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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 an 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 force [1]. For itinerant fermions, 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].

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 three-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 [42].

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.
The experiments were carried out with typically $4.2 \times 10^6$ $^{6}$Li 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 $\omega_r = 2\pi \times 100(1)$ s$^{-1}$ and $\omega_z = 2\pi \times 9.06(25)$ s$^{-1}$. The sample was evaporatively cooled at a magnetic bias field $B = 320$ G, identical to the procedure described in [22]. 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 (see below for method) to be below 5%. The temperature of the cloud was typically $0.23(3)T_F$ at 527 G with a Fermi energy of $E_F = k_BT_F = h \times 6.1$ kHz. After rapidly switching the magnetic field from 730 G to the final value in less than 350 $\mu$s, spin fluctuations were measured by speckle imaging. Optionally an appropriate rf pulse was applied directly before imaging to rotate the spin orientations along the measurement axis. Due to the use of 20 cm diameter coils outside the vacuum chamber, the inductance of the magnet coils was $330 \mu$H and the fast switching was accomplished by rapidly discharging capacitors charged to 500 V.

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 $k_Fa$ correspond to a gas of weakly bound molecules. At $k_Fa = 1.2$, 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 [10,11,13–15,18,23] 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 $k_Fa = 0.83$. 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 527 G. The variance enhancement factor reaches its maximum value of 1.6 immediately after the quench, decreasing during the 2 ms 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 [24] predicts an increase of the susceptibility by a factor of 1.5 at $k_Fa = 0.5$ and by a factor of 2 at $k_Fa = 0.8$ (i.e., no dramatic increase for $k_Fa < 1$). Therefore, our data show 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 $N_a + 2N_{mol}$ (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 5 G before releasing the atoms and imaging the cloud, converting pairs into deeply bound molecules that are completely shifted out of resonance) [25].

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 $N_a + 2N_{mol}$ 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 [26,27] into the weakly bound molecular

![Figure 2](image-url)
state. We obtain an atom loss rate $\dot{N}_a/N_a = 250$ s$^{-1}$ at 790 G in the first ms after the magnetic field switch. Assuming a three-body process we estimate the rate coefficient $L_3$ at this field to be $3.9 \times 10^{-22}$ cm$^6$ 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 $\mu$s of ramping through the strongly interacting region, indicating a lower bound of around $3 \times 10^3$ s$^{-1}$ for the loss rate which is 13% of the inverse Fermi time $E_F/h$, 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 790 G (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. Reference [28] 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 [29].

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 $k_Fa$ 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 wave vector $q = k_F/2$ and grow at a rate of up to $E_F/4h$ when the cloud is quenched sufficiently far beyond the critical interaction strength. Unstable modes with such wave vectors 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 $\xi^3$. 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 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 [30,31], spin imbalance [32,33], mass imbalance [34], lattice and band structure [35,36].

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 $\approx 100$ 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 [37], 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 twofold 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 range repulsive interactions near a Feshbach resonance. However, this Hamiltonian does not lead to ferromagnetism. This would then imply a critical temperature around or below 0.27\( T_F \), 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 \( k_{\text{F},\text{a}} = 1 \). Similar to [3], we observe features in kinetic and release energy measurements near the resonance (see Supplemental Material [38]). 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. Reference [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 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 [21,40]. 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 [41]. 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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[5] 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 \( (m\sqrt{N}/m)^2 = mN \).
[6] Potentials with a positive scattering length a have no bound state only if the effective range \( r_a \) is larger than \( a/2 \). Otherwise, the s-wave scattering amplitude \( f(k) = 1/(-1/a + r_a k^2/2 - i k) \) has a pole on the imaginary axis corresponding to a bound state.
[29] Note that the drop in molecule formation after 1 ms cannot be explained by the drop in atomic density by a factor of 2 due to conversion into molecules and to an increase in the size of the sample due to the increased repulsive interactions.