Abstract

We theoretically examine photoassociation of a two-component Fermi degenerate gas. Our focus is on adjusting the atom-atom interaction, and thereby increasing the critical temperature of the BCS transition to the superfluid state. In order to avoid spontaneous decay of the molecules, the photoassociating light must be far-off resonance. Very high light intensities are therefore required for effective control of the BCS transition.
As the field of quantum degenerate vapors emerges from its burgeoning adolescence, attention is increasingly shifting from Bose [1] to Fermi [2] systems. Beyond degeneracy itself, much of this effort is concentrated on observing the BCS transition to a superfluid state. However, such investigations are currently at an impasse: the lowest temperature that has been achieved is about a third of the Fermi temperature [3], whereas the formation of Cooper pairs requires temperatures colder by at least an order of magnitude [4]. Rather than finesse the experiments to lower the temperature of the gas further, a more fruitful approach might be to adjust the atom-atom interaction so as to raise the value of the critical temperature. Possible means for adjustment include the magnetic-field-induced Feshbach resonance [5], rf microwave fields [6], dc electric fields [7], and photoassociation [8,9]. In fact, the Feshbach resonance has been recently applied in this manner to induce condensation in the otherwise incondensable $^{85}$Rb [10], and was additionally explored (theoretically) for its usefulness in spurring the superfluid transition [11].

The purpose of this paper is to investigate the utility of photoassociation for inducing the BCS transition. We [12,13,9] and others [14] have earlier written down field theories for photoassociation of bosons. In the present case we consider instead a binary mixture of fermionic atoms, given by the fields $\phi_{\pm}(\mathbf{r})$, photoassociating into a bosonic molecule, given by the field $\psi(\mathbf{r})$. The fermions would typically be two states with different $z$ components of angular momentum in the same atom. As a result of the Pauli exclusion principle, there is no $s$-wave photoassociation for two atoms in the same internal state, but such a restriction does not apply to two different spin components.

We thus have a model Hamiltonian density governing photoassociation,

$$
\frac{\mathcal{H}}{\hbar} = -\phi_{+}^{\dagger} \frac{\hbar \nabla^{2}}{2m} \phi_{+} - \phi_{-}^{\dagger} \frac{\hbar \nabla^{2}}{2m} \phi_{-} + \psi^{\dagger} \left[ -\frac{\hbar \nabla^{2}}{4m} + \delta - \frac{1}{2} i \gamma_{s} \right] \psi \\
- \left[ \mathcal{D} \psi^{\dagger} \phi_{+} \phi_{-} + \mathcal{D}^{\ast} \phi_{+}^{\dagger} \phi_{-}^{\dagger} \psi \right] + \frac{4\pi \hbar a}{m} \phi_{+}^{\dagger} \phi_{+} \phi_{-} \phi_{-} ,
$$

where $m$ is the mass of an atom and $\delta$ is the detuning of the laser from the threshold of photodissociation. The detuning is positive when the photodissociation (inverse of photoassociation) channel is open. The coupling strength for photoassociation is $\mathcal{D}$. It may be
deduced implicitly from Refs. [12,13], and is discussed explicitly in Ref. [9]. Either way, we have

\[ |\mathcal{D}(\mathbf{r})| = \lim_{v \to 0} \sqrt{\frac{\pi \hbar^2 \Gamma(\mathbf{r})}{v \mu^2}}. \tag{2} \]

Here \( \Gamma(\mathbf{r}) \) is the photodissociation rate that a light with the intensity prevailing at \( \mathbf{r} \) would cause in a nondegenerate gas of molecules, given that the laser is tuned in such a way that the relative velocity of the molecular fragments (atoms) is \( v \), and \( \mu = m/2 \) is the reduced mass of two atoms. Because of the statistics, there is a factor of \( \sqrt{2} \) difference in Eq. (2) from the corresponding expression for identical bosons. Finally, we have included an interspecies collisional interaction governed by the \( s \)-wave scattering length \( a \) in the Hamiltonian.

Under ordinary circumstances photoassociation by absorption of a photon leads to a molecular state that is unstable against spontaneous emission. There is no particular reason why spontaneous break-up of a primarily photoassociated molecule would deposit the ensuing atoms back to the degenerate Fermi gases. A spontaneously decaying molecule is considered lost for our purposes. Correspondingly, we add to the Hamiltonian a nonhermitian term proportional to the spontaneous emission rate of the molecular state \( \gamma_s \).

The Heisenberg equation of motion for the molecular field \( \psi \) is

\[ i\dot{\psi} = \left[ -\frac{\hbar \nabla^2}{4m} + \delta - \frac{i}{2} \gamma_s \right] \psi - \mathcal{D} \phi_+ \phi_. \tag{3} \]

We assume that the detuning \( \delta \) is the largest frequency parameter in the problem, and solve Eq. (3) adiabatically for the field \( \psi \). In the process we keep the imaginary part in the energy, and obtain

\[ \psi \simeq \left[ \frac{\mathcal{D}}{\delta} + i \frac{\gamma_s \mathcal{D}}{2\delta^2} \right] \phi_+ \phi_. \tag{4} \]

Inserting into Eq. (1), we find an effective Hamiltonian density for fermions only,

\[ \frac{\mathcal{H}}{\hbar} \simeq -\phi_+^\dagger \frac{\hbar \nabla^2}{2m} \phi_+ - \phi_-^\dagger \frac{\hbar \nabla^2}{2m} \phi_- + \frac{4\pi \hbar a}{m} \phi_+^\dagger \phi_+ \phi_- + \left[ -\frac{\mathcal{D}^2}{\delta} - i \frac{\gamma_s |\mathcal{D}|^2}{2\delta^2} \right] \phi_+^\dagger \phi_+ \phi_. \tag{5} \]
Let us first ignore the decay term $\propto \gamma_s$. Equation (5) displays an added contact interaction between the two spin species, as if from the $s$-wave scattering length

$$\bar{a} = -\frac{|D|^2}{4\pi \delta \hbar}.$$  

(6)

The interaction is attractive if the detuning is positive. But an attractive interaction is exactly what is needed for the BCS transition. To simplify matters we assume here that the collisional interaction in the absence of light $\propto a$ is too weak for experiments on the BCS transition, and ignore the native collisions altogether.

The critical temperature for the BCS transition is [4]

$$T_c = T_F \exp \left[-\frac{\pi}{2k_F a} \right] = T_F \exp \left[-\frac{2\pi^2 \hbar \delta}{k_F m |D|^2} \right].$$  

(7)

Here $k_F = (3\pi^2 \rho)^{1/3}$ is the Fermi wave number for the total density of atoms $\rho$, and $T_F = \hbar^2 k_F^2 / 2mk_B$ is the corresponding Fermi temperature. Finally, using $(\rho/2)^2$ for $\phi_-^\dagger \phi_+^\dagger \phi_+ \phi_-$, we find the loss rate per atom due to spontaneous emission from photoassociated molecules,

$$\frac{1}{\tau} = \frac{\gamma_s |D|^2 \rho}{2\delta^2}.$$  

(8)

To estimate practical experimental numbers, we first note that the rate of photoassociation in a nondegenerate sample at temperature $T$ is [15,16]

$$R = \lambda_D^3 \rho e^{-\frac{\hbar \delta}{\hbar \omega}} \Gamma \equiv \rho \left( \frac{I}{\hbar \omega} \right) \kappa.$$  

(9)

Here $\lambda_D = \sqrt{2\pi \hbar^2 / \mu k_B T}$ is the thermal deBroglie wavelength, $I$ is the intensity (W cm$^{-2}$) of photoassociating light, and $\kappa$ (cm$^{-5}$) is the photoassociation rate coefficients. There may be statistics dependent numerical factors in Eq. (9). However, in the current literature such factors are usually ignored, and we write Eq. (9) accordingly.

Using Eq. (9), a calculation or a measurement of the photoassociation rate in a thermal sample may be converted into a prediction of effective scattering length, transition temperature, and lifetime in a degenerate Fermi-Dirac gas. We express the results in terms of

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\( \lambda = \lambda/2\pi \), wavelength of photoassociating light divided by \( 2\pi \), \( \epsilon_R = \hbar/(2m\lambda^2) \), familiar photon recoil frequency, and a characteristic intensity for the given photoassociation transition, \( I_0 \). This gives

\[
\frac{\dot{a}}{\lambda} = 0.0140077 \frac{I}{I_0} \frac{\epsilon_R}{\delta},
\]

\[
\frac{T_c}{T_F} = \exp \left[ -36.2478 \frac{1}{(\lambda^3 \rho)^{1/3}} \frac{\delta}{\epsilon_R} \frac{I_0}{I} \right],
\]

\[
\epsilon_R \gamma = 4 \frac{\delta^2}{\epsilon_R \gamma_s} \frac{I_0}{I} \frac{1}{\lambda^3 \rho}.
\]

The obscure numerical factors, powers of 2 and \( \pi \), are there because we want to use the characteristic intensity for photoassociation defined in Ref. [9]. For instance, if the photoassociation rate coefficient \( \kappa \) is known at a temperature \( T \) and detuning \( \delta \), the critical intensity is

\[
I_0 = \frac{\sqrt{\pi} \hbar \delta c h^4}{2\kappa m^2(k_B T)^{3/2} \lambda^2} e^{-\hbar \delta/k_B T}.
\]

Detailed microscopic calculations (or measurements) of photoassociation rates are sparse, but they exist for the fermionic isotope \(^6\text{Li} \) of lithium [17–20]. Let us consider an example already discussed in Ref. [9], transitions to the triplet vibrational state \( v' = 79 \) with the binding energy 1.05 cm\(^{-1} \). The characteristic intensity is then \( I_0 = 9.8 \) mW cm\(^{-2} \), the wavelength is \( \lambda = 671 \) nm, and the recoil frequency is \( \epsilon_R = 63.3 \times 2\pi \) kHz. We take the decay rate of the molecular state to be twice the spontaneous decay rate of the corresponding atom, so that \( \gamma_s = 12 \times 2\pi \) MHz. In our estimate we assume \( \lambda^3 \rho = 1 \), corresponding to the density \( \rho = 8.21 \times 10^{14} \) cm\(^{-3} \) that is high but not unreasonable. It would then take the intermediate detuning \( \delta = 2 \times 2\pi 10^{14} \) Hz and the intensity \( I = 460 \) MW cm\(^{-2} \) to make \( T_c = 0.1 T_F \) and \( \tau = 10 \) s.

The intensity came out very high for a continuous-wave laser, so it seems that the only potential candidate for experiments is a tightly focused, powerful CO\(_2 \) laser. Our formalism, though, is based on the assumption that the laser is close to a photoassociating resonance. We need to amend the calculations to give meaningful estimates for the CO\(_2 \) laser, whose
electric field is in practice direct current compared to the molecular transition frequencies involved.

To this effect we first note that in an ordinary two-level system one may carry out perturbation theory both within the rotating-wave approximation, and in the quasistatic limit without the rotating-wave approximation as well. The result is that the quasistatic results are obtained from the near-resonance formulas by replacing the detuning with the molecular transition frequency, $\delta \rightarrow \omega_0$, and multiplying the intensity by two, $I \rightarrow 2I$. Applying this substitution to the scattering length, at $\lambda^3 \rho = 1$ we find that the intensity required for $T_c = 0.1 T_F$ again becomes 460 MW cm$^{-2}$. With the same substitutions, the lifetime would be about 20 s. However, as the frequency of the CO$_2$ laser is $1/16$ of the resonance frequency for photoassociation, the phase space for spontaneously emitted photons is reduced, and the actual rate of spontaneous emission would be reduced by an extra factor of at least $16^2 \sim 300$. It is clear that spontaneous emission is not an issue with CO$_2$ laser excitation.

Up to this point we have only considered photoassociation with one molecular state, the triplet state with vibrational quantum number $v' = 79$. Now, in lithium as well as in other alkali atoms, most of the transition strength for dipole transitions starting from the ground state is in the $D$ lines. Just a few electronic states in a molecule then inherit most of the transition strength for photoassociation. We only consider the singlet and triplet excited manifolds in the $^6$Li dimer, for which calculations of the photoassociation matrix elements exist for all vibrational states [17–20]. It turns out that the triplet state $v' = 79$ carries about the factor 0.07 of the total transition strength for photoassociation of low-energy atoms. As one should obviously add the changes of the scattering lengths due to all molecular states, in our CO$_2$ laser example the intensity also gets multiplied by 0.07 and becomes 30 MW cm$^{-2}$.

It is, in principle, possible to tailor the scattering length by off-resonant photoassociation, and thereby effect the BCS transition in a low-temperature Fermi gas of, say, $^6$Li vapor. The required laser intensities, however, are high. As in the case of coherent photoassociation [9], the problem is not so much that the matrix elements for photoassociation are weak, but
that the primarily photoassociated molecules tend to decay spontaneously and the sample is lost. To avoid spontaneous emission, one has to go very far off resonance, which leads to challenging requirements on laser intensity. In pursuit of BCS transition by means of off-resonant photoassociation, it might be worthwhile to try and look for other ways of getting around the spontaneous emission.
REFERENCES


[20] It should be noted that the photoassociation rates calculated in Refs. [17–19] are inadvertently low by a factor of $(2\pi)^5$. 