

Evidence from Quantum Monte Carlo Simulations of Large-Gap Superfluidity and BCS-BEC Crossover in Double Electron-Hole Layers

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We report quantum Monte Carlo evidence of the existence of large gap superfluidity in electron-hole double layers over wide density ranges. The superfluid parameters evolve from normal state to BEC with decreasing density, with the BCS state restricted to a tiny range of densities due to the strong screening of Coulomb interactions, which causes the gap to rapidly become large near the onset of superfluidity. The superfluid properties exhibit similarities to ultracold fermions and iron-based superconductors, suggesting an underlying universal behavior of BCS-BEC crossovers in pairing systems.

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There are intense ongoing experimental efforts to observe superfluidity in electron-hole double layer systems, including double quantum wells in GaAs-AlGaAs heterostructures, double graphene monolayers, double graphene bilayers, and hybrid graphene-GaAs structures [1–5]. A recent very significant experimental advance by multiple groups has been the fabrication of closely spaced electron-hole double graphene bilayers with carrier densities tunable by metal gates [6–9]. Insertion of a few layers of hexagonal boron nitride between the bilayers creates an insulating barrier. This can be as thin as 1 nm while still blocking tunneling of carriers between the bilayers, permitting the electrons and holes to interact with a very strong Coulomb attraction.

Condensation of electron-hole pairs into a BEC superfluid state in double graphene bilayers has been experimentally demonstrated in the quantum Hall regime with a magnetic field [8,9], opening the way to the generation of quantum coherent macroscopic states in spatially separated two-dimensional sheets.

Room temperature superfluidity had in fact been predicted earlier in electron-hole double graphene monolayers [10], but it was later established that since a graphene monolayer remains blocked in the weakly coupled regime, strong screening always suppresses the superfluidity [11]. Double electron-hole graphene bilayers were subsequently proposed to overcome this problem [12], combining ideas for the realization of high- T_c superfluidity with the ability to move across the BCS-BEC crossover by changing the carrier densities using metal gates. This ability to tune the system into the strong-coupling regime is key to obtaining a superfluid in a solid-state device at experimentally accessible conditions [12], offering a fascinating alternative to

ultracold fermionic atoms for studying superfluid physics across the BCS-BEC crossover.

With superfluidity in cold atoms, quantum Monte Carlo (QMC) results [13–15] were from the outset integrated closely with experiments and theory to understand and control the phenomenon. QMC simulations are extremely useful in any strongly correlated condensed matter system where there is no small parameter that can be used in perturbative expansions or controllable diagrammatic approximations. For double layer electron-hole systems, QMC simulations [16,17] are of key importance since the superfluid phase arises from a complicated competition between the long-ranged Coulomb interlayer attraction and intralayer repulsion. Our work provides a comprehensive characterization of the superfluid properties of electron-hole double layers at zero temperature, and establishes the density range in which the condensate exists. Our predictions that too high a density kills superfluidity and of the existence of a BCS-BEC crossover regime at accessible intermediate densities will be extremely useful in guiding experiments. We use state-of-the-art QMC methods to determine the dependence of BCS-like effective parameters on the equal carrier densities, with the distance separating the layers fixed at the value that maximizes exciton formation [17–19].

The boundary between the crossover and BEC regimes is of great interest because the pseudogap transforms into a real gap associated with the molecular phase near this boundary [20]. The BCS-BEC crossover has been recently observed in the shallow Fermi surface pockets of iron-based superconductors [21,22].

The zero temperature BCS-BEC crossover [23,24] can be traced by following the evolution of the condensate fraction c , the average radius of the superfluid pairs r_{ex}

[25], or the superfluid gap Δ . Here we use explicit many-body wave-function-based methods to track the evolution of the superfluid with carrier density in symmetric electron-hole layers. We directly obtain total energies, condensate fractions, and pair radii, and we extract superfluid gaps and chemical potentials from total energies using BCS-like relations with effective parameters. We employ methods similar to those applied in Ref. [26] for coupled layers of dipolar fermions.

The condensate fraction c measures the fraction of carriers in the condensate [27–30]. In the BCS regime $c < 0.2$, with only a small fraction of the carriers near the Fermi surface forming the condensate, while in the BEC regime $c > 0.8$, so almost all the carriers form local molecular bosonic pairs, and condense. Using the pair radius r_{ex} , the BCS regime is characterized by $k_F r_{\text{ex}} \gg 1$, where k_F is the Fermi wave vector, while in the BEC regime the compact pairs correspond to $k_F r_{\text{ex}} < 1$. The parameter $k_F r_{\text{ex}}$ was the first to be studied in investigations of the BCS-BEC crossover in the high- T_c cuprate superconductors [31]. r_{ex} determines the correlation length of the pairs, which enters the expressions for the properties of the vortex state of the superfluid and all other quantities affected by the spatial structure of the superfluid wave function, such as Josephson and Andreev reflection effects [32].

In contrast to c and r_{ex} , the superfluid gap is experimentally straightforward to measure using angle-resolved photoemission spectroscopy (ARPES), scanning tunneling microscopy, or measurements of the specific heat. Knowledge of the evolution of the superfluid gap with the external parameters is of fundamental relevance in designing experiments to detect and characterize electron-hole superfluidity. It is possible to link the entry into the BCS-BEC crossover regime as determined by c and $k_F r_{\text{ex}}$ to $\Delta/E_F \sim 1$ at zero temperature [33]. In our calculations we also monitor the evolution of the pseudo-Luttinger wave vector k_{min} at which the k -dependent excitation energy passes through its minimum. k_{min} can be traced by ARPES, because it directly affects the shape of the remnant Fermi surface in the broken symmetry phase at zero temperature.

With our results for Δ , μ , c , and r_{ex} as functions of density, we are then in a position to follow the evolution of the system through the weak-coupling regime, the superfluid BCS-BEC crossover regime, and the BEC regime, enabling comparisons with predictions from various microscopic theories.

In our calculations we use excitonic Hartree units, $\hbar = |e| = m_e^* = \kappa 4\pi\epsilon_0 = 1$, where m_e^* is the effective electron mass and κ is the relative permittivity of the system, and we obtain energies in units of $\text{Ha}^* = (m_e^*/m_e)\kappa^{-2}\text{Ha}$ and distances in units of $a_0^* = \kappa(m_e/m_e^*)a_0$. For reference, the relative permittivity for bilayer graphene (BLG) encapsulated in few-layer hexagonal boron nitride (hBN) is $\kappa = 2$ [34], and the effective mass is $m_e^* = 0.04m_e$ [35].

We simulate a finite version of the paramagnetic, equal-mass electron-hole double layer system with parabolic single-particle energy dispersion using square simulation cells of area A subject to periodic boundary conditions, with N particles in each layer. The in-layer particle density is defined via the density parameter $r_s = \sqrt{A/(\pi N)}$. We run calculations for systems containing $N = 58$ electron-hole pairs; tests with systems of $N = 114$ electron-hole pairs show that finite-size errors are small in our results [36]. We use a fixed interlayer separation of $d/a_0^* = 0.4$, slightly greater than the largest d at which biexciton formation is favorable [17–19], and vary the density between $r_s/a_0^* = 1.75$ and 15.

At all densities considered we evaluate total energies, condensate fractions, and pair-correlation functions (PCFs) using the variational quantum Monte Carlo (VMC) method [38,39]. Wave function parameters can be optimized within the VMC method [40,41], and the accuracy of VMC expectation values depends on the quality of the resulting wave function. The more computationally costly diffusion quantum Monte Carlo (DMC) method [39,42] employs stochastic projection to extract the lowest-energy state compatible with the nodal surface of a VMC-optimized trial wave function. Once the time-step and population-control biases are eliminated [43–45], the accuracy of the DMC method depends only on the accuracy of the nodes of the trial wave function. We have performed DMC calculations at selected densities representative of the weak-coupling, crossover, and strong-coupling regimes. Thus, our DMC calculations serve as quantitative corrections to our VMC results throughout the density range considered.

We use trial wave functions of the form $\Psi = e^J D_{e\uparrow h\downarrow} D_{e\downarrow h\uparrow}$, where e^J is a Jastrow correlation factor [46,47], imposing the Kato cusp conditions [48], and $D_{e\uparrow h\downarrow}$ and $D_{e\downarrow h\uparrow}$ are pairing determinants [16,17]. In systems with an additional (up-spin) electron, we complete the corresponding determinant with a plane-wave orbital of wave vector \mathbf{k} . Details of our trial wave functions are given in the Supplemental Material [36]. We use the CASINO code for our calculations [49].

The main properties of the BCS-BEC crossover at low temperature are captured by BCS theory, as demonstrated with ultracold fermions. When the gap equation is coupled to the density equation, the excitation energy $\varepsilon(k)$ corresponding to the addition of an electron of wave vector \mathbf{k} follows the BCS dispersion relation [26],

$$\varepsilon(k) = \sqrt{(k^2/2m^* - \mu)^2 + \Delta^2}, \quad (1)$$

where μ , m^* , and Δ are the chemical potential, effective mass, and superfluid gap of the electron quasiparticle, respectively. $\varepsilon(k)$ therefore contains the parameters that characterize the superfluid state. This excitation energy can be obtained from *ab initio* total energies as

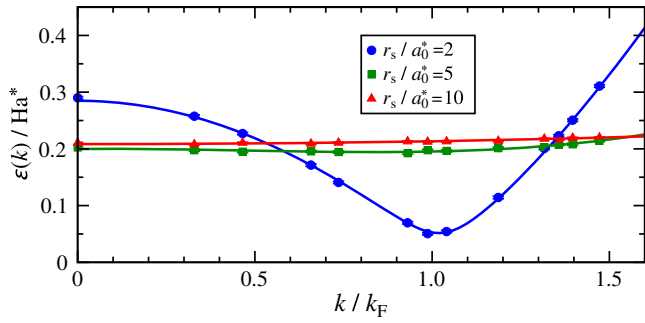


FIG. 1. DMC estimates of $\varepsilon(k)$ as a function of the magnitude k of the wave vector of the additional electron, at $r_s/a_0^* = 2, 5,$ and 10 . The solid lines are fits of the DMC data to Eq. (1).

$$\varepsilon(k) = E_A(N + 1/2; k) - E_A(N) - \mu_{\text{QMC}}(N), \quad (2)$$

where $E_A(N)$ is the energy of a system of area A containing N electron-hole pairs, $E_A(N + 1/2; k)$ is the energy of a system of area A containing N electron-hole pairs and one additional electron of associated wave vector \mathbf{k} , and $\mu_{\text{QMC}}(N) \approx \frac{1}{4}[E_A(N + 1) - E_A(N - 1)]$ is the chemical potential of the system. Note that μ_{QMC} differs from the mean-field μ due to many-body effects.

We simulate systems with $N, N + 1,$ and $N - 1$ electron-hole pairs, and systems with N electron-hole pairs and an unpaired electron at several wave vectors \mathbf{k} . We then compute $\varepsilon(k)$ at each \mathbf{k} using Eq. (2) and fit the resulting values to Eq. (1) with $m^*, \mu,$ and Δ as fitting parameters. Following tests [36], we use wave vectors such that $0 \leq k < k_{\text{cut}}$; we set $k_{\text{cut}}/k_F = 1.5$ at high densities and use larger cutoff values at low densities. Although the superfluid gap is expected to be k dependent [11,12], our calculations do not yield any significant variation of Δ with k in the ranges of k we have considered [36]. Figure 1 shows a plot of the DMC values of $\varepsilon(k)$ for densities of $r_s/a_0^* = 2, 5,$ and 10 , along with the resulting fits to Eq. (1). The fits follow the DMC data remarkably well, indicating that the BCS dispersion relation provides a robust description of the *ab initio* results.

In Fig. 2 the zero-temperature superfluid gap is reported as a function of r_s , both in excitonic Hartree units and relative to E_F . At high densities there is no superfluidity because the electron-hole pairing interaction is strongly screened [12]. Near the onset density, the electron-hole condensate is already close to the BCS-BEC crossover boundary. As r_s/a_0^* increases above 2 there is a very steep increase in the gap, which exceeds $\Delta/E_F \sim 1$ by $r_s/a_0^* \sim 3$. $\Delta/E_F > 1$ signals entry into the BCS-BEC crossover regime, so this occurs practically immediately after the onset of superfluidity. The steep rise in Δ is associated with strong screening at high densities [12]. Consequently, the weakly coupled BCS superfluidity regime, for which $\Delta/E_F \ll 1$, exists at most in a tiny range of densities.

Figure 3(a) shows μ as a function of r_s . μ becomes negative by $r_s/a_0^* \sim 6$, which signals entry into the BEC

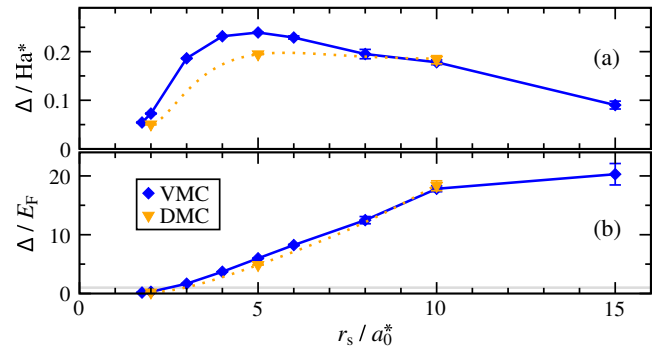


FIG. 2. Superfluid gap Δ , obtained by fitting the VMC and DMC data to Eq. (1), as a function of r_s , (a) in excitonic Hartree units, and (b) relative to E_F . The dotted lines interpolating the DMC results are intended as a guide to the eye.

regime. In Figs. 3(b) and 3(c) we plot the location of the minimum of $\varepsilon(k)$, $k_{\text{min}} = \text{argmin}_k \varepsilon(k)$, and m^* as functions of r_s . The value of k_{min}/k_F tracks the collapse of the Fermi surface, going from unity in the weak-coupling regime to zero in the BEC regime. The DMC results suggest that the Fermi surface fully collapses at a somewhat lower density than predicted by VMC.

In Fig. 3(c) the quasiparticle mass m^* has a minimum of less than the effective electron mass m_e^* near where the superfluid gap is maximal, $r_s/a_0^* \sim 5$. This is indicative of the interplay between the intralayer repulsion and the interlayer attraction, leading to quasiparticles in the superfluid state with masses $m^* < m_e^*$ for intermediate r_s . This behavior of m^* differs from theoretical and experimental findings in ultracold fermions in two dimensions, where the interaction is purely attractive. There, $m^* \gtrsim m_e^*$ always, and it varies monotonically with r_s . The quasiparticle mass of the two-dimensional electron gas is extensively discussed in Ref. [50]. Experimental measurements indicate a regime of small $r_s/a_0^* < 3$ in which the quasiparticle mass is smaller than the effective electron mass, see Fig. 4 of

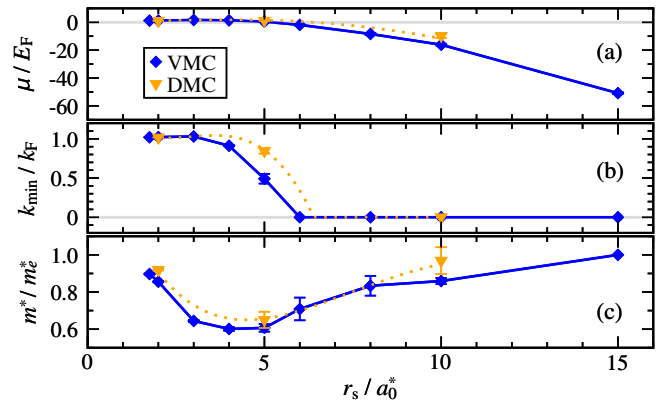


FIG. 3. (a) μ , (b) k_{min} , and (c) m^* , obtained by fitting the VMC and DMC data to Eq. (1), as functions of r_s . The dotted lines interpolating the DMC results are intended as a guide to the eye.

Ref. [50]. Thus, competition between intralayer repulsion and interlayer attraction in the electron-hole double layer can lead to small $m^* < m_e^*$, as we find.

The boundaries between the BCS, BCS-BEC crossover, and BEC regimes can be determined from the condensate fraction c , which is defined as

$$c = (A^2/N) \lim_{r \rightarrow \infty} \rho_{eh}^{(2)}(r), \quad (3)$$

where $\rho_{eh}^{(2)}(r)$ is the translational-rotational average of the two-body density matrix for electron-hole pairs [36]. We have evaluated c using the estimator of Ref. [51] which removes one-body contributions to ease extrapolation to the $r \rightarrow \infty$ limit. The results for c shown in Fig. 4(a) are consistent with the conclusions drawn from the behavior of μ . The condensate fraction is negligible for $r_s/a_0^* \lesssim 1.5$ [17]. As r_s/a_0^* increases, c grows rapidly to ~ 0.2 by $r_s/a_0^* = 2$, signaling entry into the BCS-BEC crossover regime. As r_s/a_0^* is further increased, c increases substantially and by $r_s/a_0^* = 8$ it exceeds $c = 0.8$, thus entering the BEC regime.

We also compute the translational-rotational average of the electron-hole PCF $g_{eh}(r)$ [36], which allows us to evaluate the exciton radius r_{ex} as

$$r_{ex}^2 = \int_0^{r_1} r^2 g_{eh}(r) r_s^{-2} 2\pi r dr, \quad (4)$$

where r_1 is the radius of a circle centred on a hole containing on average one electron. Figure 4(b) shows the pair radius r_{ex} for the condensate. As r_s increases, r_{ex} converges to the isolated-exciton limit, which for $d/a_0^* = 0.4$ is $r_{ex}/a_0^* = 2.002$. The values of $k_F r_{ex}$ are always less than unity, indicating pair sizes of the order of, or smaller than, the interparticle distance, confirming that the superfluidity is always in the strongly coupled crossover or BEC regimes.

In contrast with the short-range interactions typical of ultracold fermions and superconductivity in general,

electron-hole superfluidity should be affected by screening because of the long-range nature of the Coulomb pairing attraction. The nature and effectiveness of this screening has been a source of controversy in the past, with predictions from mean-field calculations ranging from negligible screening resulting in room-temperature superfluidity [10,11,52,53], to full screening by the normal state which would essentially completely suppress superfluidity [54]. Reference [55] compared the dramatically different mean-field predictions for the density dependence of the condensate fraction with the QMC values of the condensate fraction for the same system [17]. The conclusion was that the best mean-field approximation for screening was self-consistent screening by the superfluid state introduced by Lozovik [11] and applied in Ref. [12] to double bilayer graphene. Using a similar argument, Ref. [55] concluded that the vertex corrections and intralayer correlations in the superfluid state make relatively small contributions. Our present results for the superfluid gap, Fig. 2(a), are in good agreement with Fig. 3 of Ref. [55]. Our results are thus consistent with and further validate the conclusion of Ref. [55].

The trends of our QMC results are consistent with the complete suppression of superfluidity at high densities predicted in Ref. [12]. After the onset of superfluidity, $r_s/a_0^* = 1.5$, the system is in the weak-coupled BCS regime, but the condensate fraction rises rapidly and by $r_s/a_0^* = 2$ it reaches $c = 0.2$. By $r_s/a_0^* = 2$ the superfluid gap has reached $\sim E_F$. Thus the BCS regime, for which $\Delta/E_F \ll 1$, is restricted to the very small density range $1.5 < r_s/a_0^* < 2$. This confirms the effects of the highly nontrivial competition between Coulomb screening, which tends to suppress electron-hole pairing induced by the interlayer Coulomb attraction, and the opening of a large superfluid gap, which suppresses the particle-hole processes near the Fermi surface responsible for screening, thus severely weakening the screening.

When r_s is further increased, the superfluid gap first increases and then reaches a flat maximum around $r_s/a_0^* \sim 5-6$ with a very large value $\Delta/\text{Ha}^* \sim 0.2$. The large gap indicates that the superfluidity is robust with a high transition temperature. The chemical potential is still positive at this density, $\mu/E_F \sim 0.5$, the condensate fraction is ~ 0.7 , and $k_{\min}/k_F \sim 0.5$. Thus, the system retains its fermionic properties with a Fermi surface present but smeared out by the large gap, $\Delta/E_F \sim 7$.

The BEC superfluid regime is reached at larger $r_s/a_0^* \sim 8$, where the condensate fraction acquires values $c > 0.8$. In this regime μ/E_F is large and negative, the ratio $\Delta/E_F > 10$ is very large, the Fermi surface has completely collapsed, and the average pair size approaches the radius of an isolated exciton. The electron-hole superfluid can then be regarded as an ensemble of well-formed electron-hole dipoles, which are indirect excitons. The excitons will behave as a two-dimensional bosonic gas with a repulsive

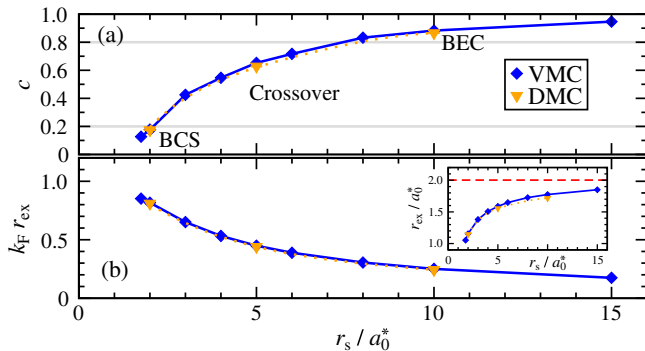


FIG. 4. VMC and DMC values of (a) c and (b) r_{ex} as functions of r_s . The inset in the bottom panel shows r_{ex} in excitonic Hartree units. The dotted lines interpolating the DMC results are intended as a guide to the eye.

interaction, with a Kosterlitz-Thouless transition [56] governing the critical temperature for superfluidity. Thus in the BEC regime the critical temperature should diminish with decreasing density.

We can thus conclude that the optimal density for experimental realization of the electron-hole condensate is around $r_s/a_0^* \sim 5$, which is deep inside the BCS-BEC crossover regime with large values of Δ/E_F . For BLG encapsulated in hBN, $r_s/a_0^* \sim 5$ corresponds to a density of $2 \times 10^{11} \text{ cm}^{-2}$, the maximum gap corresponds to $\Delta \sim 54 \text{ meV} \sim 630 \text{ K}$, and the interlayer distance $d/a_0^* = 0.4$ corresponds to 1 nm.

Our QMC results are consistent with a universal behavior of materials in the BCS, BCS-BEC, and BEC regimes not depending on the details of the microscopic interactions, and thus they point to a very general physics. The ground state properties and their evolution with coupling strength appear to be universal for (i) long-range Coulomb interactions, (ii) contact interactions in fermions [20], and (iii) spin fluctuations and phonons in iron-based superconductors [21,22,33].

Our results confirm that, unlike for fermionic superfluids with short-range pairing interactions, the BCS regime in Coulomb systems with their long-range interactions and screening is restricted to a very small range of densities. This is due to competition between screening and the superfluid gap [11,12], with strong screening suppressing the small-gap BCS regime in Coulomb systems. At high densities, the onset of superfluidity is delayed by screening, so that when the onset density is eventually reached, the pairs are relatively compact, and the superfluid gap, which rapidly becomes large both in absolute value and relative to E_F , will strongly suppress screening. Thus the system almost immediately enters the strong-coupling BCS-BEC crossover regime. For this reason the superfluidity is likely to be robust against potential detrimental effects like disorder, density imbalance, and low dimensional fluctuations, and we expect the largest gaps and highest critical temperatures not to be far from the onset density.

Supporting research data may be freely accessed by following the link in Ref. [57].

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Note added in proof.—We have become aware of a recent experimental report of the observation of electron-hole condensation in this system [58]. The density range is in extremely good agreement with our predictions.

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