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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 instability [2].

Here we investigate this system using an ultracold gas of fermionic lithium atoms, and observe that the ferromagnetic phase transition does not occur. A previous experimental study [3] employing a different apparatus found indirect evidence for a ferromagnetic phase, but did not observe the expected domain structure, possibly due to the lack of imaging resolution. Here we address this shortcoming by analyzing density and spin density fluctuations via speckle imaging [4]. When spin domains of m atoms form, the spin density variance will increase by a factor of m [5], even if individual domains are not resolved. One main result of this paper is the absence of such a significant increase which seems to exclude the possibility of a ferromagnetic state in the studied system.

The Stoner model assumes a two-component Fermi gas with a repulsive short-range interaction described by a single parameter, the scattering length. The predicted phase transition to a ferromagnetic state requires large repulsive scattering lengths on the order of the interatomic spacing. They can be realized only by short-range *attractive* potentials with a loosely bound state with binding energy $\hbar^2/(ma^2)$, with m being the atomic mass and a being the scattering length [6]. However, the repulsive gas is then by necessity only metastable with respect to decay into the bound state. Many theoretical studies of a Fermi gas with strong short-range repulsive interactions assume that the metastable state is sufficiently long-lived [7–18]. In recent Monte-Carlo simulations, the paired state is projected out in the time evolution of the system [19, 20]. Theoretical studies concluded that the pairing instability is somewhat faster than the ferromagnetic instability [21]. The second major result of

this paper is to show that pair formation occurs indeed on a very short time scale. The measured time constant of $10\hbar/E_F$ (where E_F is the Fermi energy) indicates that the metastable repulsive state will never reach equilibrium and that, even in a metastable sense, a Fermi gas with strong short-range repulsive interactions does not exist. The fast pair formation could not be observed previously due to limited time resolution [3]. Instead, a much slower second phase in the conversion of atoms to pairs was observed leading to the wrong conclusion that the unpaired atoms have a much longer lifetime.

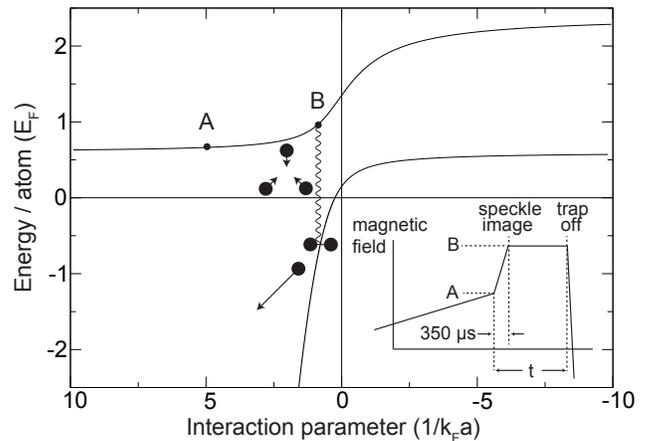


FIG. 1: Diagram showing energy levels and timing of the experiment. The upper (repulsive) and lower (attractive) branch energies, near a Feshbach resonance, are connected by 3-body collisions. In our experiment, we quickly jump from a weakly interacting Fermi gas (A) to a strongly interacting one (B)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

The experiments were carried out with typically 4.2×10^5 ^6Li atoms in each of the two lower spin states $|1\rangle$ and $|2\rangle$ confined in an optical dipole trap with radial and axial trap frequencies $\omega_r = 2\pi \times 100(1)\text{s}^{-1}$ and

$\omega_z = 2\pi \times 9.06(25)\text{s}^{-1}$. The sample was evaporatively cooled at a magnetic bias field $B = 320\text{G}$, identical to the procedure described in [23]. Then the magnetic field was slowly ramped to 730G ($k_F a = 0.35$) in 500ms . The fraction of atoms being converted to molecules during the ramp was measured (see below for method) to be below 5% . The temperature of the cloud was typically $0.23(3)T_F$ at 527G with a Fermi energy of $E_F = k_B T_F = \hbar \times 6.1\text{kHz}$. After rapidly switching the magnetic field from 730G to the final value in less than $350\mu\text{s}$, 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 20cm diameter coils outside the vacuum chamber, the inductance of the magnet coils was $330\mu\text{H}$ 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. [4]. 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.

In Ref. [4] we prepared samples on the lower branch of the Feshbach resonance, where positive values of $k_F a$ correspond to a gas of weakly bound molecules. At $k_F a = 1.2$, we observed a sixfold suppression of spin fluctuations and a fourfold enhancement of density fluctuations. Typical fluctuations in the speckle images of a non-interacting Fermi gas at $T = 0.23T_F$ amount to 5% of the average optical signal per pixel, corresponding to about 50% of Poissonian fluctuations. Those fluctuations are modified by factors between 0.2 and 1.6 due to pairing and interactions.

In this study, on the upper branch of the Feshbach resonance, the situation is reversed. For unbound atoms, as the interaction strength increases, the two spin components should develop stronger and stronger anticorrelations and enhanced spin fluctuations. Previous experimental work [3] and several theoretical studies [10, 11, 13–15, 18, 24] predicted a phase transition to a ferromagnetic state where the magnetic susceptibility and therefore the spin fluctuations diverge. Recent Monte Carlo simulations [19] predict such a divergence around $k_F a = 0.83$. We therefore expected an increase of spin fluctuations by one or several orders of magnitude, related to the size of magnetic domains.

Figure 2 shows the observed spin fluctuations enhancement compared to the non-interacting cloud at 527G .

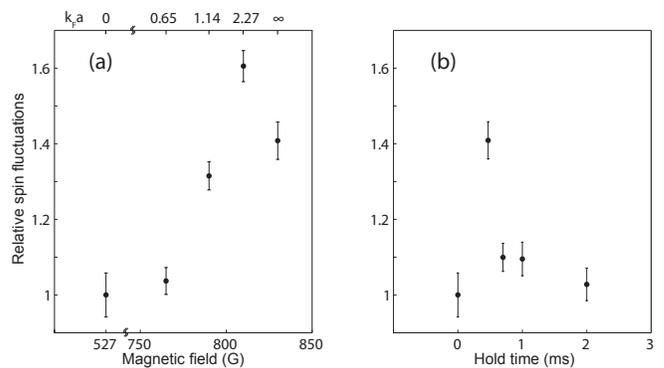


FIG. 2: Spin fluctuations (a) after $350\mu\text{s}$ as a function of magnetic field and (b) on resonance as a function of hold time scaled to the value measured at 527G . Even at strong repulsive interactions, the measured spin fluctuations are barely enhanced, indicating only short-range correlations and no domain formation. The spin fluctuations were determined for square bins of $2.6\mu\text{m}$, each containing on average 1000 atoms per spin state.

The variance enhancement factor reaches its maximum value of 1.6 immediately after the quench, decreasing during the 2ms afterward. The absence of a dramatic increase shows that no domains form and that the sample remains in the paramagnetic phase throughout. Similar observations were made for a wide range of interaction strengths and wait times. Note that first-order perturbation theory [25] predicts an increase of the susceptibility by a factor of 1.5 at $k_F a = 0.5$ and by a factor of 2 at $k_F a = 0.8$ (i.e. no dramatic increase for $k_F a < 1$). Therefore, our data shows no evidence for the Fermi gas approaching the Stoner instability.

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 $N_a + 2N_{\text{mol}}$ (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) [26].

The time evolution of the molecule production (Fig. 3) shows two regimes of distinct behavior. For times less than 1ms , we observe a considerable number of atoms converted into molecules, while the total number $N_a + 2N_{\text{mol}}$ remains constant. The initial drop in atom number becomes larger as we increase the final magnetic field, and saturates at around 50% near the Feshbach resonance.

We attribute this fast initial decay in atom number to recombination [27, 28] into the weakly bound molecular state. We obtain an atom loss rate $\dot{N}_a/N_a = 250\text{s}^{-1}$ at 790G in the first 1ms after the magnetic field switch. Assuming a three-body process we estimate the rate coefficient L_3 at this field to be $3.9 \times 10^{-22}\text{cm}^6\text{s}^{-1}$, 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 $3 \times 10^3\text{s}^{-1}$ 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) when it reaches about 50%. It seems likely that the molecule fraction has reached a quasi-equilibrium value at the local temperature, which is larger than the initial temperature due to local heating accompanying the molecule formation. Ref. [29] presents a simple model for the equilibrium between atoms and molecules (ignoring strong interactions). For phase space densities around unity and close to resonance, the predicted molecule fraction is 0.5, in good agreement with our observations [30].

For longer time scales (hundred milliseconds) we observe a steady increase of the molecule fraction to 90% for the longest hold time. This occurs due to continuous evaporation which cools down the system and shifts the atom-molecule equilibrium towards high molecule fractions. During the same time scale, a slow loss in both atom number and total number is observed caused by inelastic collisions (vibrational relaxation of molecules) and evaporation loss.

Is the rapid conversion into molecules necessarily faster than the evolution of ferromagnetic domains? Our answer is tentatively yes. First, for strong interactions with $k_F a$ around 1, one expects both instabilities (pair formation and Stoner instability) to have rates which scale with the Fermi energy E_F and therefore with $n^{2/3}$. Therefore, one cannot change the competition between the instabilities by working at higher or lower densities. According to Ref. [21] the fastest unstable modes for domain formation have a wavevector $q \approx k_F/2$ and grow at a rate of up to $E_F/4\hbar$ when the cloud is quenched sufficiently far beyond the critical interaction strength. Unstable modes with such wavevectors will develop “domains” of half a wavelength or size $\xi = \pi/q = 2\pi/k_F$ containing 5 atoms per spin state in a volume ξ^3 . This rate is comparable to the observed conversion rates into pairs of $0.13E_F$. Therefore, at best, “domains” of a few particles could form, but before they can grow further and prevent the formation of pairs (in a fully polarized state), rapid pair formation takes over and populates the lower branch of the Feshbach resonance. Based on our observations and these arguments, it seems that it is not possible to realize

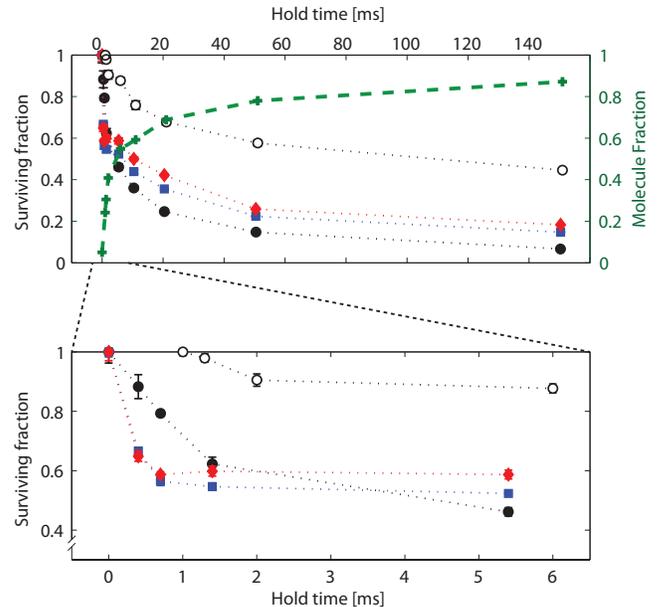


FIG. 3: Characterization of molecule formation at short and long hold times, and at different values of the interaction strength. The closed symbols, circles (black) at 790G with $k_F a = 1.14$, squares (blue) at 810G with $k_F a = 2.27$ and diamonds (red) at 818G with $k_F a = 3.5$ represent the normalized number of free atoms, the open symbols the total number of atoms including those bound in Feshbach molecules (open circles at 790G with $k_F a = 1.14$). The crosses (green) show the molecule fraction. The characteristic time scale is set by the Fermi time $\hbar/E_F = 43\mu\text{s}$, calculated with a cloud averaged Fermi energy.

ferromagnetism with strong short range interaction, and therefore the basic Stoner model cannot be realized in nature.

One possibility to suppress pair formation is provided by narrow Feshbach resonances. Here the pairs have dominantly closed channel character and therefore a much smaller overlap matrix element with the free atoms. However, narrow Feshbach resonances are characterized by a long effective range and do not realize the Stoner model which assumes short-range interactions. Other interesting topics for future research on ferromagnetism and pair formation include the effects of dimensionality [31, 32], spin imbalance [33, 34], mass imbalance [35], lattice and band structure [36, 37].

We now discuss whether ferromagnetism is possible *after* atoms and molecules have rapidly established local equilibrium. In other words, starting at $T = 0$, one could heat up the fully paired and superfluid system and create a gas of atomic quasiparticles which are similar to free atoms with repulsive interactions. Density and temperature of the atoms are now coupled. It is likely that

such a state is realized in our experiments after a few ms following the quench, until evaporative cooling converts the system into a molecular condensate over ≈ 100 ms. The possibility that such a quasiparticle gas could become ferromagnetic has not been discussed in the literature. Our experiments do not reveal any major increase in spin fluctuations which seems to exclude a ferromagnetic state. In the simplest picture, we could regard the atomic quasiparticles as free atoms, and then apply the Stoner model to them. Ferromagnetic domain formation is analogous to phase separation between the two spin components [3]. 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. Therefore, in a simple model, one may neglect dimer-atom interactions.

If the Stoner model applies to this quasiparticle gas, the next question is whether the temperature is low enough for the ferromagnetic phase transition. 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 [38], 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 $\chi(T) = \chi_0(T)/(1 - w\chi_0(T))$, where $\chi_0(T)$ 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.

Therefore, our experiment can rule out ferromagnetism for temperatures even slightly lower than the experimental temperatures. Temperatures are very difficult to measure in a transient way for a dynamic system which may not be in full equilibrium. For example, cloud thermometry requires full equilibration and lifetimes much longer than the longest trap period. 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 [4]. We measure column-integrated fluctuations that are 0.61(8) of the Poisson value which implies an effective temperature well below T_F , around 0.33(7) T_F , not much higher than our initial temperature of 0.23 T_F . Although the cloud is not in full equilibrium, an effective local temperature can still be obtained from noise thermometry.

Alternatively, we can estimate the temperature increase from the heat released by pair formation. A simple model [39] accounting for the relevant energy contributions predicts for $k_F a = 1$ that molecule fractions

of higher than 20% result in a final temperature above $0.4T_F$, an estimate which is higher than the measurement reported above. One may hope that closer to resonance many-body effects lower the released energy, however as we show in the supplement (Fig. 1 of [39]) this is not necessarily the case due to the repulsive interaction energy.

Our experiment has not shown any evidence for a possible ferromagnetic phase in an atomic gas in “chemical” equilibrium with dimers. This implies one of the following possibilities. (i) This gas can be described by a simple Hamiltonian with strong short range repulsion. However, this Hamiltonian does not lead to ferromagnetism. This would be in conflict with the results of recent quantum Monte-Carlo simulations [19, 20] and second order perturbation theory [11], and in agreement with conclusions based on Tan relations [40]. (ii) The temperature of the gas was too high to observe ferromagnetism. This would then imply a critical temperature around or below $0.2T/T_F$, lower than generally assumed. (iii) The quasiparticles cannot be described by the simple model of an atomic gas with short-range repulsive interactions due to their interactions with the paired fraction.

A previous experiment [3] 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 [3], we observe features in kinetic and release energy measurements near the resonance (see Supplemental Information [39]). 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. [3] found a maximum of the loss rate of 200 s^{-1} for a Fermi energy of 28 kHz. Our lower bound of the decay rate of $3 \times 10^3 \text{ s}^{-1}$ is 15 times faster at a five times smaller Fermi energy. Our more detailed study rules out that Ref. [3] has observed ferromagnetic behavior.

Our conclusion is that an ultracold gas with strong short range 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 [21, 41]. The Stoner criterion for ferromagnetism obtains when the effective interaction strength times the density of states is larger than one. This is at least an approximately valid criterion for multi-band lattice models [42]. We have shown here that this criterion cannot be applied to Fermi gases with short-range repulsive interactions (the basic Stoner model) since the neglected competition with pairing is crucial.

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