Second-Order Josephson Effect in Excitonic Insulators

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We show that in electron-hole bilayers with excitonic orders arising from conduction and valence bands formed by atomic orbitals that have different parities, nonzero interlayer tunneling leads to a second-order Josephson effect. This means the interlayer electrical current is related to the phase of the excitonic order parameter as $J = J_c \sin 2\theta$ instead of $J = J_c \sin\theta$ and that the system has two degenerate ground states at $\theta = 0, \pi$ that can be switched by an interlayer voltage pulse. When generalized to a three dimensional stack of alternating electron-hole planes or a two dimensional stack of chains, the ac Josephson effect implies that electric field pulses perpendicular to the layers and chains can steer the order parameter phase between the two degenerate ground states, making these devices ultrafast memories. The order parameter steering also applies to the excitonic insulator candidate Ta_2NiSe_5.

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Excitonic condensation [1–5] has been experimentally realized in electron-hole bilayers (EHBs) [6–15] where electrons in one layer pair with holes in the other layer to form excitons that condense into a single macroscopic state. In 1976, Kulik and Shevchenko [16,17] (see also Refs. [18–20]) noted that nonzero interlayer tunneling endows EHBs with a Josephson effect similar to that in superconductors. This effect was observed in 2000 by Spielman *et al.* in quantum Hall bilayers [21,22] and explained in detail in Refs. [23–26].

If the electron and hole bands are formed by atomic orbitals that transform differently under crystal symmetries, the intrinsic tunneling (hybridization) vanishes at high symmetry points of the Brillouin zone and is very small nearby, such that the excitonic insulator (EI) transition breaks a discrete symmetry [4,27–31]. In this Letter, we show that if the orbitals lie at different spatial locations, as shown in Fig. 1, a difference of symmetries (e.g., p and d orbitals) implies that the ordered state sustains a "second-order" Josephson effect as the tunneling has to create or annihilate two excitons each time. A similar effect is already well known in carefully designed superconducting Josephson junctions [32] (e.g., a 45° junction between d-wave superconductors or a junction between s- and d-wave superconductors [33–40]). We show that this effect naturally occurs in EIs, which leads to symmetry breaking degenerate ground states that are easily distinguishable and switchable. In an isolated EHB, the two ground states break parity and have opposite in-plane electrical polarization. In three dimensional (3D) stacks of coupled planes or two dimensional (2D) stacks of coupled chains (Fig. 2), the two EI states break time reversal symmetry with opposite anomalous Hall conductivity [41,42] and potentially form topologically nontrivial states. In all cases the excitonic order parameter may be "steered" by applied interlayer or interchain electric fields via the ac Josephson effect, enabling controlled switching of degenerate ground states. This order parameter steering applies as well to the EI candidate Ta_2NiSe_5 [29,43–50].

The electron-hole bilayer shown in Fig. 1(a) consists of two planes labeled 1 and 2 with the two-component

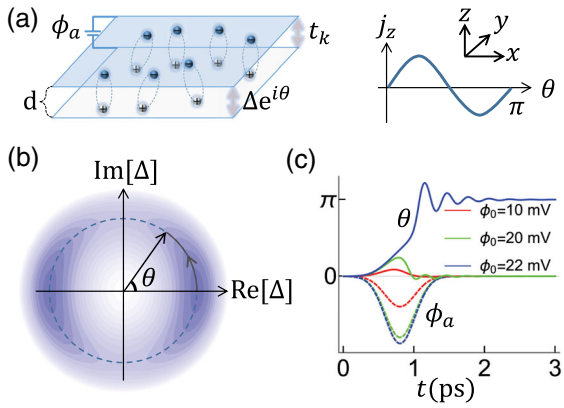

FIG. 1. (a) Schematics of the electron-hole bilayer showing electrons (−) and holes (+) and the interlayer current-phase relation in the excitonic insulating phase. (b) False color representation of the free energy on the plane of a complex order parameter where lower energy appears bluer. (c) Solid curve: time dependence of order parameter phase after a voltage pulse $\phi_a = -\phi_0 e^{-(t-t_0)^2/T_0^2}$ (dashed curve) computed from Eq. (4) with $\Delta_p = 14$ meV, $T_0 = 0.3$ ps, $\gamma = 0.3\Delta_p$, $D = 2$, and $C = 1$.

electron creation operator $\psi^\dagger = (\psi_1^\dagger, \psi_2^\dagger)$ from the two bands. The Hamiltonian is

$$H_{\mathrm{EHB}} = \sum_k \psi_k^\dagger \begin{pmatrix} \xi_1(k+A_1)+\phi_1 & e^{\mathrm{id}A_z} t_{k+A} \\ e^{-\mathrm{id}A_z} t_{k+A}^* & \xi_2(k+A_2)+\phi_2 \end{pmatrix} \psi_k + \int dr dr' V(r-r')\rho(r)\rho(r'), \quad (1)$$

where $\psi_k = \int dr e^{ikr}\psi(r)$, $\rho(r) = \psi^\dagger(r)\psi(r)$ is the density, $\xi_{1,2}(k)$ is the kinetic energy describing in-plane motion with ξ_1 dispersing upward from a minimum $-G/2$ and ξ_2 dispersing downward from a maximum $G/2$ at the same momentum $k=0$, both isotropic. (ϕ_i, A_i) is the electromagnetic (EM) potential at layer i, $A = (A_1+A_2)/2$ is the average in-plane component of the vector potential, A_z is the average out-of-plane component, and we have set $e = c = \hbar = 1$. We assume the Hamiltonian is invariant under time reversal $\hat{T}$ and in-plane inversion defined as $\hat{P}: r \to -r, (\psi_1, \psi_2)_r \to (\psi_1, -\psi_2)_{-r}$, where $r = (x, y)$, implying $\xi_{1,2}(k) = \xi_{1,2}(-k)$ and that the intrinsic interlayer tunneling satisfies $t_{-k} = t_k^* = -t_k$. Thus, one can write $t_k = i\Delta_p f_k$ where f_k is odd under $k \to -k$, $\Delta_p > 0$ is real, and the subscript "p" denotes the k-odd nature. We distinguish the Bardeen-Cooper-Schrieffer (BCS) case ($G > 0$), where the two bands cross at a Fermi momentum k_F with Fermi velocity v_F, from the Bose-Einstein condensation (BEC) case ($G < 0$), where they do not overlap. While all the equations and qualitative conclusions hold for both cases, the quantitative coefficients are presented for the analytically tractable BCS weak coupling case ($\Delta \ll G$) unless otherwise specified. Without loss of generality, we set $f_k = c_f \sin k_x$ where c_f is chosen such that $|f_{k_F}| = 1$.

To study the excitonic order, we write the model as a path integral and decompose the interaction in the electron-hole pairing channel $Z = \int D[\psi, \Delta_k, A] e^{\int d\tau dr L_0(\psi, \Delta_k, A)}$, where Δ_k is the Hubbard-Stratonovich field. The excitonic state appears as a saddle point with the order parameter $\Delta_k = \sum_{k'} V_{k-k'} \langle \psi_{2k'}^\dagger \psi_{1k'} \rangle$, where V_q is the Fourier transform of $V(r)$. For physically reasonable interactions, the energetically favored order parameter $\Delta e^{i\theta}$ has s-wave symmetry [51] so the k dependence may be neglected. The quasiparticle properties are described by replacing the term $e^{\mathrm{id}A_z} t_{k+A}$ in Eq. (1) by $\Delta e^{i\theta} + e^{\mathrm{id}A_z} t_{k+A}$ [52]. There is always an odd parity phonon [27,43,53–55] (e.g., shear motion between the two layers) that couples linearly to Δ but may be integrated out.

Integrating out the fermions, phonons, and the order parameter amplitude fluctuations, one obtains a low energy effective Lagrangian for the order parameter phase

$$L = \frac{1}{2}\nu \left\{ -(\partial_t\theta + \phi_a)^2 + v_g^2(\nabla\theta - A_a)^2 - \frac{1}{D}\Delta_p^2 \cos[2(\theta - A_z d)] \right\}, \quad (2)$$

where $(\phi_a, A_a) = (\phi_1 - \phi_2, A_1 - A_2)/2$ is the layer-antisymmetric component of the EM field [56]. The last term arises from expanding L to second order in t_k (assumed small relative to Δ or temperature), observing that terms linear in t vanish (see Ref. [57], Sec. I). An inversion even t_k would change this term to $\propto t\cos\theta$, giving rise to the usual Josephson effect [16,18,23–25]. The z-dipole density is $\rho_a = \delta L/\delta(\partial_t\theta) = -\nu(\partial_t\theta + \phi_a)$, and Eq. (2) should be supplemented by the electric field energy $\sum_q \phi_a(q)^2/[2V_{\mathrm{eff}}(q)]$ representing the dipole-dipole interactions $V_{\mathrm{eff}}(q) = (1 - e^{-dq})V_q$ [56,68]. At zero temperature, the coefficients of Eq. (2) have simple Δ-independent forms: $D = 2$ is the space dimension, ν is the density of states in the normal state at the band crossing energy, and the bare phase mode velocity is $v_g = v_F/\sqrt{2}$.

If t_k is zero, Eq. (1) conserves the charge in each plane and gives a continuous family of excitonic phases parametrized by θ, as manifested by the $U(1)$ symmetry under transformation $\theta \to \theta + \theta_0$ of the first two terms of Eq. (2). A nonzero t_k gives rise to the third term, which reduces the $U(1)$ invariance to $\hat{P}$, a Z_2 symmetry, and implies that there are two degenerate excitonic phases characterized by $\theta = 0, \pi$ [Fig. 1(b)]. The excitonic order spontaneously breaks $\hat{P}$, giving a nonvanishing in-plane electrical polarization [28,51] which in the BCS case is $P = P_{2D}[1 - \tan(\frac{1}{2}\mathrm{ArcTan}|\Delta_p/\Delta|)]\mathrm{Sign}[\Delta]/4$. Since its sign is opposite for $\theta = 0, \pi$, measuring it by an electrical circuit can distinguish the two ground states. In the BEC case [69], the polarization has a more transparent physical picture. The normal state preceding the EI phase is a semiconductor that supports excitonic modes. t_k means that these modes have oscillating in-plane electrical dipoles. In the EI phase, a mode softens and freezes as the static in-plane electrical polarization.

In spinful systems, both singlet and triplet excitonic condensates may be defined. The triplet case exhibits spin instead of charge polarization. In the pure electronic system, the two phases are degenerate at the Hartree-Fock level, but electron-lattice coupling favors the singlet state [4,27,53] (see Ref. [57], Sec. V). We focus on the more commonly studied singlet phase here.

Second-order Josephson effect and order parameter steering.—The interplane current

$$-J_z = \delta L/\delta(dA_z) = \frac{\nu}{D}\Delta_p^2 \sin(2\theta) \equiv J_c \sin(2\theta) \quad (3)$$

is periodic under $\theta \to \theta + \pi$ in contrast to the usual Josephson effect, where it is periodic only under

$\theta \to \theta + 2\pi$; the former is thus referred to as a second-order Josephson effect. Assuming a quadratic band with effective mass 0.1 m_e and $\Delta_p = 10$ meV, the critical current is estimated as $J_c \approx 4$ mA/μm^2. To observe the dc Josephson effect, one can source a current at one layer and drain it on the other layer, both on the left side of the device where the in-plane counterflow current $J_a = \nu v_g^2 \partial_x \theta$ is fixed as the boundary condition [21]. From the static limit of the Euler-Lagrange equation (charge continuity equation) implied by Eq. (2), $\nu v_g^2 \partial_x^2 \theta = J_c \sin(2\theta)$, the phase decays to the right with a decay length $l_d = \sqrt{\nu v_g^2 / J_c} \sim \sqrt{D} v_g / \Delta_p$ [70]. Thus, in a long junction, only the region within a distance l_d to the contact contributes to the Josephson current [18]. The current phase relation can be verified by applying an in-plane magnetic field to a short junction and measuring the critical Josephson current as a function of the magnetic flux Φ through it [61]. The Fraunhofer pattern $J_c(\Phi)/J_c(0) = |\frac{\sin(N\pi\Phi/(2\Phi_0))}{N\pi\Phi/(2\Phi_0)}|$ is expected where Φ_0 is the flux quantum and the frequency $N = 2$ reveals the order of the Josephson effect (see Ref. [57], Sec. IB).

To treat the order parameter steering, we focus on spatially uniform dynamics that applies to a device with gates covering the whole sample such that ϕ_a is uniform or a short EHB with side contacts. Equation (2) in the gauge $A = 0$ implies

$$\frac{1}{C}\partial_t(\partial_t\theta + \phi_a) + \gamma\partial_t\theta + \frac{1}{D}\Delta_p^2 \sin 2\theta = 0, \qquad (4)$$

where a $C \neq 1$ expresses the effect of dipole-dipole interactions (charging energy) and we have added a phenomenological damping γ. Thus, the time derivative of an interlayer voltage ϕ_a provides a force that pushes the phase to increase, meaning that a suitable voltage pulse can switch the system between ground states, as in Figs. 1(b),(c). If ϕ_a is applied by side contacts or by gates immediately adjacent to the bilayer, the external electrical circuit controls ϕ_a, which is already the total voltage across the layers, and one has $C = 1$ in Eq. (4). To climb the potential hill at $\theta = \pi/2$ with energy $\nu\Delta_p^2/4$, the threshold voltage required for a typical pulse $\phi_a = \phi_0 e^{-(t-t_0)^2/T_0^2}$ is $\phi_c \sim T_0\Delta_p^2 C/D$, giving $\phi_c \sim 25$ mV for $T_0 = 1$ ps, $\Delta_p = 10$ meV, and $C = 1$. In the limit of strong drive ($\phi_a \gg \phi_c$), the equation of motion becomes $\partial_t\theta = -\phi_a$, recovering the familiar ac Josephson effect. Note that the switching frequency scale $1/T_0$ is upper bounded by the gap Δ.

We have assumed that lattice distortions, if present, can dynamically follow the order parameter. In the opposite limit of slow lattice dynamics, one should fix the lattice distortion. For weak electron lattice coupling (ELC), the only change is that the Z_2 symmetry remains broken and the second minimum is at higher energy [71]. For a larger ELC, the second minimum no longer exists. Thus, fast phase steering can reveal the strength of the ELC.

Beyond bilayers.—The second-order Josephson effect generalizes to the 3D and 2D systems by stacking the electron-hole bilayers and chains as in Figs. 2(a),(b). The stacking is along z, and the conjugate wave vector is $k_z \in (-\pi, \pi]/(2d)$. The model is invariant under translations by the z-direction lattice constant $2d$ and reflection $z \leftrightarrow -z$ with respect to a plane containing either the electron or holes. We specialize to the short ranged density-density interaction g such that the excitonic order $\Delta_{i1/2}$ only links adjacent layers, as in Fig. 2(a), and consider mean field solutions where the amplitude Δ is spatially uniform but allows the phases $\theta_{1,2}$ on the two bonds to be different. We define the symmetric and antisymmetric phase combinations $\theta_{s,a} = (\theta_1 \pm \theta_2)/2$, whose domain is $\theta_s \in (-\pi, \pi], \theta_a \in [0, \pi)$. In the momentum basis of field operators $\psi_k^\dagger = (\psi_{1k}^\dagger, \psi_{2k}^\dagger) = \int dr \sum_j e^{i(k_\perp r + k_z j 2d)} [\psi_{j1}^\dagger(r), e^{ik_z d}\psi_{j2}^\dagger(r)]$, where $k_\perp$ is the momentum along the planes and chains, the Lagrangian reads $L = \sum_k \psi_k^\dagger(\partial_\tau + H_k)\psi_k + (2/g)|\Delta|^2$ with the mean field Hamiltonian

$$H_k = \begin{bmatrix} \xi_1(k_\perp) & \Delta(k) - i\Delta_p f_k \cos dk_z \\ \Delta(k)^* + i\Delta_p f_k \cos dk_z & \xi_2(k_\perp) \end{bmatrix}, \qquad (5)$$

where the Δ_p term is the intrinsic interlayer tunneling t_k and the order parameter is

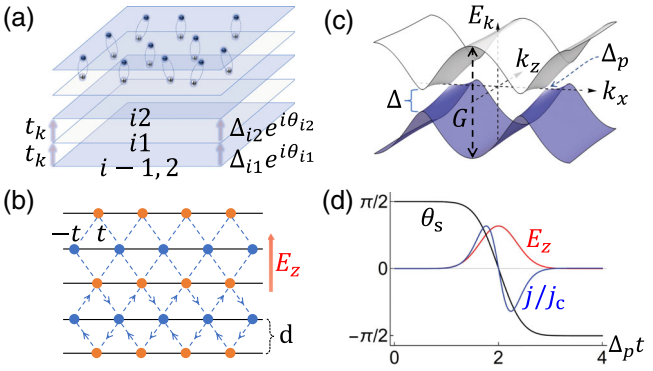

FIG. 2. (a) Schematic of the 3D stack of alternating electron (blue) and hole (unshaded) planes with pairing order parameters labeled. (b) Schematic of the 2D stack of alternating electron and hole chains. The orange and blue dots represent atomic orbitals forming the conduction and valence bands. Their different parities lead to asymmetric interchain hopping $t/-t$ [30]. Arrows represent the spontaneous circulating currents. (c) The ground state band dispersion of the 2D stack. (d) The order parameter phase dynamics (black curve) and the Josephson current (blue curve) induced by an electric field pulse $E_z(t) = E_{\max} e^{-(t-t_0)^2/T_0^2}$ (red curve) implied by Eq. (9), with $\Delta = 10\Delta_p$, $E_{\max} = 3.55\Delta_p/d$, and $T_0 = 0.5/\Delta_p$.

$$\Delta(k) = e^{i\theta_a}\Delta\cos(dk_z + \theta_s). \tag{6}$$

Our gauge choice here is that a spatially uniform electric field enters through $k \to k + A$, including the $\Delta(k)$ term.

At $\Delta_p = 0$, the energy is independent of θ_1 and θ_2. Nonzero Δ_p reduces the symmetry to $\hat{T}$ and $\hat{P}$, and the excitonic ground state turns out to spontaneously break $\hat{T}$ instead of $\hat{P}$, corresponding to $(\theta_a, \theta_s) = (0, \pm\pi/2)$, i.e., $\theta_{i1} = \theta_{i2} = \pm\pi/2$. This is verified by expanding the Lagrangian to quadratic order in Δ_p (see Ref. [57], Sec. II). Fixing $\theta_a = 0$ and in the gauge $\phi = 0$, one finds

$$L = K[\dot{\theta}_s + d\dot{A}_z, A_x] + c_\nu\Delta_p^2\cos 2\theta_s + F_0, \tag{7}$$

where K is the kinetic term that vanishes in the static limit and $F_0(|\Delta|)$ is the ground state free energy without interlayer tunneling, and we have neglected constant $O(\Delta_p^2)$ terms. The $\cos 2\theta_s$ term means a "second-order Josephson" current $j_z = j_c \sin 2\theta_s$, where $j_c = 2dc_\nu\Delta_p^2$ and $c_\nu \sim \nu$. In the equilibrium state, the total electrical polarization is zero, but there are circulating currents $j_{\text{inter},a} = \langle\sum_k(\partial_k t_k)\sin(dk_z)\sigma_1\rangle$ due to broken $\hat{T}$, as shown in Fig. 2(b). Note that this state is linearly stable to lattice distortions.

Around each of the two equilibrium configurations, expanding Eq. (7) to quadratic order in $\theta \equiv \theta_s \pm \pi/2$ and the EM fields $A_{x/z}$, one obtains the Gaussian action for θ_s fluctuations. In the low energy regime $\omega \ll \Delta_p$ and long wavelength limit $q = 0$, it reads

$$S_s = -\sum_\omega c_0(\omega)(\theta + dA_z)_{-\omega}(\theta + dA_z)_\omega + \int dtdr[c_1\theta^2 + \sigma_h(\theta + dA_z)\partial_t A_x/d] + S_{A_x^2}, \tag{8}$$

neglecting terms subleading in Δ_p. The first two terms are the kinetic and potential energies of phase fluctuations where $c_0(\omega)$ is the kinetic kernel that vanishes in the static limit and $c_1 = 2c_\nu\Delta_p^2$ for $\Delta_p \ll \Delta$. The third term gives rise to an anomalous Hall conductivity σ_h for electric fields in the $x - z$ plane, which can also be written into an "axion" form [72]. The last term is the bare optical response in the x direction.

The excitonic order leads to topologically nontrivial ground states in the BCS regime ($G > 0$). Setting $\xi_1(k) = -\xi_2(k) = \xi_k$ for simplicity, the quasiparticle dispersion is $E_k = \pm\sqrt{\xi_k^2 + |\Delta(k)|^2 + \Delta_p^2 f_k^2\cos^2(dk_z)}$. In the 2D stack of electron-hole chains, the quasiparticle is gapped with massive Dirac points at $(k_x, k_z) = (\pm k_F, 0)$ with mass $\pm\Delta_p$, as shown in Fig. 2(c). The Chern number of the valence band is $\text{Sign}[\theta_s]$ so that the system is a quantum anomalous Hall insulator [73] with quantized Hall conductivity $\sigma_h = \text{Sign}[\theta_s]e^2/h$ and chiral edge states. The kinetic kernel $c_0 = (\nu/3)\omega^2\Delta/\Delta_p$ renders the bare phase mode gap $\omega_0 \sim \Delta_p\sqrt{\Delta_p/\Delta}$. The 3D stack of electron-hole layers is a Weyl semimetal [72] with Weyl nodes at $k = (0, \pm k_F, 0)$ and anomalous Hall conductivity $\sigma_h = \text{Sign}[\theta_s](k_F/\pi)e^2/h$ (see Ref. [57] Sec. II, and Sec. V for the effect of spins). Note that the BEC regime ($G < 0$) is topologically trivial with σ_h vanishing and the minimal gap being $\sqrt{G^2 + 4\Delta^2}$, although there is nonzero ac Hall response $\sim\Delta_p$ that can be measured by Kerr rotation [neglected in Eq. (8)].

Order parameter steering by light.—In all these systems, the order parameter can be steered by electric fields perpendicular to the layers and chains, e.g., from ground state $|g\rangle$ to $\hat{P}|g\rangle$ for the bilayer and to $\hat{T}|g\rangle$ for the 3D and 2D stacks. This can be easily verified in "pump-probe" experiments since the ground states have opposite in-plane polarization in the bilayer and opposite Hall response in the stacks. The order parameter steering follows the spirit of the ac Josephson effect: the phase θ_s enters the kinetic term in Eq. (7) together with the vector potential as $\theta_s + dA_z$. This term has different forms in different regimes. For example, in the 2D stacks it behaves as $K \sim \nu|\Delta/\Delta_p|\dot{\theta}_s^2$ in the slow limit of $\dot{\theta}_s \ll \Delta_p$ and as $K \sim \nu|\Delta|\theta_s\dot{\theta}_s$ in the moderately fast case of $\Delta_p \ll \dot{\theta}_s \ll \Delta$ where we have suppressed A_z for notational simplicity. Nevertheless, upon strong electric field E_z such that the free energy potential $\cos 2\theta_s$ can be neglected, the equation of motion all reduces to $\dot{\theta}_s = d\dot{A}_z = -dE_z$, i.e., the electric field provides a force to rotate the phase θ_s so as to switch the system between the two ground states $\theta_s = \pm\pi/2$ [Fig. 2(d)]. The pulse that exactly delivers such a switch is $d\int E_z(t)dt = \pi$. For a pulse duration of 1 ps and $d = 1$ nm, the field needed is $E_z \sim 2 \times 10^4$ V/cm. For weaker fields such that the free energy potential matters, the dynamics depends on the time scale. In the case of $\Delta_p \ll \dot{\theta}_s \ll \Delta$, the equation of motion implied by Eq. (7) is simply

$$\dot{\theta}_s = \frac{\Delta_p^2}{4|\Delta|}\sin(2\theta_s) - dE_z. \tag{9}$$

The threshold field to climb over the potential barrier and switch the ground states is about $E_c \sim \Delta_p^2/|\Delta|d$, which reads $E_c \sim 10^4$ V/cm for $\Delta_p = 10$ meV, $|\Delta| = 100$ meV, and $d = 1$ nm.

Discussion.—The bilayer could be realized by, e.g., gating a suitably stacked phosphorene bilayer [74–76] or transition metal dichalcogenide bilayers [11,12] to bring the conduction band of one layer and valence band (different in symmetry under C_2 or C_3 rotations around z, respectively) of the other layer closer in energy, entering the EI phase (see Ref. [57], Sec. ID). The 3D and 2D stacks may be either natural crystals such as monolayer WTe_2 (a 2D stack of chains) [77–79] or artificial structures.

Realizations of these topological excitonic insulators [79–84] are an important research direction.

The order parameter steering also applies to the EI candidate Ta_2NiSe_5 [29,43–47]. Its basic structural unit is the Ta-Ni-Ta chain, with Ta-derived conduction band states even under reflection $\sigma_\perp: x \to -x$, while the Ni-derived valence band states are odd [29,43]. The EI state breaks $\sigma_\perp$, and while the detailed electronic structure complicates the discussion of the Josephson effect, the phase dynamics is still described by Eqs. (4) and (9), and a photon pulse perpendicular to the chains can still switch the system between its two ground states (see Ref. [57], Sec. IV). This may have already been observed [47].

Fluctuations will not destroy our qualitative conclusions. Without the $U(1)$ breaking Josephson term $\cos 2\theta$, the exciton condensate in Eq. (2) has quasi long range order at temperatures T below the Berezinskii-Kosterlitz-Thouless (BKT) temperature T_{BKT} [85,86]. According to renormalization group analysis [87], the Josephson coupling is a relevant one at $T < T_{\mathrm{BKT}}$ that renders the EI state strictly long range ordered. However, the coupling (and the Josephson current) is renormalized by fluctuations to a power $1/[1-(T/4T_{\mathrm{BKT}})]$ of its bare value (see Ref. [57], Sec. IA).

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