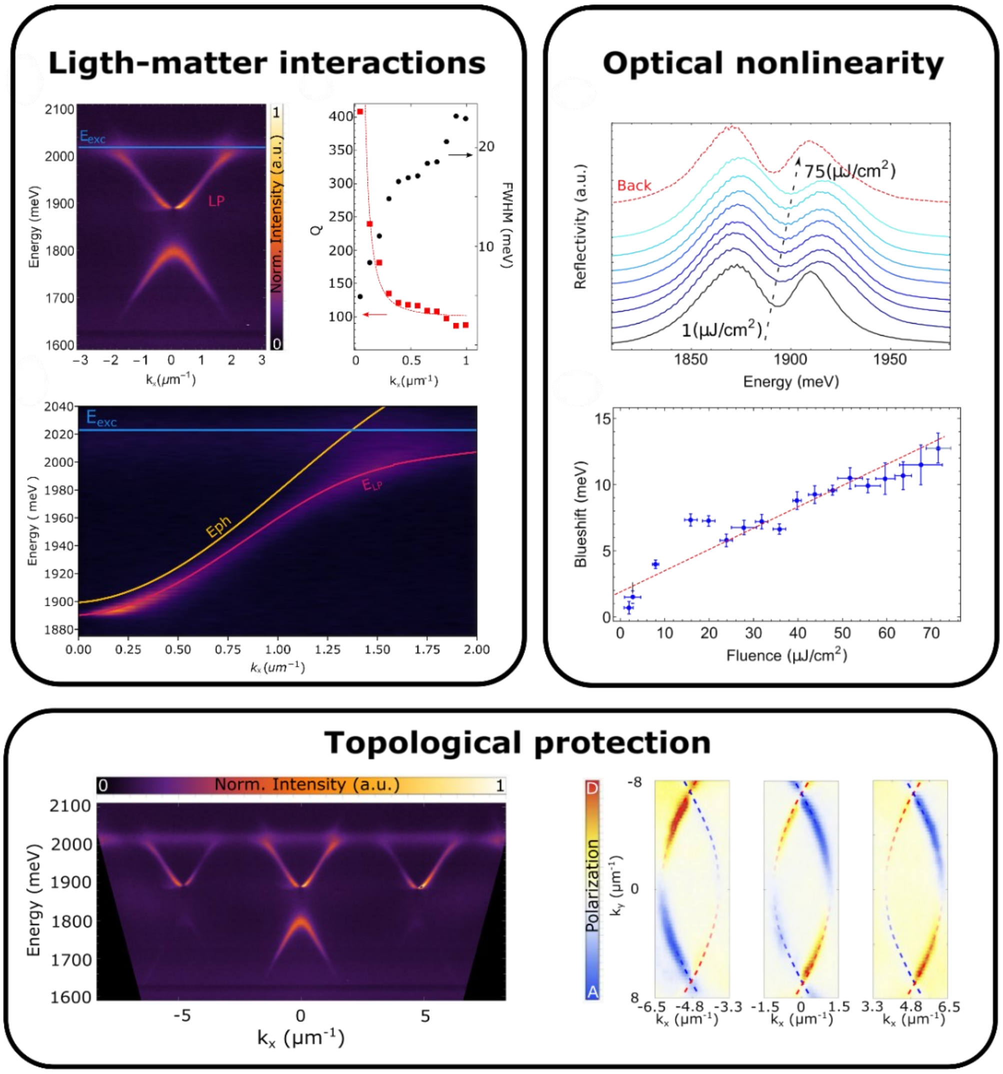
The common key in both approaches is assuring that statement (4) is true, which is not the case for a regular grating; in fact, a regular grating features twice the number of symmetry axes in the solutions proposed above; these axes will fall both in air gaps and full pillars (Fig.5 leftmost), always breaking the syllogism.

3 Experimental results

A demonstration structure was realized (see SEM images in Fig.3 rightmost) and characterized. A white light reflection measurement was performed in order to retrieve the full dispersion of the modes in the system. A non resonant measurement with a pump well above the exciton was then used in order to observe a PL dispersion and better characterized the Rabi splitting (see Fig.6 "Light-matter interactions”). Near-resonant measurements at the BIC polariton energy position were eventually performed upon sweeping pumping power, in order to observe the nonlinear response from the system (see Fig.6 “Optical nonlinearity”).

By both the reflection and PL images we were able to demonstrate a large light matter interaction strength, in terms of a large 70-80meV Rabi splitting. We could also clearly identify the remarkable divergence of the polariton Q factor when approaching the Gamma point, as expected from a BIC state. The large cooperativity which results from the merging of the high Rabi splitting and the high Q factor can be observed in its consequences on the nonlinear response: a large ~15meV blueshift was achieved on a thin linewidth, without observing any quenching effects on the slope of the nonlinear response at the highest powers explored.

As a last but interesting point, we were able to characterize the retention of the topological character for the BIC within the strong coupling regime (see Fig.6 “Topological protection”). In other words we could still clearly see the signatures associated with the topological protection of photonic BICs, namely a purely linear polarization in the farfield which whirls around the Gamma point, despite having transitioned to a fully polaritonic BIC here.



(Fig.6)

PL measurements at low pump powers show a large light-matter interaction strength jointly with an ultrathin FWHM linewidth (ultra high Q-factor). As a result large nonlinear shifts can be observed, which do not quench at high powers thanks to the BIC stability and high detuning. Topological features proper of photonic BICs can also be experimentally observed here, despite the strongly polaritonic and interacting nature of the BIC. Images partially adapted from [4].

4 Conclusions

We have here focused on some relevant points of interest from our previous work **[4]**, to which we shall refer the reader for more details. In particular, we have here highlighted how the “topological protection” often associated to photonic BICs (thoroughly described in [9]) does most of the time offer no actual protection and advantage against refractive index perturbations of both structural origin (nanofabrication inaccuracies) or optical origin (nonlinearities upon increasing pump power or mischaracterization of the device materials). In fact, topological protection in BICs from regular PhCs only assures robust existence of the BIC eigenstate at a specified momentum, but doesn’t prevent it from making sudden discontinuous jumps in energy upon small perturbations, which is indeed the case in a nonlinear system as a polariton. We have here shown a photonic design strategy to solve such an issue – we named it “deterministic placement principle” -, at the same time developing an overall device architecture and a nanofabrication approach which advance the state of the art of room temperature polariton physics in terms of a large Rabi splitting and a record cooperativity. These results, which we validated experimentally and are of further value with respect to the photonic design strategy, rely on a nontrivial embedding process for the TMD monolayer, the adoption of BSWs as elective tools for the strong coupling of 2D materials and the usage of a numerical optimization approach, essential in case of BSW modes because of their large and non-perturbatively-treatable sensitivity to the surface PhC perturbation, well different from the easier manageable character of waveguide modes.

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