Direct observation of nonlocal fermion pairing in an attractive Fermi-Hubbard gas

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The Hubbard model of attractively interacting fermions provides a paradigmatic setting for fermion pairing. It features a crossover between Bose-Einstein condensation of tightly bound pairs and Bardeen-Cooper-Schrieffer superfluidity of long-range Cooper pairs, and a "pseudo-gap" region where pairs form above the superfluid critical temperature. We directly observe the nonlocal nature of fermion pairing in a Hubbard lattice gas, using spin- and density-resolved imaging of ~1000 fermionic potassium-40 atoms under a bilayer microscope. Complete fermion pairing is revealed by the vanishing of global spin fluctuations with increasing attraction. In the strongly correlated regime, the fermion pair size is found to be on the order of the average interparticle spacing. Our study informs theories of pseudo-gap behavior in strongly correlated fermion systems.

Long-range Cooper pairs form in a Fermi gas for even the weakest attraction between fermions. With increasing interaction, fermion pairs become more tightly bound, as the system undergoes a smooth crossover from Bardeen-Cooper-Schrieffer (BCS) superfluidity toward a Bose-Einstein condensate (BEC) of molecular pairs (I-3). In the BCS limit, pair formation and the onset of superfluidity occur at the same temperature, but in the crossover, pairs are expected to form at temperatures above the critical temperature \( T_c \) for superfluidity; the onset pair-formation temperature is usually called \( T^* \). In this so-called "pseudo-gap" regime, the pair size should be on the order of the interparticle spacing and pairing strongly affected by many-body effects (4, 5). The character of this strongly correlated regime, situated between a Fermi liquid and a normal Bose liquid, is a matter of debate, whose resolution should affect understanding of other strongly coupled fermion systems, such as the high-\( T_c \) cuprates and twisted bilayer graphene (6-8). The rich physics of the BEC-BCS crossover is captured by the attractive Fermi-Hubbard model, a spin-1/2 gas of fermions hopping on a lattice with on-site interactions between unlike spins (9-17). Through a particle-hole transformation, it stands in one-to-one correspondence with the repulsive Hubbard model (18, 19), which under charge doping is believed to hold the key toward understanding high-temperature superconductivity. The model can be realized using neutral fermionic atoms in optical lattices with tunable interactions. Recent investigations have found spectral gaps (20), correlations between local pairs (21), and evidence for interspin correlations from density profiles (22).

In this work, we observe the formation and spatial ordering of nonlocal fermion pairs in the pseudo-gap regime of an attractive Hubbard gas confined to two dimensions. We use bilayer quantum gas microscopy to detect the in situ location and spin of each fermion in every experimental shot (23-25). Access to microscopic spin and density correlations reveals the formation of nonlocal pairs, the development of long-range spatial correlations between pairs, and the interplay of pair fluctuations with this density-wave order.

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Fig. 1. Atom-resolved detection of an attractive Fermi-Hubbard gas. (A) Qualitative phase diagram of the attractive Fermi-Hubbard model versus on-site attraction \( U/t \) and temperature \( T/t \) at density \( n=0.8 \) (9-13). Below a critical temperature \( T_c \), attractive fermions form a BCS or BEC superfluid (SF). In the pseudo-gap regime between \( T_c \) and pairing temperature \( T^* \), accessed in this work (white shading), increasing attraction drives pair formation, with pairs exhibiting charge density wave (CDW) and superfluid correlations. (B) Measured doublon density \( d \) (circles) at fixed density \( n \) versus \( U/t \), from the noninteracting limit \( d = (n/2)^2 \) (triangles) to the fully paired limit \( d = n/2 \) (squares), with representative images of the full density in ~20 × 20 site regions shown above. (C) Snapshot of full spin- and density readout of a strongly correlated gas at \( U/t = 8.4 (4) \) and \( T/t = 0.36 (5) \). The spin up (blue), spin down (red), and combined images (rightmost panel) are obtained via bilayer quantum gas microscopy (23, 25).
Simulating the attractive Fermi-Hubbard model

The phase diagram of the attractive Fermi-Hubbard model is shown in Fig. 1A as a function of the attractive on-site interaction strength $U$, tunneling amplitude $t$, and temperature $T$ (9–13). For weak attraction $U < t$, a BCS superfluid of long-range fermion pairs forms with $T_c = T^*$, reflecting the exponentially weak pair binding. In the opposite limit of strong attraction $U > t$, all fermions are bound into local on-site pairs below a dissociation temperature $T^* \approx U$. These pairs condense at the critical temperature of Bose-Einstein condensation $T_c$, proportional to the pair density $n_p$ and pair tunneling rate $\Delta p \sim t^2/U$. A peak of the condensation temperature $T_c/N_{\uparrow\downarrow} = \pi \sigma^2 \Delta p/t$ is expected to occur at $U/t = 6$ and density $n = 0.8$ (9–13). Above the transition temperature, superfluid correlations compete with the formation of a checkerboard charge density wave (CDW) (21). At half-filling (density $n = 1$), this competition persists down to $T = 0$ and prevents condensation. In this work we use a filling $n = 0.8$, staying in a regime where the ground state is a paired superfluid (9, 15).

We here experimentally realize the attractive Fermi-Hubbard model using a two-species gas of degenerate fermionic $^{40}$K atoms trapped within an optical lattice (23; 25). The Hubbard tunneling amplitude $t$ is controlled by the lattice depth, and the interaction strength $U$ is tuned via the magnetic field. A bilayer quantum gas microscope reveals the location of each fermion.

As a first measure of strong pairing in the attractive Hubbard gas, we detect the density $d$ of doublons (doubly occupied lattice sites) for increasing interaction strength $U/t$ across the phase diagram in Fig. 1A. At fixed density $n$, $d$ increases from the noninteracting limit $d = (n/2)^2$ of random encounters of unlike spins to the fully paired limit $d = n/2$ (Fig. 1B) (17). At intermediate attraction, strong checkerboard ordering of doublons is observed, shown in Fig. 1C at $U/t = 8.4$ (left to right), showing the formation of nonlocal pairs and on-site pairs with increasing attraction. Schematics above highlight the physics dominating spin correlations in each image, and shaded bonds suggest possible pair correlations. (B) Correlation maps $\langle m_\uparrow m_\downarrow \rangle_c$ of the magnetization $m = n_\uparrow - n_\downarrow$ at various $U/t$. (C) Total magnetization fluctuations $\sum_c \langle m_\uparrow m_\downarrow \rangle_c$ (blue circles) and on-site fluctuations (black squares) versus $U/t$. Total fluctuations equal the product of magnetic susceptibility $\chi_m$ and temperature $T$ via the fluctuation-dissipation theorem (23). Vanishing total spin fluctuations for $U/t > 6$ (orange shading) indicate full pairing and vanishing $\chi_m$. Blue shading shows quantum Monte Carlo simulations of total fluctuations at $n = 0.85$, from $T/t = 0.3$ to $T/t = 0.4$ (23). The pairing temperature $T^*$ crosses $T = 0.35t$ at $U/t = 2.5$. Nonlocal pairing is reflected in the singlet fraction per total density $s/n$ (upper inset), which scales as $-\delta^2/U^2$ at large attraction. Fluctuations at $U/t = 5.8$ (lower inset) extend beyond the interparticle spacing $1/\sqrt{\pi t}$ (dotted line). All data and error bars are obtained from bootstrapping greater than 50 images of atomic clouds with imaging loss correction (23).

**Fig. 2. Observation of nonlocal fermion pairing.** (A) Experimental snapshots of the Fermi gas at $U/t = 0$, $U/t = 5.83$, and $U/t = 8.44$ (left to right), showing the formation of nonlocal pairs and on-site pairs with increasing attraction. Schematics above highlight the physics dominating spin correlations in each image, and shaded bonds suggest possible pair correlations. (B) Correlation maps $\langle m_\uparrow m_\downarrow \rangle_c$ of the magnetization $m = n_\uparrow - n_\downarrow$ at various $U/t$. (C) Total magnetization fluctuations $\sum_c \langle m_\uparrow m_\downarrow \rangle_c$ (blue circles) and on-site fluctuations (black squares) versus $U/t$. Total fluctuations equal the product of magnetic susceptibility $\chi_m$ and temperature $T$ via the fluctuation-dissipation theorem (23). Vanishing total spin fluctuations for $U/t > 6$ (orange shading) indicate full pairing and vanishing $\chi_m$. Blue shading shows quantum Monte Carlo simulations of total fluctuations at $n = 0.85$, from $T/t = 0.3$ to $T/t = 0.4$ (23). The pairing temperature $T^*$ crosses $T = 0.35t$ at $U/t = 2.5$. Nonlocal pairing is reflected in the singlet fraction per total density $s/n$ (upper inset), which scales as $-\delta^2/U^2$ (gray line) at large attraction. Fluctuations at $U/t = 5.83$ (lower inset) extend beyond the interparticle spacing $1/\sqrt{\pi t}$ (dotted line). All data and error bars are obtained from bootstrapping greater than 50 images of atomic clouds with imaging loss correction (23).
lights the physical mechanisms that determine good agreement with theoretical predictions. The reduction in fluctuations is in the density fluctuations of each spin, and there-

An energy gap for spin excitations, which ex-

local pairs (right). (a noninteracting gas (left), to a fully paired gas with CDW order (center), to a weakly ordered gas of density response at

Pairing beyond an interaction strength

The magnetization fluctuations are directly connected to the magnetic susceptibility

Characteristic for the pseudo-gap regime is a near-complete sup-

Measuring charge correlations

The suppression of fluctuations in Fig. 2C with increasing $U/t$ signifies the development of an energy gap for spin excitations (26). Theory predicts (10–14, 27, 28) a pairing temperature $T^* = 0.25U$ in the crossover regime (23). This predicted $T^*$ crosses $T = 0.35t$ near $U/t \approx 2.5$, explaining the near-complete suppression of fluctuations beyond $U/t = 6$. The corresponding expected spin excitation gap far exceeds the two-body binding energy $E_b$, highlighting the many-body nature of pairing.

Within this regime of full pairing, the two atoms within a pair have a finite probability of being located on separate lattice sites because of quantum fluctuations, in which case they appear as isolated single atoms (called singlons). This probability is measured by the ratio of the singlon density to the total density, $s/n$ (Fig. 2C, upper inset). The observed scaling of $s/n$ with $t^2/U^2$ at strong attraction is expected from perturbation theory already for a Fermi-Hubbard double well (25, 29). At $U/t = 5.8(3)$, the nonlocal portion of the pairs amounts to ~20%. The effective size of fermion pairs can be obtained as the spatial extent of nonlocal spin fluctuations. With full pairing at $U/t = 5.8(3)$, spin fluctuations are present beyond the single-spin interparticle spacing (Fig. 2C, lower inset), indicating that fermion pairs overlap substantially.

Fig. 3. Charge density wave ordering of pairs. (A) Density correlations $\langle \hat{n}_{\mathbf{k}+\mathbf{d}} \hat{n}_{\mathbf{k}} \rangle_c$ reflect the crossover from a noninteracting gas (left), to a fully paired gas with CDW order (center), to a weakly ordered gas of local pairs (right). (B) At $U/t = 8.4(4)$, the nonlocal rectified correlations $\langle \hat{n}_{\mathbf{k+} \mathbf{d}} \hat{n}_{\mathbf{k}} \rangle_\perp$ (middle) are connected to the density fluctuations of each spin, and there-

is measured locally in our quantum gas micro-

connected to the magnetic susceptibility $\chi_m = \partial m/\partial h$, the response of the magnetization to a global magnetic field $h$, through the fluctuation-dissipation theorem $\chi_m T = \sum_\delta \langle \hat{m}_i \hat{m}_{i+\delta} \rangle_c$ (23).

An energy gap for spin excitations, which exponentially suppresses excess spins and thus $\sum_\delta \langle \hat{m}_i \hat{m}_{i+\delta} \rangle_c$, also exponentially suppresses $\chi_m$ (26).

Figure 2 reports a crossover to full fermion pairing beyond an interaction strength $U/t = 6$ at $n = 0.81(1)$ and $T/t = 0.35(5)$, determined by in situ observation of magnetization fluctu-
ations. The reduction in fluctuations is in good agreement with theoretical predictions for these parameters (12–14). Figure 2A high-

lights the physical mechanisms that determine spin fluctuations at various $U/t$. At vanishing interactions, Pauli exclusion separately reduces the density fluctuations of each spin, and there-

by also reduces total spin fluctuations. With increasing attraction, nonlocal pairs form in which spins are subject to a competition of Pauli exclusion and attraction, while deep in the on-site pair regime spin fluctuations reflect virtual hopping onto neighboring sites. From statistical averages over more than 50 spin configurations, as shown in Fig. 2A for each interaction strength, we obtain the two-dimensional (2D) magnetization correlation maps $\langle \hat{m}_i \hat{m}_{i+\delta} \rangle_c$ shown in Fig. 2B. To detect pairing, Fig. 2C presents the sum of these correlation maps, the total magnetization fluctuations, which are fully suppressed beyond $U/t=6$. Already at zero interactions, Pauli exclusion reduces total fluctuations by 68(5)% compared to the high-
temperature expectation $n(1-n/2)$. This reflects the substantial degeneracy of the Fermi gas (29). Increasing attraction reduces magnetization fluctuations further, and the fraction of unpaired spins is less than 1.5(1.8)% at $U/t = 5.8(3)$, where $\sum_\delta \langle \hat{m}_i \hat{m}_{i+\delta} \rangle_c$ gives the density of unpaired spins. This full suppression is dual to the formation of a Mott insulator for repulsive interactions (18, 19, 23, 25).
interaction \(23\), revealing that a single \(\uparrow\) atom in total repels spin \(\downarrow\) atoms on all other sites. This constitutes a strong direct signature of effective repulsion between pairs.

The development and destruction of CDW order across the phase diagram of Fig. 1 can be captured by the density response \(\chi_m\) at wave vector \(k = (\pi, \pi)\) (Fig. 3C), which reflects the prevalence of low-energy states with checkerboard order. Highlighting the power of quantum gas microscopy, this thermodynamic property can be measured in equilibrium using the fluctuation-dissipation theorem for density perturbations, 

\[
\chi_m(k) = \frac{1}{T} \sum_{\delta} \langle \hat{n}_{\delta} \hat{n}_{\delta+\delta} \rangle \cos(k \cdot \delta),
\]

and the measured uniform density compressibility \(\chi_0 = \chi_m(k = (0, 0)) = \partial \rho / \partial \mu\) (Fig. 3D) \(25\). This same method allows measurement of \(\chi_m\) throughout the Brillouin zone (Fig. 3C, inset) and provides a model-independent measurement of temperature \(T\) (Fig. 3D) \(25\). The latter enables obtaining the magnetic susceptibility \(\chi_m\) from the measured spin fluctuations without applying a magnetic field \(23, 32\). As expected from the phase diagram in Fig. 1, the peak in CDW order occurs near \(U/t = 6\). Also displayed are the interspin correlations, obtained from 

\[
\chi_{11}(k) = \frac{1}{T} \sum_{\delta} \langle \hat{n}_{\delta} \hat{n}_{\delta+\delta} \rangle \cos(k \cdot \delta).
\]

Although opposite spins are uncorrelated at \(U/t = 0\), they are seen to almost fully carry the CDW order beyond \(U/t = 6\). Because density, magnetization, and interspin correlations are related by \(\langle \hat{n}_{\delta} \hat{n}_{\delta+\delta} \rangle = -4 \langle \hat{n}_{\delta} \hat{n}_{\delta+\delta} \rangle_{\text{on}}\), the relative agreement of \(\chi_m\) and \(\chi_{11}\) illustrates the strength of density order as compared to magnetic order at \(k = (\pi, \pi)\). The pronounced CDW peak is also a signature of strong superfluid correlations within the crossover regime, as CDW correlations away from half-filling serve as a lower bound for superfluid correlations \(21, 33\).

### Polaronic correlations

Given simultaneous charge and spin measurements, we finally explore the interplay of nonlocal pair fluctuations and the CDW order of other pairs, revealing the existence of polaronic correlations in the CDW order of the attractive Hubbard model. Polaronic correlations occur in the regime of highly nonlocal pairs, where further tunneling of a separated pair can displace the CDW checkerboard or flip the sign of superfluid correlations (Fig. 4A). These tunneling events prevent the virtual delocalization of other pairs across the bonds where the order has been reversed, costing an additional \(4e^2 / U\) per bond in the strong-coupling limit and further confining spatially separated pairs \(36\). This mechanism is directly complementary \(18, 19, 23\) to the magnetic polaron mechanism of the repulsive Fermi-Hubbard model \(35, 36\), though here polaronic correlations dress the individual spins of a spatially separated fermion pair, rather than excess dopants.

In Fig. 4B, we compare the CDW correlations surrounding single spins to those present in the background. We quantify the underlying CDW strength as \(\langle (\hat{a}_{\delta} - \hat{h}_{\delta})(\hat{a}_{\delta+\delta} - \hat{h}_{\delta+\delta}) \rangle / \langle \hat{h} \rangle^2\) for a given displacement \(\delta\), which is positive for any \(\delta\) for a gas possessing checkerboard doublon-hole correlations. This underlying CDW strength peaks near an interaction strength \(U/t = 8\). By contrast, for various \(U/t\) values, the measured CDW strength is strongly reduced after conditioning on the presence of a single nearby isolated spin. This reduction substantially exceeds the lowest-order expectation of single-pair fluctuation events depicted in Fig. 4A, e.g., 25% for a displacement \(\delta = (0, 1)\) and 50% for \(\delta = (1, 1)\) or \((2, 0)\). The measurements directly reveal the spatial extent of these polaronic effects, captured by the relative change \(\Delta_{CDW}(\delta)\) of conditioned to unconditioned CDW strength (Fig. 4, C and D). Virtual pair fluctuations disturb the background CDW order and provide a direct measure of polaronic correlations.
the CDW order over a range of $-2$ sites at $U/t = 8.4(4)$, with complete reduction or even reversal of the CDW order on nearby bonds. In future work, measurements of four-point correlations (36) around pairs of spins will further elucidate the internal structure of these quantum fluctuations.

Conclusions

Our real-space observations of nonlocal fermion pairing and its interplay with CDW order illustrate the richness of the pseudo-gap regime of the attractive Hubbard model. Similar competing or intertwined orders are predicted for the repulsive Hubbard model. The methods can be extended further to study polaronic physics and superfluidity (37), pairing in momentum space as measured in bulk 2D gases (38), to detect the $\pi$ phase shift of CDW order across stripes (39), and to directly measure the BCS condensate fraction through pair correlations (40).

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SUPPLEMENTARY MATERIALS

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Supplementary Text

Figs. S1 to S9

Supplementary References (42–49)

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