Strong coupling of Bloch surface waves and excitons in ZnO up to 430 K

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Abstract
We report on the investigation and observation of Bloch surface wave polaritons, resulting from the interaction between excitons in ZnO and a Bloch surface wave supported by a distributed Bragg reflector (DBR), for temperatures up to 430 K. The samples were fabricated using pulsed laser deposition and consist of a DBR made of 6.5 layer pairs of yttria-stabilized zirconia and Al₂O₃ with a ZnO surface layer. We measured the reflectivity of transverse electric modes using a SiO₂ prism in Kretschmann–Raether configuration, giving access to high in-plane momenta. Whereas the lower polariton branch was clearly observable, the upper polariton branch was not visible, due to the strong absorption in ZnO above the excitonic resonance. By employing a coupled oscillator model for the interaction between the bare surface mode and exciton, we derived a corresponding Rabi splitting between 100–192 meV at 294 K, which decreases with increasing temperature.

1. Introduction

Exciton polaritons are quasi-particles resulting from the strong coupling of a photon to an exciton. In the strong coupling regime the lifetime of the interacting photon is large enough for it to be absorbed and re-emitted several times by an exciton, leading to a dispersion which is characterized by a level splitting [1]. The properties of the exciton–polariton comprise a mixture of those of the photon and the exciton. The photonic contribution leads to fast propagation, a light effective mass and allows optical excitation while due to the excitonic part the particles can interact with each other and the lattice (phonons) [2].

In microcavity systems, these properties enable the observation of a wide variety of physical phenomena ranging from many-body coherence like Bose–Einstein condensation [3–5], superfluidity [6] to the optical spin Hall effect, spin transport [7] and topological insulators [8], which is of interest for applications like low threshold lasers [9, 10], spinoptronic devices [11, 12] and single-photon operation [13, 14]. However, in order to reach high propagation lengths on the order of 1 mm in microcavities, typically low temperatures (<10 K) and high quality factors are required [15]. This entails a considerable production effort and is impractical for room temperature applications. Furthermore, the second DBR mirror impedes any additional structuring of the active layer for the manipulation of polariton states.

A promising alternative for the investigation of polaritons in planar microcavities [16], without the constraints mentioned above, are excitons coupled to guided modes in photonic crystals [17], specifically the Bloch surface wave (BSW), which is a lossless mode propagating along the interface between the surface layer of a distributed Bragg reflector (DBR) and the ambient medium [18].

If the periodicity of the DBR is interrupted by a surface layer of a specific thickness, a guided mode (BSW) appears within the Bragg stop band and outside the vacuum light cone, which forbids radiative coupling to the vacuum field. Thus, the BSW is evanescent on both sides of the surface-ambient interface and propagates along it [19].
Due to their high surface-sensitivity, BSW are widely investigated for sensing applications [20–24]. Furthermore, they have attracted interest for on-chip manipulation [25–27] due to their efficiently controllable long-range dynamics [28–30] and straightforward production processes.

Strong coupling of the surface photon to an exciton results in the formation of a Bloch surface wave polariton (BSWP), which inherits many of the interesting features of the bare BSW. It was reported that these particles can propagate over distances of more than 100 \( \mu m \) [31, 32], which is larger than most polaritons in planar microcavities. Furthermore, BSWP can be easily guided, manipulated and confined along the sample surface by an effective index contrast, e.g. in a ridge geometry [33] and do not suffer from metallic losses like their plasmonic counterpart [34], which renders them a valuable option for the realization of ultrafast low-loss polariton devices with a high surface sensitivity.

In order to ensure functionality at room temperature and above, the exciton binding energy must be larger than the thermal energy. While this constraint is easily met in organic materials due to their large oscillator strengths, the intrinsic disorder in such materials leads to increased inelastic scattering and emission from localized as well as delocalized states [35, 36]. In contrast, ZnO is an inorganic semiconductor exhibiting high crystal quality and well defined exciton resonances with binding energies of about 60 meV, i.e. more than twice the thermal energy at room temperature [37]. This has led to the observation of polaritons above room temperature in ZnO based microcavities [38, 39].

In this paper, we report on the observation of BSWP with ZnO as active medium for temperatures up to 430 K. A model analysis of polarization-resolved reflectivity measurements using a prism coupler allows to verify that the system is in the strong coupling regime.

2. Computation of the mode dispersion

The layered structures are modelled by a (2 \( \times \) 2)—transfer matrix formalism [40, 41]. The transfer matrix \( \hat{T} \) connects the electric field amplitudes in the ambient, \( \mathcal{E}^\pm_a \), and substrate, \( \mathcal{E}^\pm_s \), for waves moving towards (+) and away from (−) the sample:

\[
\begin{pmatrix}
\mathcal{E}^+_a \\
\mathcal{E}^-_a \\
\mathcal{E}^+_s \\
\mathcal{E}^-_s 
\end{pmatrix} = \hat{T}
\begin{pmatrix}
\mathcal{E}^+_a \\
\mathcal{E}^-_a \\
\mathcal{E}^+_s \\
\mathcal{E}^-_s 
\end{pmatrix}.
\]

The transfer matrix \( \hat{T}_i \) for a single homogeneous layer of thickness \( d_i \) with dielectric function \( \varepsilon^{(i)} \) is obtained from a propagation matrix \( P_i \) and an interface matrix \( D_i \) as

\[
\hat{T}_i = D_i P_i D_i^{-1}.
\]

with

\[
k_{\|} = \sqrt{k_0^2 \varepsilon^{(i)} - k_{\perp}^2},
\]

Here, \( k_0 = E/\hbar c \) is the vacuum wave vector, which is connected to the photon energy \( E \), speed of light \( c \) and reduced Planck constant \( \hbar \) and \( k_{\perp} \) and \( k_{\|} \) denote the orthogonal and parallel components of the wavevector with respect to the sample plane. The latter is conserved among all layers. It should be noted that \( D_i \) is generally polarisation-dependent and given here in terms of transverse electric (TE) polarisation, because only BSW modes of this polarisation are supported in our geometry. Note, the corresponding transverse magnetic (TM) mode lies below the energy of the Bragg stop band but, the outlined approach is equally valid for TM polarisation. From (2.2), the complete transfer matrix of a multilayer stack is given as the matrix product

\[
\hat{T} = D_a^{-1} \left( \prod_{i=1}^{N} \hat{T}_i \right) D_s,
\]

where the subscripts \( a \), \( s \), and \( i \) denote the ambient, the substrate and the \( i \)th layer of the structure, respectively. For a given structure and \( k_{\|} \) the mode energy, \( E \), can be obtained from the condition [40]

\[
\hat{T}_{11}(E, k_{\|}) = 0
\]

which corresponds to outgoing but no incident fields i.e. diverging reflection coefficients. The reflectivity in case of incident light from the substrate side can be calculated via \( R = |\hat{T}_{12}/\hat{T}_{11}|^2 \) [42], which corresponds to the experiments reported here.

The dispersion of BSWP, resulting from the interaction between an exciton and BSW in the surface layer, can also be described semi-classically by a coupled oscillator model that considers two interacting states.
Thus a system is characterised by a $(2 \times 2)$—Hamiltonian with non-zero off-diagonal elements \cite{42}

\[
H = \begin{pmatrix}
E_{\text{BSW}} & V \\
V & E_X
\end{pmatrix}
\]  

(2.6)

and eigenvalues

\[
E_{n}^\pm = \frac{1}{2} \left[ E_{\text{BSW}} + E_X \pm \sqrt{(E_{\text{BSW}} - E_X)^2 + 4V^2} \right].
\]  

(2.7)

Here, $E_{\text{BSW}}$ denotes the dispersion of the BSW, which is obtained numerically from (2.5). Furthermore, $E_X$ corresponds to the exciton resonance energy and $V$ is the coupling strength between the exciton and photon. The two branches of the dispersion corresponding to the $+$ and $-$ sign of the square root are termed the upper (UPB) and lower polariton branch (LPB), respectively. While the UPB approaches the exciton and BSW energy for small and large $k$-vectors, respectively, this is reversed for the LPB. Note that, in contrast to exciton–polaritons in planar microcavities, the BSWP does not have a ground state, i.e. for $k = 0$. The coupling leads to a level repulsion with a Rabi splitting defined as twice the coupling strength, i.e. $\Omega = 2V$, at the crossing point of the uncoupled dispersions. At this point, the BSWP has equal photonic and excitonic contributions.

3. Experimental methods

3.1. Sample fabrication

The multilayer structures were produced by means of pulsed laser deposition (Lambda Physik LPX Pro excimer laser, 248 nm, 600 mJ pulse energy) on top of a SiO$_2$ substrate (1 cm$^2$, CRYSTAL GmbH). In this work, we used yttria-stabilized zirconia (YSZ) and Al$_2$O$_3$ as the DBR materials, for which the high contrast in refractive indices ensures a large width of the stop band ($\sim$0.5 eV) \cite{43}. In order to obtain a smooth growth of the layers, a 200 pulse YSZ seed layer was deposited with a repetition rate of 1 Hz. The oxygen partial pressure and repetition rate were adjusted for YSZ, Al$_2$O$_3$ and ZnO to 15 Hz at 0.02 mbar, 15 Hz at 0.002 mbar and 10 Hz at 0.002 mbar, respectively. The fabrication temperature was kept constant at around 920 K resulting in a good crystal quality of the ZnO layer. Furthermore, the dielectric functions (DF) of the materials were determined by means of spectroscopic ellipsometry. In the investigated spectral region the DF of SiO$_2$, Al$_2$O$_3$ and YSZ can be modelled by means of a Cauchy function, i.e.

\[n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4}\]  

(3.1)

and the parameters are given in table 1. Their temperature dependence was linearly modelled as in \cite{38}, i.e. $n(T) = n(290\text{ K}) + 2.5 \times 10^{-5} \times n(T - 290\text{ K})$.

The layer thicknesses of the Bragg reflector are chosen such that the stop band lies in the desired energetic spectral range, around 440 nm ($\sim$ 2.8 eV) at normal incidence. This leads to the exciton resonance being contained in the stop band at large $k$, as it shifts to higher energies for increasing angles of incidence \cite{44,45}. Consequently, the thickness of the top layer is adjusted to ensure the occurrence of a surface mode.

The active medium for the excitation of BSWP was ZnO with the optical axis oriented parallel to the surface normal. The influence of the anisotropy of ZnO on the BSW dispersion was numerically found to be very weakly pronounced and is therefore neglected in the following. The complex DF of ZnO, \[\varepsilon = \varepsilon_1 + i\varepsilon_2,\] for the component perpendicular to the c-axis, \[\varepsilon_1,\] was approximated using a numerical $b$-spline function \cite{46}. Exemplarily for room temperature, the DF is shown in figure 1 and the exciton resonance around 3.3 eV is clearly visible in \[\varepsilon_2.\]

In the following, the results for a sample comprising $N_{DBR} = 6.5$ layer pairs of YSZ ($d \sim 47$ nm) and Al$_2$O$_3$ ($d \sim 59$ nm) and a ZnO surface layer ($d \sim 54$ nm) are shown. Atomic force microscopy measurements of the overall structures on an area of 1 $\mu$m$^2$ yielded average roughness values between 2–5 nm.
3.2. Reflectivity measurements

The surface modes have an in-plane momentum larger than \( k_\parallel > E/\hbar c \), with \( E \) denoting the photon energy, and therefore cannot couple to the vacuum radiation field. In order to excite and observe these states, a coupling mechanism is necessary, which, in the experiments shown here, corresponds to a prism in Kretschmann–Raether configuration \([47]\). A schematic of this configuration is depicted in figure 2(a). The samples are illuminated from the backside through a right-angle SiO\(_2\) prism with refractive index \( n_p \), which is in optical contact with the substrate using a commercially available index matching liquid (\( n \sim 1.46 \)). A light wave at an angle of incidence in air, \( \theta_i \), and with a given energy is refracted inside the prism and propagates at an angle \( \theta_p \) to the surface normal, which is given by Snell’s law

\[
\sin(\theta_p - 45^\circ) = n_p^{-1}(E) \sin(\theta_i - 45^\circ)
\]  

(3.2)

This process effectively increases the in-plane momentum of the incident light inside the prism, \( k_\parallel^p = n_p E \sin \theta_p / \hbar c \), and thus allows for coupling to the surface modes, which become visible as dips in the far-field reflectivity spectrum. The resulting reflectivity spectra for the sample at selected angles of incidence are shown in figure 2(b). The optical constants of the prism were confirmed to match those of SiO\(_2\) using ellipsometric measurements.

In order to obtain polarization resolved reflectivity and probe the mode dispersion in the multilayer structures, we measured the complete Mueller matrix spectra for angles \( \theta_i \) from 45° to 63° in steps of 1°, using a dual rotating-compensator ellipsometer with a spectral range 0.7–8.2 eV. The Mueller matrix, \( \hat{M} \), contains complete information about the state of light upon reflection from a sample \([48]\). Thus, it allows to calculate the corresponding reflectivity spectrum by assuming TE-polarized incident light, i.e. a Stokes vector \( S^i = (1, -1, 0, 0)^T \), as

\[
R_{TE} = \hat{M}_{11} - \hat{M}_{12}.
\]  

(3.3)

For measurements at temperatures between 294 K and 450 K a resistive hot plate was placed below the sample, such that the ZnO layer was closest to the heat source, cf figure 2(a). The temperature readings were
taken on the surface of a reference substrate in close proximity with smaller thermal conductivity compared to ZnO, this may have lead to underestimating the ZnO temperature, but the heating process was performed over long periods of time to minimize this effect.

The reflectivity dips were approximated as a Fano-type resonance, which allows to extract the resonance energy and broadening. Though this type of resonance originates from the investigation of autoionization spectra of He atoms [49], it is also widely used in many different light–matter systems [50], such as photonic crystal slabs [51], microcavity systems [52, 53], nano-plasmonics [54] and BSW [29]. A Fano resonance results from the interaction of a discrete state with a continuum of (degenerate) states. Classically, the Fano resonance can be seen as a continuous set of driven and coupled oscillators, which leads to opposite phases and constructive/destructive interference on both sides of the resonance and hence to an asymmetric response function [55]. For a resonance with energy $E_0$, corresponding to the eigenvalue in (2.7), and FWHM $\Gamma$ it has the following form:

$$F = \frac{(q + \eta)^2}{1 + \eta^2} \quad \text{with} \quad \eta = \frac{E - E_0}{\Gamma/2}. \quad (3.4)$$

The Fano constant $q$ is a measure of the asymmetry of the resonance and it can be considered as the ratio between resonant and non-resonant transition amplitudes in the scattering process. That is, for $|q| \sim 1$ both channels are comparable (large asymmetry). For $|q| \gg 1$ or $|q| \to 0$ resonant or direct scattering dominates and the resonance profile tends toward a Lorentzian shape, which is symmetric around $E_0$. In the region of the reflectivity dip, the spectrum was described by

$$R(E) = \alpha \cdot F(E) - R_0, \quad (3.5)$$

where a constant scaling factor $\alpha$ and an offset $R_0$ were taken into account for non-idealities and measurement inaccuracy. In the system investigated here, the resonance is viewed to arise from the interference between external incident light, which is reflected from the multilayer and absorbed by the exciton continuum (at higher energies) on the one hand and the narrow Bloch mode on the other, resembling scattering to the continuum and a discrete level, respectively. This is very close to the nanocavity levels interfering with light reflected from a photonic crystal slab, as presented in [53]. In purely photonic systems we obtain $|q| \sim 0$ and hence a symmetric resonance, which is displayed in figure B1(a). For the BSWP in ZnO, $|q|$ moves closer to 1 as the mode energy approaches the exciton transition, due to the asymmetric absorption caused by the broadening of the exciton continuum states and the small broadening of the BSW mode compared to the exciton. We characterize the temperature dependence of the Fano parameter in appendix C.

4. Results and discussion

From the measured reflectivity spectra the resonance energy and corresponding full width at half maximum (FWHM) of the BWSW modes were extracted for different angles and temperatures by analysing the spectra in terms of a Fano-type resonance, as described above. The UPB was not observable due to the strong absorption in ZnO above the band gap, which agrees with findings in planar microcavities [38, 56].

The mode energy as a function of the angle of incidence inside the prism, $\theta_p$, for the LPB is depicted in figure 3(a). The symbols correspond to the experimental values, for which the error estimation was performed applying the residual resampling method [57]. The BSW energy decreases with increasing temperature. That is, at $\theta_p = 45^\circ$ and $\theta_p = 57^\circ$ it changes by $\sim 20$ meV and $\sim 60$ meV, respectively, for an overall temperature difference of $\Delta T = 136$ K. The different energetic shift is related to the excitation and photonic composition of the BSWP. At large angles, it is predominantly composed of excitons, leading to a stronger redshift for increasing temperatures than at smaller angles and higher photonic ratio, as the redshift of the BSW energy depends only on the change in the refractive index. The light line in vacuum, given by $k_0 = E/\hbar c$, the exciton energy at room temperature and the uncoupled BSW dispersions are included in figure 3(a). The intensity overlap of the uncoupled modes and the ZnO surface layer is estimated from numerical modelling to lie between 6–22% along the dispersion, for all temperatures. An example of the mode intensity profile at 350 K is given in figure A1. The crossing point of the uncoupled modes lies at $\sim 45^\circ$. The branching behaviour of the LPB around this point, i.e. the bending of the dispersion that is expected for a coupled system, is clearly visible and it is observable up to 430 K. As mentioned above, far from the crossing point, the LPB approaches the dispersion of the BSW and the exciton, respectively, which means an increase for small and a flattening curve for large angles. This behaviour is reproduced within our accessible angular range and the dispersion of the LPB can be seen to approach the energy of the $A$-exciton in ZnO for increasing angles.
Figure 3. Dispersion of the Bloch-polariton, which decreases with increasing temperatures, displayed in (a), with the resonance energies (symbols) and fitted dispersion (solid lines), as well as the exciton energy, \(E_X\) for \(T = 294\) K, the uncoupled \(E_{BSW}\) dispersions and the vertical light line (yellow). Temperature dependence of the exciton energy (b), coupling strength (c) and background dielectric constant (d).

The observed dispersion was described by means of the coupled oscillator model and the results are depicted as the solid lines in figure 3(a), agreeing well with experimental values. A multi-sample fitting procedure was employed, such that the data points were fitted by (2.7) for all temperatures, simultaneously. Thereby, the thermal expansion of the layers was neglected, i.e. the layer thickness was assumed constant. The uncoupled BSW mode is assumed to arise due to a constant dielectric background \(\varepsilon_\infty\) of ZnO and its dispersion is found from (2.5). The fit parameters are the layer thicknesses (within a narrow confidence interval \(\pm 5\) nm), the temperature dependent coupling strength, \(V\), exciton energy, \(E_X\) and the background dielectric constant, \(\varepsilon_\infty\).

The temperature evolution of these parameters can be seen in figures 3(b)–(d). The exciton energy decreases in correspondence to the temperature dependence of the band gap from around 3.34 eV to 3.28 eV, which is in good agreement with the values reported in [38]. At room temperature, a coupling strength of \(V = 96\) meV was found and for increasing temperatures it decreases, down to \(\sim 67\) meV at 430 K. In BSWP systems based on inorganic semiconductors coupling strengths in a similar range have been observed, e.g. 22.5 meV [58] and 98 meV [31] but also values as high as 240 meV [32], due to the large oscillator strengths. However, for most organic semiconductors, the coupling is not stable at high temperatures. In ZnO-based microcavities values around 45 meV and 40 meV were reported in [38, 56]. Using active layers made from defect-free ZnO substrates, an increased coupling strength could be observed [59], i.e. values between 90–130 meV, depending on the cavity layer thickness. The differences in \(V\) compared to what is reported here can be addressed by examining the specific model used to approximate an effective ZnO DF without excitonic transitions. Assuming a constant dielectric background, \(\varepsilon_\infty\), of ZnO results in a linear dispersion of the uncoupled BSW, which crosses the exciton energy at smaller angles and thus increases the value of \(V\). If instead we used a parametric DF based on the model developed in [60] and determined in planar microcavities, where the excitonic contributions were removed empirically as in [38] and similar to [61], a coupling constant \(V \sim 46\) meV is obtained at room temperature. In this case, however, the band-to-band transitions begin to contribute at around 3.7 eV, which shifts the crossing point to higher angles. Therefore we regard these two cases as an upper and lower limit for the coupling strength. The same model dependence of the coupling strength was found by others, before [61, 62].

When describing the uncoupled optical properties by a constant dielectric background only, the estimated value for \(\varepsilon_\infty\) increases with increasing temperature as shown in figure 3(d). The values are comparable to \(\varepsilon_1\) of ZnO in the considered spectral region. In the vicinity of the exciton transitions the refractive index changes strongly and with increasing temperature these transitions are shifted to lower energies, according to the temperature dependence of the band gap energy. Therefore, the change in \(\varepsilon_\infty\) is larger in this spectral range than in the visible spectral range.

In figure 4(a) the FWHM, corresponding to the resonance, which increases for increasing temperatures, mainly due to the increased scattering with phonons, is displayed. It is expected that the mode broadening...
Figure 4. Broadening of the measured BSWP mode at different temperatures in (a), the correction function, which is obtained from the ratio of the experimental and numerical FWHM, $\Gamma_{\text{exp}}$ and $\Gamma_{\text{num}}$, with a quadratic fit ($a = 3.7 \times 10^{-3}, b = 1.75$), is shown in (b) and the open circles in (c) correspond to the corrected FWHM values.

for large angles also approaches the width of the excitonic mode [1], which, in ZnO, is approximately $\Gamma \sim 35$ meV at 290 K [38].

Here, the mode broadening is seen to exceed this value. We attribute this behaviour to the following reasons. The probing source corresponds to white light in a large spectral range (0.7–6.4 eV). Due to energy-dependent refraction inside the prism, a small angular range with varying wavelengths around $\theta_p$ is measured, which increases with the incident angle in air $\theta_i$. This is enhanced due to inhomogeneity of the surface thickness which gives an inhomogeneous broadening of the BSW mode. Furthermore, the incident light is diffracted by the surface roughness, i.e. $k_\parallel$ is shifted, such that also non-resonant parts can couple into the polariton modes [47]. The ratio between the observed mode broadening and expectations from numerical computations at room temperature can be seen in figure 4(b). It has approximately quadratic dependence on $\theta_p$ and can be used as a correction function for the observed mode broadening, for which the result is shown as the open circles in figure 4(c). Consequently, for large angles the corrected broadening approaches $\Gamma \sim 39$ meV at room temperature and a value of 52 meV at 430 K. This generally agrees with findings in [38], underestimating them at temperatures above 370 K, which we attribute to the neglected temperature dependence of the correction function. The same behaviour of the observed mode broadening was found when investigating BSW in purely photonic systems and comparing the FWHM of the reflection dips to model data (see section appendix B). This is further indication for a systematic error in our measurement setup, independent of the specific system under investigation.

Nonetheless, the Rabi splitting of the BSWP at the anti-crossing point exceeds the corresponding broadening values in the system presented here and thus the strong coupling regime is confirmed up to 430 K. This still holds considering the model dependence of the coupling strength and assuming the lower limit for the determined values, if the correction of the FWHM is taken into account, which we view as justified for the above reasons. Above 430 K, the mode structure breaks down and is not observable in experiment. In order to achieve strongly coupled BSWP at even higher temperatures, it is necessary to increase the oscillator strength and reduce the broadening of the exciton, e.g. by improving the ZnO crystal quality. The existence of BSWP at such high temperatures carries a large potential for on-chip polariton devices and integrated optics.

5. Summary

The stable formation of BSWPs is demonstrated for room temperature and up to 430 K in dielectric multilayer samples with an active ZnO surface layer. Due to the large absorption in ZnO, the UPB was not observable in the reflectivity measurements. The FWHM of the polariton modes was extracted and shown to grow with increasing temperature, but it is overestimated compared to numerical expectations. By employing a model analysis it was possible to describe the experimental data in terms of two coupled oscillators and derive a maximum coupling strength of $V = 96$ meV at room temperature, which decreases with increasing temperature, down to $V = 67$ meV at 430 K. We conclude that the system is in the strong coupling regime up to a temperature of 430 K.
Figure A1. (a) Normalized intensity of the BSW inside the structure at 350 K, with colors corresponding to the points along the dispersion as shown in (b) and schematic profile of the dielectric function of the sample (blue line). The overlap between BSW and ZnO layer is shown in (b).

Figure B1. Reflectivity spectra, (a), and corresponding BSW dispersion, (b), for a purely photonic sample. The ratio of $\Gamma_{\text{exp}}$ and $\Gamma_{\text{num}}$ with quadratic fit is shown in (c).

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A. Mode profile and ZnO-BSW overlap

The intensity profile of the calculated uncoupled BSW and its integral overlap with the ZnO layer were calculated along the dispersion. For low energies the overlap is around 5% and grows to above $\sim 20\%$ for increasing values, cf figure A1(b), which is very similar for all temperatures. This increase is mostly due to the shorter decay of the mode in the ambient, which can be seen in figure A1. This is in agreement with findings for a ZnO-based microcavity as described in [38], where an overlap between the excitons in the cavity layer and the electric field of around 10% was found along the dispersion.

Appendix B. Bloch surface wave dispersion

In figure B1 the results for a purely photonic sample consisting of $N_{\text{DBR}} = 6$ layer pairs of YSZ ($d \sim 55 \text{ nm}$) and Al$_2$O$_3$ ($d \sim 70 \text{ nm}$) and a YSZ surface layer ($d \sim 21 \text{ nm}$) are shown. The experimentally obtained mode energies are obtained from a Fano-type resonance and compared to those derived from numerical
simulation of the structure using the transfer matrix, in figures B1(a) and (b). A good agreement between the resonance energies was found.

For the broadening we find the same behaviour as mentioned in the main text. For growing $\theta_p$ the broadening of the experimentally observed modes exceeds the numerical expectation in a quadratic dependence. This supports the assumption that these deviations for both, the BSW and BSWP, are due to the discussed characteristics of the measurement set-up.

**Appendix C. Temperature dependence of $q$**

The evolution of the Fano parameter $q$ as extracted from the reflectance analysis is depicted in figure C1. For rising temperatures it increases approximately linearly for all angles. We attribute this behaviour to the increased broadening of the exciton transition due to phonon interaction.

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