From enhanced diffusion to ultrafast ballistic motion of hybrid light–matter excitations

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Transport of excitons and charge carriers in molecular systems can be enhanced by coherent coupling to photons, giving rise to the formation of hybrid excitations known as polaritons. Such enhancement has far-reaching technological implications; however, the enhancement mechanism and the transport nature of these hybrid excitations remain elusive. Here we map the ultrafast spatiotemporal dynamics of polaritons formed by mixing surface-bound optical waves with Frenkel excitons in a self-assembled molecular layer, resolving polariton dynamics in energy/momentum space. We find that the interplay between the molecular disorder and long-range correlations induced by coherent mixing with light leads to a mobility transition between diffusive and ballistic transport, which can be controlled by varying the light–matter composition of the polaritons. Furthermore, we show that coupling to light enhances the diffusion coefficient of molecular excitons by six orders of magnitude and even leads to ballistic flow at two-thirds the speed of light.

Energy and charge transport in organic materials is central to various fields, ranging from photochemistry and photosynthesis to organic electronic devices. However, due to their disordered nature, transport in molecular systems is dominated by short-range random hopping, making it a slow, diffusive (or subdiffusive) and inefficient process. Moreover, the diffusion length in organic semiconductors is typically limited to ~100 nm at most. These properties pose severe limitations when using organic materials in applications.

Recently, it has been demonstrated that transport in organic semiconductors can be enhanced by coupling to resonant photonic modes. Polaritons, the hybrid light–matter excitations that emerge, are partly excitonic and partly photonic, providing exciting opportunities for controlling the properties of materials. Specifically, due to their photonic component, their wavefunctions extend over macroscopic distances, which results in long-range coherence and facilitates energy transfer and transport phenomena. The long-range propagation of polaritons has been studied in steady-state experiments, both in molecular systems and in other materials. However, constructing a complete mechanistic understanding of cavity-enhanced transport necessitates access to the kinetics of the polaritonic motion. This can be achieved using ultrafast time-resolved microscopy techniques, as employed in studying bare organic semiconductors. Indeed, a few experiments have studied the motion of cavity polaritons, over several microns within a few picoseconds. These studies found that, while the dispersion of cavity polaritons predicted a group velocity of ~20 μm ps⁻¹, the observed velocities were surprisingly smaller than 1 μm ps⁻¹ (refs. 7, 13), pointing toward a fundamental gap in the current understanding of cavity polaritons. Furthermore, although the coupling of excitons to propagating photons is assumed to lead to ballistic motion, these initial studies suggest that this is not necessarily the case. As was shown theoretically, static molecular disorder can reduce the extent of the polaritonic wavefunctions and render the transport non-ballistic. Moreover, at non-zero temperatures, scattering due to random fluctuations can also affect transport. Considering the complexity of such systems, which can also involve dissipative and thermally activated processes in addition to elastic and inelastic scattering, the nature of polaritonic transport in organic systems is clearly far from being understood.

To close this gap and enable the practical implementation of cavity-enhanced transport, we study the ultrafast spatiotemporal
dynamics of surface-bound polaritons in a dielectric one-dimensional photonic crystal supporting ultralong propagation of ~100 μm. These measurements reveal the intricate details of polaritonic transport in molecular systems and allow us to directly obtain the various parameters that govern the motion of polaritons and their dependence on the composition of these hybrid light–matter excitations. As we show, polaritons exhibit a mobility transition between diffusive and ballistic transport, occurring as these excitations become more photon-like. We find that strong coupling counteracts the dispersive and ballistic transport, occurring as these excitations become more photon-like. We find that strong coupling counteracts the dispersive and ballistic transport, occurring as these excitations become more photon-like. We find that strong coupling counteracts the dispersive and ballistic transport, occurring as these excitations become more phonon-like. We find that strong coupling counteracts the dispersive and ballistic transport, occurring as these excitations become more phonon-like. We find that strong coupling counteracts the dispersive and ballistic transport, occurring as these excitations become more phonon-like.

Bloch wave surface polaritons

The system studied in this work is depicted in Fig. 1a (Methods). It consists of a distributed Bragg reflector (DBR) which supports electromagnetic surface modes, named Bloch surface waves (BSWs), within its bandgap and beyond the air light line\(^{44}\). These modes are strongly coupled with a dense film of an organic semiconductor (TDBC, 5,6-dichloro-2-[(5,6-dichloro-1-ethyl-3-(4-sulphobutyl)benzimidazol-2-yldiene)propenyl]-1-ethyl-3-(4-sulphobutyl)-benzimidazolium hydroxide, inner salt, sodium salt; see absorption spectrum in Extended Data Fig. 2). The DBR structure facilitates ultralong polariton propagation\(^{6,10}\), operating in either reflection or emission mode (Methods). Operating in either reflection or emission mode (Methods), the system studied in this work is depicted in Fig. 1a (Methods). It consists of a distributed Bragg reflector (DBR) which supports electromagnetic surface modes, named Bloch surface waves (BSWs), within its bandgap and beyond the air light line\(^{44}\). These modes are strongly coupled with a dense film of an organic semiconductor (TDBC, 5,6-dichloro-2-[(5,6-dichloro-1-ethyl-3-(4-sulphobutyl)benzimidazol-2-yldiene)propenyl]-1-ethyl-3-(4-sulphobutyl)-benzimidazolium hydroxide, inner salt, sodium salt; see absorption spectrum in Extended Data Fig. 2). The DBR structure facilitates ultralong polariton propagation\(^{6,10}\), operating in either reflection or emission mode (Methods). Operating in either reflection or emission mode (Methods), the system studied in this work is depicted in Fig. 1a (Methods). It consists of a distributed Bragg reflector (DBR) which supports electromagnetic surface modes, named Bloch surface waves (BSWs), within its bandgap and beyond the air light line\(^{44}\). These modes are strongly coupled with a dense film of an organic semiconductor (TDBC, 5,6-dichloro-2-[(5,6-dichloro-1-ethyl-3-(4-sulphobutyl)benzimidazol-2-yldiene)propenyl]-1-ethyl-3-(4-sulphobutyl)-benzimidazolium hydroxide, inner salt, sodium salt; see absorption spectrum in Extended Data Fig. 2).

Next, we record the energy-resolved, steady-state emission profiles of the BSWP following a localized, non-resonant excitation (Methods) and plot the results in Fig. 1c. As in previous studies\(^{6,10}\), the emission profile reflects the steady-state spatial distribution of the polaritons and therefore it provides a clear visualization of their long-range propagation. Since the delocalization of the polaritonic wavefunctions stems from their partly photonic nature, it is useful to examine the polariton propagation as a function of its photonic weight \(\alpha\)\(^{17,43}\). The decay length reaches values of more than 150 μm (see white circles in Fig. 1c), demonstrating the ultralong propagation of BSWPs\(^{6,10}\).

Ultrafast microscopy results

To reveal the mechanism of polaritonic transport, we study the dependence of the spatiotemporal dynamics on the polariton composition, using ultrafast pump–probe microscopy\(^{44}\), as illustrated in Fig. 2a (Methods and Extended Data Fig. 5). We excite the sample using a focused pump beam (2.3 eV, 6 μm diameter) and capture the evolving polariton distribution using a wide probe beam, reflected off the sample. At the excited region, the non-resonant excitation is followed by a fast energy redistribution process, populating the entire lower BSWP branch within several tens of femtoseconds\(^{49,50}\). The polaritons start migrating away from their initial positions, with each subpopulation characterized by its own energy and in-plane momentum. As evident from Fig. 1c, the various subpopulations behave in very different manners, which can arise from their energy-dependent group velocity \(V_g\) (Supplementary
Information, section 1). Moreover, the polariton propagation can also be affected by various processes, such as coherent or incoherent scattering and energy relaxation. Therefore, to understand the transport of polaritons, it is imperative to resolve them at each particular polaritonic state separately (that is, at a specific location along the lower BSWP dispersion curve in Fig. 1b). Thus, we match the probe energy and in-plane momentum to those of the polariton, by adjusting the probe incident angle θ and imaging the reflected (broadband) probe through a variable bandpass filter, as shown in Fig. 2a (Methods).

Figure 2b shows a representative sequence of pump–probe images (that is, differential reflectivity ΔR/R) measured at different pump–probe delay times τ. Here the probe energy (1.96 eV) is matched to the polaritonic state marked in Fig. 1b (white circle), with |α_{ph}|^2 = 0.82. The displayed signal is proportional to the local density of excitations in the sample, and hence these images capture the expanding polariton cloud, providing direct evidence of the enhanced energy transport over macroscopic length scales. Starting with a Gaussian-like profile with a full width at half maximum (FWHM) of 5 μm at τ = 0 ps, the polariton cloud broadens to 58 μm within 0.3 ps (expanding in the direction...
of the in-plane component of the probe). Interestingly, compared with previous time-resolved experiments on Fabry–Perot cavities, the propagation distance is an order of magnitude larger and the expansion velocity (estimated as ~180 μm ps⁻¹) is two orders of magnitude higher, and it approaches $V_g$ at the same location on the lower BSWP branch (~190 μm ps⁻¹).

The detailed dynamics of the BSWPs are provided in Fig. 3, which shows the evolving cross sections of the normalized differential signals for various photonic weights (see Extended Data Fig. 7 for representative non-normalized data). Figure 3a shows the results for the bare-exciton energy (2.13 eV or 582 nm), whereas Fig. 3b,c,d shows the evolution of the BSWP cloud for $|\alpha_{ph}|^2 = 0.48, 0.66$ and 0.82 respectively. As expected, at the bare exciton energy no expansion is observed because the diffusion length is far below our resolution. In sharp contrast, when the probe is tuned to the BSWP state with $|\alpha_{ph}|^2 = 0.48$, the polariton cloud expands with time, until it reaches a stationary width of ~14 μm at ~1.2 ps. At the same time, its peak position also shifts by ~4 μm. Both of these effects become more prominent as $|\alpha_{ph}|^2$ increases, as seen in Fig. 3c,d. For all measurements, after reaching a stationary width, the profiles exhibit a spatially uniform decay, with a lifetime of several picoseconds (Extended Data Figs. 6b and 7b). We stress that, because each data series presented in Fig. 3 corresponds to a specific subpopulation with a narrow (~15 meV) energy span and not the entire (incoherent) polariton wavepacket, the observed expansion cannot be associated with the dispersive nature of the polariton energy band. Moreover, the expansion of the polariton cloud proceeds in parallel to the build-up of the polariton population (Extended Data Fig. 7), suggesting that the expansion process involves complex population dynamics between the polaritonic states and the molecular ones.

**Unveiling the mixed nature of polaritonic transport**

Generally, the components that make up the polariton exhibit very different transport behaviour—Frenkel excitons move in a random, diffusive manner, whereas photons can propagate ballistically over large distances. Therefore, a fundamental question is: what is the transport nature of the hybrid polaritons? This question becomes even more intriguing when taking a closer look at the expansion velocities. While for $|\alpha_{ph}|^2 = 0.82$, the expansion follows the group velocity $V_g$ (Supplementary Information, section 1) as expected for purely ballistic transport, at lower photonic weights this picture changes significantly. With $|\alpha_{ph}|^2 = 0.48$, the measured velocity is ~7.6 μm ps⁻¹, 15 times smaller than $V_g$.

To answer this question and to reveal the various parameters that govern transport under strong coupling, we plot in Fig. 4a the variance $\sigma_\tau^2$ of the polariton distribution (Supplementary Information, section 2) as a function of $\tau$ (note the log–log scale). The data clearly show distinct linear regions for all photonic weights, indicating a power-law dependence on time. This trend matches the general behaviour for transport in the presence of random scattering, which is commonly captured by the generalized diffusion equation, with a solution of the form $\sigma_\tau^2 = \sigma_0^2 + D_\beta \tau^\beta$. Here $D_\beta$ is the generalized diffusion coefficient, $\sigma_0^2$ is the variance at zero delay (that is, the point spread function of the system) and $\beta$ is the order of the transport, which is directly linked to the underlying transport mechanism. A $\beta$ value of unity corresponds to diffusive transport, where the distribution expands as $\sqrt{\tau}$, while $\beta = 2$ represents ballistic transport with expansion at constant velocity. To identify the transport nature in each region, we fit the curves shown in Fig. 4a to straight lines, as marked by the black and red lines. We find that for $|\alpha_{ph}|^2 \approx 0.5$ the transport is essentially ballistic (that is, characterized by $\beta = 1$) up to $\tau = 1$ ps, at which time the polariton distribution reaches its stationary width. On the other hand, polaritons with $|\alpha_{ph}|^2 > 0.7$ exhibit a very different behaviour, with $\beta = 2$, indicating that the transport becomes ballistic due to the mixing with the photons. Interestingly, for intermediate cases, that is, with $0.6 \lesssim |\alpha_{ph}|^2 \lesssim 0.7$, the log–log plots clearly show the existence of two distinct regions, designating a crossover in the transport mechanism as the polariton cloud expands, which clearly demonstrates the mixed transport nature of polaritons. For example, for $|\alpha_{ph}|^2 = 0.6$ the expansion is initially ballistic (with $\beta = 2$) and becomes diffusive at $\tau = 0.3$ ps, where the slope changes to ~1. The results of this analysis are summarized in Fig. 4b, which shows the extracted exponents as a function of $|\alpha_{ph}|^2$, which aggregate around either $\beta = 1$ or $\beta = 2$. The sudden transition between these two values indicates that the transition between the different transport regimes occurs at a specific length/time scale, rather than passing gradually through a superdiffusive region.

By further analysing the time-dependence of the polariton cloud width, we can also extract the diffusion coefficient and the ballistic velocity (Supplementary Information, section 2), which are plotted in Fig. 5a. The diffusion coefficient increases with the photonic weight,
reaching 284 μm² ps⁻¹ at |αph|² ≃ 0.69. Recalling that the typical diffusion coefficient for excitons in semiconductors (organic and inorganic) is 10⁻³–10⁻² μm² ps⁻¹ (refs. 4, 38, 39, 42), our results signify a more than six orders of magnitude enhancement of the diffusion coefficient due to strong coupling with photons.

Similarly, the polariton expansion velocity in the ballistic regime (black circles in Fig. 5a) also increases with |αph|². For comparison, we plot the theoretical group velocity Vg, which represents the expected velocity in an ideal, perfectly homogeneous system with fully delocalized wavefunctions (Supplementary Information, section 1). At |αph|² = 0.60, the lowest photonic weight for which a ballistic regime is observed, the expansion velocity is 30 μm ps⁻¹, considerably smaller than the Vg at this photonic weight. Moreover, even for |αph|² ≥ 0.7, where the diffusive region disappears and the transport appears to be purely ballistic, the measured velocity nevertheless remains lower than Vg, indicating that even under these conditions the effects of the disorder cannot be completely ignored. As the photonic component of the polaritons continues to increase, the effect of the coherent coupling becomes larger, and the measured expansion velocity becomes closer to Vg (for the corresponding value of |αph|²). At |αph|² = 0.82 the observed velocity reaches 176 μm ps⁻¹, which is comparable to Vg and is two-thirds the speed of light in vacuum.

The complete mapping of the transport dynamics, as presented in Fig. 4a, reveals a very rich yet intuitive picture for the motion of polaritons. Starting with diffusive molecular excitons with a typical mean free path l of only several nanometres, the mixing with photons under strong coupling gives rise to collective, extended polariton wavefunctions, resulting in long-range spatial correlations and enhanced transport. However, due to the underlying molecular disorder, dynamic fluctuations and the various dissipative processes occurring in the system, these polaritonic wavefunctions are not infinitely delocalized. Under these circumstances, it should be expected that the polariton propagation will not be purely ballistic and will still be characterized by a finite mean free path, similar to pure optical waves propagating through a scattering medium under the influence of static or dynamic disorder. This mean free path should contain information about the microscopic scattering mechanisms acting on the polaritons, either static or dynamic. Initially, when the excitonic component of the polaritons is relatively large (that is, ~0.5) their mean free path, albeit much larger than that of the bare excitons, is still smaller than the resolution of our set-up. Therefore, the expansion process, as captured by the measurements, is dominated by multiple scattering, leading to the observed diffusive behaviour at all times. This diffusion, however, is enhanced relative to bare excitons, which we directly observe as the micrometre-scale propagation of these polaritons. As the photon weight is increased, the mean free path of the polaritons reaches macroscopic scale (that is, on the order of 10 μm). At this point, the initial ballistic propagation, which persists until the polaritons traverse a distance comparable to l' (when scattering events start to dominate), becomes accessible to our time-resolved microscopy. Finally, for a high enough photon weight, the mean free path becomes comparable to the length scale set by absorption, at which point the polariton propagation is dominated by dissipation, rather than random scattering. This results in purely ballistic expansion which is now limited by the loss length (which sets the final width of the polariton distribution). This becomes even more apparent when considering the intermediate cases of 0.6 ≤ |αph|² ≤ 0.69 in Fig. 4a,b for which we explicitly observe the transition between ballistic and diffusive flow occurring during the propagation. In fact, since we obtain both the ballistic velocity and the diffusion coefficient for those specific measurements (Fig. 5a), we can extract the microscopic quantities governing the transport in this intermediate range of photonic weights. The mean free time (τ*) and transport mean free path (l') are related to the diffusion coefficient and ballistic velocity by D = l'τ*/2 and l' = νgτ* (ref. 55). Using the values shown in Fig. 5a, we calculate the transport mean free path l' and the scattering rate νg = 1/τ*, which are represented in Fig. 5b by the blue circles and green crosses, respectively. The mean free path increases monotonically with the photonic weight, while the scattering rate decreases monotonically. This is consistent with the behaviour seen in Fig. 4a where the transition to diffusive transport occurs at a later time (and hence at a longer distance) as the photonic weight of the polaritons increases. This causes the diffusive regime (red lines in Fig. 4a) to gradually shrink, until it completely disappears at |αph|² = 0.71, when the mean free path becomes comparable to the absorption length. Interestingly, the values obtained for the mean free path are on the order of tens of micrometres, which is four orders of magnitude larger than the intermolecular distance, demonstrating the long-range correlation induced by strong coupling. Moreover, when the polariton scattering is dominated by excitonic processes

Fig. 5 | Polariton transport parameters. a. Diffusion coefficient (red crosses) and ballistic expansion velocity extracted from the data in Fig. 4a. The values of the diffusion coefficient were obtained from the regions with β ≥ 1 (marked by red lines in Fig. 4a) while the expansion velocity values were obtained from the regions with β ≥ 2 (black lines in Fig. 4a). The solid black line shows the theoretical group velocity, calculated from the dispersion of the lower BSWP branch, and the blue line marks the speed of light in vacuum. The black and red dashed lines show the results of the kinetic model based on thermally activated polariton scattering. b. Polariton scattering rate (green crosses) and transport mean free path (blue circles), obtained for the transport dynamics at intermediate values of |αph|² for which mixed transport behaviour is observed. The green dashed line is a linear fit of the scattering rate (that is, τ* = γph [1 - |αph|²] with γph = 80 fs, extracted by a least-squares fitting) while the blue dashed line shows the result of the kinetic model. The transport parameters are averaged over three different locations on the sample for each data series shown in Fig. 4a. The data points represent the mean values and the error bars represent the s.d. from these locations.

Article | https://doi.org/10.1038/s41563-022-01463-3
that is, static molecular disorder, exciton–phonon scattering, interaction with dark states, etc.) the scattering rate should be proportional to the excitonic fraction \( |\alpha_x|^2 = 1 - |\alpha_{ph}|^2 \) (refs. 34, 53). As seen in Fig. 5b, our results are in agreement with this trend, as indicated by the dashed green line, corresponding to \( y^* = y_x \left( 1 - |\alpha_{ph}|^2 \right) \) with \( y_x^{-1} = 80 \, \text{fs} \). While the diffusion coefficient is a macroscopic quantity characterizing the motion following multiple scattering events (that is, over time scales larger than \( \tau^* \) and length scales larger than the mean free path), it is interesting to consider again the observed velocities in the ballistic regime (black circles in Fig. 5a). As discussed above, we observe a significant discrepancy between the polariton expansion velocity \( V^* \) and the group velocity \( V_x \) extracted from the polariton dispersion, with the observed velocity being lower than \( V_x \). This reduction in velocity, in a similar manner to the scattering rate, becomes larger as the polaritons become more exciton-like. In complex media, reduced effective velocities are often observed in the single-scattering limit due to intermittent trapping of the waves in long-lived localized states, for example, due to resonant scatterers \(^{31,31}\). In the case of molecular polaritons such a process can originate from reversible population transfer between the polaritonic states and the stationary dark manifold, as observed in recent numerical simulations \(^{31}\). As described in Supplementary Information, section 3, assuming that such a back-and-forth population transfer follows an Arrhenius-type behaviour (that is, thermally activated), we find that the redution in velocity should vary with the polariton energy as \( 1 + G e^{-\Delta E/k_B T} \), where \( \Delta E = E_x - E_{ph} \) is the thermal energy (at room temperature), \( G \) is a constant and \( E_x/E_{ph} \) is the exciton/polariton energy. The effective polariton velocity calculated using this kinetic model, and the diffusion coefficient based on it (see Supplementary Information, section 3) are plotted in Fig. 5a (black and red dashed lines, respectively). Despite its simplicity, this model reproduces the experimentally observed trends reasonably well, indicating that thermally activated scattering and energy redistribution processes play an important role in polaritonic transport at room temperature.

**Conclusion**

We present a comprehensive time-resolved study of BSWPs, taking advantage of their ultralong propagation to map the enhanced transport dynamics in strongly coupled organic semiconductors. This enables us to directly quantify the mesoscopic parameters governing the mechanism of cavity-enhanced transport and their dependence on the mixing between light and matter. As we reveal, the competition between molecular-scale disorder and long-range correlations leads to mixed transport behaviour exhibiting a clear transition between diffusion and ballistic expansion. Boosting the mean free path up to tens of micrometres, cavity-enhanced transport is initially manifested as a 10\(^3\) enhancement in the diffusion coefficient and, at higher photonic fractions, when it overcomes the disorder, as fully ballistic transport. This ballistic propagation, which is observed even with an excitonic fraction as high as \(-20\%\), can reach two-thirds the speed of light. The diffusive behaviour and the clear influence of disorder on polaritonic transport calls for further experimental and theoretical exploration to understand its origins. This includes, for example, the effect of scattering due to static or dynamic disorder and polariton–phonon interactions. Our results provide crucial insight into the mechanism of cavity-enhanced transport, which is essential for engineering transport for future applications, such as organic optoelectronic devices exhibiting superior conductivity, or active photonic devices relying on the ultrafast response and ultralong propagation of polaritons \(^{30}\).

**Online content**

Any methods, additional references, NaturePortfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41563-022-01463-3.
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Methods
Sample preparation
The DBR structure was fabricated by radiofrequency sputtering alternating layers of TiO2 and SiO2 on a precleaned glass cover slip. The structure consisted of six pairs of 85 nm TiO2/120 nm SiO2 layers, topped with a 13 nm TiO2 layer which was added to tune the BSW dispersion such that it crossed the TDBC absorption wavelength (582 nm) at an incident angle of 45° (for light impinging from the prism side). The organic layer was deposited on the DBR structure using the method of spin self-assembly6. The substrate was first cleaned by sonication in a Helmanex solution, followed by rinsing with water, acetone and isopropanol, and dried with a stream of nitrogen. Then, the sample was treated with oxygen plasma to create a negatively charged surface, followed by the sequential deposition of alternating layers of cationic polyelectrolyte PDAC (poly(diallyldimethylammonium chloride) and anionic TDBC (5,6-dichloro-2-[(5,6-dichloro-1-ethyl-3-(3-sulfopropyl)-2(3H)benzimidazolidene)-1-propenyl]-1-ethyl-3-(3-sulfopropyl)benzimidazolium hydroxide, inner salt, sodium salt, Few Chemicals), beginning and ending with a cationic layer. The polyelectrolyte PDAC (high molecular weight, 20% in water, Merck) was diluted 1:400 with deionized water and the anionic TDBC molecules were dissolved in water (0.026 wt%).

Using the spin self-assembly technique, each layer was created by depositing 500 μl of the appropriate solution on the substrate using static spin coating (3,000 r.p.m., 15 s) followed by two rinses with water while spinning at low speed (500 r.p.m.) to remove any excess molecules, removing the water after each rinse by spinning at high speed (3,000 r.p.m.). The build-up of the dye layers was monitored by measuring the absorbance of TDBC after the deposition of each layer (Extended Data Fig. 1a). The absorbance increased in a linear fashion with the addition of each bilayer (Extended Data Fig. 1b). Three molecular bilayers (PDAC + TDBC) were determined to be sufficient for strong coupling with the DBR Bloch surface wave mode. Finally, the sample was sealed under a nitrogen atmosphere to prevent oxidative damage by placing a similarly sized microscope slide on top of the sample substrate (with a 1.2 mm spacer between them) and then sealing with NOA68 optical adhesive (Norland Products).

Steady-state characterization of BSWPs
An illustration of the set-up used to measure the reflectivity and emission of BSWP modes beyond the light line is shown in Extended Data Fig. 2a. In reflection mode a white-light beam is focused on the sample in Extended Data Fig. 2a. In reflection mode a white-light beam is focused on the sample. In reflection mode, the dispersion is constructed by resolving any one of the modes of the system (specifically the BSWPs) can be absorbed in the sample, and therefore the polariton dispersion is detected as a reduction in the reflected intensity. On the other hand, in emission modes, the dispersion is constructed by resolving the energy/momentum of photons emitted from the modes of the system. Note that the BSWP dispersion curves acquired through both reflection and emission coincide with each other (and also with the calculated dispersion) with no apparent blue-shift in energy. This indicates that the measurements are conducted within the linear regime, such that the effect of density-dependent polariton–polariton interactions is negligible under our experimental conditions. As shown in the inset of Extended Data Fig. 2b, the linewidth of the BSWP is as small as 4.7 meV, corresponding to a quality factor of ~400. As the BSWP energy approaches the bare exciton energy, the photonic component of the polariton is reduced, which is accompanied by a slight increase in its linewidth and reduction of the Q-factor. Extended Data Fig. 3 shows the quality factor at the lower polariton branch as a function of the polariton energy (blue circles), and its photonic contribution (red line), calculated from the measured dispersion as

\[
\frac{q}{2\pi} = \sqrt{\left(\frac{E_x}{\mu} \right)^2 + \left(\frac{E_p}{\mu} - E_p\right)^2}
\]

Here \(\mu = 71 \text{ meV}\) is the coupling constant (half of the Rabi splitting energy), \(E_x = 2.13 \text{ eV}\) is the molecular exciton energy for TDBC and \(E_p(k)\) is the polariton energy. The spatial distributions of the steady-state emission measured for the BSWP (Fig. 1c) clearly demonstrate the long-range propagation of the BSWPs. To quantify the propagation length, we plot the emission intensity profile (essentially the vertical cross section of the data in Fig. 1c) as a function of position in Extended Data Fig. 4a for polaritons with \(|\alpha|_f^2 = 0.48 \text{ and } 0.82\), marked by points A and B in Extended Data Fig. 2c (note logarithmic scale). As seen, the emission profiles exhibit an exponentially decaying tail, which can be fitted by straight lines in the semi-log plot (black lines in Extended Data Fig. 4a). Interestingly, the observed emission decay lengths match nicely with the spatial scale set by the width in momentum space (that is, \(2\pi/\Delta k\)) of the polaritonic resonance.

Extended Data Fig. 4b shows the decay length, as extracted from the steady-state emission profiles, as a function of \(|\alpha|_f^2\) (blue circles, same as in Fig. 1c), and the length obtained by calculating \(2\pi/\Delta k\) based on either the measured dispersion (red circles) or the transfer matrix simulations. Note the discrepancy at low values of \(|\alpha|_f^2\), which most probably originates from the limited spatial resolution of this particular set-up, which prevents us from accurately measuring decay length values lower than ~40 μm.

Pump–probe microscopy
To capture the full spatiotemporal dynamics of BSWPs we use a custom-built ultrafast pump–probe microscopy system, a sketch of which is presented in Extended Data Fig. 5. The optical set-up is based on our previous time-resolved polariton imaging experiments7 and adapted for measuring surface wave excitations beyond the air light line by operating the pump–probe set-up in the Kretschmann attenuated total internal reflection configuration.

We use a pulsed laser amplifier (Spitfire Ace) operating at 500 Hz with 80 fs pulses and a wavelength of 800 nm to generate a beam which is split to eventually produce both the pump and probe beams. The pump beam is produced by focusing the 800 nm light onto a sapphire plate, generating pulses of supercontinuum white light which are then filtered using a 530 nm bandpass filter (with a bandwidth of 35 nm). This beam is passed through an optical chopper which is synchronized with the femtosecond-pulsed laser to reduce its repetition rate to 250 Hz and then focused onto the sample at normal incidence using a 40× objective (Olympus; numerical aperture, 0.8). We note that the pump is injected into the sample from the molecular layer side, allowing us to pump all the BSWP modes indirectly and non-resonantly. The second portion of the amplifier output is sent into an optical parametric amplifier (Topas, Light Conversion) which converts the pulse wavelength to ~610 nm with a bandwidth of ~20 nm. This beam is then sent through a delay stage to control the time difference between the pump and probe pulses. The probe beam passes through a BK7 prism as shown in Extended Data Fig. 5 and is incident on the sample at an angle close to 45° such that its in-plane momentum is matched with that of the polaritons. To measure the expanding polariton cloud in a wide-field (that is, non-scanning) mode, the pump and probe beams are spatially overlapping in the plane of the sample, with the probed area diameter (~1 mm) being much larger than both the imaging field of view (~90 μm).
and the area excited by the pump beam (having a diameter of ~5 μm full width at half maximum). The reflected probe beam is imaged onto a scientific CMOS camera using a camera lens (Nikkor 50 mm f/1.2 AiS, Nikon) with a 52× magnification.

A plane-parallel plate (K-space translator) along with a long focal length lens (300 mm) allow us to change the incident angle around 45°. This, in conjunction with a narrow bandpass filter (5 nm bandwidth) positioned before the camera, allows us to match the energy and in-plane momentum of the probe to those of the BSWPs that we intend to probe. For a particular incident angle θ and energy $E_p$, the in-plane momentum is given by $k_x = n_p k_0 \sin \theta$, with $n_p$ being the prism refractive index and $k_0 = 2\pi E_p/\hbar c$. It should be noted that the transient spectrum of the polaritons exhibits an antisymmetric profile centred around the polariton energy (Extended Data Fig. 6a). Therefore, to prevent the spectrally integrated pump–probe signal from averaging out to zero, we red-detune the probe wavelength from the polariton resonance by 2.5 nm.

The camera integration time is set to 1.2 ms, and the camera is synchronized with the laser amplifier. The data are acquired as pairs of images, such that each pair of acquisitions captures first the probe image following a pump pulse (the ‘pumped’ image, $I^*$) followed by a probe image without excitation (the ‘unpumped’ image, $I_0$). Using these two intensity distributions, the relative transient reflection can be obtained, defined as $\Delta R/R = [I^* - I_0]/I_0 = I^*/I_0 - 1$ where $I^*/I_0$ is the pixel-wise ratio of the two intensity distributions. Extended Data Fig. 7 shows the (non-normalized) cross sections of the spatial distributions captured for $|\alpha_{ph}|^2 = 0.82$ at various times during the build-up and decay of the signal.

The pump intensity was adjusted such that the magnitude of the signal varied linearly with it, with no apparent changes in the shape of the signal, as shown in Extended Data Fig. 8. This ensures that polariton–polariton interactions, which may further complicate the dynamics, are negligible under the experimental conditions.

**Reporting summary**

Further information on research design is available in the Nature Portfolio Reporting Summary linked to this article.

**Data availability**

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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**Acknowledgements**

This research was supported by the Israel Science Foundation, grant number 1435/19 and 1993/13. The authors thank G. Markovich, T. Ellenbogen, S. Reuveni, C. Genet, G. Groenhof and A. Nitzan for useful discussions. T. S. is grateful to M. Segev for his kind support.

**Author contributions**

M.B. and T.S. conceived the idea and designed the experiments. M.B. conducted the majority of the experimental measurements, data analysis and modelling. A.S. and A.G. carried out the sample preparation. G.S. participated in the optical measurements. G.A. fabricated the dielectric DBR structures. M.B., A.G. and T.S. wrote the manuscript.

**Competing interests**

The authors declare no competing interests.

**Additional information**

Extended data is available for this paper at https://doi.org/10.1038/s41563-022-01463-3.

**Supplementary information**

The online version contains supplementary material available at https://doi.org/10.1038/s41563-022-01463-3.

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*Nature Materials* thanks Ivan Shelykh and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Characterisation of TDBC molecular layers. (a) The absorbance of the TDBC layers deposited on the DBR substrate, measured after the addition of each successive bilayer, beginning with the deposition of a layer of PDAC. (b) The value of the absorbance measured at 582 nm as a function of the number of bilayers deposited on the DBR. The black line represents a linear fit to the data.
Extended Data Fig. 2 | Steady state characterisation of BSW. (a) A sketch of the Kretschmann spectral imaging set-up used for either angle-resolved or spatially resolved reflection/emission steady-state measurements. (b,c) Dispersion of the BSW polariton modes measured via angle-resolved reflection (b) and emission (c) spectroscopy (represented by the false-colour plots). The sharp signal corresponds to the lower BSW polariton mode, while the white dashed lines represent the simulated dispersion (using the T-matrix method). The solid white lines indicate the bare BSW dispersion and the exciton energy (fixed at 2.13 eV), the red dashed line indicates the light line and the inset in (b) shows the typical linewidth of the reflection dip corresponding to the BSW resonance, as measured around 1.91 eV. Note that in the reflection measurement (b), unlike similar angle-resolved reflectivity measurements conducted on polaritons in normal Fabry–Perot cavities,7,49 here only the lower BSW polariton branch is observed. Since such reflectometry measurements are sensitive to absorption into the modes of the system the fact that the upper polariton is not observed probably results from inefficient coupling between incoming photons and the upper polariton modes via the prism. Moreover, in the emission measurements (c) the upper polariton is also missing, which is consistent with previous measurements and results from the fast, nonradiative decay of the upper polaritons49,60.
Extended Data Fig. 3 | Properties of BSWPs. Quality factor (blue circles) and photonic weight (red line) as a function of energy of the lower polariton branch. As the polariton energy shifts away from the bare exciton energy, its Q-factor (and lifetime) increases while it becomes more photon-like.
Extended Data Fig. 4 | BSWP steady state spatial profile. (a) Representative steady state emission profiles (note logarithmic scale) measured at the points A and B indicated in Extended Data Fig. 2c. The black solid lines show the exponential fits to the data. (b) Decay length as a function of photonic weight (blue circles), extracted from the exponential fit to the tails of the steady-state distributions given in Fig. 1c. The red circles and the black solid line show the decay lengths calculated as the inverse of the width in Fourier space, from both experimental reflectivity measurements and transfer-matrix simulations respectively.
Extended Data Fig. 5 | Pump–probe microscopy set-up. Detailed illustration of the pump–probe microscopy set-up, as described in Methods.
Extended Data Fig. 6 | Spectral and temporal transient response of BSWP. (a) Transient reflection spectra, measured using a pump–probe spectrometer (Helios, Ultrafast Systems) at \( \tau = 1 \) ps for probe incident angles of \( \theta \sim 43^\circ, 45^\circ \) and \( 46^\circ \). The spectra show two resonant features, similar to the transient spectra observed in strongly coupled Fabry–Perot cavities: a prominent, angle-dependent feature around the energy of the lower BSWP (corresponding to \(|\alpha_{ph}|^2\) values of 0.82, 0.66 and 0.54) and a second, weaker one that occurs at the bare exciton energy and does not show any angular dependence. The shaded regions mark the 5 nm-wide spectral band which is probed by the time-resolved imaging set-up. (b) Temporal dynamics of the spectral features observed in (a) showing the decay kinetics of the BSWP signals corresponding to \(|\alpha_{ph}|^2\) values of 0.82, 0.66 and 0.54 and at the bare exciton energy. The measured lifetime for the BSWPs are 3.8, 6.5 and 6.6 ps respectively while at the bare exciton energy we observe a lifetime of 6.6 ps.
Extended Data Fig. 7 | Long-time evolution of BSWP distribution. Horizontal cross sections of the ΔR/R distribution measured during the rise (a) and decay (b) of the signal for |\(\alpha_{\text{ph}}\)| = 0.86.
Extended Data Fig. 8 | Verification of signal linearity. Magnitude (a) and normalized cross sections (b) of the $\Delta R/R$ signal under various pump energy densities (measured at a time delay of 1 psec). The dotted lines in (a) show the 95% confidence bounds for the linear fit.
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