Correlations and Pair Formation in a Repulsively Interacting Fermi Gas

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A degenerate Fermi gas is rapidly quenched into the regime of strong effective repulsion near a Feshbach resonance. The spin fluctuations are monitored using speckle imaging and, contrary to several theoretical predictions, the samples remain in the paramagnetic phase for arbitrarily large scattering length. Over a wide range of interaction strengths a rapid decay into bound pairs is observed over times on the order of $10\,\hbar/E_F$, preventing the study of equilibrium phases of strongly repulsive fermions. Our work suggests that a Fermi gas with strong short-range repulsive interactions does not undergo a ferromagnetic phase transition.

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Many-body systems can often be modeled using contact interactions, greatly simplifying the analysis while maintaining the essence of the phenomenon to be studied. Such models are almost exactly realized with ultracold gases due to the large ratio of the de Broglie wavelength to the range of the interatomic forces [1]. For itinerant fermions with strong short-range repulsion, textbook calculations predict a ferromagnetic phase transition - the so-called Stoner model [2].

Here we simulate this system using an ultracold gas of fermionic lithium atoms, and observe that the ferromagnetic phase transition does not occur. It even appears that strong repulsive short-range interactions cannot be realized in nature. They cannot be realized by purely repulsive forces. A repulsive square well potential is characterized by a positive scattering length a smaller than or equal to the barrier radius. Strong interactions with $k_F a \geq 1$ require this radius to be comparable to the Fermi wavelength $\lambda_F = 2\pi/k_F$ and are no longer short range. Arbitrary large repulsive scattering lengths can be realized with short-range *attractive* potentials with a loosely bound state with binding energy $\hbar^2/(ma^2)$, m being the atomic mass. However, the repulsive gas is then by necessity only metastable with respect to decay into the bound state. Many theoretical studies of a repulsive Fermi gas assume that the metastable state is sufficiently long-lived [3–14]. In recent Monte-Carlo simulations, the paired state is projected out in the time evolution of the system [15, 16]. One theoretical study concluded that the pairing instability is somewhat faster than the ferromagnetic instability [17]. Here we show that pairing occurs on a very short time scale.

The experiment reported here contains two major innovations. One is the fast change of the scattering length a achieved by rapidly switching large magnetic fields near a Feshbach resonance. We observe extremely fast conversion of atoms to molecules, much faster than was observed before due to the limited time resolution of previous experiments [18]. We present arguments that this rapid molecule formation is accompanied by strong local heating which destroys the cold atomic gas even faster than the molecule formation. Since this conversion occurs on a time scale of $10\hbar/E_F$, our study indicates that the metastable repulsive state may never reach equilibrium and that, even in a metastable sense, a Fermi gas with strong short-range repulsive interactions does not exist.

The second major innovation in our experiment is the ability to precisely monitor spin fluctuations, defined as the variance of the spin density difference $(n_1 - n_2)$, by measuring the corresponding index of refraction fluctuations in the quantum gas [19, 20]. When spin domains of m atoms form, the measured phase noise increases by a factor of m. This is illustrated by a simplified model assuming Poissonian fluctuations in a given probe volume within the atom sample. With on average N atoms in this volume one would measure a standard deviation in the atom number of $\sqrt{N}$. However, if the atoms formed clusters each made of m atoms the standard deviation of the number of clusters would be $\sqrt{N/m}$, leading to a variance in atom number of $(m\sqrt{N/m})^2 = mN$.

One can characterize the onset of ferromagnetism by a divergence in the correlation length ξ for aligned spins. After a quench across a ferromagnetic phase transition, ξ increases with time to the size of the system, establishing long-range order [21]. This buildup of correlations is reflected in the spin fluctuations, which can be expressed as an integral over the pair correlation function $g^{(12)}(r)$ between spin up and down particles:

$$\mathrm{Var}(N_1 - N_2) = \mathrm{Var}(N_1) + \mathrm{Var}(N_2) - \frac{2\langle N_1\rangle\langle N_2\rangle}{V}\int_V dr(g^{12}(r) - 1) \quad (1)$$

where $\langle N_1 \rangle$ and $\langle N_2 \rangle$ are the average numbers of atoms in state 1 and 2 in a volume V. The fluctuations are enhanced due to ferromagnetic correlations by a factor $(\xi/\lambda_F)^3$, for $\xi^3 \ll V$, which is also the number of atoms in a domain, in agreement with the argument given above. As a result, spin fluctuations provide a very sensitive method for characterizing the onset of magnetic order and the buildup of correlations since there is a strong enhancement even for microscopic domains of only a few particles.

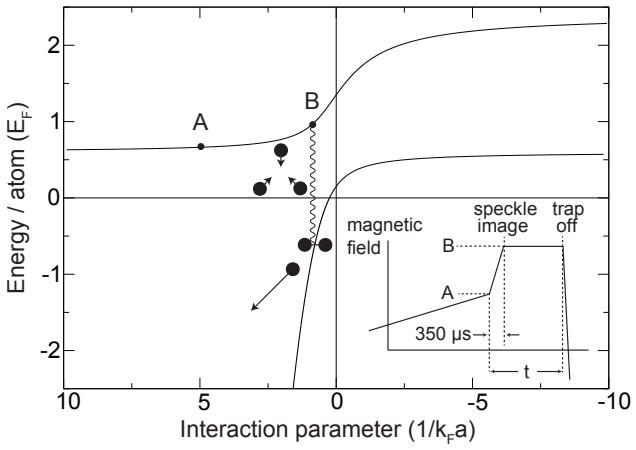

FIG. 1: Diagram showing energy levels and timing of the experiment. The upper (repulsive) and lower (attractive) branch energies, near a Feshbach resonance, are connected by 3-body collisions. In our experiment, we quickly jump from a weakly interacting Fermi gas (A) to a strongly interacting one (B) with a rapid magnetic field change. At (B), speckle imaging is used to look for magnetic domains in the sample. Molecule formation (population of the lower branch) is studied as a function of hold time. Adapted from [22].

The experiments were carried out with typically 4.2×10^5 ^{6}Li atoms in each of the two spin states $|1\rangle$ and $|2\rangle$ confined in an optical dipole trap with radial and axial trap frequencies $\omega_r = 2\pi \times 100(1)\mathrm{s}^{-1}$ and $\omega_z = 2\pi \times 9.06(25)\mathrm{s}^{-1}$. The sample was evaporatively cooled at a magnetic bias field $B = 320G$, identical to the procedure described in [19]. Then the magnetic field was slowly ramped to 730 G ($k_Fa = 0.35$) in 500 ms. The fraction of atoms being converted to molecules during the ramp was measured to be below 5 percent. The temperature of the cloud was typically $0.23(3)\,T_F$ at 527 G with a Fermi energy of $E_F = k_BT_F = h \times 6.1\,\mathrm{kHz}$. After rapidly switching the magnetic field from 730 G to the final value in less than 350 μs, spin fluctuations were measured by speckle imaging. Optionally an appropriate RF pulse was applied directly before imaging to rotate the spin orientation along the measurement axis. Due to the use of 20 cm diameter coils outside the vacuum chamber, the inductance of the magnet coils was 330 μH and the fast switching was accomplished by rapidly discharging capacitors charged to 500V.

Experimentally, spin fluctuations are measured using the technique of speckle imaging described in Ref. [20]. For an appropriate choice of detuning, an incident laser beam experiences a shift of the refractive index proportional to the difference between the local populations of the two spin states N_1 and N_2. Spin fluctuations create spatial fluctuations in the local refractive index and imprint a phase pattern into the incoming light, which is then converted into an amplitude pattern during propagation. The resulting spatial fluctuations in the probe laser intensity are used to determine the spin fluctuations in the sample.

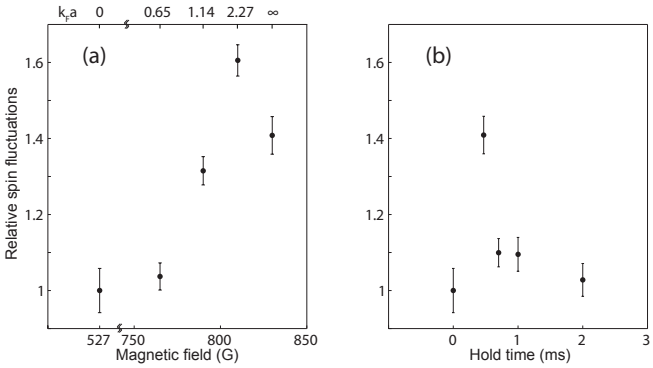

FIG. 2: Spin fluctuations (a) after 350 μs as a function of magnetic field and (b) on resonance as a function of hold time scaled to the value measured at 527G. Even at strong repulsive interactions, the measured spin fluctuations are barely enhanced, indicating only short-range correlations and no domain formation. The spin fluctuations were determined for square bins of 2.6 μm.

Figure 2 shows the observed spin fluctuations compared to the non-interacting cloud at 527 G. The main result of this paper is the absence of any major enhancement due to strong repulsive forces. The observed spin fluctuations remain within a factor of two of their original value and demonstrate that the sample remains in the paramagnetic phase for a wide range in the strength of interactions and wait time.

Before we can fully interpret these findings, we have to take into account the decay of the atomic sample on the upper branch of the Feshbach resonance into bound pairs. We characterize the pair formation by comparing the total number of atoms and molecules $N_\mathrm{a}+2N_\mathrm{mol}$ (determined by taking an absorption image after ballistic expansion at high magnetic field where molecules and atoms have the same absorption resonance) to the number of free atoms (determined by rapidly sweeping the magnetic field to 5G before releasing the atoms and imaging the cloud, converting pairs into deeply bound molecules that are completely shifted out of resonance)

[23].

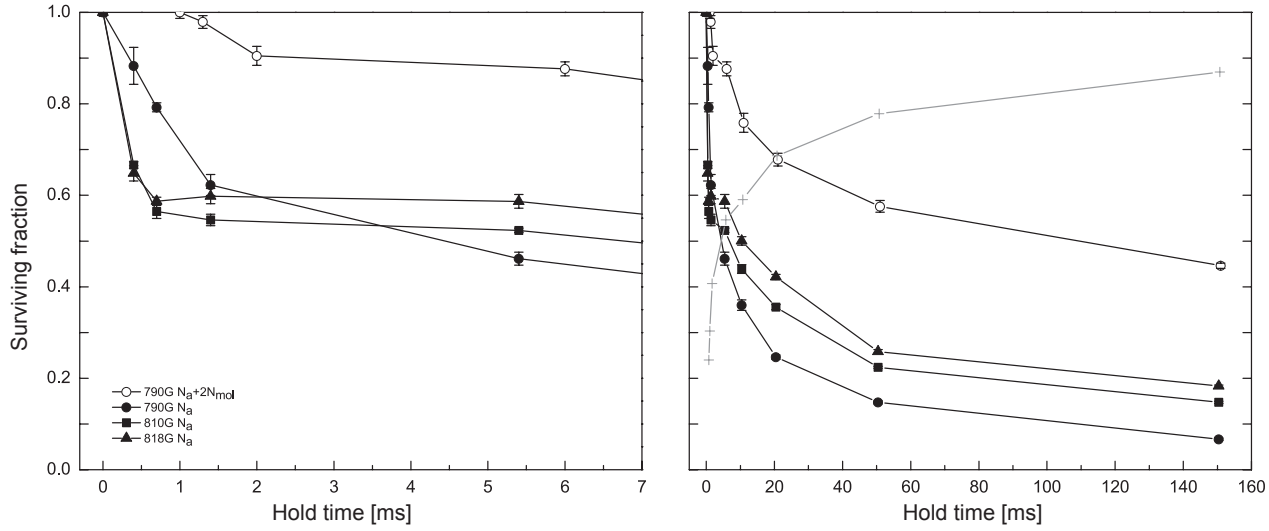

FIG. 3: Characterization of molecule formation at short and long times, and at different values of the magnetic field. The closed symbols represent the normalized number of free atoms, the open symbols the total number of atoms including those bound in Feshbach molecules. The crosses show the molecule fraction.

The time evolution of the molecule production (Fig. 3) shows two regimes of distinct behavior. For times less than 1 ms, we observe a considerable number of atoms converted into molecules, while the total number $N_{\rm a}+2N_{\rm mol}$ remains constant. The initial drop in atom number becomes larger as we increase the final magnetic field, and saturates at around 40 percent near the Feshbach resonance.

We attribute this fast initial decay in atom number to three-body recombination [24, 25] into the weakly bound molecular state. We obtain an atom loss rate $\dot{N}_a/N_a = 250\,{\rm s}^{-1}$ at 790G in the first 1ms after the magnetic field switch, and from this we estimate the three body formation coefficient L3 at this field to be $3.9\times10^{-22}\,{\rm cm}^6\,{\rm s}^{-1}$, though the interaction is already sufficiently strong for many-body effects to be significant. For stronger interactions, about 30% of atom loss occurs already during the relevant 100 μs of ramping through the strongly interacting region, indicating a lower bound of around $3\times10^3\,{\rm s}^{-1}$ for the loss rate which is 13% of the inverse Fermi time $E_F/\hbar$, calculated with a cloud averaged Fermi energy.

After the first millisecond, the molecule formation rate slows down, by an order of magnitude at a magnetic field of 790G (and even more dramatically at higher fields). This can be only partially attributed to a drop in atomic density by a factor of two due to conversion into molecules and to an increase in the size of the sample due to the increased repulsive interactions. In addition, it seems likely that the molecule fraction approaches a quasi-equilibrium value at the local temperature, which may have increased due to local heating accompanying the molecule formation. A slow loss in both atom number and total number is observed on a time scale of hundreds of milliseconds and is caused by inelastic collisions (vibrational relaxation of molecules) and evaporation loss. Continuous evaporation cools down the system, and therefore the molecule fraction steadily increases and reaches 90 % for the longest hold time.

Does a partial conversion of the cloud into molecules suppress a possible ferromagnetic phase transition? Ferromagnetic domain formation is analogous to phase separation between the two spin components [18]. Since dimers interact equally with the two spin components, one might expect that even a noticeable dimer fraction should not suppress the tendency of the atomic gas to form domains (other than by increasing the local temperature). In this case, our experiment excludes a ferromagnetic instability on ms time scales. However, it is possible that the unpaired atoms have different properties in the presence of a large pair fraction and then our window to observe domain formation would be shorter.

Pair formation heats the sample by transferring the binding energy and excess kinetic energy of the molecules to the remaining atoms, and also by creating holes in the Fermi sea. For small values of $k_F a$, the total energy release per molecule is $\hbar^2/ma^2 + 2E_F$. From the energy per particle for an ideal homogeneous degenerate Fermi gas $E = 0.6E_F[1 + (5\pi^2/12)(T/T_F)^2]$ we obtain

$$\frac{T}{T_F} = \sqrt{\frac{4\eta}{\pi^2}\left(1 + \frac{1}{(k_F a)^2}\right)} \qquad (2)$$

for the final temperature with a small molecule fraction η (assuming initially η=0 and T=0). If we evaluate this result at $k_F a$=1, where the two-body binding energy is $2E_F$, we obtain that molecule fractions of higher than 20% result in a final temperature above $0.4T_F$. One may hope that closer to resonance many-body effects lower the released energy, however as we show in the supplement [26] this is not necessarily the case due to the repulsive interaction energy.

We attempted to measure the temperature after the hold time near the Feshbach resonance by quickly switching the magnetic field to weak interactions at 527 G and then performing noise thermometry using speckle imaging [20]. We measure column-integrated fluctuations that are 0.61(8) of the Poisson value, this implies an effective temperature well below T_F, around 0.33(7) T_F. Although the cloud is not in full equilibrium, an effective local temperature can still be obtained from noise thermometry.

Available theoretical treatments do not predict an exact maximum transition temperature to the ferromagnetic state and obtain an unphysical divergence for large scattering lengths. Since the only energy scale is the Fermi temperature, one would expect a transition temperature which is a fraction of the Fermi temperature [27], higher or around the temperature scale probed in our experiments. However, even above the transition temperature, the susceptibility is enhanced. A simple Weiss mean field or Stoner model leads to the generic form of the susceptibility $\xi(T) = \xi_0(T)/(1 - w\xi_0(T))$, where $\xi_0(T)$ is the Pauli susceptibility of the non-interacting gas and w the interaction parameter. This formula predicts a two-fold increase in the susceptibility even 50 % above the transition temperature, which is well within the sensitivity of our measurements.

A previous experiment [18] reported evidence for ferromagnetism by presenting non-monotonic behavior of atom loss rate, kinetic energy and cloud size when approaching the Feshbach resonance, in agreement with predictions based on the Stoner model. Our measurements confirm that the properties of the gas strongly change near $k_F a = 1$. Similar to Ref. [18], we observe near the resonance features in kinetic and release energy measurements (see Supplemental Information [26]). However, the behavior is more complex than that captured by simple models. The atomic fraction decays non-exponentially (see Fig. 3), and therefore an extracted decay time will depend on the details of the measurement such as time resolution. Ref. [18] found a maximum of the loss rate of $200\,\mathrm{s}^{-1}$ for a Fermi energy of 28 kHz. Our lower bound of the decay rate of $3 \times 10^3\,\mathrm{s}^{-1}$ is 15 times faster at a five times smaller Fermi energy. Our more detailed study rules out that Ref. [18] has observed ferromagnetic behavior.

Our conclusion is that an ultracold gas with effectively repulsive interactions near a Feshbach resonance remains in the paramagnetic phase. The fast formation of molecules and the accompanying heating makes it impossible to study such a gas in equilibrium, confirming predictions of a rapid conversion of the atomic gas to pairs [17, 28]. Therefore, Nature does not realize a strongly repulsive Fermi gas with short range interaction, and the widely used Stoner model is unphysical.

We want to raise now the hypothetical question if the gas would show a ferromagnetic phase transition if the molecule formation were absent. Recent Monte-Carlo simulations [15, 16] came to a positive answer when the molecular state was projected out of the simulation. The discussion above suggests that molecule formation and heating of the cloud give us a temporal window of several ms to observe a possible tendency towards domain formation. Since theoretical predictions for domain formation after a quench across the ferromagnetic phase transition predict a faster formation of domains [17, 26], our results suggest the absence of a ferromagnetic instability, in contrast to predictions of Monte-Carlo simulations [15, 16] and second order perturbation theory [7], and in agreement with conclusions based on Tan relations [29].

Interesting topics for future research include the effects of dimensionality [30], spin imbalance [31, 32], mass imbalance [33], and band structure [34, 35] on ferromagnetism and pair formation.

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