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Towards quantum magnetism with ultracold atoms

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Abstract. At ICAP we presented the efforts and progress at MIT towards using ultracold
atoms for the realization of various forms of quantum magnetism. These efforts include a study of
fermions with strong repulsive interactions in which we obtained evidence for a phase transition
to itinerant ferromagnetism, the characterization of cold atom systems by noise measurements,
and a new adiabatic gradient demagnetization cooling scheme which has enabled us to realize
temperatures of less than 350 picokelvin and spin temperatures of less than 50 picokelvin in
optical lattices. These are the lowest temperatures ever measured in any physical system.

1. Introduction
Over the last 25 years, the field of ultracold atoms has focused on motion: beam slowing, cooling
atoms to low temperatures, population of a single motional state (Bose-Einstein condensation,
atom lasers), superfluid motion of bosons and fermion pairs, and the suppression of motion
in the superfluid-to-Mott-insulator transition. The next challenge, when all motion is frozen
out, is ordering of an internal degree of freedom which can be mapped onto spin ordering. A
bosonic or fermionic mixture of two or more quantum states can form magnetic phases, such as
ferromagnetically or antiferromagnetically ordered states or spin liquids.

A major experimental challenge is to reach the low temperature and entropy scales required
to observe these phenomena, and also to develop techniques of preparation and detection of
spin-ordered phases. In this paper, we discuss the efforts of our group at MIT towards this
goal. This includes a study of a two-component system of fermions with repulsive interactions,
for which a phase transition to itinerant ferromagnetism has been predicted, the development
of noise measurements to characterize new quantum phases, and a new refrigeration technique
which works “natively” in the two-component Mott insulator and is capable of cooling below
the critical temperature for spin ordering in an optical lattice.

2. Itinerant ferromagnetism in a gas of ultracold atoms
Ferromagnetism of delocalized (itinerant) fermions occurs due to repulsive interactions and the
exchange energy which reduces the interaction energy for spin polarized domains due to the
Pauli exclusion principle. At a critical interaction, given by the so-called Stoner criterion [1],
the system spontaneously develops domains and becomes ferromagnetic. This, together with a
suitable band structure in a periodic lattice, explains why certain metals, like iron and nickel,
are ferromagnetic. The simplest models for ferromagnetism assume a gas of fermions with
repulsive interactions, and predict, in mean-field and second-order approximation, the onset
Figure 1. Atom loss rate as a probe for local spin polarization, for different temperatures. (a) \( T/T_F = 0.55 \) (dashed curve), (b) \( T/T_F = 0.22 \) (dotted curve), and \( T/T_F = 0.12 \) (solid curve). The atom loss rate (due to molecule formation) increases for increasing strength of interactions, until the two components of the Fermi gas separate in domains, suppressing the loss. The maximum of the loss rate occurs close to the onset of ferromagnetism. Higher temperatures appear to suppress ferromagnetism.

of ferromagnetism [2–4]. However, there has been no proof or experimental observation for ferromagnetism in a Fermi gas.

We have studied a gas of ultracold fermionic lithium atoms and increased the strength of repulsive interactions by tuning an external magnetic field close to a Feshbach resonance. We observe non-monotonic behaviour of lifetime, kinetic energy and size (see Fig. 1). This provides strong evidence for the Stoner instability, i.e., a phase-transition to a ferromagnetic state [5]. This experiment can be regarded as a quantum simulation of a simple Hamiltonian (the hard core Fermi gas), for which even the existence of a phase transition has not been proven.

In principle, the gas has several options when the strength of interactions is increased. It can form a ferromagnetic state which would have macroscopic domains or it can form a non-magnetic state where the two spin states are anti-correlated (so to speak they form microscopic fluctuating domains). Both states may show similar signature in loss rate and kinetic energy [6]. Recent Monte-Carlo simulations compared the energies of these two scenarios and concluded that for sufficiently strong interactions the ferromagnetic state is favoured [7, 8]. The question remains whether this state forms faster than the paired state (which is a strongly interacting molecular condensate) which occupies the lower branch of the Feshbach resonance and has therefore lower energy. The prediction is that both instabilities (the ferromagnetic one and the pairing instability) have similar rates of formation with the pairing instability somewhat faster [9]. More experimental and theoretical work is needed to fully answer the question of whether a gas of fermions with short range interactions can undergo a phase transition to a ferromagnetic state.

In the case of itinerant ferromagnetism, the fermions still occupy different motional states which contribute to the total entropy. Therefore, the total entropy per particle can be higher than \( k_B \ln 2 \), which is an upper bound for the spin entropy of a magnetically ordered two-component system.
Figure 2. Comparison of observed variances (black dots) with a theoretical model (solid line) and the observed atom number (connected gray dots), at three different temperatures (a, b, and c), showing 50, 40, and 15% suppression of density fluctuations, respectively. Noise thermometry is implemented by fitting the observed fluctuations, resulting in temperatures \( T/T_F \) of 0.23, 0.33, and 0.60, in good agreement with temperatures obtained by fitting the shape of the expanded cloud.

3. Study of Density Fluctuations in Fermi gases

One frontier in the field of ultracold atoms is the realization of quantum systems with strong interactions and strong correlations. Many properties of strongly correlated systems cannot be deduced from mean density distributions. This has drawn interest toward novel ways of probing cold atoms, e.g., via RF spectroscopy, Bragg and Raman scattering, interferometric methods and by recording density correlations. Further insight into quantum systems is obtained by looking not only at expectation values, but also at fluctuations.

We have studied density fluctuations and spin fluctuations of Fermi gases. This work is motivated by the connection between density (spin) fluctuations and compressibility (magnetic susceptibility) through the fluctuation-dissipation theorem. Many interesting phases of matter have a clear signature in the compressibility or susceptibility. The crossover to a Mott or band insulator is characterized by the onset of an incompressible phase. At a ferromagnetic phase transition the susceptibility diverges, whereas in a transition to a paired or antiferromagnetic phase the susceptibility becomes exponentially small in the ratio of the pair binding energy (or antiferromagnetic gap) to the temperature.

First, we have validated our technique for determining the compressibility by applying it to the ideal Fermi gas. In recent years, several classic experiments have observed different manifestations of Pauli suppression in Fermi gases. Here we study density profiles of an ideal Fermi gas and observe Pauli suppression of density fluctuations (atom shot noise) for cold clouds deep in the quantum degenerate regime [10, 11].

For temperatures well below degeneracy, we observed strong suppression for probe volumes containing more than 10,000 atoms. Measuring the level of suppression provides noise thermometry which remains sensitive down to low temperatures since the fluctuations scale linearly in temperature (see Fig. 2).

This method was applied to strongly interacting fermions along the BEC-BCS crossover [12]. Spin fluctuations are observed by directly measuring the difference in densities for the two spin states. This was done by using a probe laser which had equal detuning from both states, but with opposite signs.
Figure 3. (a) The ratio $\chi/\kappa$, (b) the normalized susceptibility $\chi$, and (c) the normalized compressibility $\kappa$ in the BEC-BCS crossover. The variances derived from sequences of images are converted into thermodynamic variables using the measured temperatures and a calibration factor determined from the noninteracting gas. The vertical line indicates the onset region of superfluidity, as determined via condensate fraction measurements. The curves show theoretical zero temperature estimates based on 1st (dotted) and 2nd order (solid) perturbative formulas obtained from Landau’s Fermi liquid theory integrated along the line of sight, and results from a Monte Carlo calculation (dashed) for the compressibility in a homogeneous system [15].

The fluctuations in the atomic cloud led to an observed speckle pattern. The atoms imprint a phase shift into the transmitted beam. Upon propagation, this dispersive signal is transformed into an intensity speckle signal. Since the depth of the cloud is larger than the Rayleigh range associated with the resolution of the imaging systems, the transformation from phase signal to intensity signal takes place already within the atomic cloud. The random density fluctuations lead to a speckle pattern corresponding to the imaging resolution.

This new sensitive method easily resolves a tenfold suppression of spin fluctuations below shot noise due to pairing. Compressibility and magnetic susceptibility are determined from the measured fluctuations (see Fig. 3). They reproduce the expected qualitative behaviour: for the sample at unitarity and on the BEC side the spin susceptibility is strongly suppressed relative to the compressibility. This reflects the fact that the atoms form bound molecules or generalized Cooper pairs. The spin susceptibility should be exponentially small in the binding energy, while the enhanced compressibility reflects the bosonic character of the molecular condensate. This new technique is directly applicable to studying pairing and magnetic ordering of two-component gases in optical lattices [13, 14].

4. Spin gradient demagnetization cooling of lattice-trapped ultracold atoms

Magnetic ordering can occur for delocalized (itinerant) particles, as described above, and for localized particles. Many interesting new quantum phases are predicted for ultracold fermions and bosons localized in optical lattices. One reason for the substantial excitement surrounding this topic is the possibility that the physics of high-temperature d-wave superconductivity in the cuprates [16] is nothing else but the physics of a doped antiferromagnetic Hubbard model [17]. A doped antiferromagnetic Hubbard model is realized in a two-component fermionic Mott insulator at sufficiently low entropy and temperature. It looks promising to use the tools and precision of atomic physics to attack this important, long-standing, and open question of the origin of pairing in high-$T_c$ superconductors.
However, the transition temperature to superfluidity for a doped Hubbard model is much lower than the transition for magnetic ordering. Therefore, for the time being, the experimental focus is on the achievement of spin ordering in optical lattices. This goal can be pursued in both fermionic and bosonic systems [18, 19]. In fact, both systems can in principle realize a large class of spin Hamiltonians, and each offers advantages and disadvantages for certain Hamiltonians. For example, bosonic atoms usually favour ferromagnetic phases (unless strongly spin dependent lattices and/or interactions are added), whereas fermionic atoms tend to order antiferromagnetically. All magnetically ordered phases require very low temperatures and entropy. The spin entropy per particle has to be lower than $k_B \ln 2$ for a two-component system. One advantage of bosonic systems is that lower entropies are experimentally accessible. In our group, we pursue quantum simulations of both fermionic and bosonic Hubbard models. In this section we discuss our efforts towards spin ordering in bosonic Mott insulators.

It has been shown that two-component bosonic Mott insulators support nontrivial magnetic phases [18, 19]. Exchange-stabilized correlated phases such as the XY ferromagnetic state or the antiferromagnetic state typically have a Curie or Néel temperature on the order of $1/k_B$ times the exchange energy. In an atomic Mott insulator, this energy is proportional to $J^2/U$, where $J$ is the tunneling energy and $U$ is the interaction energy. In a typical system (for example, $^{87}\text{Rb}$ in a lattice with a period of 532 nm and a depth sufficient to produce a Mott insulator), the resulting critical temperature for spin ordering is on the order of 200 picokelvin [20, 21]. This temperature is very cold even for the world of ultracold atoms, and in particular is lower than any temperature ever measured in any system. Clearly, new techniques of thermometry and cooling are required in order to make progress in the quest to observe spin-ordering in optical lattices.

Our approach to this problem has been to make use of the unique properties of the two-component Mott insulator to enable new methods of thermometry and cooling. Here we will discuss these unique properties before going on to discuss a recently demonstrated refrigeration technique which depends on them. The two-component Mott insulator supports the same spectrum of particle-hole excitations as the one-component Mott insulator. At sufficiently low temperatures, these excitations are localized at concentric ellipsoidal shells at the boundary between two Mott domains of definite occupation number [21, 22]. This entropy segregation, which has often been referred to as the “icing” on the “wedding cake” of the Mott insulator, has given rise to a number of proposed cooling schemes [23–30], many of which work by differentially addressing and removing the entropy-rich regions of the Mott insulator. Despite the great excitement surrounding the quest for lower temperatures in the Mott insulator, none of these techniques have been realized experimentally. One reason for this may be the difficulty of controlling and addressing the entropy-rich layers. For typical trap frequencies, atom numbers, and temperatures, the width of these layers is often as little as one or two lattice sites, and the energy spectrum of the particle-hole excitations cannot be easily controlled without greatly deforming the trapping potential.

However, the two-component Mott insulator supports an additional spectrum of excitations, more amenable to experimental control, which are not present in its one-component incarnation. These can be thought of as spin excitations. In a finite magnetic field gradient, the two spins will be pulled to opposite sides of the trap, assuming they have different magnetic moments. At zero temperature there will be a zero-width boundary between these two spin-purified regions. The spin excitations consist of atoms with spin $a$ crossing into the spin-$b$ region, and vice versa. The crucial point is that the energy of these excitations is linearly proportional to the applied magnetic field gradient. Since the applied gradient can be easily changed, this gives the experimenter a “handle” on the energy scale of the spin excitations which is not available in the case of particle-hole excitations. It is this “handle” on the energy spectrum of spin excitations which enables the new methods of thermometry and refrigeration that our group has
Figure 4. Illustrations of the new cooling technique. Left: A cartoon of spin gradient demagnetization cooling. The top image is of a sample with many particle-hole excitations but no spin excitations, and the bottom image is of a sample with no particle-hole excitations but many spin excitations. These are intended to represent the sample before and after reduction of the gradient. Right: Calculated effect of spin gradient demagnetization cooling on the entropy distribution. These are the results of the theoretical calculations discussed in Ref. [32]. All images are cuts through the centre of the atomic cloud, and all images are at approximately the same total entropy. The applied gradient is high in a, moderate in b, and low in c. The pumping of entropy from the particle-hole excitations (the rings) to the spin excitations (the stripe in the middle) is clearly visible.

recently developed. For example, at finite temperatures, there will exist a region of mixed spin between the two pure-spin regions. The width of this region will depend on the ratio between the temperature and the applied field gradient. This allows the experimenter to adjust the gradient so that the temperature one wishes to measure will correspond to an easily measurable width of the mixed-spin region. We have used this effect to demonstrate a new kind of thermometry called spin gradient thermometry [31]. The experimental simplicity of this technique is a direct result of the ability to tune the energy of spin excitations using the gradient.

Our group has also recently used the two-component Mott insulator to realize a new method of refrigeration. This technique, called spin gradient demagnetization cooling, works in a lattice and uses the spin excitations discussed above as a refrigerant. It is in some ways locally analogous to adiabatic demagnetization refrigeration in condensed matter systems [33–35]. We believe that it is capable of cooling a two-component Mott insulator of rubidium atoms below the critical temperature for spin ordering.

The new cooling method is applied to an optically trapped cloud of cold atoms in a mixture of two internal states with different magnetic moments. As discussed above, application of a suitable magnetic field gradient will result in partial spatial segregation of the two spin components, with a mixed region in the centre. This region, whose width is proportional to the temperature, comprises a spectrum of soft and easily measurable spin excitations, the energy of which can be tuned by adjusting the strength of the magnetic field gradient. The spin system can be regarded as isolated from all other degrees of freedom when the gradient is changed on a time scale faster than the spin relaxation rate. This regime enables the realization of spin distributions with a very low positive (or, if the sign of the gradient is changed, negative) temperature. Contrastingly, when the gradient is changed adiabatically, the spin system is fully equilibrated. In this regime, reduction of the gradient reduces the energy of the spin degrees
of freedom, causing entropy to flow into the mixed-spin region from other degrees of freedom. This entropy redistribution (visible in Fig. 4) lowers the temperature of the whole system [32].

This technique is locally analogous to adiabatic demagnetization refrigeration, but there are important differences between the two techniques. For example, all prior realizations of demagnetization cooling (including one in an ultracold gas of chromium [35]) required spin-flips. Spin flip collisions in atomic gases are usually very slow, except for atoms with high magnetic moments such as chromium. However, spin gradient demagnetization cooling, because it uses a magnetic field gradient instead of a spatially homogeneous field, proceeds via spin transport in a system with fixed magnetization. This makes it well suited to ultracold atomic samples in optical lattices. On a practical level, it is also easier to achieve very small magnetic field gradients than very small magnetic fields. In our system, an energy resolution of $k_B \times 25$ picokelvin can be relatively easily achieved with a gradient of 1 mG/cm and an optical resolution of 5 µm, while similar energy resolution in a homogeneous system would require control of the magnetic field at the microgauss level (assuming a magnetic moment of one Bohr magneton).

We have used this method to prepare isolated spin distributions at positive and negative spin temperatures of ±50 picokelvin, and to reduce the temperature of an apparently equilibrated sample of rubidium atoms in a Mott insulating state to 350 picokelvin [36]. These are the lowest temperatures ever measured in any system. This new cooling method not only opens up a previously inaccessible temperature regime, but also provides a realistic path to the observation of magnetic quantum phase transitions in optical lattices.

5. Conclusion
The physics of phase transitions in spin systems is extremely rich and has led to many important insights into the nature of phase diagrams. It is safe to predict that this will be an important application for ultracold atoms, probably as important as the previous studies of superfluidity in bosonic and fermionic gases, if it is possible to prepare such systems using the tools of atomic physics. The major obstacle is the realization of practical and efficient cooling schemes which work at picokelvin temperatures in optical lattices. What is needed is a technique which works as well as the magneto-optical trap at microkelvin temperatures or evaporative cooling at nanokelvin temperatures. If the solution is as simple as these techniques, then there is a lot of excitement ahead of us!

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