

Optical Signatures of Dynamical Excitonic Condensates

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We theoretically study dynamical excitonic condensates occurring in bilayers with an imposed chemical potential difference and in photodoped semiconductors. We show that optical spectroscopy can experimentally identify phase-trapped and phase-delocalized dynamical regimes of condensation. In the weak-bias regime, the trapped dynamics of the order parameter's phase lead to an in-gap absorption line at a frequency almost independent of the bias voltage, while, for larger biases, the frequency of the spectral feature increases approximately linearly with bias. In both cases, there is a pronounced second-harmonic response. Close to the transition between the trapped and freely oscillating states, we find a strong response upon application of a weak electric probe field, compare the results to those found in a minimal model description for the dynamics of the order parameter's phase, and analyze the limitations of the latter.

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Introduction—Excitons, due to their bosonic nature, can undergo condensation akin to the condensation of Cooper pairs in superconductors [1–5]. The typically light exciton mass implies the possibility of coherence at high temperatures. While several materials were proposed to host excitonic condensation in the ground state [6–9], progress in the fabrication of heterostructures [10–12] and in ultrafast techniques [13–15] has opened the possibility of stabilizing *dynamical excitonic condensates*, i.e., nonequilibrium quantum states where excitons condense in a coherent macroscopic state. In these, the excitonic population is maintained by a nonequilibrium process involving supplying electrons and holes either by optical excitation [15–20] or by separately contacting gates to each layer in bilayer structures with interlayer excitons [21–28]. In the latter setup, hybridization of intervalley excitons with opposing dipoles was recently reported [29]. In either case, the nonequilibrium stabilization of the excitonic phase implies an interlayer chemical potential difference that may drive a time dependence of the condensate.

One recurring problem in the field is how to experimentally detect the coherence of dynamical excitonic condensation. Recent observations of photoinduced absorption edge enhancement in time-resolved optical spectroscopy [16] and fine structure in the angle-resolved photoemission [13] were interpreted as signatures of dynamical condensation. Dynamical excitonic condensates can host characteristic phenomena like excitonic fluids [25,26], perfect Coulomb drag [27,28,30], and coherent photoluminescence [31,32]. While measuring transport properties using Coulomb drag is challenging, recent studies claim to observe the perfect Coulomb drag [27]—however, without superfluidity [28].

Several theoretical studies analyzed the properties of dynamical excitonic condensates assuming no intrinsic tunnelling between layers, leading to effects like the band-gap renormalization due to injected charge carriers [24,33] and oscillating polarization (photocurrent) with a pump tunable frequency [17,34] as well as dynamical instabilities of the excitonic condensate [35,36]. However, the approximation of perfect decoupling is not always physically reasonable, and weak tunneling will break the U(1) order parameter symmetry, which can induce finite interlayer resistance in Coulomb drag experiments [28] and interesting dynamical effects, including the ac Josephson effect [37] and tunability between dark and bright excitons [38], and leads to the exciton-driven band-gap

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renormalization [39]. Bright interlayer excitons are particularly interesting, since they provide in-plane electromagnetic polarization, which acts as an antenna for out-of-plane emission or absorption of photons.

Therefore, it would be highly desirable to have direct experimental signatures of dynamical exciton condensates similar to early interference measurements of photoluminescence in heterostructures within the Hall regime [32]. In this Letter, we show that the optical response of dynamical excitonic condensate carries important information about the dynamics of the order parameter's phase exhibiting a sharp transition between the trapped and the freely rotating oscillations with an increasing number of excited charge carriers. We use time-dependent dynamical mean-field theory (tdDMFT) to evaluate the optical response of the system, as a function of nonequilibrium drive in the trapped and deconfined phases and, in particular, in the vicinity of the transition. The general features of this instability can be understood within an effective Landau-Ginzburg theory of the order parameter phase; however, microscopic dynamics show qualitatively different dynamics close to the instability.

Model—We will restrict to a minimal model capturing the essence of a coupled bilayer by considering one band in each of the layers, which we label as v (valence) and c (conduction) band electrons. Taking into account band inversion and a local interband interaction U gives rise to the following Hamiltonian:

$$\begin{aligned}\hat{H}_{\text{hop}} &= -t_h \sum_{\langle i,j \rangle} (c_i^\dagger c_j - v_i^\dagger v_j) \\ \hat{H}_{\text{loc}} &= \frac{\Delta\epsilon}{2} \sum_i (c_i^\dagger c_i - v_i^\dagger v_i) + t_{\text{hyb}} \sum_i (v_i^\dagger c_i + c_i^\dagger v_i) \\ &\quad + U \sum_i \left(v_i^\dagger v_i - \frac{1}{2} \right) \left(c_i^\dagger c_i - \frac{1}{2} \right).\end{aligned}\quad (1)$$

Here, $\langle i, j \rangle$ denotes nearest neighbors, $\Delta\epsilon$ is the energetic distance between the noninteracting bands, and t_{hyb} is the strength of the interlayer tunneling. $\hat{H}_{\text{hop}}$ describes intra-layer hopping, whereas $\hat{H}_{\text{loc}}$ contains all interlayer couplings. To include interaction effects in a nonequilibrium setup, we will solve the problem within tdDMFT [40,41] on the infinitely connected Bethe lattice using the second-order self-consistent expansion as an impurity solver; see Ref. [42] for details. In order to be able to unambiguously resolve tunneling-induced in-gap features in the optical conductivity, we choose $U = 5$ and $\Delta\epsilon = 0.6$ in the following, which gives rise to a large optical gap. All energy (time) scales are measured in units of (inverse) hoppings t_h (t_h^{-1}), and the equilibrium temperature is given by $k_B T = 0.05 t_h$. We expect the conclusions of this Letter to be qualitatively unchanged under the inclusion of a spinful description [47,48] and weak disorder. We use the

simplest gauge-invariant formulation of the electron-light coupling [49–53] leading to intraband Peierls substitution $-t_h c_i^\dagger c_j \mapsto -t_h e^{iA(t)} c_i^\dagger c_j$. We also assume that the interband transition is dipole active, leading to an additional Hamiltonian term:

$$\hat{H}_{\text{dip}}(t) = -DE_{\text{pr}}(t)(v_i^\dagger c_i + c_i^\dagger v_i) =: -E_{\text{pr}}(t)\hat{P}, \quad (2)$$

where $E_{\text{pr}} = -\partial_t A$ is the applied in-plane electric field and $\hat{P}$ is the corresponding in-plane polarization operator. The total energy is, thus, given by $E \equiv \langle \hat{H}_{\text{hop}} + \hat{H}_{\text{loc}} + \hat{H}_{\text{dip}} \rangle$. This coupling implies that the Hamiltonian is not centrosymmetric, which could be realized by, e.g., s orbitals in one layer and p orbitals in the other whose centers are not aligned. We take $D = 0.1$ in this Letter. The total induced current $j = j_{\text{intra}} + j_{\text{inter}}$ consists of an interband component $j_{\text{inter}} = \partial_t \langle \hat{P} \rangle$ and the intraband contribution coming from the Peierls phase in the kinetic term of Eq. (1) [41,42,54]. The explicit dipolar coupling D is necessary for phase mode signatures to be visible in the optical spectrum, and both the bare contribution and contribution from the intralayer excitons would lead only to the renormalization of the dipolar strength [55,56]. We will evaluate the optical response in and out of equilibrium directly from the response current generated by a short electric field pulse [57–59] as $\sigma(t_{\text{pr}}, \omega) = \text{FT}[j](t_{\text{pr}}, \omega) / \text{FT}[E_{\text{pr}}](t_{\text{pr}}, \omega)$, where $j(t)$ is the current responding to the field $E_{\text{pr}}(t) = E_{\text{pr}}^0 e^{-(t-t_{\text{pr}})^2/(2\sigma_{\text{pr}}^2)}$. Here, we have defined the Fourier transform as $\text{FT}[X](t_{\text{pr}}, \omega) = \int_0^{t_{\text{cut}}} X(t_{\text{pr}} + s) e^{-(i\omega + \eta)s} ds$. The resulting data depend only weakly on the probe pulse width σ_{pr} , so we always choose $\sigma_{\text{pr}} = 0.2 t_h^{-1}$. Furthermore, we use $E_{\text{pr}}^0 = 10^{-2}$ and $t_{\text{cut}} = 80 t_h^{-1}$ as a typical upper time limit for the Fourier transform, and η is a broadening factor.

Equilibrium and voltage-biased states—The Hamiltonian (1) with $t_{\text{hyb}} = 0$ separately conserves the number of electrons in each band, implying invariance under changes in the U(1) phase conjugate to the difference in population between the two bands. At low T , the symmetry may be spontaneously broken via a transition into an excitonic insulator state with the order parameter $\Delta \equiv \langle v_i^\dagger c_i \rangle = |\Delta| e^{i\theta}$, where the energy is independent of θ giving rise to gapless fluctuations. A $t_{\text{hyb}} \neq 0$ explicitly breaks this internal U(1) symmetry, generically implying that $|\Delta| \neq 0$ and the energy depends on the value of θ resulting in a gapped phase mode [22,60–62]. In Fig. 1(a), we show the equilibrium value of $|\Delta|$ as a function of the band splitting $\Delta\epsilon$ for $t_{\text{hyb}} = 0$ and $t_{\text{hyb}} \neq 0$. In the former case, there is a critical value of band splitting $|\Delta\epsilon|$ beyond which $|\Delta| = 0$; if $t_{\text{hyb}} \neq 0$, $|\Delta| \neq 0$ for all $|\Delta\epsilon|$. Figure 1(b) shows the equilibrium linear response optical conductivity for $U = 5$ and $\Delta\epsilon = 0.6$. At $t_{\text{hyb}} = 0$, the main feature is an interband transition for $\omega \approx 3 t_h$. The gap increases as t_{hyb}

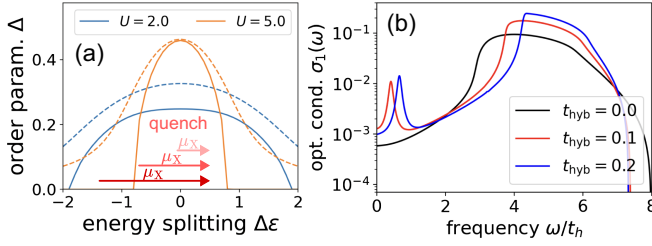


FIG. 1. (a) Equilibrium order parameter as a function of the orbital energy splitting $\Delta\epsilon$; solid lines have $t_{\text{hyb}} = 0$, dashed lines $t_{\text{hyb}} = 0.1$. The arrow sketches the quench protocol (darker colors represent larger values of μ_X). (b) Real part of the equilibrium optical conductivity $\sigma_1(\omega)$ for different values of the interlayer tunneling t_{hyb} and $U = 5$, $\Delta\epsilon = 0.6$. The low-energy peaks appearing for $t_{\text{hyb}} > 0$ correspond to a massive phase mode.

increases, and an in-gap feature arising from the gapped phase mode appears. Such a mode is a consequence of phase coherence. In this situation, the strength of the in-gap feature is set by the dipolar matrix element D .

Next, we turn to the bilayer system in the presence of either a bias voltage or photoexcited states with quasistationary carrier distribution [13,15,16,63,64]. In both cases, the state is described by an interorbital chemical potential difference μ_X . We follow the protocol where the equilibrium state is prepared with $\Delta\epsilon = \Delta\epsilon_0 - \mu_X$ and then suddenly changed to $\Delta\epsilon = \Delta\epsilon_0$ at time $t = 0$ as marked in Fig. 1(a). It was shown in Ref. [17] that such a protocol leads to efficient real-time preparation of states with biased chemical potentials. In the absence of interlayer tunneling, $t_{\text{hyb}} = 0$, the number of excited carriers $N_X = \sum_i (c_i^\dagger c_i + v_i v_i^\dagger)$ is a conserved quantity for all times. If $0 < t_{\text{hyb}} \ll t_h$, N_X oscillates coherently around a nonzero mean and is conserved only on average. We confirmed numerically that these oscillations are undamped for $U = 5$ such that they give rise to a long-lived prethermalized state. In particular, the correction to the Hartree-Fock self-energy from the second Born approximation is small in this regime due to the large gap size, which also justifies the time-dependent mean-field approximation used in previous studies [17,62,65]. Further reduction of the interband interaction U leads to a decay of the oscillations of N_X and to a breakdown of the biased state [42].

Order parameter dynamics—We begin our discussion of the postquench dynamics with the order parameter. Expectations about the behavior of its phase $\theta(t)$ may be formulated based on a simple time-dependent Landau-Ginzburg theory [42]. Note that in the microscopic simulations the full dynamics of both the amplitude and the phase of the order parameter are recorded, which serve as unbiased reference for the current effective description. Then, the effective Hamiltonian for $\theta(t)$ and for our quench setup is given by

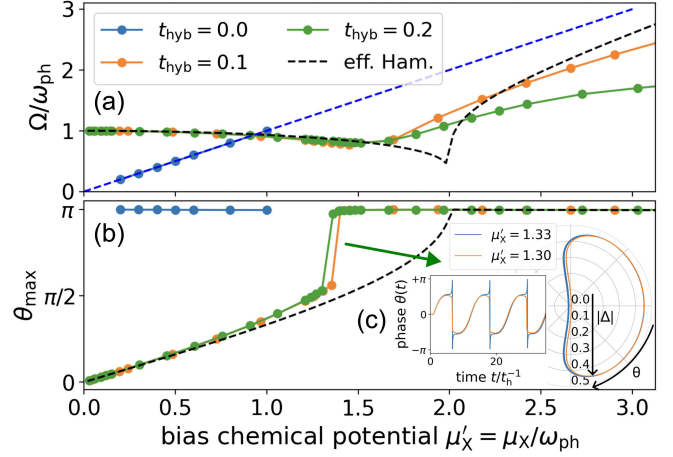


FIG. 2. (a) Phase mode frequency (confined regime) or velocity (deconfined regime) Ω_{ph} and (b) maximal amplitude $\theta_{\text{max}} = |\max_t \theta(t)|$ of the phase mode $\theta(t)$ versus the exciton chemical potential μ_X for $U = 5$, $\Delta\epsilon = 0.6$, and varying values of t_{hyb} . The dashed lines are computed from H_{eff} in (3). For data with $t_{\text{hyb}} > 0$, μ_X and Ω_{ph} are rescaled by the respective equilibrium phase mode frequency $\omega_{\text{ph}}(t_{\text{hyb}})$. (c) Real-time and phase space trajectories for two values of $\mu_X/\omega_{\text{ph}} = 1.30, 1.33$ ($t_{\text{hyb}} = 0.2$) slightly below and above the trapping transition, respectively.

$$H_{\text{eff}} = \frac{1}{2} \dot{\theta}^2 + \omega_{\text{ph}}^2 \cos(\theta), \quad \begin{cases} \theta(t=0) = 0, \\ \dot{\theta}(t=0) = \mu_X, \end{cases} \quad (3)$$

where ω_{ph} is the frequency of the phase mode oscillations about the equilibrium value; it has a somewhat complicated dependence on microscopic parameters [42]. The voltage bias μ_X enters via the initial condition $\dot{\theta}(t=0) = \mu_X$; i.e., it provides an initial kick to the phase mode. The response to the probe pulse is introduced by adding a dipolar excitation term $H_{\text{eff,dip}} = \gamma E_{\text{pr}}(t) \cos(\theta)$, where γ is proportional to the dipolar matrix element D . If $\omega_{\text{ph}} = 0$, θ performs a free rotation of the form $\theta(t) = \mu_X t$, which is a direct consequence of the U(1) phase translation symmetry present in this case (massless Goldstone mode). However, for $\omega_{\text{ph}} > 0$, the phase mode moves in a cosinusoidal potential and may remain trapped in the minimum around its initial value $\theta(0) = 0$ if its energy is too small. We may, therefore, anticipate a transition from—in general, nonharmonic—oscillations with $\theta_{\text{max}} := |\max_t \theta(t)| < \pi$ at small μ_X to a winding phase motion at large μ_X . In Fig. 2(a), we plot the frequency $\Omega_{\text{ph}} = (2\pi/T_{\text{ph}})$, based on the phase mode periodicity $\theta(t + T_{\text{ph}}) = \theta(t)$ or $\dot{\theta}(t + T_{\text{ph}}) = \dot{\theta}(t)$ and in Fig. 2(b) the maximum value θ_{max} of the phase oscillation ($\theta_{\text{max}} = \pi$ for the winding case). The dashed lines corresponding to the minimal model indeed show that, for $\mu_X < \mu_{X,c} = 2\omega_{\text{ph}}$, the phase mode is trapped. The frequency Ω_{ph} softens upon approaching $\mu_{X,c}$ from below because of the nonharmonic parts of the cosinusoidal

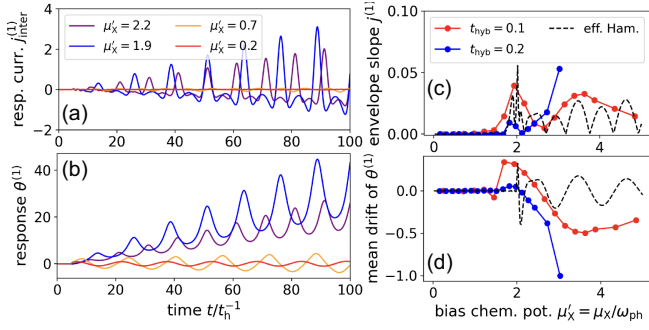


FIG. 3. (a) The interlayer current response $j_{\text{inter}}^{(1)}(t)$ and (b) the order parameter's response $\theta^{(1)}(t)$ of the dynamical condensate after the application of a weak Gaussian-shaped probe pulse. (c) Slope of a linear fit to the maxima of $j_{\text{inter}}^{(1)}(t)$ (envelope) and (d) slope of a linear fit to the phase response of the order parameter $\theta^{(1)}$ versus biased chemical potential μ_X . System parameters are $U = 5$, $\Delta\epsilon = 0.6$, and $t_{\text{hyb}} = 0.1$, and for the effective Hamiltonian model we assume $|\Delta| = 0.4$.

potential. For $\mu_X > 2\omega_{\text{ph}}$, $\theta(t)$ is winding and $\Omega_{\text{ph}} \sim \mu_X$ in the regime $\mu_X \gg 2\omega_{\text{ph}}$.

Turning to the tDMFT simulations, represented by the colored dots in Fig. 2, we confirm the phase rotation with frequency μ_X for $t_{\text{hyb}} = 0$. If $t_{\text{hyb}} > 0$, the predicted softening of Ω_{ph} at small values of μ_X is present as well. However, the untrapping transition occurs sharply at a chemical potential smaller than $2\omega_{\text{ph}}$. To shed more light on this behavior, we plotted $\theta(t)$ and the $|\Delta| - \theta$ phase space trajectory in Fig. 2(c) for values of μ_X slightly below and slightly above the transition. In the phase space picture, the shape of the trajectory is strongly squeezed along the $|\Delta|$ direction close to $|\Delta| = 0$. This explains the possibility of a sudden strong change of the range of explored angles. One can roughly reproduce the squeezed trajectories as equipotential lines within the equilibrium Landau-Ginzburg potential [42].

Optical response: Currents—We will now argue that the two dynamical regimes can also be distinguished in optical measurements. Using $j_{\text{inter}}(t) = \partial_t \langle P \rangle = 2D((\partial_t |\Delta|) \cos(\theta) - |\Delta| \dot{\theta} \sin(\theta))$, Fig. 3(a) shows the interlayer response current $j_{\text{inter}}^{(1)}(t) = (j_{\text{inter}}(t)|_{E_{\text{pr}}^0 > 0} - j_{\text{inter}}(t)|_{E_{\text{pr}}^0 = 0})/E_{\text{pr}}^0$, obtained from the tDMFT simulation for values of μ_X in both the trapped and the winding regime of the phase mode. In the former case, the response current remains small, while it grows strongly with a linearly growing envelope in the latter case. A more quantitative analysis of this observation yields Fig. 3(c), in which the slope of the envelope of $j^{(1)}(t)$ is plotted. In contrast to the jump for the phase of the order parameter θ in Fig. 2(b), the physically observable current response grows more smoothly but with a significant increase of the slope beyond the critical $\mu_{X,c}$; see Fig. 3. The growth of the interlayer response current is connected to the phase

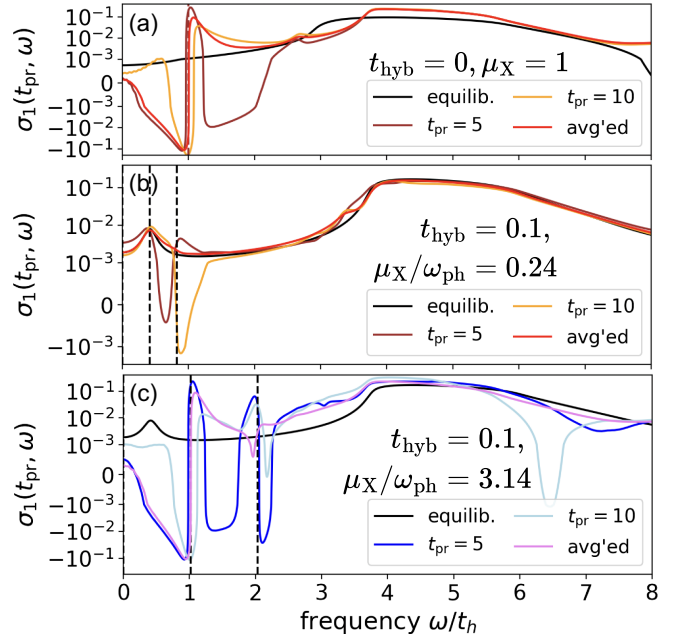


FIG. 4. Nonequilibrium optical conductivity $\sigma_1(t_{\text{pr}}, \omega)$ for (a) decoupled layers $t_{\text{hyb}} = 0$ after a quench of size $\mu_X = 1$ and for coupled layers with $t_{\text{hyb}} = 0.1$ in the (b) phase-trapped, $\mu_X' = 0.24$ ($\mu_X = 0.1$), and (c) phase-delocalized, $\mu_X' = 3.14$ ($\mu_X = 1.3$), regimes at different delay times t_{pr} . Dashed lines show the main oscillation frequency of $\theta(t)$, which agrees with the equilibrium phase mode peak position only in the confined case, and its second harmonics. The used broadening for the Fourier transform is $\eta = 10/t_{\text{cut}}$.

response of the order parameter, $\theta^{(1)}(t)$, shown in Fig. 3(b). In the winding phase mode regime, we find $\theta^{(1)}(t)$ oscillating around a drifting mean, while it oscillates around zero in the trapped regime. Figure 3(d) displays the drift velocity as a function of μ_X and shows qualitative agreement with the effective model. Hence, the probe-induced current's behavior goes hand in hand with the strong growth of the order parameter's phase such that these quantities can detect the presence of the dynamical transition.

Optical response: Conductivities—Finally, we discuss the Fourier-transformed total response current and the time-dependent optical conductivities $\sigma(t_{\text{pr}}, \omega)$. We begin with the case of decoupled layers $t_{\text{hyb}} = 0$, shown in Fig. 4(a) exemplarily for a value of $\mu_X = 1$. One finds that the dynamical phase $\theta(t) = \mu_X t$ indeed shows up in the (non-equilibrium) optical conductivity $\sigma_1(t_{\text{pr}}, \omega)$ as an additional peak at $\omega = \mu_X$. In particular, the position of the peak is independent of the precise probe time delay t_{pr} , while the optical weight depends on it (within a period of $(2\pi/\mu_X)$) and can become negative as observed in other contexts [61,66,67]. This can be schematically understood from the dynamics of the response current in Fig. 3(a), since the Fourier transform of a current $j(t) \sim t \sin(\nu t)$ has components $\pm \partial_\omega \delta(\omega \mp \nu)$. Negative optical conductivities indicate the system's tendency for stimulated emission of photons at a given

frequency. We also show the period-averaged optical conductivity $\bar{\sigma}(\omega) = (1/T_{\text{ph}}) \int_{t_0}^{t_0+T_{\text{ph}}} \sigma(t_{\text{pr}}, \omega) dt_{\text{pr}}$ as a measure of average absorption and emission in a typical stroboscopic experimental setup.

Optical conductivities at finite interlayer tunneling $t_{\text{hyb}} = 0.1$ are shown in Fig. 4(b) for the trapped-phase and Fig. 4(c) for the phase-delocalized regime, respectively. In the trapped state with $\mu_X/\omega_{\text{ph}} = 0.24$ ($\mu_X = 0.1$), we find a peak appearing at the equilibrium phase mode frequency ω_{ph} which is different from μ_X . In contrast, in the regime with complete phase winding, $\mu_X/\omega_{\text{ph}} = 3.14$ ($\mu_X = 1.3$), $\sigma_1(t_{\text{pr}}, \omega)$ does not peak at the phase mode frequency but instead at a larger frequency corresponding to the main oscillating frequency of the order parameter's phase. Again, these signals can have positive or negative weight, depending on the probe time. The period-averaged optical conductivity for the trapped phase in Fig. 4(b) remains positive at all frequencies. In both regimes, second harmonics at $2\Omega_{\text{ph}}$ appear—see dashed lines in Figs. 4(b) and 4(c)—in agreement with previous studies [61]. Not only are in-gap states in the optical response, therefore, a signature of the *dynamical excitonic condensation*, but their scaling with the strength of the excitation μ_X allows one, in addition, to distinguish between different dynamical regimes of excitonic order.

Conclusions—We studied the optical properties of a dynamical exciton condensate as realized either in bilayer structures with an applied voltage bias or photodoped semiconductors. The dynamical condensate may be in one of two states, characterized by the long-time behavior of the phase of the condensate amplitude. In the weakly nonequilibrium state, the phase is confined, oscillating about the value that minimizes the free energy. In the strongly nonequilibrium state, the phase is deconfined, linearly increasing with time. We used nonequilibrium dynamical mean-field theory to understand the two states, the transition between them, and the observable signatures in the linear response optical conductivity. At small voltage bias μ_X , the optical conductivity shows a sharp peak at the energy of the equilibrium phase mode excitation. At large voltage bias μ_X , the low-energy optical peak decouples from the phase mode frequency, and a wide regime of stimulated optical emission appears. The evolution of the in-gap peak with increasing driving strength, thus, provides experimental access to detect the state of dynamical condensate.

An important direction for future research concerns the nature of spontaneous emission in driven condensates, which would require one to solve the coupled evolution of the electromagnetic environment and the correlated solid. A further extension is to modify the electromagnetic environment by placing the material in a cavity in order to understand if the superradiant phase proposed in effective theories [22,38] is consistent with the microscopic

evolution presented in this Letter. Finally, nonlinear responses—such as the order of the Josephson effect [37] and nonlinear optical or Raman responses [48,68]—are highly sensitive to microscopic details. It is, therefore, important to extend the current microscopic framework to more realistic systems, such as two-dimensional van der Waals heterostructures driven by external voltages (e.g., MoSe₂/WSe₂ bilayers [27,28]) or layered transition metal chalcogenides driven by optical pulses (e.g., 1T-TiSe₂ [69,70] or Ta₂NiSe₅ [71,72]). This would help elucidate how material-dependent properties [47,48] modify the general picture presented in this Letter and motivate future experiments to test the proposed mechanism.

The NESSi package [73] was used for the calculations in this Letter.

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- [1] D. Jérôme, T. M. Rice, and W. Kohn, Excitonic insulator, *Phys. Rev.* **158**, 462 (1967).
 - [2] L. V. Keldysh and A. N. Kozlov, Collective properties of excitons in semiconductors, *Zh. Eksp. Teor. Fiz.* **54**, 978 (1968) [*Sov. J. Exp. Theor. Phys.* **27**, 521 (1968)].
 - [3] A. V. Balatsky, Y. N. Joglekar, and P. B. Littlewood, Dipolar superfluidity in electron-hole bilayer systems, *Phys. Rev. Lett.* **93**, 266801 (2004).
 - [4] J. Eisenstein and A. H. MacDonald, Bose–Einstein condensation of excitons in bilayer electron systems, *Nature (London)* **432**, 691 (2004).
 - [5] J. Eisenstein, Exciton condensation in bilayer quantum hall systems, *Annu. Rev. Condens. Matter Phys.* **5**, 159 (2014).
 - [6] P. Littlewood, P. Eastham, J. M. J. Keeling, F. Marchetti, B. Simons, and M. Szymanska, Models of coherent exciton condensation, *J. Phys. Condens. Matter* **16**, S3597 (2004).

- [7] Y. Wakisaka, T. Sudayama, K. Takubo, T. Mizokawa, M. Arita, H. Namatame, M. Taniguchi, N. Katayama, M. Nohara, and H. Takagi, Excitonic insulator state in Ta_2NiSe_5 probed by photoemission spectroscopy, *Phys. Rev. Lett.* **103**, 026402 (2009).
- [8] K. Seki, Y. Wakisaka, T. Kaneko, T. Toriyama, T. Konishi, T. Sudayama, N. L. Saini, M. Arita, H. Namatame, M. Taniguchi, N. Katayama, M. Nohara, H. Takagi, T. Mizokawa, and Y. Ohta, Excitonic Bose-Einstein condensation in Ta_2NiSe_5 above room temperature, *Phys. Rev. B* **90**, 155116 (2014).
- [9] C. Monney, H. Cercellier, F. Clerc, C. Battaglia, E. F. Schwier, C. Didiot, M. G. Garnier, H. Beck, P. Aebi, H. Berger, L. Forró, and L. Patthey, Spontaneous exciton condensation in 1T-TiSe_2 : BCS-like approach, *Phys. Rev. B* **79**, 045116 (2009).
- [10] A. K. Geim and I. V. Grigorieva, Van der Waals heterostructures, *Nature (London)* **499**, 419 (2013).
- [11] K. S. Novoselov, A. Mishchenko, A. Carvalho, and A. H. C. Neto, 2D materials and van der Waals heterostructures, *Science* **353**, aac9439 (2016).
- [12] C. Jin, E. Y. Ma, O. Karni, E. C. Regan, F. Wang, and T. F. Heinz, Ultrafast dynamics in van der Waals heterostructures, *Nat. Nanotechnol.* **13**, 994 (2018).
- [13] V. Pareek, D. R. Bacon, X. Zhu, Y.-H. Chan, F. Bussolotti, N. S. Chan, J. P. Urquiza, K. Watanabe, T. Taniguchi, M. K. L. Man, J. Madéo, D. Y. Qiu, K. E. J. Goh, F. H. da Jornada, and K. M. Dani, Driving non-trivial quantum phases in conventional semiconductors with intense excitonic fields, *arXiv:2403.08725*.
- [14] F. Boschini, M. Zonno, and A. Damascelli, Time-resolved ARPES studies of quantum materials, *Rev. Mod. Phys.* **96**, 015003 (2024).
- [15] M. Reutz, G. S. M. Jansen, and S. Mathias, Probing excitons with time-resolved momentum microscopy, *Adv. Phys. X* **9**, 2378722 (2024).
- [16] Y. Murotani, C. Kim, H. Akiyama, L. N. Pfeiffer, K. W. West, and R. Shimano, Light-driven electron-hole Bardeen-Cooper-Schrieffer-like state in bulk GaAs, *Phys. Rev. Lett.* **123**, 197401 (2019).
- [17] E. Peretto, D. Sangalli, A. Marini, and G. Stefanucci, Pump-driven normal-to-excitonic insulator transition: Josephson oscillations and signatures of BEC-BCS crossover in time-resolved ARPES, *Phys. Rev. Mater.* **3**, 124601 (2019).
- [18] E. Peretto, S. Bianchi, and G. Stefanucci, Time-resolved ARPES spectra of nonequilibrium excitonic insulators: Revealing macroscopic coherence with ultrashort pulses, *Phys. Rev. B* **101**, 041201(R) (2020).
- [19] D. Schmitt, J. P. Bange, W. Bennecke, A. AlMutairi, G. Meneghini, K. Watanabe, T. Taniguchi, D. Steil, D. R. Luke, R. T. Weitz *et al.*, Formation of moiré interlayer excitons in space and time, *Nature (London)* **608**, 499 (2022).
- [20] J. P. Bange, D. Schmitt, W. Bennecke, G. Meneghini, A. AlMutairi, K. Watanabe, T. Taniguchi, D. Steil, S. Steil, R. T. Weitz, G. S. M. Jansen, S. Hofmann, S. Brem, E. Malic, M. Reutz, and S. Mathias, Probing electron-hole Coulomb correlations in the exciton landscape of a twisted semiconductor heterostructure, *Sci. Adv.* **10**, eadi1323 (2024).
- [21] X. Zhu, P. B. Littlewood, M. S. Hybertsen, and T. M. Rice, Exciton condensate in semiconductor quantum well structures, *Phys. Rev. Lett.* **74**, 1633 (1995).
- [22] P. Littlewood and X. Zhu, Possibilities for exciton condensation in semiconductor quantum-well structures, *Phys. Scr.* **1996**, 56 (1996).
- [23] M. H. Szymanska and P. B. Littlewood, Excitonic binding in coupled quantum wells, *Phys. Rev. B* **67**, 193305 (2003).
- [24] M. Xie and A. H. MacDonald, Electrical reservoirs for bilayer excitons, *Phys. Rev. Lett.* **121**, 067702 (2018).
- [25] Z. Wang, D. A. Rhodes, K. Watanabe, T. Taniguchi, J. C. Hone, J. Shan, and K. F. Mak, Evidence of high-temperature exciton condensation in two-dimensional atomic double layers, *Nature (London)* **574**, 76 (2019).
- [26] L. Ma, P. X. Nguyen, Z. Wang, Y. Zeng, K. Watanabe, T. Taniguchi, A. H. MacDonald, K. F. Mak, and J. Shan, Strongly correlated excitonic insulator in atomic double layers, *Nature (London)* **598**, 585 (2021).
- [27] P. X. Nguyen, L. Ma, R. Chaturvedi, K. Watanabe, T. Taniguchi, J. Shan, and K. F. Mak, Perfect Coulomb drag in a dipolar excitonic insulator, *Science* **388**, 274 (2025).
- [28] R. Qi, A. Y. Joe, Z. Zhang, J. Xie, Q. Feng, Z. Lu, Z. Wang, T. Taniguchi, K. Watanabe, S. Tongay, and F. Wang, Perfect Coulomb drag and exciton transport in an excitonic insulator, *Science* **388**, 278 (2025).
- [29] X. Liu, N. Leisgang, P. E. Dolgirev, A. A. Zibrov, J. Sung, J. Wang, T. Taniguchi, K. Watanabe, V. Walther, H. Park, E. Demler, P. Kim, and M. D. Lukin, Optical signatures of interlayer electron coherence in a bilayer semiconductor, *Nat. Phys.* (2024), [10.1038/s41567-025-02971-0](https://doi.org/10.1038/s41567-025-02971-0).
- [30] D. Nandi, A. Finck, J. Eisenstein, L. Pfeiffer, and K. West, Exciton condensation and perfect coulomb drag, *Nature (London)* **488**, 481 (2012).
- [31] A. L. Ivanov, P. B. Littlewood, and H. Haug, Bose-Einstein statistics in thermalization and photoluminescence of quantum-well excitons, *Phys. Rev. B* **59**, 5032 (1999).
- [32] L. V. Butov, A. C. Gossard, and D. S. Chemla, Macroscopically ordered state in an exciton system, *Nature (London)* **418**, 751 (2002).
- [33] Y. Zeng and A. H. MacDonald, Electrically controlled two-dimensional electron-hole fluids, *Phys. Rev. B* **102**, 085154 (2020).
- [34] Y. Murakami, M. Schüler, S. Takayoshi, and P. Werner, Ultrafast nonequilibrium evolution of excitonic modes in semiconductors, *Phys. Rev. B* **101**, 035203 (2020).
- [35] R. Hanai, P. B. Littlewood, and Y. Ohashi, Non-equilibrium properties of a pumped-decaying Bose-condensed electron-hole gas in the BCS-BEC crossover region, *J. Low Temp. Phys.* **183**, 127 (2016).
- [36] R. Hanai, P. B. Littlewood, and Y. Ohashi, Dynamical instability of a driven-dissipative electron-hole condensate in the BCS-BEC crossover region, *Phys. Rev. B* **96**, 125206 (2017).
- [37] Z. Sun, T. Kaneko, D. Golež, and A. J. Millis, Second-order Josephson effect in excitonic insulators, *Phys. Rev. Lett.* **127**, 127702 (2021).
- [38] Z. Sun, Y. Murakami, F. Xuan, T. Kaneko, D. Golež, and A. J. Millis, Dynamical exciton condensates in biased electron-hole bilayers, *Phys. Rev. Lett.* **133**, 217002 (2024).

- [39] Y. Zeng, V. Crépel, and A. J. Millis, Keldysh field theory of dynamical exciton condensation transitions in nonequilibrium electron-hole bilayers, *Phys. Rev. Lett.* **132**, 266001 (2024).
- [40] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions, *Rev. Mod. Phys.* **68**, 13 (1996).
- [41] H. Aoki, N. Tsuji, M. Eckstein, M. Kollar, T. Oka, and P. Werner, Nonequilibrium dynamical mean-field theory and its applications, *Rev. Mod. Phys.* **86**, 779 (2014).
- [42] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/58r8-cpzn> for additional info on the numerics, further supporting figures and details on the Ginzburg-Landau model, which includes Refs. [43–46].
- [43] P. Werner, H. U. R. Strand, S. Hoshino, and M. Eckstein, Ultrafast switching of composite order in A_3C_{60} , *Phys. Rev. B* **95**, 195405 (2017).
- [44] S. Giuli, A. Amaricci, and M. Capone, Mott-enhanced exciton condensation in a Hubbard bilayer, *Phys. Rev. B* **108**, 165150 (2023).
- [45] A. M. J. Schakel, Time-dependent Ginzburg-Landau theory and duality, in *Topological Defects and the Non-Equilibrium Dynamics of Symmetry Breaking Phase Transitions*, edited by Y. M. Bunkov and H. Godfrin (Springer, Dordrecht, 2000), pp. 213–238.
- [46] Z. Sun and A. J. Millis, Bardasis-Schrieffer polaritons in excitonic insulators, *Phys. Rev. B* **102**, 041110(R) (2020).
- [47] G. Mazza, M. Rösner, L. Windgätter, S. Latini, H. Hübener, A. J. Millis, A. Rubio, and A. Georges, Nature of symmetry breaking at the excitonic insulator transition: Ta_2NiSe_5 , *Phys. Rev. Lett.* **124**, 197601 (2020).
- [48] B. Chatterjee, J. Mravlje, and D. Golež, Collective modes and Raman response in Ta_2NiSe_5 , *Phys. Rev. B* **111**, L121106 (2025).
- [49] T. B. Boykin, R. C. Bowen, and G. Klimeck, Electromagnetic coupling and gauge invariance in the empirical tight-binding method, *Phys. Rev. B* **63**, 245314 (2001).
- [50] D. Golež, M. Eckstein, and P. Werner, Multiband nonequilibrium GW + EDMFT formalism for correlated insulators, *Phys. Rev. B* **100**, 235117 (2019).
- [51] J. Li, D. Golež, G. Mazza, A. J. Millis, A. Georges, and M. Eckstein, Electromagnetic coupling in tight-binding models for strongly correlated light and matter, *Phys. Rev. B* **101**, 205140 (2020).
- [52] M. Schüler, J. A. Marks, Y. Murakami, C. Jia, and T. P. Devereaux, Gauge invariance of light-matter interactions in first-principle tight-binding models, *Phys. Rev. B* **103**, 155409 (2021).
- [53] O. Dmytruk and M. Schiró, Gauge fixing for strongly correlated electrons coupled to quantum light, *Phys. Rev. B* **103**, 075131 (2021).
- [54] P. Werner, J. Li, D. Golež, and M. Eckstein, Entropy-cooled nonequilibrium states of the Hubbard model, *Phys. Rev. B* **100**, 155130 (2019).
- [55] H. Yu, Y. Wang, Q. Tong, X. Xu, and W. Yao, Anomalous light cones and valley optical selection rules of interlayer excitons in twisted heterobilayers, *Phys. Rev. Lett.* **115**, 187002 (2015).
- [56] D. A. Ruiz-Tijerina and V. I. Fal'ko, Interlayer hybridization and moiré superlattice minibands for electrons and excitons in heterobilayers of transition-metal dichalcogenides, *Phys. Rev. B* **99**, 125424 (2019).
- [57] C. Shao, T. Tohyama, H.-G. Luo, and H. Lu, Numerical method to compute optical conductivity based on pump-probe simulations, *Phys. Rev. B* **93**, 195144 (2016).
- [58] M. Eckstein and M. Kollar, Theory of time-resolved optical spectroscopy on correlated electron systems, *Phys. Rev. B* **78**, 205119 (2008).
- [59] Z. Lenarčič, D. Golež, J. Bonča, and P. Prelovšek, Optical response of highly excited particles in a strongly correlated system, *Phys. Rev. B* **89**, 125123 (2014).
- [60] R. Guseinov and L. Keldysh, Nature of the phase transition under the conditions of an “excitonic” instability in the electronic spectrum of a crystal, *Zh. Eksp. Teor. Fiz.* **63**, 2255 (1972) [*J. Exp. Theor. Phys.* **36**, 1193 (1973)].
- [61] D. Golež, Z. Sun, Y. Murakami, A. Georges, and A. J. Millis, Nonlinear spectroscopy of collective modes in an excitonic insulator, *Phys. Rev. Lett.* **125**, 257601 (2020).
- [62] Y. Murakami, D. Golež, T. Kaneko, A. Koga, A. J. Millis, and P. Werner, Collective modes in excitonic insulators: Effects of electron-phonon coupling and signatures in the optical response, *Phys. Rev. B* **101**, 195118 (2020).
- [63] L. Keldysh, The electron-hole liquid in semiconductors, *Contemp. Phys.* **27**, 395 (1986).
- [64] H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 2009).
- [65] Y. Murakami, D. Golež, M. Eckstein, and P. Werner, Photoinduced enhancement of excitonic order, *Phys. Rev. Lett.* **119**, 247601 (2017).
- [66] G. Chiriacò, A. J. Millis, and I. L. Aleiner, Negative absolute conductivity in photoexcited metals, *Phys. Rev. B* **101**, 041105(R) (2020).
- [67] D. Golež, L. Boehnke, H. U. R. Strand, M. Eckstein, and P. Werner, Nonequilibrium GW + EDMFT: Antiscreening and inverted populations from nonlocal correlations, *Phys. Rev. Lett.* **118**, 246402 (2017).
- [68] T. Kaneko, Z. Sun, Y. Murakami, D. Golež, and A. J. Millis, Bulk photovoltaic effect driven by collective excitations in a correlated insulator, *Phys. Rev. Lett.* **127**, 127402 (2021).
- [69] S. Mathias, S. Eich, J. Urbancic, S. Michael, A. Carr, S. Emmerich, A. Stange, T. Popmintchev, T. Rohwer, M. Wiesenmayer *et al.*, Self-amplified photo-induced gap quenching in a correlated electron material, *Nat. Commun.* **7**, 12902 (2016).
- [70] C. Monney, M. Puppini, C. W. Nicholson, M. Hoesch, R. T. Chapman, E. Springate, H. Berger, A. Magrez, C. Cacho, R. Ernstorfer, and M. Wolf, Revealing the role of electrons and phonons in the ultrafast recovery of charge density wave correlations in $1T - TiSe_2$, *Phys. Rev. B* **94**, 165165 (2016).
- [71] K. Katsumi, A. Alekhin, S.-M. Souliou, M. Merz, A.-A. Haghighirad, M. Le Tacon, S. Houver, M. Cazayous, A. Sacuto, and Y. Gallais, Disentangling lattice and electronic instabilities in the excitonic insulator candidate Ta_2NiSe_5 by nonequilibrium spectroscopy, *Phys. Rev. Lett.* **130**, 106904 (2023).

- [72] S. Mor, M. Herzog, D. Golež, P. Werner, M. Eckstein, N. Katayama, M. Nohara, H. Takagi, T. Mizokawa, C. Monney, and J. Stähler, Ultrafast electronic band gap control in an excitonic insulator, *Phys. Rev. Lett.* **119**, 086401 (2017).
- [73] M. Schüler, D. Golež, Y. Murakami, N. Bittner, A. Herrmann, H. U. Strand, P. Werner, and M. Eckstein, NESSi: The non-equilibrium systems simulation package, *Comput. Phys. Commun.* **257**, 107484 (2020).