Threshold of molecular bound state and BCS transition in dense ultracold Fermi gases with Feshbach resonance

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Abstract. We consider the normal state of a dense ultracold atomic Fermi gas in the presence of a Feshbach resonance. We study the BCS and the molecular instabilities and their interplay, within the framework of a recent many-body approach. We find surprisingly that, in the temperature domain where the BCS phase is present, there is a non zero lower bound for the binding energy of molecules at rest. This could give an experimental mean to show the existence of the BCS phase without observing it directly.

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Experimental progress in the study of ultracold Fermi gases has proceeded quite recently at a very fast pace. Working in the vicinity of a Feshbach resonance which allows to cover a very wide range of scattering length by sweeping the magnetic field across the resonance, several groups dealing either with ^{40}K [1,2] or ^{6}Li [3–5] have been able to vary the interatomic interaction in such a way that the system goes essentially from a weakly attractive atomic Fermi gas to a dilute gas of diatomic molecules. The clear observation of long lived molecules is one of the very positive outcomes of these experiments. More generally these experiments have shown that it is easy to modify the scattering length a, and consequently interactions, by shifting the magnetic field rapidly enough so that the system does not have time at all to adjust to this change. Hence it is experimentally feasible to prepare the gas in an out-of-equilibrium situation, then to study its evolution from a metastable state and in particular the manifestation of various instabilities.

In this paper we study, throughout the $a-T$ phase diagram, the two instabilities which arise in a normal Fermi gas due to an attractive interaction, namely the molecular and the BCS instabilities. We find an unexpected interplay between these two instabilities. In particular, at temperatures where the BCS phase is present, there is a nonzero threshold for the binding energy of molecules at rest. In other words it should not be possible to observe a molecule with zero binding energy, in contrast to the standard situation for two atoms in vacuum where this binding energy is zero right at the resonance $a^{-1} = 0$. Hence quite

unexpectedly this link between BCS and molecular properties would provide a signature of the presence of the BCS phase just by looking at the molecular spectroscopic properties, which could be easier to observe experimentally than the BCS phase itself.

Actually we have quite recently considered the effect of the Fermi sea on the molecular bound state associated with a Feshbach resonance, when this is the only instability. We note that, since the Fermi sea is responsible for the formation of Cooper pairs, which are some kind of molecules, it is somewhat natural that it does also affect the molecular properties. We have shown that the presence of the Fermi sea shifts the location of the appearance of the molecular state toward positive values for the scattering length [6]. Instead of appearing right at the Feshbach resonance as in vacuum, the molecule appears for an inverse scattering length a^{-1} larger than a positive threshold. Physically this happens because, due to the Fermi sea and Pauli exclusion, the occupation of a number of plane wave states making up the wavefunction of the (large) molecule is prohibited. So building the molecular state is more difficult than in vacuum. Naturally this exclusion effect decreases as T increases since the statistical occupation of states gets lower. The above picture is for molecules with zero momentum for the center of mass. When the molecule has a nonzero total momentum, the adverse effect of the Fermi sea is smaller and for fast molecules there is essentially no effect of the Fermi sea on the molecular bound state formation. This shift of the threshold for appearance of the molecular states is in qualitative agreement with experimental observations of strong losses in ^{6}Li appearing much below the location of the Feshbach resonance [7–9]. However the interpretation of these experiments is obviously complex, with in

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particular dynamical effects, and it remains to be seen how strong is experimentally the role of the effect that we have just described.

Here we will use, as in the above study, the result obtained recently [10] for the scattering amplitude due to a Feshbach resonance, modified by the presence of the dense Fermi gas. Although obtained within a quite general formalism, this result makes use of strong simplifications. Nevertheless it is clearly worth exploring simple physical approximations before going to more elaborate schemes. First the irreducible vertex is taken merely as the simple scattering amplitude of two isolated atoms. Second only Pauli exclusion is taken into account for the effect of the Fermi gas. Nevertheless the resulting scattering amplitude is quite non trivial. In particular, in contrast with the vacuum case, it depends on the total momentum of the scattering atoms because of Pauli exclusion. Here we will concentrate on the case where the total momentum is zero since it displays the strongest manifestation of Pauli exclusion. Also any background scattering is omitted for simplicity.

The expression [10] for the full vertex $\Gamma(\omega)$ in the particle-particle channel, which, except for a factor $\pi/2k_0$, is just the inverse f^{-1} of the effective scattering amplitude, is given by (we take $\hbar = 1$):

$$-\frac{2\pi^2}{mk_0}\frac{1}{\Gamma(\omega)} = \frac{1}{\lambda} - \frac{\bar{\omega}}{\bar{W}} + I(\bar{\omega})$$

$$I(\bar{\omega}) = \int_0^\infty dx \left[1 - \frac{x^2}{x^2 - \bar{\omega}/2}\tanh\frac{x^2-1}{2\bar{t}}\right]. \tag{1}$$

The (positive) chemical potential $\mu \equiv k_0^2/2m$ is our energy scale and k_0 our wavevector scale. We have introduced the reduced wavevector $x = k/k_0$, reduced energy $\bar{\omega} = \omega/\mu$ and reduced temperature $\bar{t} = T/\mu$. In contrast with reference [10], the origin for the energy ω of the two atoms is taken as usual at the bottom of the continuous energy spectrum for free particles. The coupling constant λ is related to a by $\lambda = -2k_0a/\pi$. The reduced width of the Feshbach resonance $\bar{W} = m^2|w|^2/\pi^2k_0$ is related [10] to the matrix element w corresponding to the Feshbach coupling between the open and the closed channel. In the strongly explored case of ^{6}Li, $\bar{W}$ is quite large and in equation (1) we will neglect $\bar{\omega}/\bar{W}$ in the following.

As it is well-known the appearance of the molecular bound state will show up as a pole in the vertex, and therefore it will be found by writing that the r.h.s. of equation (1) is zero. We note that the BCS instability itself appears also as such a pole. The imaginary part of equation (1) is zero at the chemical potential $\omega = 2\mu$ and writing that the real part is zero leads to:

$$\frac{1}{a} = \frac{2}{\pi}\int_0^\infty dk \left[1 - \frac{k^2}{k^2 - 2m\mu}\tanh\frac{k^2 - 2m\mu}{4mT}\right] \tag{2}$$

which is just the standard BCS equation with our notations (notice that $-1/\lambda$ is changed into $-1/\lambda + 2/\bar{W}$ if we take the $\bar{W}$ term into account).
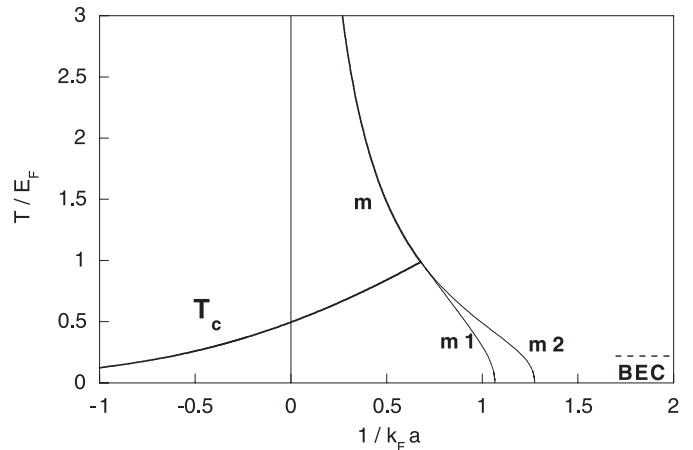

Fig. 1. Critical temperature T_c of the BCS transition as a function of the inverse scattering length a^{-1}. Threshold $a_m^{-1}(T)$ for formation of zero momentum molecules (curve labelled m) as a function of a^{-1}, for negative chemical potential. Threshold $a_{m1}^{-1}(T)$ (curve labelled $m1$) for the appearance of a molecular instability (at zero momentum) for positive chemical potential. Threshold $a_{m2}^{-1}(T)$ (curve labelled $m2$) for the appearance of a single molecular instability (at zero momentum) for positive chemical potential (see text).

It is easily seen from equation (2) that the BCS transition is also found in the region $a > 0$, that is beyond the location of the Feshbach resonance in vacuum. This situation has already been considered by Milstein et al. [11] and by Ohashi and Griffin [12] within a phenomenological fermion-boson model, and it is actually a clear ingredient of the BCS-BEC crossover. It seems at first surprising to see BCS pairing for $a > 0$ since in this case one has an effective repulsion between atoms. But one has rather to consider that the tendency to form a bound state is an increasing function of a^{-1} (taken algebraically), so that going to $a > 0$ makes it even easier to form BCS pairs. On the other hand BCS pairing occurs in the Fermi sea: the BCS pole appears for a positive energy $\omega = 2\mu > 0$. Hence the BCS transition stops when $\mu = 0$. Leggett [13] has already pointed out that, for the excitation spectrum at $T = 0$, there is a qualitative change when one crosses $\mu = 0$. In our approximation, the chemical potential is merely related to the one-species atom number n by the free particle relation $n = (1/2\pi^2)\int dk k^2[e^{(k^2/2m-\mu)/T} + 1]^{-1}$. Instead of n, we introduce k_F defined by $n = k_F^3/6\pi^2$ and $E_F = k_F^2/2m$. From equation (2) we find the critical temperature T_c as a function of the scattering length a. In Figure 1 we plot T_c/E_F as a function of $1/k_Fa$. The end point (T_0, a_0) is found at $T_0/E_F \simeq 0.99$ and $1/k_Fa_0 \simeq 0.68$.

When a^{-1} is increased beyond this point we expect to find a molecular instability. Naturally this is what happens but there is a qualitative change. Indeed molecular states correspond to negative values of the energy ω, for which the r.h.s. of equation (1) is always real so we are no longer forced to require that its imaginary part is zero. Hence we do not need to take a specific energy, and for a fixed temperature T, we expect to find the binding energy $|\omega|$ increasing with a^{-1}, just as in the absence of the
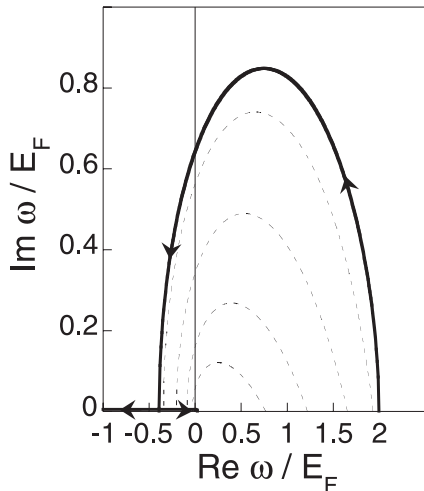
Fig. 2. Trajectory of the BCS pole in the upper complex ω plane for $T = 0$ (full curve) and for $\bar{t} = 0.2, 0.5, 1.0, 2.0$ (successive dashed curves). The arrows indicate how the pole(s) moves when a^{-1} is increasing.

Fermi sea. Actually this is the case that we have explored recently [6] for $\mu < 0$, which means $T > T_0$, and we have found that the molecular state with zero binding energy does not form for $a^{-1} = 0$ as in vacuum, but rather for a positive value of $a_m^{-1}(T)$ which increases when T decreases. Naturally when for fixed T we increase a^{-1} beyond $a_m^{-1}(T)$, the molecular binding energy increases from zero. We have plotted $1/k_F a_m(T)$ in Figure 1 and we see that it meets the above curve for the BCS transition at the end point. This is natural since this point corresponds to cancel the r.h.s. of equation (1) for $\omega = 0$ and $\mu = 0$, so it belongs to the two curves. Hence there is a clear link between the molecular shift and the existence of the BCS phase.

It is now interesting to study in the same way what happens when we increase a^{-1} at fixed T for $\mu > 0$, that is $T < T_0$. More specifically we will follow the pole of $\Gamma(\omega)$ which gives rise to instability. When, for fixed T, we start from a^{-1} large and negative there is no instability, until by increasing a^{-1} (algebraically) we reach the BCS instability when we touch the $T_c(a^{-1})$ curve. As it is well-known this means that the corresponding pole, which was in the lower ω complex plane has moved up and has reached the real ω-axis. Quite generally, since a^{-1} is real, we find the trajectory of the pole by writing that the imaginary part in equation (1) is zero, that is $\text{Im} I(\bar{\omega}) = 0$. The resulting trajectory in the ω complex plane is shown in Figure 2 for various temperatures.

We see naturally that, after crossing the real axis, the pole goes deep into the upper complex plane, corresponding to the fact that, when we enter more into the BCS phase domain, the normal state becomes more and more unstable. However when a^{-1} is further increased, the pole goes back toward the real ω-axis, eventually reaching it on the semi-axis $\omega < 0$. This happens for a positive value $a_{m1}^{-1}(T)$ shown in Figure 1. At this stage the normal state has no longer, strictly speaking, a BCS type instability. It

is rather a molecular type instability, characterized by a pole on the negative energy axis. However what happens next is rather surprising. When a^{-1} is increased beyond $a_{m1}^{-1}(T)$, the pole splits into two poles, one with increasing ω, the other one with decreasing ω. This is most easily seen at $T = 0$, where one finds from equation (1) for the position of the pole $|\bar{\omega}| = 2r^2$:

$$-\frac{1}{\lambda} \equiv \frac{\pi}{2k_F} a^{-1} = 2r \arctan r - \frac{\pi}{2} r + 2. \qquad (3)$$

Starting from 2 at $r = 0$, the r.h.s. of equation (3) decreases, reaches a minimum $-1/\lambda_{min} = 1.67$ for $\bar{\omega} = -0.39$ ($r = 0.44$), then increases and behaves asymptotically as $(\pi/2)r$. The minimum gives the value of $a_{m1}^{-1}(T = 0)$ for which the BCS pole reaches the negative ω-axis at $\bar{\omega} = -0.39$. When a^{-1} is increased beyond $a_{m1}^{-1}(0)$, one finds two solutions for $\bar{\omega}$ around the minimum. One of them decreases, in agreement with the expected increase of a molecular binding energy. But surprisingly the other one increases and goes rapidly to $\omega = 0$, where it disappears (more precisely it goes in the lower complex plane along the negative ω-axis, where it has no direct physical manifestation). This disappearance occurs for $-1/\lambda = 2$. We call $a_{m2}^{-1}(T = 0) = 4k_F/\pi$ the corresponding value of a^{-1}. The lower pole is then at $\bar{\omega} = -2$. Beyond $a_{m2}^{-1}(0)$ there is only one solution, corresponding to the continuation of the lower energy solution. Hence we recover the standard molecular situation of a single pole with a binding energy increasing with a^{-1}.

In order to understand physically this strange situation it is worthwhile to note that, in the absence of a Fermi sea, the r.h.s. of equation (3) would merely be $(\pi/2)r$, leading to the usual result $|\omega| = 1/ma^2$ for the molecular binding energy. Hence the Fermi sea contribution is $2r \arctan r - \pi r + 2 = 2 \int_0^1 dx\, x^2/(x^2 + r^2)$. Therefore the non analytic decrease, near $\bar{\omega} = 0$, of the r.h.s. of equation (3), responsible for the existence of the two poles, is produced by the Fermi sea. This is specifically due to the $-\pi r$ term. Now this term is easily linked to the density of states for positive energy, proportional to $\omega^{1/2}$. Indeed if we evaluate the above integral for small and positive ω (with infinitesimal positive imaginary part), we find $2R \ln[(1 - R)/(1 + R)] + i\pi R + 2$ with $R = (\bar{\omega}/2)^{1/2}$, and the r.h.s. of equation (3) is obtained from the analytical continuation of this last result through the upper complex plane toward the negative ω-axis. Actually all the information about this Fermi sea contribution is contained in the imaginary part $i\pi R$ since the real part can be recovered through Kramers-Kronig relations. Now, for small ω, this imaginary part is positive in contrast with the free particle result $\text{Im} I(\bar{\omega}) = -i(\pi/2)R$. The overall result [10] is $\text{Im} I(\bar{\omega}) = i(\pi/2)R$.

Clearly the physical interpretation of this change of sign is that, below the Fermi energy, we deal with hole-like excitations while above the Fermi energy we have the standard particle-like excitations: in order to create excited states below the Fermi energy we have to remove particles since Pauli exclusion forbids to add them. This leads to the conclusion that the bound state with very

small binding energy we have found above, just detached from the Fermi sea, has a hole-like character. In other words it corresponds to a molecular state formed by two atomic holes (absence of atoms) rather than to a standard molecule formed by two atoms. It is a kind of anti-molecule. We can come to this conclusion more rapidly by thinking of the situation we would have for a Fermi energy going to infinity. All the low energy states we could consider other than the ground state would be hole-like, since the only thing which could be done would be to remove particles. In particular we would have only hole-like molecules. Naturally we interpret the lower energy pole as a standard particle-like molecular state, since in particular it will be the only one present for $a^{-1} > a_{m2}^{-1}(0)$. On the other hand, if we think of decreasing a^{-1} below $a_{m2}^{-1}(0)$, we will have two poles, one being particle-like and the other one hole-like. Therefore when they merge at $a_{m1}^{-1}(0)$ the corresponding pole has necessarily a mixed particle-hole nature. But this is rather natural since this pole is a BCS-type pole as we have seen. Indeed it is well-known that the BCS instability has this mixed nature (we can think of it as the formation of pairs of particles, or as well as pairs of holes), as it is reflected in the nature of the low energy single particle excitations in BCS theory. Hence it is rather natural that when the BCS pole, after moving in the upper complex plane, reaches the negative ω-axis, it has still this mixed nature. This makes also likely that, when the two poles separate, they have actually also a mixed nature, with a full particle or hole character only reached at a_{m2}^{-1}.

The experimental observability of such a hole-like molecule is unclear. Indeed a hole around zero energy is quite deep below the Fermi energy and corresponds to a high energy excited state. It is likely that the lifetime of such a state will be quite short due to particle-hole recombination processes. These would appear because of the strong interactions of the atoms in Fermi sea, but they are not present in our approximate treatment since we evaluate $I(\bar{\omega})$ without taking into account these interactions. We would have to go beyond this theoretical level to make them appear. Similarly it is likely that the molecular states with partial hole-like nature will have a too short lifetime to be directly observed. Hence the physical situation for $a_{m1}^{-1} < a^{-1} < a_{m2}^{-1}$ is quite unclear and it is not obvious how this domain will survive in improved theories. In particular the existence of two poles may suggest a more complex instability, or a forbidden domain with phase separation. On the other hand for $a^{-1} > a_{m2}^{-1}$ we are back to a simple physical situation. We have just a single pole corresponding to the formation of standard molecular state. However the remarkable point is that the binding energy $|\bar{\omega}|$ is always larger than 2 (i.e. $|\omega| > 2E_F$). This means that it is impossible to observe a molecular state with zero binding energy, in contrast to the classical case of a dilute gas for which the binding energy is zero at the Feshbach resonance $a^{-1} = 0$. Even if we believe it is possible to observe short lived molecules in the range $a_{m1}^{-1} < a^{-1} < a_{m2}^{-1}$, this result remains valid as we have seen above. The lower bound for the molecular binding

energy will just be smaller. Finally we have for simplicity limited our explicit quantitative study to the $T = 0$ case, but from Figure 2 the same results will be qualitatively valid for all the range $0 \leq T \leq T_0$. The lower bound for the molecular binding energy will decrease with increasing T and go to zero for $T = T_0$. Since T_0 is also the maximum temperature for the existence of the BCS phase, we reach the surprising conclusion that, whenever the BCS phase is present, we can not observe a molecule with zero binding energy. This offers an indirect way to demonstrate the presence of the BCS phase.

We have naturally to be quite specific with respect to the above statement. In this paper we have only studied the instabilities arising in a normal Fermi gas, with in particular no molecules already present. Therefore when we consider this gas for $a^{-1} > a_{m2}^{-1}(T)$, we deal with an out of equilibrium situation since at equilibrium a sizeable fraction of the gas should be under molecular form. Therefore we have in mind an experiment where, starting from the $a^{-1} < 0$ side of the Feshbach resonance, one would very rapidly change a^{-1} by acting on the magnetic field and then observe the binding energy of the first few molecules appearing in the gas. We could also worry about the effect of the Bose-Einstein condensation of molecules. However in our simple picture the critical temperature for this BEC is [11] $T_{BEC} = 0.218T_F$, so the effect we have considered could be at least observed in the range $T_{BEC} < T < T_0$. Also the instabilities we have considered are all at zero wavevector, just as the dominant BCS instability itself. So the molecules we considered have zero total momentum. For nonzero momentum the effects will be weaker, and we expect them to disappear at some wavevector, whose inverse is of order of the Cooper pair size, just as the BCS instability itself.

Finally we have naturally to consider that our theoretical approach is clearly not quantitatively accurate, since for example our value for the BCS critical temperature T_c is just the standard one [14], and does not contain lower order fluctuation effects [15] nor higher orders and self-energy effects [16] including lifetime effects (in particular these effects will reduce [15,16] markedly T_c and the temperature of the end point (T_0, a_0), and thereby strongly increase the excitation lifetime in the vicinity of this point). However we believe that our results remain qualitatively valid under much more general conditions. Indeed we see from Figure 1 that the nonzero threshold for molecular binding energy that we have found is ultimately linked to the trajectory of the BCS pole in the upper complex plane. But the existence of this pole, and its trajectory in the upper complex plane are quite general features of any theoretical description. Since we naturally expect to find molecules for very large a^{-1}, this pole has to go back to the real negative ω-axis, as we have found, which implies again the nonzero molecular binding energy whenever the BCS phase is present. In other words the (qualitative) topology of our results should remain valid even if they are quantitatively modified.

We stress again that we have here only looked at the instability properties of the normal state, even in regions

where it is not the equilibrium state. We have found that, when a^{-1} goes from $-\infty$ to $+\infty$ across the phase diagram, these properties do not change smoothly, but rather that the behaviour changes on some lines that we have identified. One may wonder if the same is not true for the equilibrium phase, and in particular for the ground state in the superfluid domain for which a quite natural interpolating hypothesis [13,17] is a BCS-like ground state whatever the scattering length, since this wavefunction is known to describe properly the dilute molecular ground state [18] as well as the weak coupling BCS state. In this case one would rather expect a single instability to appear corresponding to the pair formation, so it might be that this BEC-BCS crossover is not as smooth as suggested by this picture, and that the actual physics is more complex. However our results have no direct bearing on this question, since we have explored only the normal phase, not the superfluid. We just note that, in the superfluid state, the effect of Pauli exclusion will still be present.

References

1. C.A. Regal et al., Nature **424**, 47 (2003)
2. C.A. Regal et al., Phys. Rev. Lett. **92**, 083201 (2004)
3. J. Cubizolles et al., Phys. Rev. Lett. **91**, 240401 (2003)
4. S. Jochim et al., Phys. Rev. Lett. **91**, 240402 (2003)
5. K.E. Strecker et al., Phys. Rev. Lett. **91**, 080406 (2003)
6. R. Combescot, New J. Phys. **5**, 86 (2003)
7. K. Dieckmann et al., Phys. Rev. Lett. **89**, 203201 (2002)
8. K.M. O'Hara et al., Science **298**, 2179 (2002)
9. T. Bourdel et al., Phys. Rev. Lett. **89**, 020402 (2003)
10. R. Combescot, Phys. Rev. Lett. **91**, 120401 (2003)
11. J.N. Milstein et al., Phys. Rev. A **66**, 043604 (2002)
12. Y. Ohashi, A. Griffin, Phys. Rev. Lett. **89**, 130402 (2002)
13. A.J. Leggett, J. Phys. (Paris) **41**, C7 (1980)
14. H.T.C. Stoof et al., Phys. Rev. Lett. **76**, 10 (1996)
15. L.P. Gorkov, T.K. Melik-Barkhudarov, Sov. Phys. JETP **13**, 1018 (1961)
16. R. Combescot, Phys. Rev. Lett. **83**, 3766 (1999)
17. P. Nozières, S. Schmitt-Rink, J. Low Temp. Phys. **59**, 195 (1985)
18. L.V. Keldysh, A.N. Kozlov, Zh. Eksp. Teor. Fiz. **54**, 978 (1968) [Sov. Phys. JETP **27**, 521 (1968)]