True exciton condensate of one-dimensional electrons through interwire tunneling

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(Dated: September 30, 2016)

We theoretically predict that a true bilayer exciton condensate, characterised by off-diagonal long range order and global phase coherence, can be created in one-dimensional solid state electron systems. The mechanism by which this happens is to allow for a single particle hybridization of electron and hole populations, which breaks the continuous symmetry associated with particle number conservation in each individual layer and hence invalidates the Mermin-Wagner theorem. Electron–hole interactions then amplify this tendency towards off-diagonal long range order, enhancing the condensate properties by more than an order of magnitude over the non-interacting limit. The absence of screening in one-dimensional systems means that the predicted critical temperature associated with the formation of a substantial condensate fraction will be in the range of hundreds of Kelvin.

Excitons are composite bosons formed from paired electrons and holes. They can be produced either by optical pumping of carriers from the valence band into the conduction band, or by positioning two independent populations of electrons and holes in their respective ground states (GS) close to each other, so that the mutual Coulomb interaction gives rise to a many-body instability, driving the system into a new excitonic GS. We call this latter type “bilayer excitons”. Under certain conditions, these composite bosons may condense into a Bose-Einstein condensate characterised by off-diagonal long range order and a global coherent phase. Such a condensate has been observed for optically pumped excitons \cite{1}, and bilayer excitons in the quantum Hall regime \cite{2}. In zero magnetic field, it is proposed that bilayer condensates may have many applications in devices, including ultra-low power transistors \cite{3}, and heat exchangers \cite{4}.

However, a condensate of bilayer excitons in zero magnetic field has never been observed in an experiment \cite{5–9}. There are several possible reasons for this. One is that the critical temperature associated with the many-body instability is simply too low, due to strong screening of the inter-layer Coulomb interaction \cite{10}. Another is that charged impurity disorder has a major effect in two-dimensional materials, as it destroys the nesting of the Fermi surfaces required for the electron–hole pairing to occur \cite{11,12}. Both of these possibilities could be mitigated by producing an exciton condensate in one-dimensional (1D) bilayers, using e.g. two parallel nanowires \cite{13}. Screening is known to be less strong in 1D systems \cite{14,15}, implying that the inter-layer interaction would be more effective in 1D. Robustness against disorder is set by the size of the order parameter, so the absence of screening enhances the stability of the condensate in this way as well. Finally, if devices utilizing these advantages were to be fabricated, such 1D implementations would be smaller and allow more options for design.

A major obstacle to any condensate of quantum particles in 1D is the Mermin-Wagner (MW) theorem. This prohibits off-diagonal long range order (ODLRO) by spontaneous breaking of a continuous symmetry due to the enhancement of quantum fluctuations in 1D \cite{16}. In this work, we show that for 1D bilayer excitons, a very weak single particle tunneling between the two layers can lead to a true exciton condensate (EC) with ODLRO as the tunneling \textit{explicitly} breaks the continuous number conservation symmetry and thus the MW theorem no longer applies. Electron–hole attractions can then strongly feed into this small tendency towards ODLRO, resulting in large enhancements of all properties of the EC, which is a true many body condensate. The EC ground state is characterized by one large and one small excitation gap, both of which can be probed experimentally.

To demonstrate this, we employ highly accurate density-matrix renormalization group (DMRG) numerics to compute the ground and thermal state of the many-body system. We show that the smaller gap sets the temperature scale on which crossover to an EC near the ground state occurs. We also describe experimental probes of the condensate by determining the nonlinear DC current-voltage characteristic of an interlayer transport measurement, and compute the density of states that would be probed in an STM experiment. Finally, we compute the ground states for systems with realistic length- and energy-scales and show that the EC can be realized at high temperatures after accounting for long range electron–electron interactions.

We consider a generic setup, two parallel quasi-one-dimensional electron systems (“wires”, hence), shown schematically in Fig. 1a. Gates shift the wire bands such that the minimum of the conduction band for the upper wire is below the maximum of the conduction band for the hole-like lower wire. Weak interwire tunnelling $t_{\perp}$ results in a joint chemical potential and, in the absence
of interactions, the opening of a small single electron gap $\delta_{sp} = 2t_\perp$ at the avoided level-crossing (Fig. 1(b)). Including interactions, and considering a 1D space with $2M$ lattice points ($M$ points for each wire - these can correspond either to real physical atoms in a 1D lattice or a discretized continuous 1D space) to be compatible with DMRG from the beginning, this system is described by the Hamiltonian

$$\hat{H} = \hat{H}_u + \hat{H}_t + \hat{H}_\mu + \hat{H}_{IWI} + \hat{H}_{IWT}$$

(1)

with the individual terms

$$\hat{H}_u = -\sum_{x=1}^M t_w (\hat{c}^\dagger_{xw} \hat{c}_{x+1w} + h.c)$$

$$+ \sum_{x,y=1}^M U_w(|x-y|)\hat{n}_{xw}\hat{n}_{yw},$$

(2)

and

$$\hat{H}_\mu = \frac{1}{2} \sum_{x=1}^M \mu_{\text{diff}}(\hat{n}_{xy} - \hat{n}_{xt}).$$

(3)

Here, the wire-index is $w \in \{u,l\}$, $\hat{c}_{xw}$, $\hat{c}^\dagger_{xw}$ are electron field annihilation and creation operators at site $x$ in wire $w$, respectively, $\hat{n}_{xw} = \hat{c}^\dagger_{xw} \hat{c}_{xw}$, and $U_w = U_l$ is intrawire electron–electron interaction strength. The opposite band curvatures imply $t_u = -t_l \geq t > 0$, and the chemical potential difference $\mu_{\text{diff}}$ is used to tune the electron densities inside each wire. The interwire (IW) terms are

$$\hat{H}_{IWI} = \sum_{x,y=1}^M U_{ul}(|x-y|)\hat{n}_{xu}\hat{n}_{yl},$$

(4)

$$\hat{H}_{IWT} = -t_\perp \sum_{x=1}^M (\hat{c}^\dagger_{xu} \hat{c}_{xl} + h.c).$$

(5)

where $U_{ul}$ is the IW interaction strength. To simplify the analysis and keep the required computational effort under control, we restrict the setup to spinless electrons, as could be achieved, for example, by the use of an external magnetic field.

It is the IW-tunneling that enables exciton condensation in 1D. A particle-hole transformation for the hole-wire shows $\hat{H}_{IWT}$ serving as bias field for electron–hole pairs. Without interactions, the ODLRO due to $\hat{H}_{IWT}$ is a trivial single particle effect brought on by the opening of the single-particle gap $\delta_{sp}$. In the following, we show that the interwire repulsion between electrons $\hat{H}_{IWI}$ can feed strongly into this tendency towards ODLRO and lead to a truly many body EC, with a massive enhancement of EC properties such as the temperature below which the system is close to the EC ground state, as well as the response to applying interwire current and voltage. To accomplish this requires the use of DMRG (the gold standard for simulating strongly correlated 1D systems [17]), to treat the effects of non-perturbative $U_{u,l}$ and $U_{ul}$. To exemplify the key features of the EC in 1D, we first treat a model system, where electrons have no intrawire and purely local interwire repulsion, i.e. $U_{ww} = U_{ll} = 0$ and $U_{ul}(|x-y|) = U_\perp \delta_{xy}$. Afterwards, we show that in the presence of strong and long-ranged $U_{u,l}(|x-y|)$, a non-trivial and measurable EC still forms.

For the model system we calculate the ground states of $\hat{H}$ and their exciton correlations $C_{\text{ex}}(x) = \langle \hat{c}^\dagger_{(x_u),(y_l)} \hat{c}^\dagger_{(x_l),(y_u)} \rangle$ for a grid of values for $t_\perp$ and $U_{\perp}$ while fixing the density in the electron wire at 10%. In Fig. 2, we plot $C_{\text{ex}}(x)$ for $U_{\perp} = 2t$, and this is the central result of this Letter. The ODLRO is characterised by the exciton correlator settling at a finite value at long distances. It is clear that when $t_\perp = 0$, this does not happen. In fact, the decay here goes with a power law $C_{\text{ex}}(x) \propto x^{-K_u-1/K}$, as predicted by bosonization and MW (see the Supplementary Materials, and Ref. [18]). In contrast, when $t_\perp \neq 0$ the exciton correlator remains finite at large $x$ indicating the presence of ODLRO and a stable EC. Decreasing $t_{\perp}$ by an order of magnitude only halves the strength of the ODLRO. We also observe the equivalent of the pen-
FIG. 2. (a) Spectral function of $G_w^R(x, \omega)$ model system with $t_\perp = 0.001t$, $U_\perp = 2t$, exhibiting the large gap $\Delta$. (b) Scaling of $\Delta$ with $t_\perp$, for $U_\perp = 0.25t$ (dark blue), $U_\perp = 0.5t$ (bright red), $U_\perp = 0.75$ (yellow), $U_\perp = t$ (violet) $U_\perp = 1.25t$ (green), $U_\perp = 1.5t$ (light blue), $U_\perp = 2t$ (dark red) for the model system. (c) Spectral function of $\chi_{J_\perp}(\omega)$ for the model system with $t_\perp = 0.001t$, for $U_\perp = 2t$ (blue line), $U_\perp = 1.5t$ (green dotted), $U_\perp = t$ (red dash-dotted), and $U_\perp = 0$ (black dashed), which has only $\delta_{sp} = 0.002t$. Arrows indicate position of peak at $\omega = \delta$. (d) Scaling of $\delta$ with $t_\perp$, for $U_\perp = 0.001t$ (dark blue), $t_\perp = 0.0025t$ (bright red), $t_\perp = 0.005t$ (yellow), $t_\perp = 0.01t$ (violet) $t_\perp = 0.025t$ (green), $t_\perp = 0.05t$ (light blue), $t_\perp = 0.1t$ (dark red) for the model system. (e) Dependence of real-space EC order parameter $A$, as a fraction of its ground state value, on inverse temperature $\beta$ for the model system with $U = 2t$, $t_\perp = 0.01t$. Once $\beta > 1/\delta$, the system approaches ground state properties exponentially fast in $\beta$. (f) DC I-V characteristic of a model system with $t = 1eV$, $U_\perp = 2t$, $t_\perp = 0.01t$ (blue) and $t_\perp = 0.001t$ (red), showing both dissipationless and dissipative regimes (see text).

The real space order parameter $A = \langle c_{0u}^\dagger c_{0l} \rangle$ of the EC is boosted over the value for free fermions which is set entirely by $t_\perp$. This order parameter also quantifies the ODLRO, since $C_{xx}(x) \rightarrow A^2$ when $x \rightarrow \infty$.

This enhancement and dominance of the 1D EC physics by the IW interaction $U_\perp$ is seen in experimental observables. Fundamentally, the 1D EC is characterised not by one gap, but by two, which we label $\delta$ and $\Delta$. With $t_\perp$ being by far the smallest parameter, generally $\delta \ll \Delta$ holds. The large gap $\Delta$ can be measured using a scanning tunneling microscopy experiment which probes the retarded Greens function

$$G_w^R(x, \omega) = \langle c_{xw}^\dagger(\omega - \hat{H} + E_{GS} + i\eta)^{-1}c_{xw} \rangle + \langle c_{xw}^\dagger(\omega + \hat{H} - E_{GS} + i\eta)^{-1}c_{xw} \rangle. \quad (6)$$

Perturbative weak coupling renormalization group (RG) analysis predicts $\Delta \propto U_\perp^{1/(2-2K_u)}$ (see the Supplementary Material and Ref. [18]). Numerically we find that $\Delta$ actually appears to cross over from one power law scaling in $U_\perp$ to another one as $U_\perp$ increases, with the crossover region depending weakly on $t_\perp$. Crucially, the numerics also give access to the dependence of $\Delta$ on $t_\perp$ (which perturbative RG does not) and this is shown in Fig. 2b. This reveals two regimes for the physics of the 1D EC.

One, at very small $t_\perp/t$, has $\Delta$ almost independent of $t_\perp$. This is the regime where the physics is almost completely dominated by electron–hole interactions and this is the cleanest form of a 1D many body EC. The other regime, into which the first one crosses over once $t_\perp/t > 0.005$ is marked by significant $t_\perp$ dependence of $\Delta$ and a noticeable decrease of order parameter ratio in Fig. 1d (which still remains large at large $U_\perp/t$ however).

The large gap $\Delta$ is present even when $t_\perp = 0$ and there is no EC. On the other hand, the small gap $\delta$ is different. This appears as the first peak of the spectral function of the interwire current susceptibility

$$\chi_{J_\perp}(\omega) = \langle \hat{J}_\perp(\omega - \hat{H} + E_{GS} + i\eta)^{-1}\hat{J}_\perp \rangle, \quad (7)$$

as shown in Fig. 2c, and could be probed via optical conductivity measurements. Here, $\hat{J}_\perp = \frac{1}{M} \sum_{x=1}^M \langle \hat{e}_{xu}^\dagger \hat{e}_{xl} - \text{h.c.} \rangle$ is the discretized operator for interlayer current. This gap naturally only appears when $t_\perp \neq 0$ and is key for establishing the EC. As a consequence we observe that the EC order parameter $A \propto |\text{Im}[\chi_{J_\perp}(\delta)]|^\gamma$ once $U_\perp$ becomes the dominant energy scale, where $\gamma$ is independent of $t_\perp$. Another key significance of $\delta$ is that it sets the temperature scale beyond which the 1D excitons will be essentially in the EC ground state, as shown in Fig. 2c. Using the auxiliary space approach of DMRG [17] we compute the thermal state $\text{Tr}(e^{-\beta \hat{H}})$ of the model system and find that all quantities connected to ODLRO of the 1D EC become exponentially close to the ground state value as soon as $\beta$ increases above $\delta^{-1}$. We stress that there is strictly
speaking no phase transition from an EC to a non-EC state as $\beta$ decreases (c.f. Fig. 2f). Instead, the presence of any finite IW tunneling means that technically there is ODLRO at any $\beta$, but it may just be too small to measure. In this the 1D EC behaves analogously to a magnetic material inside a weak external magnetic field.

We have calculated $\chi_{\perp}(\omega)$ in the real-frequency domain (using the GMRES approach within DMRG [19]) on the isolated system. In this way, we cannot see DC interlayer current in response to applying $\hat{J}_\perp$ at $\omega = 0$, as this would require an external bath to dissipate energy. Instead, we show the existence of non-dissipative DC interlayer current at small voltages, which is the hallmark property for technological applications of an EC, and which is a direct consequence of the EC state. At higher voltage, this interlayer current becomes dissipative. Both regimes are captured in Fig. 2f, where we considered the rate of macroscopic tunneling $\Gamma = eV/2\pi$ ($V$: the interlayer voltage) from the original to the new ground state when $I\hat{J}_\perp$ is added to $\hat{H}$. We do this by computing the decay of occupation from the original ground state through calculation of the imaginary time Green’s function $\langle GS|e^{-\tau(\hat{H}+\hat{T}_\perp)}|GS\rangle \propto e^{-\Gamma\tau}$ using time dependent DMRG. The result agrees very well with the qualitative prediction of the singular relationship $I \propto -(\log V)^{-1}$, as Fig. 2f depicts.

Going beyond the model system to study the degree to which realistic systems of 1D electrons can enter the 1D EC state requires consideration of long range interactions between electrons. This means studying a screened Coulomb repulsion in 3D coordinates $\mathbf{x}$,

$$U(|\mathbf{x} - \mathbf{x}'|) = \frac{e^{-|\mathbf{x} - \mathbf{x}'|/\Lambda}}{4\pi \varepsilon_{\text{eff}} |\mathbf{x} - \mathbf{x}'|},$$

(8)

where $\varepsilon_{\text{eff}}$ denotes the effective dielectric screening in between $\mathbf{x}$ and $\mathbf{x}'$ and $\Lambda$ the effective screening range. The ratio of intra- to interlayer repulsion can be boosted (to compensate for the larger interlayer distances compared to intralayer ones) to aid pairing of electrons and holes by choosing a strong dielectric $\varepsilon_{\text{sub}}$ for the substrate and a weak one $\varepsilon_{\text{sp}}$ for the spacer (c.f. Fig. 1f) and is a trick that is generic to any bilayer setup. The 1D system has a unique advantage in that there is no intrinsic screening and so all screening is derived from the environment (for example, from the back gates) and could thus be tuned [14, 15]. Moreover, in 1D, the IW interactions will be much more effective at forming electron–hole bound states than in higher dimensional systems. Picking parameters $t = 0.25eV$, $\varepsilon_{\text{sub}} = 16\varepsilon_0$, $\varepsilon_{\text{sp}} = \varepsilon_0$ (i.e. a vacuum spacer), $\Lambda = 0.48nm$, IW distance $d_{IW} = 1nm$, and a distance between lattice points equal to the carbon–carbon bond length in graphene, 0.142nm, we find that each individual wire has low energy, long wavelength properties characterized by a Tomonaga-Luttinger liquid parameter $K = 0.66$ due to the intrawire interactions being strong and long ranged. For comparison, the model system had $K = 1$. To further demonstrate the many body nature of the resulting 1D EC we compare with the benchmark in which we switch off $\hat{H}_{IW}$ while keeping the same $t_{\perp}$. We find that for these strongly interacting systems, where in fact $U_{ul} = U_l > U_{ul}$ at short distances, $\delta$ is now almost entirely set by $U_{ul} = U_l$ and depends only very weakly on $U_{ul}$ and $t_{\perp}$. This is shown in Fig. 3a and b. At the same time $\delta$ is strongly boosted by the intrawire interactions, reaching $\delta \approx 236K$, and therefore experimental verification of our prediction is highly likely. That this system realizes that is evidenced not only by the strong boost of $\chi_{\perp}$ over the “benchmark” case with $d_{IW} = 10nm$ (factor of 20) but also of $A$ (factor of 6).

Our DMRG simulations for other values of $\varepsilon_{\text{sub}}$, $\varepsilon_{\text{sp}}$, $t$ and $d_{IW}$ show that in trying to find optimal parameter regimes for an EC there is a need to balance $U_{ul}$ and $U_{ul}$ against each other. While it may be tempting to boost $U_{ul}$ over $U_{ul}$ by increasing the ratio $\varepsilon_{\text{sub}}/\varepsilon_{\text{sp}}$, the long ranged nature of $U_{ul}$ in conjunction with the interwire tunneling can quickly cause electrons to all crowd into one wire to rapid the interaction energy. For this reason, large ratios of $U_{ul}/U_{ul}$ are only possible if $U_{ul} < t$ at the same time.

In conclusion, we have demonstrated that a single particle tunneling between spinless electron and hole wires removes the restrictions of MW and allows for a true bilayer condensate in one dimension characterised by off-diagonal long range order and global phase coherence. Using higher-performing DMRG, it would be possible to introduce spinful electrons and go beyond the single chain model of 1D ECs. Recently developed massively-parallelized DMRG would even be capable of performing analogous calculations directly on a full physical system, such as narrow carbon nanoribbons, for a fully quantitative ab-initio analysis of their potential to support 1D ECs.

We thank Nordita for support. A.K. thanks Thierry Giamarchi for helpful discussions. D.S.L.A. thanks ERC...
project DM-321031 for financial support.

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SUPPLEMENTARY MATERIAL

Though treating $\hat{H}_{\text{HWT}}$ and $\hat{H}_{\text{HWT}}$ perturbatively will not address physically relevant systems, together with the associated bosonization framework [16] it has some use for interpreting the numerical results at strong coupling. After a particle–hole transformation $\hat{c}_{\pm} \rightarrow (-1)^{x} \hat{c}_{\pm}$ on $\hat{H}_0$, we bosonize $\hat{H}_0 + \hat{H}_1$, by retaining only the long wavelength excitations around the Fermi points $\pm k_F$ in both wires, approximating $\hat{c}_{\pm w} \approx U R e^{i(k_F x - \phi_w(x) + \theta_w(x))} + U L e^{-i(k_F x - \phi_w(x) - \theta_w(x))}$, where $\phi_w(x), \phi_{\pm w}(x)$ are conjugate field operators, $\phi_w(x), \partial_x \phi_{\pm w}(x)$ encode the long wavelength density and phase fluctuations in wire $w$ respectively and $U_L, U_R$ are the Klein factors that preserve anticommutation relations. Thus, $\hat{H}_{u,l}$ becomes quadratic in $\partial_x \phi_{u,l}(x), \partial_x \phi_{\pm u,l}(x)$ and its long wavelength properties are parametrized by just two numbers, the Tomonaga-Luttinger liquid (TLL) parameters $v_{u,l}$ and $K_{u,l}$. The TLL parameters $K_{u,l}$ encode the strength and range of $U_{u,l}$ respectively and if $U_{u,l} = 0$, then $K_{u,l} = 1$. The stronger and more long ranged $U_{u,l}$, the further below 1 the value of $K_{u,l}$ will drop. But while isolated 1D electrons have no intrinsic screening (and thus $K = 0$), 1D wires in an environment receive screening from the outside, resulting in $0 < K < 1$. Since we assume $|t_u| = |t_l|, U_u = U_l$, and $k_F$ being the same for both wires, we have $K_u = K_l = K$ in the following. Now adding $\hat{H}_{\text{HWT}}$ as perturbation to $\hat{H}_0 + \hat{H}_1$, its bosonized form in momentum space decomposes into a forward scattering part, with terms proportional to $U_{u,l}^2 = U_u(q = 0)$ and a backscattering contribution proportional to $U_{u,l}^2 = U_u(q = 2k_F)$. The forward scattering term can be incorporated into $\hat{H}_n + \hat{H}_1$ exactly, at the price of a canonical transformation to the symmetric and antisymmetric modes of the two wires $\phi_{\pm u,l} = (\phi_u + \phi_l)/\sqrt{2}, \theta_{\pm u,l} = (\theta_u - \theta_l)/\sqrt{2}$. This results in $\hat{H}_n + \hat{H}_1 \rightarrow \hat{H}_n + \hat{H}_1$, where $\hat{H}_n$ are again TLL Hamiltonians, and in perturbation theory their TLL parameters are $K_{\pm u,l} = (K - 2) \mp U_{u,l}^2 m_u/(2h k_F K)^{1/2}$. However, the backscattering part can at best be treated using the perturbative renormalization group (pRG), and the same holds for the bosonized version of $\hat{H}_{\text{HWT}}$. As in Ref. [15], to second order of momentum space pRG this yields

$$\frac{dU_{u,l}^2}{ds} = 2(1 - K_u) U_{u,l}^2, \quad \frac{dt_{u,l}}{ds} = \frac{(4 - K_u - K_l)}{2} t_{u,l}$$

(9)
Thus, both $\hat{H}_{IW1}$ and $\hat{H}_{IW-T}$ are relevant perturbations for a very wide range of parameters (e.g. $\hat{H}_{IW1}$ is so for any repulsive $U_{ul}$), and their associated couplings both flow to nonperturbative values, outside the range of any pRG. The validity of Eq. (9) is constrained further because the $U_{ul}^{R}$ may flow to its fixed point before $t_{\perp}$, locking $\hat{\phi}_{\alpha}$ to a fixed value and making the pRG equation for $t_{\perp}$ obsolete. If, against these objections, a straight extrapolation of Eq. (9) is performed, it would predict one gap $\Delta$ for fluctuations of $\hat{\phi}_{\alpha}$, with scaling $\Delta \sim (U_{ul}^{B})^{2/(4-K_{a}+K_{a}^{-1})}$, and another, $\delta$, for fluctuations of $\hat{\theta}_{s}$, with scaling $\delta \sim t_{\perp}^{1/2(1-K_{a})}$, the most relevant bosonized operators inside $\hat{H}_{IW1}$ and $\hat{H}_{IW-T}$ being compatible.