Superfluid transitions in bosonic atom-molecule mixtures near Feshbach resonance

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We study bosonic atoms near a Feshbach resonance, and predict that in addition to a standard normal and atomic superfluid phases, this system generically exhibits a distinct phase of matter: a molecular superfluid, where molecules are superfluid while atoms are not. We explore zero- and finite-temperature properties of the molecular superfluid (a bosonic, strong-coupling analog of a BCS superconductor), and study quantum and classical phase transitions between the normal, molecular superfluid and atomic superfluid states.

Experimental realizations and coherent manipulation of trapped degenerate gases \(^1\ 2\) is leading to exciting possibilities for studies of quantum liquids in previously unexplored (e.g., extremely coherent and nonequilibrium) regimes. Magnetic field-induced Feshbach resonance (FBR) in ultracold atom collisions allows fine tuning of interactions in these quantum fluids, and was recently used to create a degenerate mixture of coherently-coupled alkali atoms and their diatomic molecules \(^3\).

In this Letter we study phases and phase transitions that take place in bosonic atom-molecule mixtures. Our main contribution is the prediction of a thermodynamically distinct “molecular superfluid” (MSF) phase, that, as illustrated in Figs. \(^1\ 2\) ubiquitously intervenes between the “normal” (N) and “atomic superfluid” (ASF) phases. Molecular superfluidity [and accompanying off-diagonal long-range molecular order (ODLRO)] distinguishes MSF from the normal state, and the absence of atomic superfluidity from the ASF, in which both bosonic atoms and molecules display ODLRO. If atomic and molecular components can be imaged independently \(^4\), in a harmonic trap MSF should be easily identifiable by a sharp Bose-Einstein condensation (BEC) peak in the molecular density profile and a broad, seemingly normal, thermal atomic cloud.

As a conventional superfluid, MSF is characterized by a (molecular) acoustic second-sound mode. However, MSF also exhibits a *gapped*, Bogoliubov-like mode, derived from unpaired atom excitations. MSF ground state (bosonic analog of the BCS state) exhibits strong (atom and molecule) pairing correlations that in a trap should be observable in the atomic density-density correlation function. Experimentally, MSF should be accessible by tuning temperature, atomic density (or number), and detuning \(\nu\). The MSF-ASF transition is in the \((d + 1)\)- and \(d\)-dimensional Ising universality classes for \(T = 0\) and finite \(T\), respectively, and is *reentrant* as a function of detuning \(\nu\) and density \(n\). The tricritical point, where N, MSF and ASF meet, exhibits nontrivial and, to our knowledge, unexplored quantum critical behavior for \(d < 4\). We now sketch derivation of these results.

Near a FBR a bosonic atom-molecule system is characterized by the grand-canonical Hamiltonian \(\hat{H}_\mu = \hat{H} - \mu N\) \(^5\)

\[
\hat{H}_\mu = \int d^d x \left[ \sum_{\sigma=1}^2 \left( \hat{\psi}_\sigma \hat{\psi}_\sigma^\dagger \hat{h}_\sigma + \frac{g_\sigma}{2} \hat{\psi}_\sigma \hat{\psi}_\sigma^\dagger \hat{\psi}_\sigma \hat{\psi}_\sigma^\dagger \right) + g_{12} \hat{\psi}_1^\dagger \hat{\psi}_2^\dagger \hat{\psi}_2 \hat{\psi}_1 - \alpha \left( \hat{\psi}_1^\dagger \hat{\psi}_1^\dagger \hat{\psi}_2 + \text{h.c.} \right) \right] \tag{1}
\]

where \(\hat{\psi}_\sigma(x), \hat{\psi}_\sigma^\dagger(x)\) are bosonic field operators for atoms \((\sigma = 1)\) and molecules \((\sigma = 2)\), \(\hat{h}_\sigma = -(\hbar^2/2m_\sigma) \nabla^2 - \mu_\sigma\) are the corresponding single particle Hamiltonians (focusing for concreteness on the case of a homogeneous trap) with effective chemical potentials \(\mu_1 = \mu\) and \(\mu_2 = 2\mu - \nu\). Chemical potential \(\mu\) tunes the average total number of atoms (whether free or bound into molecules) to \(N\), and detuning \(\nu\) is related to the energy of a molecule at rest, that can be experimentally controlled with a magnetic field. In the dilute gas limit
$g_1, g_2, g_{12}$ are proportional to the 2-body s-wave atom-atom, atom-molecule and molecule-molecule scattering lengths, respectively, and $\alpha$ characterizes coherent atom-molecule interconversion rate, encoding that molecules are composed of two atoms.

The mean-field phase diagram as a function of $\mu_{1,2}$ and $\beta = 1/k_B T$ can be worked out by minimizing the imaginary-time ($\tau$) coherent-state action $S = \int_0^\beta d\tau \int d^d x \sum_{\sigma=1}^2 \left[ \psi_\sigma^* \hat{h}_\tau \psi_\sigma + H_\mu(\psi_\sigma^*, \psi_\sigma) \right]$. Simple analysis leads to three thermodynamically distinct phases (Fig. 1): (i) “normal” (N): $\Psi_{10} \equiv \langle \psi_1 \rangle = 0$, $\Psi_{20} \equiv \langle \psi_2 \rangle = 0$, (ii) “molecular superfluid” (MSF): $\Psi_{10} = 0, \Psi_{20} \neq 0$, (iii) “atomic superfluid” (ASF): $\Psi_{10} \neq 0, \Psi_{20} \neq 0$. Condensed atoms cause $\alpha$ to act as an effective field on the molecular order parameter $\Psi_{20}$, so an equilibrium phase in which atoms are condensed, but molecules are not, is forbidden 3.

We now examine in more detail these phases and corresponding phase transitions. Phase N is stable for $\mu_{1,2} < 0$, with $\mu$ determined by the total atom constraint $n = n_1 + 2n_2$, which in the non-interacting limit, appropriate to a dilute weakly interacting gas, is given by:

$$ n = \frac{1}{\Lambda^2} \left[ f_{d/2} (e^{\beta \mu}) + \frac{2(2d+2)}{2} f_{d/2} (e^{\beta(2\mu - \nu)}) \right], \quad (2) $$

where $\Lambda_T = h/\sqrt{2\pi m_1 k_B T}$ is the thermal de Broglie wavelength and $f_{d/2}(z) = \sum_{n=1}^{\infty} z^n/n^{d/2} (|z| < 1)$ is the extended zeta-function.

The N-ASF transition line $T_{c1}(n, \nu)$ occurs at $\nu = 0$ for $\nu > 0$, while the N-MSF line $T_{c2}(n, \nu)$ occurs at $\mu - 2\nu = 0$ for $\nu < 0$ (see Fig. 2). Using the appropriate asymptotics of $f_\alpha(z)$, one obtains from (2):

$$ T_{c\sigma}(n, \nu) = \begin{cases} T_{c0} \left[ 1 + a_\sigma \left( \frac{|\nu|}{k_B T_{c0}} \right)^{2-d/2} \right], & |\nu| \ll k_B T_{c0} \\ T_{c\sigma}^\infty = b_\sigma c^{d/2} T_{c0}, & |\nu| \gg k_B T_{c0} \end{cases}, \quad (3) $$

with $c = 1 + 2^{d-2}/2$, $a_1 = 2^{d+4}/2 |\Gamma(2-d)|/d \zeta(d/2)$, $a_2 = 2^{-d} a_1$, $b_1 = 1$, $b_2 = 2^{1-d+2}/d$ and $T_{c0} = (h^2/2\pi m_1 k_B) [n/c(x/d/2)]^{2/d} T$ the transition temperature at the tricritical point $\nu = 0$.

In the neighborhood of $T_{c1}$ the “massive” molecular field $\hat{\psi}_2$ decouples at low energies (can be safely integrated out of the partition function, leading to an effective quartic coupling $g_1 \rightarrow g_1 \equiv g_1 - 2x^2/|\mu_2|$), and the N-ASF transition is identical to that of a single-component system, continuous so long as $g_1 > 0$. At $T = 0$, the N-ASF transition takes place at vanishing atom density (the N phase is simply a vacuum of atoms), and although nontrivial, is exactly soluble 3, corresponding to a build-up of atomic superfluid (with condensate density $n_{10} = |\Psi_{10}|^2 \sim |\mu|^{2\beta}$, with mean-field result $\beta = 1/2$ for $d > 2$, and $\beta = d/4$ for $d < 2$) as the trap is loaded. At $T \neq 0$ the N-ASF transition lines in the usual $d$-dimensional XY-universality class 7.

Similarly, in the neighborhood of $T_{c2}$, $\hat{\psi}_1$ decouples and the resulting N-MSF transitions are in the same universality classes discussed above. The full phase boundary is illustrated in Fig. 2. In 3d it exhibits a square-root singularity at the tricritical point and for $|\nu| \rightarrow \infty$ asymptotes to the single-component BEC temperatures $T_{c\sigma}$.

To study the MSF phase, we separate $\hat{\psi}_\sigma = \hat{\psi}_{\sigma 0} + \phi_\sigma$ into classical condensate fields $\hat{\psi}_{\sigma 0}$ (with $\Psi_{10} = 0$ inside MSF) and fluctuations about it. Within MSF it is sufficient to expand $H_\mu$ to second order in fluctuations $\phi_\sigma$, which leads to $H_{\mu} = E^{(0)}(\Psi_{20}) + H^{(2)}$ with:

$$ E^{(0)} = \int d^d x \left( \sum_{\sigma=1}^2 \hat{\psi}_{\sigma 0} \hat{h}_2 \psi_{\sigma 0} + \frac{g_2}{2} |\psi_{20}|^4 \right) \quad (4) $$

$$ H^{(2)} = \int d^d x \left[ \sum_{\sigma=1}^2 \left( \hat{\phi}_\sigma \hat{\phi}_\sigma + \frac{1}{2} \left( \lambda_0 \phi_- \phi_+^* + h.c. \right) \right) \right], $$

in which $\hat{h}_2 = \hat{h}_\sigma + r_1$, $r_1 = g_{12} |\Psi_{20}|^2$, $r_2 = g_{22} |\Psi_{20}|^2$, $\lambda_1 = -2\alpha |\Psi_{20}|^2$, and $\lambda_2 = g_{22} |\Psi_{20}|^2$. The linear term in $\phi_\sigma^\dagger \phi_\sigma$ vanishes automatically by the self-consistent choice of $\Psi_{20}$ as the true minimum of the free energy. To lowest order this gives

$$ n_{20} = |\Psi_{20}|^2 = (2\mu - \nu)/g_2, $$

which coincides with the minimum of $E^{(0)}$ and allows us to eliminate $\mu$ in favor of $\nu$ and $n_{20}$.

For a homogeneous system, $H^{(2)}$ may be diagonalized by Fourier transformation $\phi_\sigma = V^{-1/2} \sum_k e^{i k \cdot x} \phi_{\sigma k}$, followed by independent Bogoliubov transformations on atoms and molecules to new boson operators $\gamma_{\sigma,k}, \gamma_{\sigma,k}^\dagger$:

$$ \hat{\gamma}_{\sigma,k} = u_{\sigma,k} \hat{a}_{\sigma,k} + v_{\sigma,k} \hat{a}_{\sigma,-k}^\dagger, $$

$$ |u_{\sigma,k}|^2 = 1 + |v_{\sigma,k}|^2 = \frac{1}{2} \left( \frac{\tilde{E}_{\sigma,k}}{E_{\sigma,k}} + 1 \right) $$

$$ \hat{H}^{(2)} = \sum_{\sigma,k} E_{\sigma,k} \left( \gamma_{\sigma,k}^\dagger \gamma_{\sigma,k} - |v_{\sigma,k}|^2 \right), \quad (6) $$

FIG. 2: Phase diagram for a bosonic atom-molecule mixture in $d = 3$, expressed in terms of detuning $\nu$ and temperature $T$. It illustrates a finite-$T$ tricritical point at $T_{c0}$ and a quantum critical point at $\nu_c(n,0)$. In the weakly-interacting limit appropriate to experiments the ratio $T_{c1}/T_{c2} = 2^{5/3}$.
in which \( \bar{\sigma}_{\sigma,k} = \gamma_{\sigma,k} - \mu_{\sigma} + r_{\sigma}, \gamma_{\sigma,k} = h^2k^2/2m_{\sigma} \) and 
\[ E_{\sigma,k} = \sqrt{\frac{\bar{\epsilon}_{\sigma,k}^2}{\epsilon_{\sigma,k}} - |\lambda_{\sigma}|^2}, \] 
and \( v_{\sigma,k}, u^*_{\sigma,k} \) have the same phase as \( \lambda_{\sigma} \). Using (4) one obtains 
\[ E_{1,k} = \sqrt{|\epsilon_{1,k} - \nu/2 - (g_{2}/2 - g_{12})n_{20}|^2 - 4\alpha^2n_{20}}, \]
\[ E_{2,k} = \sqrt{\frac{\bar{\epsilon}_{2,k}^2}{\epsilon_{2,k}} + 2g_{2}n_{20} \epsilon_{2,k}}. \] (7)

The MSF ground state is defined as vacuum of atomic and molecular Bogoliubov quasi-particles \( \gamma_{\sigma,k} |\text{MSF} \rangle = 0 \). It is easy to show that it is given by:
\[ |\text{MSF} \rangle = e^{\Psi_{20}^0 \hat{a}_0^{1}_0} \prod_{\sigma,k} e^{-\chi_{\sigma,k} \hat{a}^\dagger_{\sigma,k} \hat{a}^{\dagger}_{\sigma,k} - 0}, \] (8)

which is a coherent state in the \( k = 0 \) molecular mode and shows BCS-like pairing correlations between time-reversed \( (k, -k) \) atomic and molecular single particle states. The amplitude \( \chi_{\sigma,k} = \nu_{\sigma,k}/\nu_{\sigma,k} \) is the Fourier transform of the wavefunction of \( k \neq 0 \) atom (\( \sigma = 1 \)) and molecule (\( \sigma = 2 \)) pairs.

As in a single-component SF the relation (5) (and more fundamentally, the Goldstone theorem (11)) ensures that MSF exhibits a gapless sound mode \( E_{2,k} \approx \hbar s_{k} \), for small \( k \), with \( v_{0} = \sqrt{\frac{g_{2}n_{20}/2m_{1}}}, \) corresponding to collective, long wavelength oscillations of the molecular condensate. The resulting \( |k \rangle \) singularity in \( \chi_{2,k} \) induces a long ranged power law tail in the Fourier transform \( \chi_{2}(r) \sim 1/r^{d+1} \). In addition, we find a gapped excitation branch \( E_{1,k} \), atomic-like excitation modes, whose spectrum and eigenmodes \( (\gamma_{1,k}, \gamma_{1,k}^\dagger) \) do not even carry a definite atom number) are qualitatively distinct from unpaired atomic excitations, \( \hat{a}_{1,k}^{\dagger} \hat{a}_{1,k} \) of the N phase. The condition of being in the MSF is that the atomic gap \( E_{\text{gap}}^{(1)} = \sqrt{\varepsilon_{\pm}^{*}} \), where
\[ \bar{\epsilon}_{\pm} = -\nu/2 - (g_{2}/2 - g_{12})n_{20} \pm 2\alpha\sqrt{n_{20}}, \] (9)
remains positive. Correspondingly, the atomic pair wavefunction \( \chi_{1}(r) \sim e^{-r/\xi_{1}} \) decays exponentially at large distance. The correlation length \( \xi_{1} = \hbar/\sqrt{2m_{-}} \) characterizes atom-pair size, and diverges as \( E_{\text{gap}}^{(1)} \) vanishes. The MSF-ASF transition takes place when the size of atom-pairs becomes comparable to the intermolecular separation, and \( \xi_{1} \) is the diverging size of the coherent exchange loops of atoms between overlapping pairs.

In the present quadratic approximation, the free energy is given by
\[ f = -\mu_{2}|\Psi_{20}|^2 + \frac{g_{2}}{2}|\Psi_{20}|^4 - \frac{1}{2V} \sum_{\sigma,k} (\bar{\sigma}_{\sigma,k} - E_{\sigma,k})^2 \]
\[ + \frac{k_{B}T}{V} \sum_{\sigma,k} \ln(1 - e^{-E_{\sigma,k}/k_{B}T}). \] (10)

The condition of fixed density \( n = -(\partial f/\partial \mu)n_{20} \),
\[ n = 2n_{20} + \frac{1}{V} \sum_{\sigma,k} \sigma \left( u_{\sigma,k}^2 + u_{\sigma,k}^{*2} e^{E_{\sigma,k}/k_{B}T - 1} \right), \] (11)
where the first term under the summation represents the \( T = 0 \) interaction-induced condensate depletion, may be used to determine the condensate density \( n_{20}(n, \nu, T) \). At \( T = 0 \), in the weakly-interacting limit \( (\alpha^2n \ll 1) \), in 3d we find:
\[ n_{20} = \frac{1}{2}n - \frac{2^{(d-2)/2} \Gamma(d+1/2)}{\Gamma(d/2)} \left( \frac{n_{20}}{d \pi^{1/2}} \right)^{d/2} \left[ \frac{(\nu_{\sigma} - \bar{n}_{\sigma})^{1/2}}{2^{d+1} \Gamma(d/2)} - \frac{1}{T_{d}} \left( \frac{\bar{\alpha} \sqrt{n}}{\nu_{\sigma} - \bar{n}_{\sigma}} \right) \right], \] (12)
where \( \bar{n}_{\sigma} = n_{2} - 2a_{12}, a_{2}, a_{12} \) are molecule-molecule and atom-molecule scattering lengths, respectively, defined by \( g_{2}/2 = 4\pi\hbar^2 a_{12}/m_{2} \), the “detuning density” is \( \nu_{\sigma} = m_{1}|\nu|/(\pi\hbar^2 a_{12}) \), \( a = 3/2 \bar{\alpha}_{\sigma} a_{12}/\bar{\alpha}_{12} \), and \( \mathcal{I}(y) = y^{2} \int_{0}^{\infty} dx d^{3}x/dx^{2}[(1 + x^{2})^{2} - y^{2}]^{-3/2} \) is a scaling function describing additional molecular-condensate depletion due to atom pairs. Equation (12), together with the vanishing of \( \varepsilon_{-} \), determines the MSF-ASF phase boundary. At \( T = 0 \), approximating \( n_{20} \approx n/2 \), one finds:
\[ \nu_{c}(n) \approx -(g_{2}/2 - g_{12})n - 2\alpha \sqrt{n}, \] (13)

which is illustrated for \( g_{2} > g_{12} \) in Fig. 1. At finite \( T \), in the noninteracting limit, \( u_{\sigma,k} = 0 \) and \( u_{\sigma,k} = 1 \), molecular condensate density reduces to
\[ n_{20}(\nu, T) \approx \frac{n}{2} \left[ 1 - \left( \frac{T}{T_{c2}} \right)^{3/2} \right] = \frac{1}{2\lambda_{T}^{3/2}} \left( e^{-|\nu|/2k_{B}T} \right), \] (14)
with the standard BEC result (first term) corrected by depletion due to thermal atomic excitations (last term), that is exponentially small at large \( |\nu| \) and low \( T \).

From the structure of the interconversion (\( \alpha \)) term in \( \bar{H}_{0} \), it is clear that it is a Z_{2} (Ising) symmetry that is broken at the MSF→ASF transition (11). We now show that for a homogeneous trap, both the \( T = 0 \) and finite \( T \) MSF-ASF transitions lie in the Ising universality class. This can be most easily seen from the coherent state action, which, when expressed in terms of real and imaginary parts of the atomic field \( \psi_{1} = \psi_{R} + i\psi_{I} \), in MSF phase reduces to:
\[ S = \int_{0}^{\beta} d\tau \int d^{3}x \left[ -2i\psi_{I} \hbar \partial_{\tau} \psi_{R} + \psi_{R} (\bar{\hbar}_{1} - 2\alpha \bar{\Psi}_{20}) \psi_{R} \right. \]
\[ \left. + \psi_{I} (\bar{\hbar}_{1} + 2\alpha \Psi_{20}) \psi_{I} \right] + S_{\text{int}} \] (15)
where \( S_{\text{int}} \) terms are not essential for our argument. In this form it is clear that in the presence of the molecular condensate, \( |\Psi_{20}| > 0 \), positive \( \alpha \) reduces the \( O(2) \) symmetry to \( Z_{2} \), with \( \psi_{R} \) reaching criticality before \( \psi_{I} \). Because canonically conjugate field \( \psi_{I} \) remains “massive” at the critical point [defined by where the coefficient of \( \psi_{I}^{2} \) vanishes, consistent with (15)], it can be safely integrated.
out and leads to a $d+1$-dimensional (Lorentz-invariant) action even in the scalar order parameter $\psi_R$. Therefore, as asserted above, the $T = 0$ MSF-ASF transition is in the $(d+1)$-dimensional Ising universality class. The Ising transition is well studied, and leads to the following predictions \[10\]. For $d = 3$, up to logarithmic corrections, the mean-field theory derived above will be accurate. On the other hand in 2d, MSF-ASF exponents are nontrivial but are well-known. For example, standard scaling arguments predict:

\[ n_{10} \sim |\nu - \nu_c|^{2\beta_I}, \quad E_{\text{gap}}^{(1)} \sim |\nu - \nu_c|^{2\nu_I}, \]

where $\beta_I \approx 0.31$, $z_I = 1$, and $\nu_I \approx 0.63$ are 3d Ising exponents. These, together with the relevance of $T$ at this quantum critical point also imply a universal shape of the MSF-ASF phase boundary $\nu_c(n, T) \sim \nu_c(n, 0) + a T^{1/\nu_I}$ in Fig. 2. One may hope that when long-lived molecular condensates are produced, nontrivial behavior of $E_{\text{gap}}^{(1)}(\nu)$ maybe observed in Ramsey fringe experiments \[3\].

Although from symmetry point of view ASF state is quite conventional, it also exhibits a set of interesting features not found in a single-component SF. For example, because it is a discrete (Ising) symmetry that distinguishes ASF from MSF, it will exhibit a gapped (Ising) order parameter, because it is a discrete (Ising) symmetry that distinguishes ASF from MSF, it will exhibit a gapped (Ising) order parameter. For example, standard scaling arguments predict:

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Another experimentally interesting feature of ASF is that a seemingly standard $2\pi$ vortex in atomic condensate $\Psi_{20}$ will generically split into two $\pi$ vortices (see Fig. 4) confined by a domain wall of length

\[ r_0 = \sqrt{h^2 n_{20}^{3/2}/2m a_n^{3/2}}, \]

that diverges as ASF$\rightarrow$MSF phase boundary is approached \[12\]. This arises because in large $\alpha$ limit a molecular condensate. Such double molecular vortex is unstable to two fundamental $2\pi$ molecular vortices, that, in 2d repel logarithmically, but are confined linearly inside the ASF phase.

Finally, standard renormalization-group analysis shows that upper-critical dimensions for the tricritical point (dominated by scaling of $\alpha$) are $d_{uc}^c = 6$ and $d_{uc}^e = 4$, and therefore should display nontrivial critical properties \[12\].

We plan to present analysis of these problems as well as generalization to experimentally relevant harmonic trap in a future publication.

To conclude, we have studied a mixture of bosonic atoms and their diatomic molecules and predicted an existence of a molecular superfluid phase, qualitatively distinct from the normal and atomic superfluid states. Ising transition MSF-ASF should be observable in such systems as a function of detuning $\nu$, temperature and/or density.

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\[11\] In the “hard-spin” $\psi_\alpha = e^{i\theta_\alpha}$ description, interconversion term reduces to $\alpha \cos(2\theta_1 - \theta_2)$, locking atomic phase to the phase of the molecular condensate up to $\pi$, corresponding to the unbroken $Z_2$ symmetry of the MSF. In terms of a vector order parameter, MSF is characterized by a finite quadrupole moment ($l = 2$ angular harmonic, akin to nematic order in liquid crystals), and ASF by a vector order parameter ($l = 1$ harmonic).

\[12\] L. Radzihovsky, unpublished.