Regular Article

Radio-frequency association of molecules: an assisted Feshbach resonance

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Received 26 June 2009

Published online 6 November 2009 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2009

Abstract. We develop a theoretical model to describe the radio-frequency (rf) induced coupling of a pair of colliding atoms to a Feshbach molecule when a magnetic field arbitrarily far from the Feshbach resonance is modulated in time. We use the dressed atom picture, and show that the coupling strength in presence of rf is equal to the Feshbach coupling strength multiplied by the square of a Bessel function. The argument of this function is equal to the ratio of the atomic rf Rabi frequency to the rf frequency. We experimentally demonstrate this law by measuring the rate of rf-association of molecules using a Feshbach resonance in d wave collisions between ultra-cold chromium atoms.

PACS. 34.50.-s Scattering of atoms and molecules – 67.85.-d Ultracold gases, trapped gases

1 Introduction

In a Feshbach resonance, pairs of colliding atoms are resonantly coupled to a molecular bound state. The difference of energy between the molecular state and the pair of atoms can be tuned thanks to the Zeeman effect, so that one can resonantly modify the scattering cross-section between atoms [1]. Another important use of Feshbach resonance is to produce ultra-cold molecules, either by pulsing a magnetic field [2], or by ramping a magnetic field through the resonance [3]. In this context, radio frequency (rf) oscillating magnetic fields have first been employed to dissociate Feshbach molecules and precisely measure their binding energy [4], and then to produce molecules by associating atom pairs near a Feshbach resonance [5,6] or to transfer ground state molecules between states of different vibrational quantum numbers [7]. Thus rf has become one of the tools used for the production of weakly bound Feshbach molecules. These molecules can then be coherently transferred to their rovibrational ground state at high phase-space densities [8], with important perspectives for the study of dipolar gases [9], quantum information [10], or metrology [11].

Coupling between different molecular potentials is not provided by rf, which merely induces precession of the total spin of a molecule. This is in contrast with photoassociation techniques, where the electric dipole operator couples two different molecular potentials [12]. Rf alone does not couple two different vibrational states of a given molecular potential either, because of their orthogonality. Rf association therefore necessarily relies on a coupling between two different molecular potentials (which also provides Feshbach resonances). Such coupling can be due to the spin exchange interaction, or is directly related to the spin (e.g. spin dipole interaction).

It is thus generally considered that the proximity of a Feshbach resonance is needed for rf association to be efficient, as the coupling of the colliding atoms in a given molecular potential to the bound state of another molecular potential is then maximal. Indeed, an increasing number of experiments using rf to associate molecules [13,14] are performed very close to a Feshbach resonance. Theoretical interpretations of the results of these experiments exist, but they rely on numerics, and are very sensitive to the system under study [13–16]. Moreover, although it has been noted that the efficiency of rf association decreases when one gets further away from resonance [17], it has up to now never been shown generically how close one needs to be from the resonance to produce molecules, and how efficient association is, as a function of the rf power and frequency.

In this paper, we show that the dressed-atom picture provides generic answers to these questions. Here we do not address the interesting many-body question of the molecular conversion fraction as a function of phase space density [18], but focus instead on the two-body problem. When the Zeeman effect is linear, rf association can be seen as a photon-assisted Feshbach resonance. We use the

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dressed atom picture to derive simple analytical laws for rf association: when a rf field is applied parallel to the static field the rf coupling strength of the pairs of colliding atoms to the molecular state is equal to the Feshbach coupling strength times the square of a Bessel function of the first kind, when \hbar times the rf frequency ω is equal to the difference of energy between the open and the bound channel. The argument of the Bessel function is the ratio between the rf Rabi frequency Ω and ω . Association of molecules can thus be performed arbitrarily far from the Feshbach resonance provided the available rf Rabi frequency is of the order of ω .

The quantization of the rf field present in the dressed atom picture is in fact not necessary to understand the physics of rf association. We show hereafter, using a Floquet approach, that Bessel functions arise as a consequence of the time-modulation of the eigenenergies of the system by the periodic modulation of the magnetic field: the rf-assisted Feshbach resonance corresponds to a Feshbach resonance between one side-band of the initial eigenstate and the molecular bound state. The situation is similar to the case of Rydberg atoms interacting in the presence of modulated electric fields [19], or to the case of photon-assisted tunneling: in solid-states systems (tunneling between two superconductors [20], in semi-conductor superlattices [21], between quantum dots [22]), or more recently in the study of Bose-Einstein condensates in optical lattices [23]. In all these different systems, an oscillating field creates side bands to the eigenstates, and new resonances occur, whose strength are set by Bessel functions.

We also provide experimental evidence for our theoretical results, using ultra-cold trapped chromium atoms near a Feshbach resonance in d wave collisions. Although in our experiment rf association produces very few molecules (due to weak coupling and short molecular lifetime [24]), our model also applies in the more favorable cases of large s wave resonances (such as in [5,6,14]).

2 Theoretical model

For sake of simplicity, although quantization of the rf field is not required to derive our results, our theoretical description of rf-association uses the dressed atom picture [25]. The Hamiltonian describing the interaction of a pair of atoms with a linear rf field parallel to a static field can be written in the following way:

$$H = H_{mol} + \hbar \omega_0 S_z + \hbar \omega a^+ a + \lambda S_z (a + a^+) + H_{dd}.$$
 (1)

 H_{mol} is the molecular Hamiltonian with no magnetic dipole-dipole interactions. $\hbar\omega_0S_z$ describes the Zeeman effect, which is assumed to be linear; $\hbar\omega_0 = g_J\mu_B B$, with g_J the Landé factor, μ_B the Bohr magneton, and B the magnetic field. $H_{rf} = \hbar \omega a^{\dagger} a + \lambda S_z(a + a^{\dagger})$ accounts for the energy of the linear rf field of angular frequency ω parallel to z, and its coupling to the molecules. We define the Rabi frequency by $2\lambda\sqrt{N}=\hbar\Omega$, where the number of rf photons N is assumed to be large ($\hbar\Omega = g_J \mu_B B_{rf}$, where

 B_{rf} is the amplitude of the rf field). H_{dd} is the dipoledipole interaction Hamiltonian. The key point is that the first part of the Hamiltonian $H_{mol} + \hbar \omega_0 S_z$ commutes with H_{rf} [25]. In absence of H_{dd} the diagonalization of H is exact (irrelevant of the rf power), and the eigenstates appear as a series of manifolds of dressed states:

$$\left|\widetilde{X,N}\right\rangle = T_X^+ \left|X,N\right\rangle$$
 (2)

where X denotes the internal state of the molecule. $T_X = \exp\left(-\frac{M_X\lambda}{\hbar\omega}(a-a^+)\right)$ is a field translation operator, with M_X the spin projection of state X along the axis z. The eigen-energies of the dressed states $|X,N\rangle$ are $W_X = E_X + M_X \hbar \omega_0 + N \hbar \omega - \frac{M_X^2 \lambda^2}{\hbar \omega}$, where E_X is the eigen-energy of state X without rf or magnetic field. The

term $\frac{M_X^2 \lambda^2}{\hbar \omega}$ is negligible, as $N \gg 1$. We apply this model to an isolated Feshbach resonance, between molecular states $|A\rangle = |\psi_{\epsilon}\rangle$ (the incoming channel with collision energy ϵ) and $|B\rangle = |\psi_{bound}\rangle$ of respective spin projections M_A and M_B . The Feshbach resonance (without rf) occurs at a magnetic field such that $E_A - E_B + (M_A - M_B) \hbar \omega_0 = 0$. The strength of the coupling is set by a Fermi golden rule: $\Gamma_0(\epsilon) = 2\pi$ $\psi_{bound}|H_{dd}|\psi_{\epsilon}>|^2$ [26].

In presence of rf, we find that additional resonances occur when eigenstates belonging to different rf manifolds cross, i.e. when $(E_A - E_B + (M_A - M_B) \hbar \omega_0 +$ $(N_A - N_B) \hbar \omega$) is equal to zero. The strength of the resonance coupling between those two dressed states is given by an equation similar to the one used for $\Gamma_0(\epsilon)$:

$$\Gamma_{N_A,N_B}(\epsilon) = 2\pi \left| \left\langle \widetilde{A, N_A} \right| H_{dd} \left| \widetilde{B, N_B} \right\rangle \right|^2
= 2\pi \left| \left\langle A \right| H_{dd} \left| B \right\rangle \left\langle N_A \right| T_A T_B^+ \left| N_B \right\rangle \right|^2
= \Gamma_0(\epsilon) \left| \left\langle N_A \right| e^{\frac{(M_A - M_B)^{\lambda}}{\hbar \omega} (a - a^+)} \left| N_B \right\rangle \right|^2. (3)$$

We follow a calculation generalizing the one in [25], which consists of expanding the exponential in series, and recognizing the series expansion of Bessel functions when the photon number is very large. We then find that:

$$\Gamma_{N_A,N_B}(\epsilon) = \Gamma_0(\epsilon) \left(J_{N_B-N_A} \left(\frac{(M_A - M_B) \Omega}{\omega} \right) \right)^2$$
(4)

where J_N is the Nth Bessel function. This is the main result of this paper. When $\omega > \Gamma_0(\epsilon)$, these Feshbach resonances between dressed states are separated, and the rf coupling strength to the Feshbach molecule with the exchange of $N_B - N_A = M$ photons is proportional to $\left(J_M\left(\frac{(M_A - M_B)\Omega}{\omega}\right)\right)^2$. Rf association can hence be simply interpreted as the results of a Feshbach resonance between dressed states. As $J_M(x)^2 < 1$, the formation of molecules using rf is always less efficient than when setting the magnetic field at resonance without rf.

The occurrence of a Bessel function in equation (4) can also be understood by treating the rf field as a classical field. Let us consider for sake of simplicity the case of a transition between $M_A=-1$ and $M_B=0$ (an extension to arbitrary M_A and M_B is straightforward). As the Zeeman effect is linear, in presence of rf the eigenstate $|A\rangle$ is phase modulated at the rf frequency, whereas $|B\rangle$ is not. The phase time dependent factor $\exp\left(i\left(\frac{(E_A-\hbar\omega_0)t}{\hbar}-\frac{\Omega}{\omega}\sin(\omega t)\right)\right)$ can be developed as $\sum_N(-i)^NJ_N\left(\frac{\Omega}{\omega}\right)\exp(i\left(\frac{(E_A-\hbar\omega_0)t}{\hbar}+N\omega t\right))$, following a Floquet expansion. In the interaction between $|A\rangle$ and $|B\rangle$ in presence of rf, resonances occur when $E_B=E_A-\hbar\omega_0+N\hbar\omega$, which corresponds to a resonant coupling between $|A\rangle$ and $|B\rangle$ with the absorption of N photons. The amplitude of this coupling is therefore set by the fraction of the entrance channel wavefunction evolving at frequency E_B/\hbar , i.e. $J_N\left(\frac{\Omega}{\omega}\right)$. The Bessel function in equation (4) thus appears as the consequence of a phase modulation of the difference of energy between the eigenstates.

3 Experimental results

We now apply the above formalism to the case of the Feshbach resonance in ultra-cold chromium atom collisions studied in [24]. In this resonance, first described in [27], the incoming open channel is a l=2partial wave. $|A\rangle = |S=6, M_S=-6, l=2, m_l=1\rangle \times$ $F_{\epsilon}(R)$ is resonantly coupled to a molecular bound state $|B\rangle = |S = 6, M_S = -5, l = 0\rangle \times F_{bound}(R)$ through dipole-dipole interactions, at a magnetic field of $B_{res} =$ $8.155 \,\mathrm{G.}\, F_{\epsilon}(R)$ and $F_{bound}(R)$ are the radial wavefunctions of respectively the incoming channel and the bound molecular state. At low temperatures, coupling of the states $|A\rangle$ and $|B\rangle$ is strongly inhibited because of the centrifugal barrier in the incoming channel, which reduces the overlap between $F_{\epsilon}(R)$ and $F_{bound}(R)$. Given the experimental atomic density, the coupling rate is then much smaller than the collisionally limited life-time of the bound state, so that association of molecules at the Feshbach resonance simply translates into losses, with a loss rate solely determined by the Feshbach coupling strength itself [24]:

$$\frac{\dot{n}}{n} = -\alpha \left(n\Lambda_{dB}^3 \right) \Gamma_0(\epsilon_0) \exp(-\epsilon_0/k_B T) \equiv -K_2^0(\epsilon_0) n \quad (5)$$

where $\Lambda_{dB} = \frac{h}{\sqrt{2\pi m k_B T}}$ is the thermal de Broglie wavelength, $\alpha = 6\sqrt{2}$ is a numerical factor, and $\epsilon_0 = g_J \mu_B (B - B_{res})$ is the energy of the bound molecular state relative to the dissociation limit of the incoming channel. $g_J \approx 2$ for chromium atoms in the ⁷S₃ state. $\Gamma_0(\epsilon) \propto \epsilon^{5/2}$ describes the Feshbach coupling, and $K_2^0(\epsilon_0)$ is the associated loss parameter. Analysing losses near this Feshbach resonance is thus a good means to directly measure $\Gamma_0(\epsilon)$. We emphasize the difference with usual s wave Feshbach resonances, where $\Gamma_0(\epsilon)$ can be deduced from the value of the magnetic field resonance width (larger than $k_B T$, as opposed to our case), and the background scattering length [3].

In our experiments, we use rf fields with a frequency $\omega/2\pi$ close to ϵ_0/h . As in [24], we analyse losses, deduce a rf dependent loss parameter $K_2(\Omega,\omega,\epsilon_0)$ and relate it to

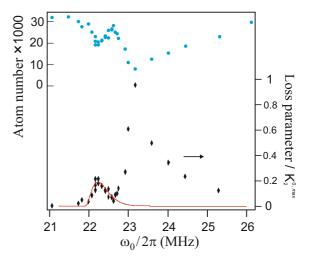


Fig. 1. (Color online) Losses as a function of the Larmor frequency $\omega_0/2\pi$. We report: (up, left) blue circles: the number of atoms after 8 s hold time in the static magnetic field corresponding to ω_0 , in presence of a 900 kHz frequency rf field; (down, right) black diamonds: the corresponding loss parameter divided by the maximum loss parameter without rf. The red solid line corresponds to the results from a fit of the data without rf using equation (5) (see Fig. 2), shifted by 900 kHz, and multiplied by $(J_1(1.0))^2$.

the rf-assisted coupling parameter, using equation (5). As the rf-assisted coupling strength is related to the Feshbach coupling strength (Eq. (4)), we therefore conclude that

$$K_2(\Omega, \omega, \epsilon_0) = K_2^0(\epsilon_0 + \hbar\omega) \left(J_1(\Omega/\omega)\right)^2.$$
 (6)

 $K_2(\Omega, \omega, \epsilon_0)n$ gives the rate of rf association of molecules. We experimentally demonstrate this result below.

In the experiments reported here, we follow the procedure described in [24]. We load cold ground state chromium atoms in a one-beam optical dipole trap and polarize them in the lowest energy state $|S=3,m_S=-3\rangle$. Forced evaporation is performed by transferring power from this horizontal dipole trap to a crossed vertical beam. At the end of this process, the atom number is between 50 000 and 60 000 and the temperature is 7 μ K. At this point we tune a static homogeneous magnetic field B near B_{res} . We use rf spectroscopy to calibrate the magnetic field with an uncertainty of 2 mG.

We report in Figure 1 the number of atoms remaining in the trap after 8 s in presence of an rf magnetic field with a fixed frequency $\omega/2\pi=900$ kHz and $\Omega/\omega=1.0$, as a function of the Larmor frequency $\omega_0/2\pi$ associated with the static field. When the pairs of colliding atoms are coupled to the bound molecular state, we observe losses, because the molecular state has a very short lifetime, as explained above. In this loss spectrum we observe two peaks: one due to the Feshbach resonance (which we will refer to as the 'Feshbach peak') and a smaller one shifted to lower magnetic fields (the 'rf peak'). This latter peak results from the coupling between the closed and open molecular channels by the rf field, with the *emission* of one rf photon.

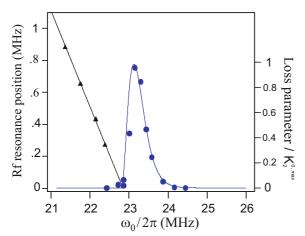


Fig. 2. (Color online) (Right) Loss parameter without rf (blue circles). The blue solid line is given by a fit according to equation (5). The salient point gives the value of the magnetic field at the Feshbach resonance. (Left) rf spectroscopy of the Feshbach resonance (black triangles): the position of the rf resonant losses, as given by a fit according to equation (5), varies linearly with the static magnetic field. The black solid line is a linear fit of slope one.

We also plot in Figure 1 the corresponding loss parameter, deduced by comparing the initial number of atoms to the number of atoms remaining in the trap after 8 s, assuming a two body loss process. The lineshapes of the rf peak and of the Feshbach resonance without rf, which is plotted in Figure 2, are very similar. The separation between the two lineshapes is exactly the rf frequency. The agreement with equation (6) is excellent.

Concerning the 'Feshbach peak', we find that the line-shape of the loss parameter in presence of rf is distorted (Fig. 1) as compared to when the rf is off (Fig. 2). In particular, more losses are observed at high magnetic fields. We do not account for these larger losses; they may be induced by the coupling between the closed and open molecular channels associated with the *absorption* of one or more rf photons. We have not studied this feature in detail, and the rest of the paper concentrates on the rf peak.

Tuning B to a value below B_{res} , we measure losses as a function of the rf frequency. We then observe rf-induced losses centered at rf frequencies depending on B. For a given B, we fit the rf loss curve using a K_2 of the form given in equation (5), and we report in Figure 2 the fitted position of the resonance (i.e. using ϵ_0 as a free parameter) as a function of B. We find that the resonant rf frequency varies linearly with B. A linear fit of slope one to the rf resonance frequency as a function of B (in MHz) predicts that the 'rf peak' merges with the 'Feshbach peak' at B = 8.157 G. This value corresponds within a very good accuracy to the previously measured position of the Feshbach resonance [24]. Losses in presence of rf are thus related to population of the molecular bound state involved in the Feshbach resonance. The linear dependence versus B of the relative Zeeman energy between the closed and the open channel is expected, as the value of $\Gamma_0(\epsilon)$ is

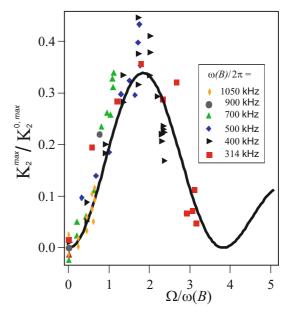


Fig. 3. (Color online) rf-assisted maximum loss parameter K_2^{max} as a function of $\Omega/\omega(B)$ for different resonant rf frequencies, corresponding to various magnetic fields B. K_2^{max} is scaled to the maximum loss parameter measured at the Feshbach resonance without rf. The solid line is $J_1(\Omega/\omega)^2$.

typically 10 Hz at 8 μK [24], much smaller than the typical detuning from resonance: $\Gamma_0(\epsilon)/\omega \approx 10^{-5}$. We therefore expect to be in the regime where our theoretical model is valid. We underline that we have used detunings which are much larger than the ones which can be found in the literature; for example in [17] we estimate that $\Gamma_0(\epsilon)/\omega$ is between 0.1 and 1.

We now turn to the quantitative analysis of the amplitude of rf induced losses. The 'rf peak' is maximum for an rf frequency $\omega(B)$ which varies linearly with B. This frequency corresponds to the maximum of the lineshape described by equation (6). We find that the corresponding loss parameter K_2^{max} depends both on Ω and on $\omega(B)$. We plot K_2^{max} as a function of $\Omega/\omega(B)$ in Figure 3 for different resonant rf frequencies $\omega(B)$. In this figure, we have divided K_2^{max} by the maximum loss parameter measured at the Feshbach resonance without rf, $K_2^{0,max} = 4 \times 10^{-20} \mathrm{m}^3 \mathrm{s}^{-1}$. Within the experimental error bars, this scaled loss parameter exactly fits equation (6), with no adjustable parameter: Ω is measured by observing Rabi oscillations at resonance for any ω .

4 Discussion

Our framework, supported by the experimental evidence of Figure 3, treats rf association as a Feshbach resonance between dressed states. Other observations validate this treatment. In particular, we have found that all the peculiar features observed for the Feshbach resonance [24] also apply to the case of the rf-resonant losses: for example, the dynamics of the rf-induced atom decay is well described

assuming a two-body loss parameter; both the temperature dependence and the lineshape of the rf-resonant loss parameter also are, to within the experimental uncertainty, identical to the ones of the Feshbach resonance. This indicates that the rf association process is qualitatively similar to the coupling to the molecular bound state at the Feshbach resonance itself. The advantage of using rf fields to associate molecules rather than using magnetic fields sweeps near the Feshbach resonance itself simply relies on the larger versatility of rf components (e.g. smaller switching and ramping times).

With sufficient rf power, one should also observe Feshbach resonances assisted by multiphoton processes. For a given magnetic field, a process with the emission of two photons happens at $\hbar\omega(B)/2$. We indeed observed such resonances. These are unfortunately more difficult to analyze, mainly because at large rf power the rf amplifier produces non negligible power in the second harmonics, and two-photon processes at $\omega(B)/2$ coexist with one-photon processes at $2(\omega(B)/2)$.

The agreement between our data and equation (6) is good up to $\Omega/\omega \approx 4$, but breaks down for higher Ω/ω . In practice, given our rf amplifier, $\Omega/\omega > 4$ corresponds to $\omega < 300$ kHz, for which we observe shifts of the molecular level, and of the Feshbach resonance, as a function of rf power. Similar disagreement between the Bessel-function predictions and observations was mentioned in [28] in the case of resonant dipole-dipole collisional energy transfer of K Rydberg atoms, and was also attributed to AC Stark shifts.

5 Conclusion

In conclusion, we have demonstrated a simple formula for the strength of coupling of colliding atoms to a molecular bound state when a magnetic field arbitrarily far from a Feshbach resonance is modulated in time. This formula is valid as long as the Zeeman shift is linear and the rf frequency is larger than the resonance width. We show that it is possible to efficiently associate molecules far from a Feshbach resonance, provided an rf Rabi frequency similar to the rf frequency is achievable. In our experiment, the detuning of the magnetic field from the Feshbach resonance was up to 10^5 times the resonance width. We have derived these results assuming that coupling is provided by dipolar interactions, and that there is no hyperfine structure. However, the model can readily be extended to any other coupling, and the results remain valid in presence of hyperfine structure, provided that the Zeeman effect is linear over a range of fields set by the amplitude of rf modulation.

There is an analogy between the present work and previous studies of electric field resonant dipole-dipole collisional energy transfer in Rydberg atoms in presence of microwave [19]. In that case, the micro-wave assisted cross-section for collisional energy transfer is also set by Bessel functions. As stated above, the situation also resembles the case of photon-assisted tunneling in solid-state systems [20–22] or in Bose-Einstein condensates [23]. One

difference between our results and the ones found in [19] is that our results involve the square of Bessel function, and not the absolute value. Our situation is described by a Fermi golden rule, because the rate of decay of the molecules is much larger than the rate at which they are produced, whereas the absolute value occurs in the regime of strong (Rabi) coupling. The situation in [23] may be an interesting case of an intermediate situation between the coherent Rabi coupling and the Fermi golden rule dissipative regime (also known as sequential tunneling in the framework of solid-state physics), and it should be possible to study a similar cross-over by working with broader Feshbach resonances.

LPL is Unité Mixte (UMR 7538) of CNRS and of Université Paris Nord. We acknowledge financial support Ministère de l'Enseignement Supérieur et de la Recherche (CPER), IFRAF (Institut Francilien de Recherche sur les Atomes Froids) and the Plan-Pluri Formation (PPF) devoted to the manipulation of cold atoms by powerful lasers.

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