

Keldysh Field Theory of Dynamical Exciton Condensation Transitions in Nonequilibrium Electron-Hole Bilayers

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Recent experiments have realized steady-state electrical injection of interlayer excitons in electron-hole bilayers subject to a large bias voltage. In the ideal case in which interlayer tunneling is negligibly weak, the system is in quasiequilibrium with a reduced effective band gap. Interlayer tunneling introduces a current and drives the system out of equilibrium. In this work we derive a nonequilibrium field theory description of interlayer excitons in biased electron-hole bilayers. In the large bias limit, we find that p -wave interlayer tunneling reduces the effective band gap and increases the effective temperature for intervalley excitons. We discuss possible experimental implications for InAs/GaSb quantum wells and transition metal dichalcogenide bilayers.

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Introduction.—Excitons are bosonic bound states of conduction band electrons and valence band holes in semiconductors. The possibility of Bose-Einstein condensation of excitons was first proposed [1,2] over sixty years ago. It was later realized [3] that condensation of interlayer excitons in bilayer two-dimensional systems has striking experimental consequences including counterflow superfluidity and Josephson-like tunneling peaks [4,5]. Equilibrium interlayer exciton condensation has been experimentally established in quantum Hall bilayers [6–10]. Equilibrium exciton condensation in the absence of a magnetic field has been theoretically studied in a number of contexts [11–19], but has so far remained elusive experimentally in conventional semiconductor systems despite much effort [20–24].

Group-VI transition metal dichalcogenides (TMDs) with chemical formula MX_2 (where $M = \text{Mo, W}$ and $X = \text{S, Se, Te}$) are a class of two-dimensional semiconductors that host strongly bound excitons [25–29] and can be stacked in various combinations. When two TMD layers are stacked, electrons from one layer and holes from the other layer form interlayer excitons that are strongly bound even with thin insulating barriers separating the electron and hole layers. Interlayer excitons in TMD bilayers have long lifetimes and electrically tunable properties [30–33]. If separate contacts are made on the electron and hole layers [24,34–37], the chemical potentials of carriers in the two layers are controlled separately, and their difference, the bias voltage, controls the exciton chemical potential [38,39]. When the exciton chemical potential exceeds the lowest bound state energy of electron-hole pairs, interlayer excitons are electrically injected into the bilayer system and undergo Bose-Einstein condensation (BEC) at

low enough temperatures. Excitonic insulating states in TMD bilayers have been established in recent experiments by compressibility measurements [34,35] and drag measurements [36,37].

If tunneling between layers is negligible, the potential difference required to maintain a nonzero steady state exciton density can be gauged away, so the system is equivalent to an equilibrium electron-hole bilayer with a reduced effective band gap. Nonzero interlayer tunneling introduces a tunneling current [24,38] that drives the system out of equilibrium, leading to new physics [40] different from that of driven-dissipative condensates [41–45]. In this Letter we present a microscopic theory of the nonequilibrium exciton condensate based on a Keldysh nonequilibrium field theory [42,46–48] that includes the effects of both a bias voltage and interlayer tunneling.

In TMD bilayers, because the conduction and valence band extrema are located at the $\pm K$ valleys, the functional form of interlayer tunneling depends on the local stacking registry [49–52]. In this Letter we focus on the experimentally relevant case of angle-aligned TMD homobilayers in which interlayer tunneling is uniform in space, and assume in our explicit calculations p -wave interlayer tunneling that applies to most of the high-symmetry stacking registries of TMDs as well as InAs/GaSb quantum wells [53–55]. Different from s -wave tunneling, p -wave interlayer tunneling produces a potential landscape that is second order in the phase angle of the exciton field, leading when no bias voltage is applied to a second-order Josephson effect [56] that breaks the interlayer phase symmetry down to \mathbb{Z}_2 from $U(1)$. We find that in the large bias limit, the system is described by an effective action in which interlayer $U(1)$ phase symmetry is

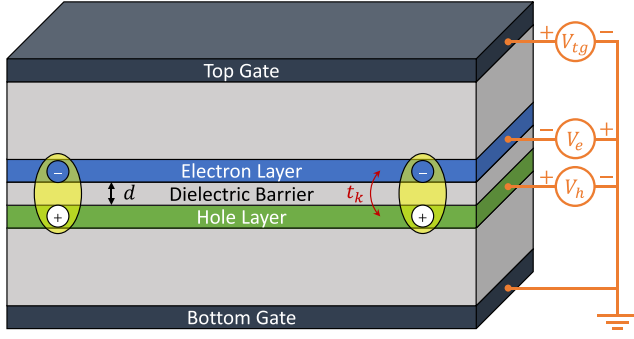


FIG. 1. An electrically controlled electron-hole bilayer. A negative voltage $-V_e$ and a positive voltage $+V_h$ are applied on the electron and hole layers, respectively, to inject electrons and holes into the system. The chemical potential of interlayer excitons $\mu_x = \mu_c - \mu_v = e(V_e + V_h)$ is set by the bias voltage between layers. The bottom gate is grounded, and the top gate voltage V_{tg} produces a perpendicular electric field that tunes the band gap. The gray regions represent dielectric layers.

effectively restored, but p -wave interlayer tunneling leads to a reduced effective band gap and an increased effective temperature for intervalley excitons.

Model.—We consider an electron layer and a hole layer separated by a weakly conducting barrier as shown in Fig. 1. Experimentally the system is controlled in two ways: by tuning the top or bottom gate potential difference (fundamentally an equilibrium effect) and by connecting the electron and hole layers to reservoirs held at different chemical potentials, enabling injection, and removal of carriers. The system is described by the Hamiltonian $H = H_0 + H_t + H_C$, where

$$H_0 = \sum_{\tau b k} \xi_{b k} a_{\tau b k}^\dagger a_{\tau b k} \quad (1)$$

describes the kinetic energy of conduction band electrons and valence band holes. Here $\tau = \pm$ is the valley index and $b = c, v$ is the band (layer) index. $\xi_{c k} = k^2/2m_e^* + E_g/2$ and $\xi_{v k} = -k^2/2m_h^* - E_g/2$ describe the dispersion of the conduction and valence bands, where m_e^* and m_h^* are the effective masses of electrons and holes and E_g is the band gap that can be tuned by a perpendicular electric field produced by the difference between top and bottom gate voltages. The next term in the Hamiltonian

$$H_t = \sum_{\tau k} t_{\tau k} a_{\tau c k}^\dagger a_{\tau v k} + \text{H.c.} \quad (2)$$

describes interlayer tunneling arising from hybridization of electron and hole wave functions in the two layers. A nonzero $t_{\tau k}$ explicitly breaks the U(1) symmetry of the model associated with charge conservation in each layer. The momentum and valley dependence of $t_{\tau k}$ depends on symmetries of the system and is crucial for our upcoming

results. For most of the high-symmetry stacking registries of angle-aligned TMD homobilayers, direct tunneling is forbidden by rotational symmetry [50,51], leading to p -wave interlayer tunneling

$$t_{\tau k} = v_t(\tau k_x + i k_y). \quad (3)$$

This form of interlayer tunneling also applies to InAs/GaSb quantum wells [53–55], in which case τ is the spin index. In this Letter we focus on p -wave interlayer tunneling and briefly discuss other forms of interlayer tunneling at the end. The Coulomb interaction term

$$H_C = \frac{1}{2A} \sum_{bb'\tau\tau'} \sum_{kk'q} V_{bb'}(q) a_{\tau b, k+q}^\dagger a_{\tau' b', k'-q}^\dagger a_{\tau' b' k'} a_{\tau b k}, \quad (4)$$

where A is the system area, distinguishes intralayer ($b = b'$) and interlayer ($b \neq b'$) interactions but neglects intervalley scattering due to the large momentum transfer required. Electron-hole exchange interactions [57–61] are also neglected due to the suppression of current matrix elements between electrons and holes in different layers.

By tuning the electrochemical potential of the electron layer $\mu_c = eV_e$ near the bottom of the conduction band and the hole layer $\mu_v = -eV_h$ near the top of the valence band, electrons and holes are injected into the system and form interlayer excitons. The chemical potential of interlayer excitons $\mu_x = \mu_c - \mu_v = e(V_e + V_h)$ is set by the bias voltage between two layers.

Keldysh action.—We derive a nonequilibrium field theory that describes a biased electron-hole bilayer with interlayer tunneling based on the Keldysh formalism [42,46–48], outlining here the procedure to obtain the Keldysh action and presenting the main results, with detailed derivations left for the Supplemental Material [62]. We express the model as a path integral along a closed time path C that starts from the distant past, proceeds to the distant future, and then returns to the starting point. The generating function is

$$Z = \text{Tr} \left\{ \rho_0 \mathcal{T}_C \exp \left[-i \int_C dt H(t) \right] \right\} / \text{Tr}(\rho_0), \quad (5)$$

where \mathcal{T}_C is the contour ordering operator along C , and ρ_0 is the density matrix of the system in the distant past which we take as the equilibrium distribution of decoupled electron and hole layers: $\rho_0 = e^{-(H_0 - \mu_c N_c - \mu_v N_v)/T}$, where $N_b = \sum_{\tau k} a_{\tau b k}^\dagger a_{\tau b k}$ is the number of electrons in each layer. For notational convenience Eq. (5) is written for a closed system; coupling to leads is included in the theory as the imaginary (dissipative) part of inverse Green's functions as detailed in the Supplemental Material [62]. To derive a theory of excitons, we perform a Hubbard-Stratonovich transformation of interlayer electron-hole interactions and introduce the electron-hole pairing fields $\Delta_{kq}^{\tau\tau'}$, where k and

q are, respectively, the relative momentum and center-of-mass momentum of an electron-hole pair, and τ, τ' are the valley indices of electrons and holes. A nonzero value of Δ reflects spontaneous breaking of interlayer U(1) symmetry associated with formation of the exciton condensate. The k dependence of the pairing fields is irrelevant to the low-energy physics we discuss and is eliminated by projecting the Δ fields onto the $1s$ -exciton basis by defining

$$\Delta_{kq}^{\tau\tau'} = \frac{1}{A} \sum_{k'} V_{cv}(k-k') \varphi_{k'} \Phi_q^{\tau\tau'}, \quad (6)$$

where φ_k is the $1s$ -exciton wave function that is the lowest-energy solution of the eigenvalue equation

$$\frac{k^2}{2m} \varphi_k - \frac{1}{A} \sum_{k'} V_{cv}(k-k') \varphi_{k'} = -E_b \varphi_k. \quad (7)$$

Here, $m = m_e^* m_h^* / (m_e^* + m_h^*)$ is the reduced mass of an electron-hole pair and the exciton binding energy E_b is defined as the absolute value of the $1s$ -exciton energy. The $1s$ -exciton fields Φ have two valley indices, one for electrons and the other for holes, and we express them

in terms of a four-component spinor (Φ^μ) defined as $\Phi^{\tau\tau'} = (\sum_\mu \Phi^\mu \tau_\mu / \sqrt{2})^{\tau\tau'}$, where τ_0 and $\tau_{1,2,3}$ are the 2×2 identity and Pauli matrices in valley space [15]. In this notation Φ^0, Φ^3 are intravalley exciton fields and Φ^1, Φ^2 are intervalley exciton fields. Integrating out the fermion fields, we obtain an effective action in terms of the $1s$ -exciton fields Φ . Following the convention widely used in the literature on Keldysh field theory [42,46–48], we transform the forward (+) and backward (−) branches of the Φ fields into the *classical* (c) and *quantum* (q) fields defined as

$$\begin{pmatrix} \Phi^c \\ \Phi^q \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} \Phi^+ + \Phi^- \\ \Phi^+ - \Phi^- \end{pmatrix}. \quad (8)$$

The generating function is now expressed as the functional integral

$$Z = \int D[\Phi^q, \Phi^c] e^{iS[\Phi^q, \Phi^c]}. \quad (9)$$

Expanding the action in powers of Φ and in powers of interlayer tunneling we find the leading term

$$S_0[\bar{\Phi}, \Phi] = \frac{1}{A} \sum_q \int \frac{d\omega}{2\pi} \text{Tr} \left[\left(\omega - \frac{q^2}{2M} - E_g + E_b + i\gamma \right) \Phi_q^{q\dagger}(\omega) \Phi_q^c(\omega) + \text{c.c.} + ig(\omega - \mu_x) \coth \frac{\omega - \mu_x}{2T} \Phi_q^{q\dagger}(\omega) \Phi_q^q(\omega) \right], \quad (10)$$

which describes free excitons with energy $E_g - E_b + q^2/2M$ (where the exciton mass $M = m_e^* + m_h^*$) and chemical potential μ_x at temperature T . While the quadratic coefficients take the stated form only in the dilute exciton regime (BEC regime) $|E_g - E_b - \mu_x| \ll E_b$ and in the frequency range $|\omega - \mu_x| \ll E_b$, the overall form of Eq. (10) is general and we expect that our qualitative results apply to a larger parameter regime. The imaginary coefficients γ and g describe coupling of excitons to leads. Fluctuation-dissipation theorem implies $\gamma = g(\omega - \mu_x)$. In

the absence of interlayer tunneling, the bias voltage μ_x can be absorbed into ω and the system is equivalent to an unbiased bilayer with a reduced band gap $E_g - E_b - \mu_x$. Excitons spontaneously form and undergo BEC at low enough temperatures when $E_g - E_b < \mu_x$. Below the transition the exciton fields have semiclassical solutions of the form $\Phi^c(t) = |\Phi^c| e^{-i\mu_x t}$ with amplitude determined by the ratio of quadratic and quartic coefficients of the action.

P -wave interlayer tunneling gives rise to a second-order Josephson action of the form

$$S_J[\bar{\Phi}, \Phi] = \frac{1}{A} \sum_{i=1,2} \sum_q \int \frac{d\omega}{2\pi} [-c_J \Phi_{-q}^{q,i}(-\omega) \Phi_q^{c,i}(\omega) - c_J \Phi_{-q}^{c,i}(-\omega) \Phi_q^{q,i}(\omega) + ig_J \Phi_{-q}^{q,i}(-\omega) \Phi_q^{q,i}(\omega) + \text{c.c.}], \quad (11)$$

in which the intervalley exciton fields Φ^1, Φ^2 at frequency ω are coupled to those at frequency $-\omega$. Because of this coupling, the bias voltage μ_x cannot be absorbed into ω and the system is out of equilibrium as shown in the Supplemental Material [62]. Diagrammatic representations of S_0 and S_J are shown in Figs. 2(a) and 2(b).

If interlayer tunneling is s wave, a first-order Josephson term proportional to $\Phi^q(\omega = 0)$ exists. Second-order terms

of the form $\Phi(-\omega)\Phi(\omega)$ also exist and the coefficients are equal for all valley components. For p -wave interlayer tunneling (3), in contrast, angular momentum conservation implies that first-order Josephson terms vanish and that second-order terms are nonzero only for intervalley exciton fields Φ^1, Φ^2 . The second-order Josephson action (11) produces an energy landscape with explicit dependence on the phase angle $\theta = \arg \Phi$ of the form $E_J \sim \cos 2\theta$ that

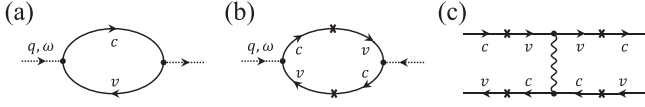


FIG. 2. Diagrammatic representations of the effective exciton action (a), (b) and an electron-hole scattering process due to interlayer tunneling (c). The solid and dotted curves represent fermion and boson fields, respectively, the crosses represent interlayer tunneling, and the wavy line represents Coulomb interaction. Panels (a) and (b), respectively, represent the free exciton action S_0 and Josephson action S_J .

breaks the $U(1)$ phase symmetry down to \mathbb{Z}_2 . The interlayer tunneling current satisfies the second-order Josephson relation $I \sim \sin 2\theta$ [56]. For an unbiased electron-hole bilayer below the BEC transition, the exciton fields are static and the system picks one of the two preferred phase angles that differ by π as the ground state implying Ising-type phase transitions of the exciton fields. Because the Josephson action (11) involves only intervalley exciton fields, intervalley excitons are energetically favored over intravalley excitons by pinning the phase at one of the two preferred phase angles, in agreement with mean-field theory results in the context of InAs/GaSb quantum wells [54,55].

Large bias limit.—In the absence of interlayer tunneling, the phase of the exciton field rotates at a constant frequency $\omega = \mu_x$. Interlayer tunneling leads to a potential landscape that explicitly breaks the $U(1)$ phase symmetry. The interplay between the $U(1)$ symmetry breaking term that traps the phase of the condensate and the bias voltage that drives a rotating phase gives rise to interesting nonequilibrium physics that is different from previous work on driven-dissipative condensates [41–45]. For small bias voltage μ_x , the exciton condensate is a static one with its phase trapped at one of the potential minima. Above a threshold bias voltage $\mu_x \sim c_J$ the condensate becomes a dynamical one with rotating phase. The transition from static to dynamical condensates is schematically shown in Fig. 3. If the bias voltage is much larger than the Josephson energy scale c_J , the phase-dependent energy landscape is swept rapidly by the rotating fields at approximately constant frequency $\omega \approx \mu_x$. Instead of Josephson effects, the Josephson action produces an average effect on the exciton fields and $U(1)$ symmetry is effectively restored.

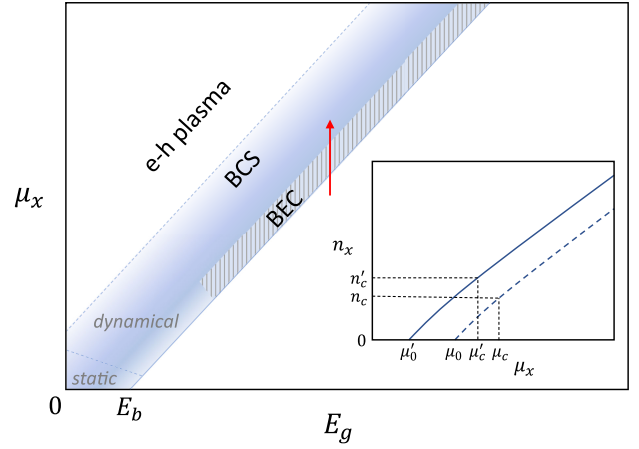


FIG. 3. Schematic phase diagram of a biased electron-hole bilayer with interlayer tunneling. E_g is the interlayer band gap, E_b is the binding energy of interlayer excitons, and μ_x is the bias voltage. The system is out of equilibrium when $\mu_x \neq 0$. The blue region represents the region in which exciton condensation occurs, and the color scale represents the strength of excitonic coherence. As the exciton density increases, the condensate undergoes a BEC-BCS crossover [63,64] and then a Mott transition [65–68] to an electron-hole plasma. The hatched area represents the large-bias BEC regime in which our theory applies. Inset: schematic plot of exciton density n_x as μ_x increases along the red arrow. The solid and dashed curves are the n_x - μ_x curves with and without interlayer tunneling, respectively. $\mu_0 = E_g - E_b$ is the threshold bias voltage for injection of excitons in the absence of interlayer tunneling, μ_c and n_c are the critical bias voltage and critical density for the occurrence of BKT transition, and μ'_0, μ'_c , and n'_c are the corresponding quantities in the presence of interlayer tunneling.

To make the above statement more precise, we note that weak interlayer tunneling ($c_J \ll \mu_x$) acts as a small perturbation that does not significantly affect the frequency of phase rotation. Thus the physically active fields are $\Phi(\omega \approx \mu_x)$ with a frequency range determined by c_J . The Josephson action S_J couples the physically active fields $\Phi(\omega \approx \mu_x)$ to the frozen degrees of freedom $\Phi(-\omega \approx -\mu_x)$. Since the $\Phi(-\omega)$ fields are trivially gapped, we can integrate them out at the quadratic level and obtain an effective action for the $\Phi(\omega)$ fields:

$$S_1[\bar{\Phi}, \Phi] = \frac{1}{A} \sum_{i=1,2} \sum_q \int \frac{d\omega}{2\pi} [\varepsilon \bar{\Phi}_q^{q,i}(\omega) \Phi_q^{c,i}(\omega) + \text{c.c.} + i\lambda \bar{\Phi}_q^{q,i}(\omega) \Phi_q^{q,i}(\omega)], \quad (12)$$

where the ω integral is defined over the small frequency range $|\omega - \mu_x| \lesssim c_J$. Equation (12) suggests that interlayer tunneling produces an extra contribution to both the c - q and q - q quadratic terms for intervalley exciton fields. The

c - q coefficient $\varepsilon > 0$ is an effective decrease of the band gap (or enhancement of the exciton binding energy), while the q - q coefficient λ implies an effective increase of temperature $\delta T = \lambda/2g$. An order-of-magnitude estimate

of the coefficients yields $\varepsilon \sim (mv_t^2)^2 E_b^3 / \mu_x E_g^3$ and $\delta T \sim (mv_t^2)^2 E_b^5 / \mu_x E_g^5$.

Physically the action (12) originates from the electron-hole scattering process illustrated by the diagram in Fig. 2(c), where an electron and a hole tunnel to the other layer, scatter by interlayer Coulomb potential, and then tunnel back to their original layers. Such scattering process enhances the effective electron-hole interactions and increases the exciton binding energy. For s -wave excitons with p -wave interlayer tunneling, the net contribution is nonzero only when the electron and hole are from opposite valleys so that angular momentum is conserved in the scattering process. Another equivalent point of view [40] is that p -wave interlayer tunneling leads to a Pondermotive force that favors intervalley excitons in the large-bias and low-density limit. This process breaks the degeneracy between intravalley and intervalley excitons and lowers the degeneracy of the ground state manifold either from $S^1 \times S^3$ to $S^1 \times S^1$ or from $S^1 \times S^2 \times S^2$ to $S^1 \times \mathbb{Z}_2$, depending on the sign of the exchange quartic term [15] (see Supplemental Material [62]). Because of the repulsion between intravalley and intervalley excitons, the ground state consists of only intervalley excitons even when the bias voltage is above the threshold value for intravalley excitons.

The effective temperature increase that shows up as an extra contribution to the q - q coefficient is physically a fluctuating force on the intervalley exciton fields and breaks the fluctuation-dissipation theorem. In our case it is the $\Phi(-\omega)$ fields that act as an extra fluctuating force on the $\Phi(\omega)$ fields, with coupling strength proportional to interlayer tunneling amplitude. The effective temperature $T_{\text{eff}} = T + \delta T$ is the temperature that controls the thermal distribution of intervalley excitons, and is the one that relates response to correlation functions of intervalley exciton fields. The emergence of an effective temperature is common in the Keldysh field theory analysis of driven-dissipative systems [43–45,69,70].

Nonequilibrium effects.—The binding energy of interlayer excitons in few-layer hBN separated TMD bilayers is typically $E_b \sim 100$ meV and decreases with the interlayer distance d . The band gap $E_g \sim 1$ eV is an order of magnitude larger than E_b , but can be tuned by a displacement field produced by the difference between top and bottom gate voltages. Altogether, the ratio $\delta T / \varepsilon \sim (E_b / E_g)^2 \sim 0.01$ is a small number, which seems to suggest that the increase of effective temperature is a negligible effect.

A finer look at the nonequilibrium effects unveils that, despite the small E_b / E_g ratio, the nonequilibrium dissipative term δT can be as important as the ε term. To see this, we sketch in the inset of Fig. 3 the density of intervalley excitons n_x as a function of the bias voltage μ_x . The gap reduction for intervalley exciton discussed above shifts the $n_x - \mu_x$ curve to the left by $\delta\mu_0 = \varepsilon$. Exciton condensation occurs when the temperature is below the Berezinskii-Kosterlitz-Thouless (BKT) transition temperature [14,71–73]

$$T_{\text{BKT}} \approx 1.3 \frac{n_x}{M}, \quad (13)$$

with M the exciton mass and n_x the exciton density. In other words, the critical exciton density for the occurrence of BKT transition at temperature T is $n_c \approx MT/1.3$. Because of the effective temperature increase, the critical density increases by $\delta n_c \approx M\delta T/1.3$. Since the $n_x - \mu_x$ curve is approximately linear at small exciton densities, with the slope approximately given by [15,38] the geometric capacitance $C = e^2 \partial n_x / \partial \mu_x \approx \varepsilon / d$ of the bilayer, the critical bias voltage in the presence of interlayer tunneling decreases by

$$\delta\mu_c = \delta\mu_0 - \frac{e^2}{C} \delta n_c \approx \varepsilon - \frac{Me^2 d}{1.3\varepsilon} \delta T. \quad (14)$$

For a TMD bilayer with a few-layer hBN dielectric spacer, $M \approx m_e$, $d \approx 2$ nm, $\varepsilon \approx 5\varepsilon_0$ (here m_e is the free electron mass and ε_0 is the vacuum permittivity), we estimate the prefactor of δT in Eq. (14) to be around 73. Since ε and δT also differ by 2 orders of magnitude, the expression in Eq. (14) can be either positive or negative in realistic systems, and its sign can be tuned by a displacement field that changes the ratio E_b / E_g .

Discussion.—We have shown in this Letter that when interlayer tunneling takes the p -wave form (3), the degeneracy between intravalley and intervalley excitons is lifted. If a large bias voltage is applied between the electron and hole layers, the U(1) symmetry breaking caused by interlayer tunneling is averaged out by the fast rotating exciton fields. The main effects of interlayer tunneling are the reduction of effective band gap and increase of effective temperature for intervalley excitons.

The assumption of p -wave interlayer tunneling (3) is crucial for our results and deserves further discussion. Our theory applies to InAs/GaSb quantum wells and angle-aligned TMD homobilayers with four of the six high-symmetry stackings (R_h^h, R_h^x, H_h^h , and H_h^x [50,51]), interlayer tunneling is p wave and our theory is directly applicable. When interlayer tunneling is s wave (e.g., TMD homobilayers with R_h^M or H_h^M stacking), a nonzero first-order Josephson term ($\propto \Phi(\omega = 0)$) exists for intravalley excitons, leading to nonzero static exciton density even before the condensation transition occurs. While the tunneling-induced static excitons are not coupled to the high-frequency exciton fields at quadratic level, electrostatic repulsion between excitons leads to an effective gap increase for excitons in both valleys.

For TMD heterobilayers or homobilayers with a nonzero twist angle, the two layers form a moiré pattern with spatially varying local stacking registry. A proper treatment of general TMD bilayers needs to take account of the momentum shift between conduction and valence bands [49,52,74,75] and is left for future work. Excitonic coherence between shifted bands leads to density wave states that break translational symmetry [55,76,77]. In a simple

intuitive picture, excitons in a moiré potential are localized near one of the high-symmetry stacking sites [52,78], and the effects of interlayer tunneling are determined by the local stacking registry.

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- [1] J. M. Blatt, K. Böer, and W. Brandt, Bose-Einstein condensation of excitons, *Phys. Rev.* **126**, 1691 (1962).
- [2] L. Keldysh and A. Kozlov, Collective properties of excitons in semiconductors, *Sov. Phys. JETP* **27**, 521 (1968), http://www.jetp.ras.ru/cgi-bin/dn/e_027_03_0521.pdf.
- [3] Y. E. Lozovik and V. Yudson, A new mechanism for superconductivity: Pairing between spatially separated electrons and holes, *Zh. Eksp. Teor. Fiz.* **71**, 738 (1976), http://www.jetp.ras.ru/cgi-bin/dn/e_044_02_0389.pdf.
- [4] M. M. Fogler and F. Wilczek, Josephson effect without superconductivity: Realization in quantum Hall bilayers, *Phys. Rev. Lett.* **86**, 1833 (2001).
- [5] A. Stern, S. M. Girvin, A. H. MacDonald, and N. Ma, Theory of interlayer tunneling in bilayer quantum Hall ferromagnets, *Phys. Rev. Lett.* **86**, 1829 (2001).
- [6] J. Eisenstein and A. MacDonald, Bose-Einstein condensation of excitons in bilayer electron systems, *Nature (London)* **432**, 691 (2004).
- [7] J. Eisenstein, Exciton condensation in bilayer quantum Hall systems, *Annu. Rev. Condens. Matter Phys.* **5**, 159 (2014).
- [8] I. B. Spielman, J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, Resonantly enhanced tunneling in a double layer quantum Hall ferromagnet, *Phys. Rev. Lett.* **84**, 5808 (2000).
- [9] M. Kellogg, J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, Vanishing Hall resistance at high magnetic field in a double-layer two-dimensional electron system, *Phys. Rev. Lett.* **93**, 036801 (2004).
- [10] E. Tutuc, M. Shayegan, and D. A. Huse, Counterflow measurements in strongly correlated GaAs hole bilayers: Evidence for electron-hole pairing, *Phys. Rev. Lett.* **93**, 036802 (2004).
- [11] 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).
- [12] H. Min, R. Bistritzer, J.-J. Su, and A. MacDonald, Room-temperature superfluidity in graphene bilayers, *Phys. Rev. B* **78**, 121401(R) (2008).
- [13] A. Perali, D. Neilson, and A. R. Hamilton, High-temperature superfluidity in double-bilayer graphene, *Phys. Rev. Lett.* **110**, 146803 (2013).
- [14] M. Fogler, L. Butov, and K. Novoselov, High-temperature superfluidity with indirect excitons in van der Waals heterostructures, *Nat. Commun.* **5**, 4555 (2014).
- [15] F.-C. Wu, F. Xue, and A. H. MacDonald, Theory of two-dimensional spatially indirect equilibrium exciton condensates, *Phys. Rev. B* **92**, 165121 (2015).
- [16] Y. Zeng, N. Wei, and A. H. MacDonald, Layer pseudospin magnetism in a transition metal dichalcogenide double-moiré system, *Phys. Rev. B* **106**, 165105 (2022).
- [17] S. Conti, M. Van der Donck, A. Perali, F. M. Peeters, and D. Neilson, Doping-dependent switch from one- to two-component superfluidity in coupled electron-hole van der Waals heterostructures, *Phys. Rev. B* **101**, 220504(R) (2020).
- [18] S. Conti, S. Saberi-Pouya, A. Perali, M. Virgilio, F. M. Peeters, A. R. Hamilton, G. Scappucci, and D. Neilson, Electron-hole superfluidity in strained si/ge Type II heterojunctions, *npj Quantum Mater.* **6**, 41 (2021).
- [19] S. Conti, A. Perali, A. R. Hamilton, M. V. Milošević, F. m. c. M. Peeters, and D. Neilson, Chester supersolid of spatially indirect excitons in double-layer semiconductor heterostructures, *Phys. Rev. Lett.* **130**, 057001 (2023).
- [20] A. F. Croxall, K. D. Gupta, C. A. Nicoll, M. Thangaraj, H. E. Beere, I. Farrer, D. A. Ritchie, and M. Pepper, Anomalous coulomb drag in electron-hole bilayers, *Phys. Rev. Lett.* **101**, 246801 (2008).
- [21] J. A. Seamons, C. P. Morath, J. L. Reno, and M. P. Lilly, Coulomb drag in the exciton regime in electron-hole bilayers, *Phys. Rev. Lett.* **102**, 026804 (2009).
- [22] R. Gorbachev, A. Geim, M. Katsnelson, K. Novoselov, T. Tudorovskiy, I. Grigorieva, A. MacDonald, S. Morozov, K. Watanabe, T. Taniguchi *et al.*, Strong coulomb drag and broken symmetry in double-layer graphene, *Nat. Phys.* **8**, 896 (2012).
- [23] G. W. Burg, N. Prasad, K. Kim, T. Taniguchi, K. Watanabe, A. H. MacDonald, L. F. Register, and E. Tutuc, Strongly enhanced tunneling at total charge neutrality in double-bilayer graphene-WSe₂ heterostructures, *Phys. Rev. Lett.* **120**, 177702 (2018).
- [24] 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).
- [25] K. F. Mak, C. Lee, J. Hone, J. Shan, and T. F. Heinz, Atomically thin MoS₂: A new direct-gap semiconductor, *Phys. Rev. Lett.* **105**, 136805 (2010).
- [26] K. He, N. Kumar, L. Zhao, Z. Wang, K. F. Mak, H. Zhao, and J. Shan, Tightly bound excitons in monolayer WSe₂, *Phys. Rev. Lett.* **113**, 026803 (2014).
- [27] G. Wang, A. Chernikov, M. M. Glazov, T. F. Heinz, X. Marie, T. Amand, and B. Urbaszek, Colloquium: Excitons in atomically thin transition metal dichalcogenides, *Rev. Mod. Phys.* **90**, 021001 (2018).
- [28] K. F. Mak and J. Shan, Photonics and optoelectronics of 2D semiconductor transition metal dichalcogenides, *Nat. Photonics* **10**, 216 (2016).
- [29] E. C. Regan, D. Wang, E. Y. Paik, Y. Zeng, L. Zhang, J. Zhu, A. H. MacDonald, H. Deng, and F. Wang, Emerging exciton physics in transition metal dichalcogenide heterobilayers, *Nat. Rev. Mater.* **7**, 778 (2022).

- [30] H. Fang, C. Battaglia, C. Carraro, S. Nemsak, B. Ozdol, J. S. Kang, H. A. Bechtel, S. B. Desai, F. Kronast, A. A. Unal *et al.*, Strong interlayer coupling in van der Waals heterostructures built from single-layer chalcogenides, *Proc. Natl. Acad. Sci. U.S.A.* **111**, 6198 (2014).
- [31] P. Rivera, J. R. Schaibley, A. M. Jones, J. S. Ross, S. Wu, G. Aivazian, P. Klement, K. Seyler, G. Clark, N. J. Ghimire *et al.*, Observation of long-lived interlayer excitons in monolayer MoSe₂-WSe₂ heterostructures, *Nat. Commun.* **6**, 6242 (2015).
- [32] L. A. Jauregui, A. Y. Joe, K. Pistunova, D. S. Wild, A. A. High, Y. Zhou, G. Scuri, K. De Greve, A. Sushko, C.-H. Yu *et al.*, Electrical control of interlayer exciton dynamics in atomically thin heterostructures, *Science* **366**, 870 (2019).
- [33] K. F. Mak and J. Shan, Opportunities and challenges of interlayer exciton control and manipulation, *Nat. Nanotechnol.* **13**, 974 (2018).
- [34] 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).
- [35] R. Qi, A. Y. Joe, Z. Zhang, Y. Zeng, T. Zheng, Q. Feng, E. Regan, J. Xie, Z. Lu, T. Taniguchi *et al.*, Thermodynamic behavior of correlated electron-hole fluids in van der Waals heterostructures, [arXiv:2306.13265](https://arxiv.org/abs/2306.13265).
- [36] 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, [arXiv:2309.14940](https://arxiv.org/abs/2309.14940).
- [37] R. Qi, A. Y. Joe, Z. Zhang, J. Xie, Q. Feng, Z. Lu, Z. Wang, T. Taniguchi, K. Watanabe, S. Tongay *et al.*, Perfect Coulomb drag and exciton transport in an excitonic insulator, [arXiv:2309.15357](https://arxiv.org/abs/2309.15357).
- [38] Y. Zeng and A. H. MacDonald, Electrically controlled two-dimensional electron-hole fluids, *Phys. Rev. B* **102**, 085154 (2020).
- [39] M. Xie and A. H. MacDonald, Electrical reservoirs for bilayer excitons, *Phys. Rev. Lett.* **121**, 067702 (2018).
- [40] Z. Sun, Y. Murakami, T. Kaneko, D. Golež, and A. J. Millis, Dynamical exciton condensates in biased electron-hole bilayers, [arXiv:2312.06426](https://arxiv.org/abs/2312.06426).
- [41] L. M. Sieberer, S. D. Huber, E. Altman, and S. Diehl, Dynamical critical phenomena in driven-dissipative systems, *Phys. Rev. Lett.* **110**, 195301 (2013).
- [42] L. M. Sieberer, M. Buchhold, and S. Diehl, Keldysh field theory for driven open quantum systems, *Rep. Prog. Phys.* **79**, 096001 (2016).
- [43] K. Dunnett and M. H. Szymańska, Keldysh field theory for nonequilibrium condensation in a parametrically pumped polariton system, *Phys. Rev. B* **93**, 195306 (2016).
- [44] M. F. Maghrebi and A. V. Gorshkov, Nonequilibrium many-body steady states via Keldysh formalism, *Phys. Rev. B* **93**, 014307 (2016).
- [45] E. G. Dalla Torre, S. Diehl, M. D. Lukin, S. Sachdev, and P. Strack, Keldysh approach for nonequilibrium phase transitions in quantum optics: Beyond the Dicke model in optical cavities, *Phys. Rev. A* **87**, 023831 (2013).
- [46] L. V. Keldysh *et al.*, Diagram technique for nonequilibrium processes, *Sov. Phys. JETP* **20**, 1018 (1965), http://www.jetp.ras.ru/cgi-bin/dn/e_020_04_1018.pdf.
- [47] A. Kamenev, *Field Theory of Non-Equilibrium Systems* (Cambridge University Press, Cambridge, England, 2023).
- [48] A. Altland and B. D. Simons, *Condensed Matter Field Theory* (Cambridge University Press, Cambridge, England, 2010).
- [49] Y. Wang, Z. Wang, W. Yao, G.-B. Liu, and H. Yu, Interlayer coupling in commensurate and incommensurate bilayer structures of transition-metal dichalcogenides, *Phys. Rev. B* **95**, 115429 (2017).
- [50] P. Rivera, H. Yu, K. L. Seyler, N. P. Wilson, W. Yao, and X. Xu, Interlayer valley excitons in heterobilayers of transition metal dichalcogenides, *Nat. Nanotechnol.* **13**, 1004 (2018).
- [51] G.-B. Liu, D. Xiao, Y. Yao, X. Xu, and W. Yao, Electronic structures and theoretical modelling of two-dimensional group-VIB transition metal dichalcogenides, *Chem. Soc. Rev.* **44**, 2643 (2015).
- [52] H. Yu, G.-B. Liu, J. Tang, X. Xu, and W. Yao, Moiré excitons: From programmable quantum emitter arrays to spin-orbit-coupled artificial lattices, *Sci. Adv.* **3**, e1701696 (2017).
- [53] C. Liu, T. L. Hughes, X.-L. Qi, K. Wang, and S.-C. Zhang, Quantum spin Hall effect in inverted type-II semiconductors, *Phys. Rev. Lett.* **100**, 236601 (2008).
- [54] F. Xue and A. H. MacDonald, Time-reversal symmetry-breaking nematic insulators near quantum spin Hall phase transitions, *Phys. Rev. Lett.* **120**, 186802 (2018).
- [55] Y. Zeng, F. Xue, and A. H. MacDonald, In-plane magnetic field induced density wave states near quantum spin Hall phase transitions, *Phys. Rev. B* **105**, 125102 (2022).
- [56] Z. Sun, T. Kaneko, D. Golež, and A. J. Millis, Second-order Josephson effect in excitonic insulators, *Phys. Rev. Lett.* **127**, 127702 (2021).
- [57] H. Yu, G.-B. Liu, P. Gong, X. Xu, and W. Yao, Dirac cones and Dirac saddle points of bright excitons in monolayer transition metal dichalcogenides, *Nat. Commun.* **5**, 3876 (2014).
- [58] M. M. Glazov, T. Amand, X. Marie, D. Lagarde, L. Bouet, and B. Urbaszek, Exciton fine structure and spin decoherence in monolayers of transition metal dichalcogenides, *Phys. Rev. B* **89**, 201302(R) (2014).
- [59] T. Yu and M. W. Wu, Valley depolarization due to intervalley and intravalley electron-hole exchange interactions in monolayer MoS₂, *Phys. Rev. B* **89**, 205303 (2014).
- [60] F. Wu, F. Qu, and A. H. MacDonald, Exciton band structure of monolayer MoS₂, *Phys. Rev. B* **91**, 075310 (2015).
- [61] D. Y. Qiu, T. Cao, and S. G. Louie, Nonanalyticity, valley quantum phases, and lightlike exciton dispersion in monolayer transition metal dichalcogenides: Theory and first-principles calculations, *Phys. Rev. Lett.* **115**, 176801 (2015).
- [62] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.132.266001> for detailed derivation of the Keldysh action for excitons and discussion of nonequilibrium effects.
- [63] C. Comte and P. Nozieres, Exciton Bose condensation: The ground state of an electron-hole gas-I. Mean field description of a simplified model, *J. Phys.* **43**, 1069 (1982).
- [64] P. Nozieres and S. Schmitt-Rink, Bose condensation in an attractive fermion gas: From weak to strong coupling superconductivity, *J. Low Temp. Phys.* **59**, 195 (1985).

- [65] N. F. Mott, The basis of the electron theory of metals, with special reference to the transition metals, *Proc. Phys. Soc. London Sect. A* **62**, 416 (1949).
- [66] N. Mott, Metal-insulator transitions, *Contemp. Phys.* **14**, 401 (1973).
- [67] W. F. Brinkman and T. M. Rice, Electron-hole liquids in semiconductors, *Phys. Rev. B* **7**, 1508 (1973).
- [68] D. Guerci, M. Capone, and M. Fabrizio, Exciton Mott transition revisited, *Phys. Rev. Mater.* **3**, 054605 (2019).
- [69] A. Mitra, S. Takei, Y. B. Kim, and A. J. Millis, Nonequilibrium quantum criticality in open electronic systems, *Phys. Rev. Lett.* **97**, 236808 (2006).
- [70] A. Mitra, I. Aleiner, and A. J. Millis, Semiclassical analysis of the nonequilibrium local polaron, *Phys. Rev. Lett.* **94**, 076404 (2005).
- [71] J. M. Kosterlitz and D. J. Thouless, Ordering, metastability and phase transitions in two-dimensional systems, *J. Phys. C* **6**, 1181 (1973).
- [72] A. Filinov, N. V. Prokof'Ev, and M. Bonitz, Berezinskii-Kosterlitz-Thouless transition in two-dimensional dipole systems, *Phys. Rev. Lett.* **105**, 070401 (2010).
- [73] Although Eq. (13) was obtained for equilibrium systems, since U(1) symmetry is effectively restored in the large bias limit, Eq. (13) is applicable but with T and n_x modified by nonequilibrium effects [Eq. (12)].
- [74] R. Bistritzer and A. H. MacDonald, Moiré bands in twisted double-layer graphene, *Proc. Natl. Acad. Sci. U.S.A.* **108**, 12233 (2011).
- [75] F. Wu, T. Lovorn, E. Tutuc, I. Martin, and A. H. MacDonald, Topological insulators in twisted transition metal dichalcogenide homobilayers, *Phys. Rev. Lett.* **122**, 086402 (2019).
- [76] P. Rickhaus, F. K. de Vries, J. Zhu, E. Portoles, G. Zheng, M. Masseroni, A. Kurzmann, T. Taniguchi, K. Watanabe, A. H. MacDonald *et al.*, Correlated electron-hole state in twisted double-bilayer graphene, *Science* **373**, 1257 (2021).
- [77] A. Kogar, M. S. Rak, S. Vig, A. A. Husain, F. Flicker, Y. I. Joe, L. Venema, G. J. MacDougall, T. C. Chiang, E. Fradkin *et al.*, Signatures of exciton condensation in a transition metal dichalcogenide, *Science* **358**, 1314 (2017).
- [78] F. Wu, T. Lovorn, and A. H. MacDonald, Theory of optical absorption by interlayer excitons in transition metal dichalcogenide heterobilayers, *Phys. Rev. B* **97**, 035306 (2018).