Photo-excitation measurement of Tan’s contact for a strongly interacting Fermi gas

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We derive theoretically an exact relation between Tan’s universal contact and the photo-excitation rate of a strongly interacting Fermi gas, in the case of optically transferring fermionic pairs to a more tightly bound molecular state. Our deviation generalizes the relation between Tan’s contact and the closed-channel molecular fraction found earlier by Werner, Tarruell and Castin in Eur. Phys. J. B \textbf{68}, 401 (2009). We use the relation to understand the recent low-temperature photo-excitation measurement in a strongly interacting \textsuperscript{6}Li Fermi gas [Liu et al., arXiv:1903.12321] and show that there is a reasonable agreement between theory and experiment close to the unitary limit. We propose that our relation can be applied to accurately measure Tan’s contact coefficient at finite temperature in future experiments.

I. INTRODUCTION

In 2008, in a series of seminal works\cite{1,2,3}, Shina Tan presented a set of elegant exact relations, showing that the short-range, large-momentum and high-energy behaviors of a strongly correlated atomic gas, both statically and dynamically, can be universally governed by a coefficient. These relations together with the coefficient, namely Tan’s relations and Tan’s contact coefficient, pave an entirely new direction to understand complicated quantum many-body systems\cite{4,5,6}.

To date, Tan relations have been experimentally verified in both Fermi gases\cite{7,8} and Bose gases\cite{9}. The key Tan’s contact coefficient has also been measured in a number of ways\cite{10,11,12}, following different Tan relations\cite{13,14,15,16,17,18,19,20,21}. In particular, great efforts have been taken to accurately determine the contact coefficient of a unitary Fermi gas with an infinitely large s-wave scattering length, by using Bragg spectroscopy\cite{11,12} and radio-frequency (rf) spectroscopy\cite{13}. At finite temperature, the measured contact of a homogeneous unitary Fermi gas clearly shows a dramatic change at the superfluid transition temperature, providing an unambiguous signature for the onset of the superfluid transition\cite{12,13}.

In this work, we would like to propose that photo-excitation, in which a fermionic pair absorb a photon to form an excited molecule, may give an alternative, potentially more accurate method to measure finite-temperature Tan’s contact for a strongly interacting Fermi gas at the crossover from a Bose-Einstein condensate (BEC) to another excited molecular bound state. Our deviation generalizes the relation between Tan’s contact and the closed-channel transition temperature at the entire BEC–BEC crossover, which remains elusive so far.

Our proposal is based on the derivation of a new exact Tan relation for the photo-excitation rate, if we optically transfer fermionic pairs to a more tightly bound molecular state\cite{22,23}. In the limit of a weak laser intensity or small Rabi coupling, our relation recovers the known relation between Tan’s contact and the closed-channel molecular fraction, predicted earlier by Werner, Tarruell and Castin\cite{13}. The advantage of our new relation is that it also works at a moderately large laser intensity, where the accuracy of the photo-excitation measurement could be greatly enhanced. This leads to a more accurate way to measure Tan’s contact at finite temperature. As an application, we use our relation to better understand a recent photo-excitation measurement in a strongly interacting \textsuperscript{6}Li Fermi gas at the University of Science and Technology of China (USTC), Shanghai\cite{24,25}. We show that the puzzling huge discrepancy between theory and experiment, at about two orders of magnitude, can be reasonably resolved, although there is a still a factor of three difference found on the BCS side at the largest magnetic field considered in the experiment.

The rest of the paper is organized as follows. In the next chapter (Sec. II), we sketch the photo-excitation scenario and present the model Hamiltonian. In Sec. III, we derive the Tan relation for the photo-excitation rate. In Sec. IV, we discuss the experimental relevance, first on analyzing the recent photo-excitation measurement and then on the proposal for future finite-temperature contact measurements. Finally, we conclude in Sec. V.

II. MODEL HAMILTONIAN

We consider the photo-excitation experiments at Rice\cite{24}, Ulm\cite{25}, and USTC Shanghai\cite{26}, all of which use a strongly interacting \textsuperscript{6}Li Fermi gas near the Feshbach resonance at $B_0 \approx 832.18$ G, where fermionic pairs are in the superposition of a stable, ground molecular bound state $X^1\Sigma_g^+(v = 38)$. A resonant laser transition is used to photo-excite fermionic pairs in the admixture to another excited molecular bound state $A^1\Sigma_u^+(v' = 68)$, which suffers from the spontaneous-emission loss at the rate $\gamma$. In the case that this is the dominant loss channel, one can determine the photo-excitation rate from the loss rate of the system, i.e., $-\dot{N}/N$. The above mentioned Fermi gas system can be well described by a two-channel model Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}} + \mathcal{H}(am) + \mathcal{H}(m)$,
where \[ \begin{align*}
\mathcal{H}_0^{(a)} &= \sum_{k\sigma} (\epsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma}, \\
\mathcal{H}_{\text{int}}^{(a)} &= \frac{u_0}{V} \sum_{kk'q}\left[ c_{kq}^\dagger c_{-kq}^\dagger - c_{-kq} c_{kq} \right] + \frac{\gamma}{2} \sum_{kk'q} \left[ c_{kq}^\dagger c_{-kq} + c_{-kq}^\dagger c_{kq} \right], \\
\mathcal{H}_{\text{am}}^{(m)} &= \frac{g_0}{V} \sum_{kkq} \left[ \phi_{gq}^\dagger \phi_{cq}^\dagger \mathcal{M}_q \left( \phi_{gq}, \phi_{cq} \right) \right], \\
\mathcal{H}^{(m)} &= \sum_q \left( \phi_{gq}^\dagger \phi_{cq}^\dagger \right) \mathcal{M}_q \left( \phi_{gq}, \phi_{cq} \right),
\end{align*} \]

describe respectively the kinetic Hamiltonian of atoms, the background interaction Hamiltonian of atoms with the interaction strength \(u_0\), the coupling between atoms and ground-state molecules \(g_0\), and the molecules in both ground and excited molecular states. Here, \(\epsilon_k \equiv \hbar^2 k^2/(2m)\) is the free dispersion relation, \(\mu\) is the chemical potential of atoms and, \(\phi_{gq}, \phi_{cq}\) are the annihilation field operators of atoms (with spin \(\sigma = \uparrow, \downarrow\)), ground-state molecules and excited-state molecules, respectively. In the last Hamiltonian for molecules, \(\mathcal{M}_q\) is a 2 by 2 matrix,

\[ \mathcal{M}_q = \begin{bmatrix} \epsilon_q - 2\mu + \delta_{g0} & \Omega/2 \\ \Omega/2 & \epsilon_q - 2\mu + \delta_{c0} - \Delta - i\frac{\pi}{2} \end{bmatrix}, \]

where \(\Omega\) and \(\Delta\) are the Rabi coupling and detuning of the photo-excitation laser, respectively, \(\gamma\) is loss rate, and \(\delta_{g0}\) and \(\delta_{c0}\) are the bare detunings of the molecular states.

The subscript “0” in various parameters indicates that these bare parameters are to be renormalized and related to some physical observables \[28\, 29\]. For example, \(u_0\) will be expressed in terms of the background s-wave scattering length \(a_{bg}\), \(g_0\) will be replaced by the width of the Feshbach resonance \(W\), and finally \(\delta_{g0}\) and \(\delta_{c0}\) will correspond to the detunings of the closed-channels. The parameter renormalization has been discussed in detail in the literature \[28\, 29\]. It is very easy to implement: we can simply do the following replacements in final equations derived, i.e.,

\[ u_0 \to \mu = \frac{4\pi\hbar^2}{m} a_{bg}, \]
\[ g_0 \to g = \sqrt{\frac{4\pi\hbar^2}{m}} a_{bg} W_{\mu g}, \]
\[ \delta_{g0} \to \delta_g = \mu_{ag} (B - B_0), \]
\[ \delta_{c0} \to \delta_c = \mu_{ac} (B - B_0). \]

For \(^6\text{Li}\) atoms near the Feshbach resonance at \(B_0 \approx 832.18 \text{ G}, a_{bg} \approx -1582 a_0 \) \((a_0 = 0.0529177 \text{ nm})\), \(W = -262.3 \text{ G}, \mu_{ag} = 2\mu_a - \mu_g \approx 2\mu_B\) (\(\mu_B\) the Bohr magneton) is the difference in the magnetic moments of atoms and of molecules for the ground state \[30\], and similarly \(\mu_{ac} = 2\mu_c - \mu_e \approx 2\mu_B\). For the excited molecular state, the loss rate \(\gamma \approx 2\pi \hbar \times 11.8 \text{ MHz}\). The Rabi coupling \(\Omega\) is typically at the order of \(2\pi \times 1.0 \text{ MHz}\). In addition, to convert the Zeeman field to frequency, we could use \(\mu_B \times 1 \text{G} \approx 2\pi \hbar \times 1.3996 \text{MHz}\).

### III. UNIVERSAL TAN RELATION FOR PHOTO-EXCITATION

According to Tan, Braaten and Platter \[34\], the loss rate of a quantum many-body system with short-range inter-particle interactions at the number of particles \(N\) is given by,

\[ \dot{N} \equiv \frac{dN}{dt} = \frac{\hbar \left\{ -\text{Im} (B) \right\}}{2\pi m |a(B)|^2 \mathcal{I}}. \]

Here, \(a(B)\) is the s-wave scattering length at a given Zeeman field \(B\) and \(\mathcal{I}\) is the contact coefficient of the system. The loss rate equation can be intuitively understood from Tan’s adiabatic relation,

\[ \frac{dE}{d(1/a)} = -\frac{\hbar^2}{4\pi m} \mathcal{I}, \]

which relates the total energy \(E\) of the many-body system to the contact coefficient \(\mathcal{I}\). Due to the coupling to the dissipative excited molecular state, the total energy acquires a relatively small imaginary part \(E - i\mathcal{I}/2\), where \(\mathcal{I}/\hbar = -dN/dt\) can be understood as the loss rate of the system, since the time evolution of the system can be given by \(\exp[-i (E - i\mathcal{I}/2) t/\hbar]\). Correspondingly, the scattering length also has a small imaginary part. From the adiabatic relation, we have,

\[ -\frac{E_i}{2} \approx \text{Im} [dE] = -\frac{\hbar^2}{4\pi m} \text{Im} [d(1/a)], \]

which immediately leads to the loss rate equation \[34\].

Let us now calculate the scattering length \(a(B)\) for the experimental photo-excitation process. Taking into account the molecule-mediated attraction, the total (un-renormalized) interaction between two atoms can be written into the form,

\[ U(\mathbf{q}, i\nu_n) = u_0 + g_0 \varphi^{(0)}(\mathbf{q}, i\nu_n), \]

where \(\varphi^{(0)}(\mathbf{q}, i\nu_n)\) is the non-interacting Green function of molecules in the ground-state channel and is the 11-component of the 2 by 2 matrix,

\[ \varphi^{(0)}(\mathbf{q}, i\nu_n) = [i\nu_n - \mathcal{M}_q]^{-1}. \]

By explicitly working out the inverse of the matrix, we obtain that,

\[ \varphi^{(0)}(\mathbf{q}, i\nu_n) = \frac{1}{\left( \nu_n - \epsilon_{\mathbf{q}}/2 + 2\mu - \delta_{g0} - \frac{\Omega^2/4}{\nu_n - \epsilon_{\mathbf{q}}/2 + 2\mu - \delta_{c0} + \Delta + i\gamma/2} \right)^{-1}}. \]

The last term in the bracket gives the conventional Stark shift to the ground-state molecules due to the coupling to the excited molecular state. In the vacuum, where we set the chemical potential \(\mu = 0\), the s-wave scattering
length $a(B)$ is then given by,

$$\frac{4\pi\hbar^2 a(B)}{m} = u_0 + g_0^2 \varphi_g^{(0)}(0,0),$$

$$= u - \frac{g^2}{\delta_g + \Omega^2/\left[4(-\delta_e + \Delta + i\gamma/2)\right]}.$$  

In the last step, we have replaced the un-regularized parameters with the regularized ones. In the absence of the Rabi coupling, i.e., $\Omega = 0$, it is easy to check that we recover the well-known expression for the scattering length near a Feshbach resonance,

$$a_s(B) = \frac{m}{4\pi\hbar^2} \left[ u - \frac{g^2}{\delta_g} \right] = a_{bg} \left( 1 - \frac{W}{B - B_0} \right).$$  

Here, the subscript “$s$” in $a_s(B)$ indicates the $s$-wave scattering length without Rabi coupling. In the presence of Rabi coupling, instead we would obtain,

$$a(B) = a_{bg} - \frac{a_{bg}\Gamma}{\delta_g + \Omega^2/\left[4(-\delta_e + \Delta + i\gamma/2)\right]}.$$  

where $\Gamma \equiv \mu_{ag} W$ is the characteristic energy related to the width of the Feshbach resonance. It is straightforward to obtain,

$$-\frac{\text{Im} a(B)}{|a(B)|^2} = \left( \frac{\Gamma}{a_{bg}} \right) \frac{\left[ \Omega^2/(2\gamma) \right]}{(\delta_g - \Gamma)^2 + c^2 \left[ \Omega^2/(2\gamma) \right]^2},$$

where the coefficient

$$c = 1 - \frac{4(\delta_e - \Delta)(\delta_g - \Gamma)}{\Omega^2}.$$  

By substituting it into the loss rate expression (10), we find that,

$$-\frac{\dot N}{N} = \frac{\hbar I}{2\pi m N} \left( \frac{\Gamma}{a_{bg}} \right) \frac{\left[ \Omega^2/(2\gamma) \right]}{(\delta_g - \Gamma)^2 + c^2 \left[ \Omega^2/(2\gamma) \right]^2}.$$  

By recalling that

$$\delta_g - \Gamma = -\frac{\Gamma}{1 - a_{bg}/a_s},$$  

and defining

$$R_s = \frac{\hbar^2}{ma_{bg}\Gamma},$$

$$I = 4\pi N k_F F \left( \frac{1}{k_F a_s} \right),$$

we finally arrive at,

$$-\frac{\dot N}{N} = \hbar^{-1} k_F R_s F \left( \frac{1}{k_F a_s} \right) L(\Delta),$$

where the line-shape function of the photo-excitation laser $L(\Delta)$ takes the form,

$$L(\Delta) = \frac{\Omega^2/\gamma}{[1 - a_{bg}/a_s]^2 + \left[ \Omega^2 - 4(\delta_e - \Delta)(\delta_g - \Gamma) \right]^{2}}$$

Under the resonant condition $\delta_e = \Delta$ and with a weak laser intensity $\Omega^2 \ll \gamma\Gamma$, the line-shape function is

$L(\Delta) = (1 - a_{bg}/a_s)^2 \Omega^2/\gamma$. As $\hbar^{-1} \Omega^2/\gamma$ is the effective decay rate of the ground molecular state for a resonant transition, see, i.e., Eq. (19), we may rewrite

$$-\frac{\dot N}{N} = Z \frac{\Omega^2}{\hbar\gamma},$$

where $Z$ is the molecular fraction in the ground molecular state and hence is given by

$$Z = k_F R_s F \left( \frac{1}{k_F a_s} \right) \left[ 1 - \frac{a_{bg}}{a_s} \right]^2 = \frac{I R_s}{4\pi N} \left[ 1 - \frac{a_{bg}}{a_s} \right]^2.$$

Thus, in the limit of a weak laser intensity, we recover the universal relation that links the closed-channel molecular fraction to the contact coefficient, firstly pointed out by...
The magnetic field dependence of the closed-channel fraction $Z$ of a strongly interacting $^6$Li Fermi gas at the density characterized by a Fermi temperature $T_F = 0.45 \mu K$. The low-temperature data from USTC Shanghai group (symbols) are compared with our zero-temperature theoretical predictions using the contact obtained from, a Gaussian pair fluctuation theory (solid line) and a perturbation theory (dashed line).

Werner et al. Our loss rate equation Eq. (26) does not have the weak probe restriction. It holds as long as the assumption of a single $s$-wave scattering length $a_s(B)$ is applicable.

**IV. EXPERIMENTAL RELEVANCE**

A. Photo-excitation measurement at USTC Shanghai

In a recent experiment at USTC Shanghai, the laser intensity is weak and the Rabi coupling is measured to be $\Omega^2/\gamma \approx 2\pi \times 0.136(1) \text{ MHz}$. This is much smaller than the characteristic energy of the Feshbach resonance width, i.e., $\Gamma = \mu_{bg} W = 2 \mu_B \times 262.3G \approx 2 \pi \hbar \times 734 \text{ MHz}$. Therefore, for resonant excitation ($\delta_s = \Delta$) we may use the weak-intensity result,

$$\frac{\dot{N}}{N} = k_FR_s \mathcal{F} \left( \frac{1}{k_F a_s} \right) \left[ 1 - \frac{a_{bg}}{a_s} \right]^2 \frac{\Omega^2}{\hbar \gamma}, \quad (30)$$

to analyze the experimental data and to calculate the closed-channel fraction $Z$ in the ground molecular state according to Eq. (29). In Fig. 1 we present the density dependence of the closed-channel fraction $Z$ in the unitary limit $B = 832 \text{ G (a)}$ and on the BCS side $B = 925 \text{ G (b)}$, experimentally measured at USTC Shanghai and theoretically calculated by using Eq. (29) with the contact obtained either from a Gaussian pair fluctuation theory or from a perturbation theory. The latter is only applicable in the deep BCS limit. We find a good agreement between theory and experiment in the unitary limit. However, on the BCS side, there is about a factor of three difference. In Fig. 2, we show the comparison as a function of the magnetic field. The agreement seems to become increasingly worse when we increase the magnetic field.

We do not fully understand why there is about a factor of three difference on the BCS side. A possible source for the discrepancy is the Rabi coupling $\Omega$, which is only experimentally calibrated on the BEC side and is then assumed to be invariant across the BEC-BCS crossover. Mathematically, the Rabi coupling is given by the overlap between the two wave-functions of the bound molecular states, i.e.,

$$\Omega = \left< \psi_{\nu,=68}(S = 0) \right| \vec{d} \cdot \vec{E}_L \left| \psi_{\nu,=38}(S = 0) \right>, \quad (31)$$

where $\vec{d}$ is the transition dipole and $\vec{E}_L$ is the laser field for photon excitation. This wave-function overlap could change notably on the BCS side, as the ground molecular state becomes increasingly affected by the admixture with atoms. A few-body calculation is needed, i.e., following the theoretical photoassociation work [22], in order to fully understand the magnetic dependence of the Rabi coupling near a Feshbach resonance.

B. Contact measurement at finite temperature

To avoid the complications due to the magnetic field dependence in the Rabi coupling, we may perform the photo-excitation measurement at a fixed magnetic field and consider different temperatures. To calibrate $\Omega^2/\gamma$, we can go to the strong laser intensity regime, in the sense that $\Omega^2/\gamma$ can be enlarged to be comparable to $\Gamma$, as long as there is no significant heating. This will considerably increase the experimental resolution for the loss rate measurement.

In greater detail, by increasing the laser intensity $I$ to increase the Rabi coupling (i.e., $\Omega^2 = \alpha I$), under the resonant condition we may first confirm the predicted line shape in Eq. (27),

$$L(\Delta) = 2\Gamma \frac{\Omega^2}{(2\gamma \Gamma)} \frac{1}{[1 - a_{bg}/a_s]^2 + [\Omega^2/(2\gamma \Gamma)]^2}, \quad (32)$$

which takes a maximum $2\Gamma(1 - a_{bg}/a_s)$. At a given magnetic field, the detunings $\Delta_s, \delta_g, \delta_e$ and $a_s(B)$ are known precisely and the zero-temperature contact has been also determined to a reasonable accuracy. Using these knowledge, we can determine the proportional factor $\alpha$ and hence calibrate the Rabi coupling $\Omega$. Then, working at a fixed Rabi coupling, we may tune the temperature of the system and consequently measure the temperature dependence of the contact coefficient. As the contact coefficient changes significantly across the superfluid phase transition, we can ultimately determine the critical temperature of a strongly interacting Fermi gas across the BEC-BCS crossover.
V. CONCLUSIONS

In summary, we have derived a new universal relation for the photon-excitation measurement of a strongly interacting Fermi gas near a Feshbach resonance. We have shown that the determination of the photo-excitation rate can directly give Tan’s contact coefficient. In the limit of a weak laser intensity, our relation reduces to the well-known relation for the closed-channel molecular fraction, derived earlier by Werner, Tarruell and Castin [15]. With a strong laser intensity, we anticipate that the photon-excitation measurement can have improved experimental resolution and hence provide an accurate way to measure the temperature dependence of the contact coefficient and also the critical temperature at the BEC-BCS crossover.

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[1] S. Tan, Energetics of a strongly correlated Fermi gas, Ann. Phys. 323, 2952 (2008).
[2] S. Tan, Large momentum part of a strongly correlated Fermi gas, Ann. Phys. 323, 2971 (2008).
[3] S. Tan, Generalized Virial Theorem and Pressure Relation for a strongly correlated Fermi gas, Ann. Phys. 323, 2987 (2008).
[4] E. Braaten and D. Kang, Short-Time Operator Product Expansion for rf Spectroscopy of a Strongly Interacting Fermi Gas, Phys. Rev. Lett. 104, 223004 (2010).
[5] E. Taylor and M. Randeria, Viscosity of strongly interacting quantum fluids: Spectral functions and sum rules, Phys. Rev. A 81, 053610 (2010).
[6] S. Giorgini, L. P. Pitaevskii, and S. Stringari, Theory of ultracold atomic Fermi gases, Rev. Mod. Phys. 80, 1215 (2008).
[7] M. Randeria and E. Taylor, Crossover from Bardeen-Cooper-Schrieffer to Bose-Einstein Condensation and the Unitary Fermi Gas, Annu. Rev. Condens. Matter Phys. 5, 209 (2014).
[28] G. M. Falco and H. T. C. Stoof, Atom-molecule theory of broad Feshbach resonances, Phys. Rev. A 71, 063614 (2005).

[29] X.-J. Liu and H. Hu, Self-consistent theory of atomic Fermi gases with a Feshbach resonance at the superfluid transition, Phys. Rev. A 72, 063613 (2005).

[30] G. Zürn, T. Lompe, A. N. Wenz, S. Jochim, P. S. Julienne, and J. M. Hutson, Precise Characterization of $^6$Li Feshbach Resonances Using Trap-Sideband-Resolved RF Spectroscopy of Weakly Bound Molecules, Phys. Rev. Lett. 110, 135301 (2013).

[31] H. Hu, X.-J. Liu, and P. D. Drummond, Equation of state of a superfluid Fermi gas in the BCS-BEC crossover, Europhys. Lett. 74, 574 (2006).

[32] H. Hu, P. D. Drummond, and X.-J. Liu, Universal thermodynamics of strongly interacting Fermi gases, Nat. Phys. 3, 469 (2007).

[33] P. Pellegrini, M. Gacesa, and R. Côté, Giant Formation Rates of Ultracold Molecules via Feshbach-Optimized Photoassociation, Phys. Rev. Lett. 101, 053201 (2008).