Trimers, Molecules, and Polarons in Mass-Imbalanced Atomic Fermi Gases

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We consider the ground state of a single ''spin-down'' impurity atom interacting attractively with a ''spin-up'' atomic Fermi gas. By constructing variational wave functions for polarons, molecules, and trimers, we perform a detailed study of the transitions between these dressed bound states as a function of mass ratio $r = m_\uparrow/m_\downarrow$ and interaction strength. Crucially, we find that the presence of a Fermi sea *enhances* the stability of the p-wave trimer, which can be viewed as a Fulde-Ferrell-Larkin-Ovchinnikov molecule that has bound an additional majority atom. For sufficiently large r, we find that the transitions lie outside the region of phase separation of the imbalanced Fermi gas and should thus be observable in experiment, unlike the well-studied equal-mass case.

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The spin-imbalanced Fermi gas has received much attention recently owing to its elegant realization in ultracold atomic gases. The ability to tune both the interspecies interaction and the spin polarization has allowed cold-atom experiments to access the rich phase diagram of the spin-imbalanced system [1]. However, as experiments become ever more precise, a challenge for theory is to accurately determine the existence of exotic phases in the regime of strong correlations.

One approach that has proven useful at high polarizations is to consider the problem of a *single* spin-down impurity atom immersed in a Fermi gas of spin-up atoms [2–4]. Such a scenario is just one example of the canonical ''polaron'' problem, the solution of which is used to construct the low-energy behavior of the many-body system. Moreover, this limit of full polarization contains some of the critical points of the full zero-temperature phase diagram; e.g., it features the tricritical point that marks the existence of the spatially homogeneous superfluid phase for *all* polarizations [5]. Thus, an analysis of the high-polarization limit allows one to characterize parts of the topology of the whole phase diagram.

An important feature of the single-impurity problem is that it can exhibit binding transitions where the impurity changes its statistics and/or effective mass for a sufficiently strong attractive interaction. Thus far, the focus has been on equal masses $m_\downarrow = m_\uparrow$, where it has been shown that the impurity undergoes a first-order transition from a polaron (an impurity dressed with particle-hole excitations) to a molecule (an impurity bound to a single majority fermion) [4]. Here we demonstrate that a richer variety of impurity-atom ground states can be obtained when the masses are unequal, with $m_\downarrow < m_\uparrow$. In this case, the presence of a spin-up Fermi sea can favor a dressed molecule with *nonzero* ground-state momentum, corresponding to the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superfluid phase in the limit of extreme imbalance. However, we find that the FFLO molecule usually binds another majority atom to form a p-wave trimer. Thus, as one increases the density of majority spins from the limit of zero density, one finds that the stability of the trimer is initially *enhanced* by the Fermi sea.

In this Letter, we map out the ground-state phase diagram for the dressed impurity as a function of mass ratio $r = m_\uparrow/m_\downarrow$ and interaction strength. We also determine whether or not our dressed-impurity phase diagram is thermodynamically stable in the highly imbalanced Fermi gas, by estimating the onset of phase separation in this limit by using the results of previous quantum Monte Carlo (QMC) simulations. In contrast to the equal-mass case, we find that the polaron-trimer binding transitions sit *outside* of the phase-separated region and should thus be experimentally accessible.

In the following, we use the Hamiltonian for a two-component (we call the two species $\uparrow$ and $\downarrow$) atomic Fermi gas interacting via a wide Feshbach resonance:

$$H = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \frac{g}{V} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} c^\dagger_{\mathbf{k}\uparrow} c^\dagger_{\mathbf{k}'\downarrow} c_{\mathbf{k}'+\mathbf{q}\downarrow} c_{\mathbf{k}-\mathbf{q}\uparrow}, \quad (1)$$

where $\epsilon_{\mathbf{k}\sigma} = \frac{\mathbf{k}^2}{2m_\sigma}$ (we set $\hbar = 1$), V is the system volume, and g is the strength of the attractive contact interaction. The s-wave scattering length a_s is then obtained via the prescription $\frac{m_r V}{4\pi a_s} = \frac{V}{g} + \sum_{\mathbf{k}}^{\Lambda} \frac{1}{\epsilon_{\mathbf{k}\uparrow} + \epsilon_{\mathbf{k}\downarrow}}$, where the ''reduced mass'' $\frac{2}{m_r} = \frac{1}{m_\downarrow} + \frac{1}{m_\uparrow}$ and Λ is a UV cutoff that can be sent to infinity at the end of the calculation. Note that since the $\downarrow$ impurity is distinguishable from the $\uparrow$ Fermi sea, our results in the single-impurity limit are relevant to both Fermi gases and Bose-Fermi mixtures.

We construct variational wave functions for the single-impurity atom immersed in a Fermi sea by considering different numbers of particle and particle-hole pair

excitations upon the Fermi sea. Previous studies have shown this approach to be reasonably accurate [6]. The novelties of our work are to thoroughly explore the case of unequal masses, to allow the dressed impurity to have nonzero momentum, and to consider the possibility of trimers as well as polarons and molecules. We also considered tetramers at angular momentum $L = 1$, but they did not bind anywhere for the parameters we considered.

We adopt the following nomenclature for the different trial wave functions: A subscript n refers to a state with at most n operators acting on the noninteracting Fermi sea, and a bracketed momentum refers to the total momentum of the state. For example, the polaronic state $P_3(\mathbf{Q})$ will contain the impurity atom and at most one particle-hole pair on top of the Fermi sea, with a total momentum $\mathbf{Q}$. In our explicit calculations we concentrate on states with at most one particle-hole pair, since these are known to provide a good approximation for the impurity energy when $m_\uparrow = m_\downarrow$ [6–9] and going to higher order greatly increases the numerical effort. Thus, the wave function for the polaron is [2,6]

$$|P_3(\mathbf{Q})\rangle = \alpha^{(\mathbf{Q})} c^\dagger_{\mathbf{Q}\downarrow}|\mathrm{FS}\rangle + \sum_{\mathbf{k},\mathbf{q}} \beta^{(\mathbf{Q})}_{\mathbf{k}\mathbf{q}} c^\dagger_{\mathbf{Q}+\mathbf{q}-\mathbf{k}\downarrow} c^\dagger_{\mathbf{k}\uparrow} c_{\mathbf{q}\uparrow}|\mathrm{FS}\rangle,$$

where $|\mathrm{FS}\rangle$ is a Fermi sea of majority atoms, filled up to momentum $k_{F\uparrow}$. The presence of a Fermi sea implies that the spin-up hole momentum $|\mathbf{q}| \equiv q < k_{F\uparrow}$ and the spin-up particle momentum $\mathbf{k}$ satisfies $k_{F\uparrow} < k < \Lambda$. Likewise, for the molecule, the wave function is [7–9]

$$|M_4(\mathbf{Q})\rangle = \sum_{\mathbf{k}} \gamma^{(\mathbf{Q})}_{\mathbf{k}} c^\dagger_{\mathbf{Q}-\mathbf{k}\downarrow} c^\dagger_{\mathbf{k}\uparrow}|\mathrm{FS}\rangle + \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} \delta^{(\mathbf{Q})}_{\mathbf{k}\mathbf{k}'\mathbf{q}} c^\dagger_{\mathbf{Q}+\mathbf{q}-\mathbf{k}-\mathbf{k}'\downarrow} c^\dagger_{\mathbf{k}\uparrow} c^\dagger_{\mathbf{k}'\uparrow} c_{\mathbf{q}\uparrow}|\mathrm{FS}\rangle.$$

Finally, for the trimer, we set $\mathbf{Q} = 0$ and approximate the wave function as being insensitive to the hole momentum $\mathbf{q}$ by using its value at $\mathbf{q} = 0$ (this approach has been studied for the polaron [6] and molecule [8,9] and can be shown to give an upper bound to the binding energy):

$$|T_5(0)\rangle = \sum_{\mathbf{k}_1,\mathbf{k}_2} \tau_{\mathbf{k}_1\mathbf{k}_2} c^\dagger_{-\mathbf{k}_1-\mathbf{k}_2\downarrow} c^\dagger_{\mathbf{k}_1\uparrow} c^\dagger_{\mathbf{k}_2\uparrow}|\mathrm{FS}\rangle + \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k},\mathbf{q}} \eta_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k},0} c^\dagger_{\mathbf{q}-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}\downarrow} c^\dagger_{\mathbf{k}_1\uparrow} c^\dagger_{\mathbf{k}_2\uparrow} c^\dagger_{\mathbf{k}\uparrow} c_{\mathbf{q}\uparrow}|\mathrm{FS}\rangle.$$

Here, we set the total angular momentum to be $L = 1$ and choose $L_z = 0$, in which case the bare part is $\tau_{\mathbf{k}_1\mathbf{k}_2} = \hat{k}_1 \cdot \hat{z} F(k_1, k_2, \hat{k}_1 \cdot \hat{k}_2) - \hat{k}_2 \cdot \hat{z} F(k_2, k_1, \hat{k}_1 \cdot \hat{k}_2)$ with any function F [10]. Unlike the polaron and molecule, which both have $L = 0$ in the ground state, we find that this $L = 1$ odd-parity trimer is always the lowest energy state throughout the portion of the $\{1/k_{F\uparrow}a_s, r\}$ phase diagram where the trimer is stable. We have not yet explicitly looked at trimer states with nonzero momentum, but we expect the trimer to have $\mathbf{Q} = 0$ in its ground state.

In the limit $k_{F\uparrow} \to 0$, this $L = 1$ trimer becomes the 3-particle bound state in a vacuum, for which analytical solutions have been found [11]. Here, it has been shown that the trimer is bound relative to a molecule and an extra particle for $r > r_{C1} \cong 8.17$. However, for $r > r_{C2} \cong 13.6$, the energy of the trimer is no longer finite in the limit $\Lambda \to \infty$. This shows up as a wave function with weight at increasingly high momenta as one approaches r_{C2} from below. This critical r_{C2} is independent of $k_{F\uparrow}$, since it relies on high momenta. Thus, the results for the trimer are always cutoff-dependent once $r > r_{C2}$.

We solve for the ground-state energy of a given normalized wave function $|\psi\rangle$ by minimizing the energy $E = \langle\psi|H|\psi\rangle$ with respect to the amplitudes α, β, γ, δ, τ, and/or η. One can generally sum over one of the momenta and solve a self-consistent equation for the resulting function, where symmetries play a crucial role in simplifying the calculation. Details of the trimer calculation and comments on the accuracy of the results are in the supplemental information section [10].

By determining the minimum energies of the wave functions $P_3(\mathbf{Q})$, $M_4(\mathbf{Q})$, and $T_5(0)$, we constructed an approximation to the ground-state phase diagram of the impurity atom, as depicted in Fig. 1. Here, we find that the polaron phase always has its lowest energy at zero momentum. By contrast, a very small region in Fig. 1 exists where the molecule has *nonzero* momentum in the ground state. We identify this as FFLO since it can be shown that a repulsive, dilute gas of these molecules forms a spatially modulated condensate [12].

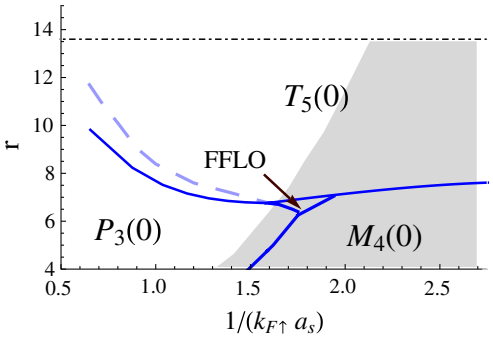

FIG. 1 (color online). The ground-state phase diagram as a function of mass ratio r and interaction strength $1/k_{F\uparrow}a_s$ for the polaron (P_3), molecule (M_4), and trimer (T_5) wave functions. The FFLO region corresponds to M_4 with nonzero momentum; the momentum of the FFLO molecule is approximately $k_{F\uparrow}$ at the P_3-T_5-FFLO triple point and goes continuously to zero at the FFLO-$M_4(0)$ transition line, coming in as a square root near the transition. The T_5-M_4 boundary (full [blue] line) approaches the three-body transition $r_{C1} \simeq 8.17$ in the limit $1/k_{F\uparrow}a_s \to \infty$, as expected. Above the dashed-dotted ($r = r_{C2}$) line, the results for T_5 become cutoff-dependent and are therefore no longer universal. The shaded region marks where the system is unstable to phase separation. The dashed (blue) line marks the transition line where a metastable polaron rapidly binds into a FFLO molecule which then binds into a trimer.

The FFLO state has a very natural interpretation in this limit: Having a molecule with $\mathbf{Q} = 0$ requires the impurity atom to have momentum $k > k_{F\uparrow}$, which is disfavored due to its high kinetic energy as r increases. However, as r is increased farther, it quickly becomes favorable for the FFLO molecule to bind another majority fermion and form a p-wave trimer at zero total momentum. Indeed, at large enough $1/(k_{F\uparrow}a_s)$, the transition is directly from the $\mathbf{Q} = 0$ molecule to the trimer, without any intervening FFLO state. A surprising result is that, as one approaches unitarity, the Fermi surface actually favors the trimer over the molecule and we have a direct transition from polaron to trimer. We expect this to be a robust result since our approximation for T_5 underestimates the trimer binding energy.

How are these impurity-atom ground states distinct from one another? For the polaron, the ground-state wave function (including dressing with an arbitrarily large number of particle-hole pairs) has a nonzero weight $Z = |\alpha^{(\mathbf{Q})}|^2$ for the "bare" impurity atom. We find a positive effective mass m^* for the polaron, implying that Z is only nonzero for $\mathbf{Q} = 0$: A polaron at nonzero $\mathbf{Q}$ can lower its momentum by emitting a particle-hole pair. This weight Z is an "order parameter" for the polaron state.

For the molecule $Z = 0$, but a fully dressed ground state will have a nonzero weight $Z_M = \sum_{\mathbf{k}} |\gamma_{\mathbf{k}}^{(\mathbf{Q})}|^2$ for the bare molecule at one or more values of $\mathbf{Q}$. The molecule is FFLO when this nonzero weight appears at $Q > 0$. Thus again we can consider this nonzero Z_M, as well as the momentum Q where it occurs, as the order parameters of the molecule states. Similarly, the trimer phase has nonzero $Z_T = \sum_{\mathbf{k}_1\mathbf{k}_2} |\tau_{\mathbf{k}_1\mathbf{k}_2}|^2$ for the bare trimer.

Next, we examine the nature of the binding transitions. For the case of equal masses, the matrix elements connecting the polaron and molecule go to zero at the transition point due to the transition involving a "decay" into at least four particles [4]. Thus, one can have a long-lived molecule (polaron) existing on the polaron (molecule) side of the transition, with the lifetime $\Gamma^{-1} \sim (\Delta E)^{-9/2}$ diverging faster than the inverse of the energy difference ΔE as one approaches the unbinding transition [13]. This first-order binding transition occurs when the momenta of the two ground states do not differ by $k_{F\uparrow}$. For example, when the molecule and polaron both have their minimum energy at $\mathbf{Q} = 0$, for the molecule to decay into a polaron, it must create two particles and a hole, by momentum conservation. This 4-particle process leads to a first-order transition with discontinuous jumps in the order parameters. Referring to Fig. 2, this follows from the displacement of the molecule dispersion from the polaron + particle continuum by $k_{F\uparrow}$. The same considerations apply to the zero-momentum trimer unbinding into a zero-momentum molecule plus a particle.

However, if the molecule ground state has $Q = k_{F\uparrow}$, then it can unbind in a continuous fashion into either a $Q = 0$ polaron plus a particle (see Fig. 2) or a $Q = 0$ trimer plus a hole. This behavior shows up in the decay rates of the excited states on either side of the transition, which we estimate by using Fermi's golden rule like in Ref. [13]. We find that the unbinding of a FFLO molecule into a trimer or polaron has decay rate $\Gamma \sim \Delta E$ as $\Delta E \to 0$, where, once again, ΔE is the energy difference between the two phases near the transition. Therefore, this is a marginal case where the lifetime Γ^{-1} diverges as fast as the inverse of ΔE. The decay of a trimer or polaron into a FFLO molecule is even faster, with Γ being finite as $\Delta E \to 0$, since the momentum of the final-state FFLO molecule can lie anywhere on the Fermi surface and thus the phase space is enlarged. Finally, the transition from the zero-momentum molecule to the FFLO molecule is not an unbinding transition, and we find that $\mathbf{Q}$ generally moves continuously away from zero at this transition.

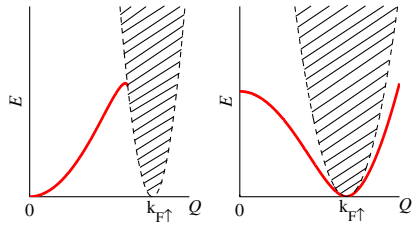

FIG. 2 (color online). Schematics of two scenarios for a molecule unbinding into a polaron + particle. The solid (red) lines represent the molecule dispersion $E(Q)$, and the shaded regions correspond to the polaron + particle (two-body) continuum. When both the molecule and polaron have their minimum energies at $\mathbf{Q} = 0$ (left), the transition is first-order. However, we have a continuous transition (where the bound state fully "mixes" with the continuum) when the molecule has ground-state momentum $Q = k_{F\uparrow}$ (right).

A direct transition from the trimer to the polaron and vice versa would be first-order, since this is a three-body decay: the trimer (polaron) "shedding" two particles (holes). Here the decay rate $\Gamma \sim (\Delta E)^2$ close to the transition, similar to the behavior of a Fermi liquid quasiparticle. This implies that a quench at fixed r from the polaron to trimer phase leads to a metastable polaron that becomes unstable to forming a FFLO molecule (see Fig. 1). If the difference between the FFLO and trimer energies is small here, then there may exist a metastable FFLO state, a scenario which is worthy of investigation.

In experiments on highly imbalanced Fermi gases, one has a nonzero number density $n_\downarrow$ of minority atoms, and the system must be stable against phase separation in order for the single-impurity transitions to be observable. For equal masses, QMC predicts that the single-impurity transition occurs in the phase-separated regime [14], and, indeed, the experimentally measured disappearance of the polaron [15] agrees well with the onset of phase separation between superfluid (SF) and normal (N) phases rather than with the polaron-molecule transition for a single impurity. To estimate the onset of phase separation for general mass ratios, we impose the following coexistence conditions on the pressures P and chemical potentials μ in each phase:

$P_{\rm SF} = P_N$, $\mu_\sigma^{\rm SF} = \mu_\sigma^N$, and $\mu_\downarrow^N = E_b$, where E_b is the binding energy of the impurity immersed in the Fermi gas. Fortunately, for mass ratios $r \geq 1$, the superfluid is likely to be unpolarized ($n_\uparrow = n_\downarrow$) or only weakly polarized at this onset point, and thus we can exploit the QMC equation of state for the unpolarized superfluid. At unitarity, this equation of state has been shown to be relatively insensitive to mass ratio [16], while in the BCS and Bose-Einstein condensation limits, the equation of state is that of a weakly attractive Fermi gas and a weakly repulsive Bose gas, respectively. Thus, we have the pressure and the average chemical potential:

$$P_{\rm SF} = \frac{2^{5/2} m_r^{3/2}}{15\pi^2} \varepsilon_F^{5/2} g(1/k_F a_s, r), \tag{2}$$

$$\mu_{\rm SF} \equiv \frac{\mu_\uparrow^{\rm SF} + \mu_\downarrow^{\rm SF}}{2} = \varepsilon_F f(1/k_F a_s, r), \tag{3}$$

where the functions $g(1/k_F a_s, r)$ and $f(1/k_F a_s, r)$ are determined via numerical interpolation between the known limits, while $k_F = (6\pi^2 n_\uparrow^{\rm SF})^{1/3}$ and $\varepsilon_F = k_F^2/2m_r$.

Clearly, the pressure and spin-up chemical potential in the fully polarized normal phase are known exactly:

$$P_N = \frac{m_r^{3/2}}{15\pi^2}(1+r)^{3/2}(\mu_\uparrow^N)^{5/2} = \frac{m_r^{3/2}}{15\pi^2}\frac{2^{5/2}}{1+r}\varepsilon_{F\uparrow}^{5/2}, \tag{4}$$

where $\varepsilon_{F\uparrow} = (k_{F\uparrow})^2/2m_r$. To determine $\mu_\downarrow^N$, we estimate the binding energy E_b by using the P_3 wave function. Note that at large r, where the polaron is no longer the ground state, this E_b will be an underestimate, and so the calculated onset position $1/k_{F\uparrow}a_s$ will be lower than the exact result in this limit. By applying coexistence conditions, we arrive at the set of equations

$$\left(\frac{k_{F\uparrow}}{k_F}\right)^5 = (1+r) g(1/k_F a_s, r), \tag{5}$$

$$g(1/k_F a_s, r) = (1+r)^{3/2}\left(\frac{f(1/k_F a_s, r)}{1 + (1+r)\frac{E_b}{\varepsilon_{F\uparrow}}}\right)^{5/2}. \tag{6}$$

For equal masses, our calculation gives $1/k_{F\uparrow}a_s \simeq 0.75$, which agrees well with fixed-node QMC calculations [14].

This onset line determines the region of phase separation near unitarity, and this is plotted in Fig. 1. For sufficiently large r, we see that the polaron-trimer transition extends well outside of the regime of phase separation and thus should be observable in the polarized gas. This allows one to investigate first-order binding transitions and the possible metastable states discussed earlier.

A remaining question is what happens to the trimer phase at a finite density of spin-down atoms. In 1D, it is known that a trimer phase exists for $n_\downarrow/n_\uparrow = 1/2$, provided the interactions are sufficiently large [17]. Here, we expect a Fermi liquid of trimers for $n_\downarrow/n_\uparrow \ll 1/2$. However, it is possible that the FFLO phase will eventually beat the trimer phase as $n_\downarrow/n_\uparrow$ is increased. One may also have a mixture of trimers, polarons, and/or molecules as we approach the binding transition.

Experimentally, one can explore our phase diagram by using two atomic species with unequal masses or by artificially increasing the effective mass of the spin-up atoms with a spin-dependent optical lattice. ^{40}K-^{6}Li mixtures are the favored choice, and, in Fig. 1, their mass ratio ($r \simeq 6.7$) just touches the bottom parts of the trimer and FFLO regions. Since each phase has a different effective mass, one can distinguish between them by determining the effective mass using the low-lying compression modes of the gas, as in Ref. [18]. Another possibility is to directly measure the order parameter: Reference [15] has successfully measured Z in the polaron phase by using rf spectroscopy, but it remains an open question whether or not this method can be extended to study Z_M or Z_T.

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