P H Y S I C A L   R E V I E W   L E T T E R S

Spontaneous Dissociation of Long-Range Feshbach Molecules

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We study the spontaneous dissociation of diatomic molecules produced in cold atomic gases via magnetically tunable Feshbach resonances. We provide a universal formula for the lifetime of these molecules that relates their decay to the scattering length and the loss rate constant for inelastic spin relaxation. Our universal treatment as well as our exact coupled channels calculations for ^{85}Rb dimers predict a suppression of the decay over several orders of magnitude when the scattering length is increased. Our predictions are in good agreement with recent measurements of the lifetime of ^{85}Rb$_2$.

The discovery of Feshbach resonances in cold gases provides the unique opportunity to widely tune the interatomic interactions via magnetic fields near a singularity of the scattering length. This singularity is due to the near degeneracy of the very small collision energy of the cold atoms with the binding energy of an extremely loose diatomic molecular state for magnetic fields at which the scattering length is positive. A number of recent experiments, as summarized in the accompanying experimental Letter of Thompson *et al.* [1], have taken advantage of this virtual energy match to populate this molecular state. The interacting pairs of atoms in the first studies of these cold Feshbach molecules [2,3] were subject to deeply inelastic spin relaxation collisions involving the deexcitation of at least one of the colliding atoms. Spin relaxation can affect both free [4] and bound atom pairs.

In this Letter we predict the spontaneous dissociation of Feshbach molecules due to spin relaxation. We provide a universal treatment of the molecular decay by which we relate the molecular lifetime τ to the spatial extent of the bound state wave function and the loss rate constant for inelastic spin relaxation collisions. Both the size of the molecule and the loss rate constant exhibit a pronounced resonance enhancement at the singularity of the scattering length. General considerations on the nature of this enhancement allow us to predict the functional form of the magnetic field dependence of τ. We show that the near resonant molecular lifetime increases with increasing scattering lengths and can therefore be varied over several orders of magnitude in related experiments. We demonstrate the predictive power of our universal treatment in a comparison with exact coupled channels calculations of the lifetime of the ^{85}Rb dimers produced in the experiments of Refs. [1,2,5]. The exact calculations show that τ varies in the remarkable range between only 100 μs and roughly 30 ms in the experimental range of magnetic field strengths. Our studies indicate that it is the large spatial extent of the near resonant bound state wave function that

can provide the crucial stability of the Feshbach molecules with respect to their spontaneous decay.

In the following, we shall consider pairs of atoms in a cold gas that interact via *s* waves. We denote the binary scattering channel of a pair of asymptotically free atoms as the entrance channel and choose the zero of energy at its dissociation threshold. This threshold is determined by the Zeeman energy of the separated atoms. The Hamiltonian H of the relative motion can then be divided into the contribution H_{cl} of the closed channels with dissociation thresholds at or above zero energy (including the entrance channel), the contribution H_{op} of the open channels with negative threshold energies, as well as the weak coupling H_{int} between the closed and open channels:

$$
H = \begin{pmatrix} H_{op} & H_{int} \\ H_{int}^\dagger & H_{cl} \end{pmatrix}. \tag{1}
$$

We shall consider magnetic field strengths on the side of positive scattering lengths of a zero energy resonance (i.e., a singularity of the scattering length). The large positive values of the scattering length imply the existence of a weakly bound (metastable) molecular state ϕ_{-1}^{cl}, which corresponds to the highest excited vibrational ($v = -1$) bound state of H_{cl}. The wave function ϕ_{-1}^{cl} thus fulfills the stationary Schrödinger equation $H_{cl}\phi_{-1}^{cl} = E_{-1}^{cl}\phi_{-1}^{cl}$, where E_{-1}^{cl} is the negative binding energy. Fermi's golden rule determines the exponential decay of the Feshbach molecule by a rate

$$
\gamma = -\frac{2}{\hbar}\mathrm{Im}[\langle \phi_{-1}^{cl}|H_{int}^\dagger G_{op}(E_{-1}^{cl} + i0)H_{int}|\phi_{-1}^{cl}\rangle]. \tag{2}
$$

Here $G_{op}(E_{-1}^{cl} + i0) = (E_{-1}^{cl} + i0 - H_{op})^{-1}$ is the Green's function of the open binary scattering channels, whose energy argument $E_{-1}^{cl} + i0$ indicates that E_{-1}^{cl} is approached from the upper half of the complex plane. As $|E_{-1}^{cl}|$ is much smaller than typical transition energies, it can be neglected in the argument of the Green's function.

Fermi's golden rule also determines the loss rate constant for inelastic spin relaxation collisions to be

$$K_2 = -\frac{4}{\hbar}(2\pi\hbar)^3 \mathrm{Im}[\langle \phi_{0,\mathrm{cl}}^{(+)}|H_{\mathrm{int}}^\dagger G_{\mathrm{op}}(i0)H_{\mathrm{int}}|\phi_{0,\mathrm{cl}}^{(+)}\rangle]. \quad (3)$$

Here $\phi_{0,\mathrm{cl}}^{(+)}$ is the zero energy scattering state associated with the Hamiltonian H_{cl}, which fulfils the stationary Schrödinger equation $H_{\mathrm{cl}}\phi_{0,\mathrm{cl}}^{(+)} = 0$. The prefactors in Eq. (3) correspond to a cold thermal gas of identical bosons. The associated rate equation reads [4] $\dot{N}(t) = -K_2 \langle n(t)\rangle N(t)$, where $N(t)$ is the number of atoms and $\langle n(t)\rangle$ is their average density.

At magnetic field strengths in the vicinity of a zero energy resonance, $\phi_{0,\mathrm{cl}}^{(+)}$ and ϕ_{-1}^{cl} are determined by their wave functions in the entrance channel and in a closed channel strongly coupled to it [6,7]. This strong interchannel coupling is due to the near degeneracy of the energy $E_{\mathrm{res}}(B)$ of a closed channel vibrational state (the bare Feshbach resonance level) ϕ_{res} with the dissociation threshold of the entrance channel. The restricted two-body Hamiltonian H_{cl} of the entrance channel and the closed channel can be effectively described by:

$$H_{\mathrm{cl}} = \begin{pmatrix} -\frac{\hbar^2}{m}\nabla^2 + V_{\mathrm{bg}}(r) & W(r) \\ W(r) & -\frac{\hbar^2}{m}\nabla^2 + V_{\mathrm{cl}}(B,r) \end{pmatrix}. \quad (4)$$

Here r denotes the relative distance between the atoms and m is twice their reduced mass, B is the magnetic field strength, $V_{\mathrm{bg}}(r)$ is the background scattering potential of the entrance channel, and $W(r)$ provides the interchannel coupling. The closed channel potential $V_{\mathrm{cl}}(B,r)$ supports the resonance state, i.e., $[-\hbar^2\nabla^2/m + V_{\mathrm{cl}}(B,r)]\phi_{\mathrm{res}}(r) = E_{\mathrm{res}}(B)\phi_{\mathrm{res}}(r)$. The dissociation threshold of $V_{\mathrm{cl}}(B,r)$ is determined by the energy of a pair of noninteracting atoms in the closed channel. The relative Zeeman energy shift between the two channels as well as the bare energy $E_{\mathrm{res}}(B)$ can be tuned by varying the magnetic field strength B. We denote by B_{res} the magnetic field strength, at which $E_{\mathrm{res}}(B)$ crosses the dissociation threshold energy of the entrance channel, i.e., $E_{\mathrm{res}}(B_{\mathrm{res}}) = 0$. The bare energy $E_{\mathrm{res}}(B)$ varies virtually linearly in B, and an expansion about B_{res} yields $E_{\mathrm{res}}(B) = \mu_{\mathrm{res}}(B - B_{\mathrm{res}})$. Here μ_{res} is the difference in magnetic moment between the Feshbach resonance state and a pair of atoms in the entrance channel.

Under the assumption that the spatial configuration of a pair of atoms in the closed channel is restricted to the resonance state $\phi_{\mathrm{res}}(r)$, the dressed highest excited vibrational bound state ϕ_{-1}^{cl} of H_{cl} is given by [7]:

$$\phi_{-1}^{\mathrm{cl}} = \frac{1}{\mathcal{N}}\begin{pmatrix} G_{\mathrm{bg}}(E_{-1}^{\mathrm{cl}})W\phi_{\mathrm{res}} \\ \phi_{\mathrm{res}} \end{pmatrix}. \quad (5)$$

Here $G_{\mathrm{bg}}(E_{-1}^{\mathrm{cl}}) = (E_{-1}^{\mathrm{cl}} + \hbar^2\nabla^2/m - V_{\mathrm{bg}})^{-1}$ is the Green's function associated with the entrance channel, and the factor $1/\mathcal{N}$ assures that ϕ_{-1}^{cl} is unit normalized. The associated binding energy E_{-1}^{cl} is determined by [7]

$E_{-1}^{\mathrm{cl}} = E_{\mathrm{res}}(B) + \langle \phi_{\mathrm{res}}|WG_{\mathrm{bg}}(E_{-1}^{\mathrm{cl}})W|\phi_{\mathrm{res}}\rangle$. The strong coupling between the entrance channel and the bare Feshbach resonance state $\phi_{\mathrm{res}}(r)$ also determines the zero energy scattering state $\phi_{0,\mathrm{cl}}^{(+)}$ of H_{cl} by the general formula [7]:

$$\phi_{0,\mathrm{cl}}^{(+)} = \begin{pmatrix} \phi_{0,\mathrm{bg}}^{(+)} + G_{\mathrm{bg}}(0)W\phi_{\mathrm{res}}A(B) \\ \phi_{\mathrm{res}}A(B) \end{pmatrix}. \quad (6)$$

Here $\phi_{0,\mathrm{bg}}^{(+)}$ denotes the bare zero energy state associated with the background scattering, i.e., $[-\hbar^2\nabla^2/m + V_{\mathrm{bg}}(r)]\phi_{0,\mathrm{bg}}^{(+)}(r) = 0$, and the amplitude $A(B)$ in Eq. (6) is given by [7]:

$$A(B) = -\frac{\langle \phi_{\mathrm{res}}|W|\phi_{0,\mathrm{bg}}^{(+)}\rangle}{E_{\mathrm{res}}(B) + \langle \phi_{\mathrm{res}}|WG_{\mathrm{bg}}(0)W|\phi_{\mathrm{res}}\rangle}. \quad (7)$$

The scattering length $a(B)$ can then be obtained from the asymptotic behavior $(2\pi\hbar)^{-3/2}[1 - a(B)/r]$ of the entrance channel component of Eq. (6) at large interatomic distances r. This yields

$$a(B) = a_{\mathrm{bg}}\left(1 - \frac{\Delta B}{B - B_0}\right), \quad (8)$$

where $\Delta B = (2\pi\hbar)^3 |\langle \phi_{\mathrm{res}}|W|\phi_{0,\mathrm{bg}}^{(+)}\rangle|^2 m/(4\pi\hbar^2 a_{\mathrm{bg}}\mu_{\mathrm{res}})$ is the resonance width and a_{bg} is the background scattering length. The singularity of $A(B)$ thus determines the measurable position B_0 of the zero energy resonance, which is distinct from the zero B_{res} of the bare energy $E_{\mathrm{res}}(B)$ by the shift $B_0 - B_{\mathrm{res}} = -\langle \phi_{\mathrm{res}}|WG_{\mathrm{bg}}(0)W|\phi_{\mathrm{res}}\rangle/\mu_{\mathrm{res}}$ of the denominator on the right-hand side of Eq. (7).

At near resonant magnetic field strengths the binding energy is determined by $E_{-1}^{\mathrm{cl}} = -\hbar^2/(ma^2)$ (cf., e.g., Refs. [2,5,7]). The energy argument of the Green's function in Eq. (5), therefore, vanishes at large positive scattering lengths. In this limit the background scattering contribution $\phi_{0,\mathrm{bg}}^{(+)}$ to the right-hand side of Eq. (6) also becomes negligible for interatomic distances on the length scale set by the range of the interchannel coupling H_{int} in Eqs. (2) and (3). Equations (5) and (6) thus coincide up to a factor $\mathcal{N}A(B)$, and Eqs. (2) and (3) yield $K_2(B)/\gamma(B) = 2(2\pi\hbar)^3|A(B)|^2\mathcal{N}^2(B)$. The near resonant amplitude $A(B)$ is readily obtained from the resonance width and shift of Eq. (8), while the normalization constant $\mathcal{N}^2(B) = (\mu_{\mathrm{res}}\Delta B)ma_{\mathrm{bg}}a(B)/(2\hbar^2)$ can be determined from $\langle \phi_{-1}^{\mathrm{cl}}|\phi_{-1}^{\mathrm{cl}}\rangle = 1$ [7]. The molecular lifetime $\tau(B) = 1/\gamma(B)$ is then given by the universal formula

$$\tau(B) = 4\pi a^3(B)/K_2(B) = [4/K_2(B)]4\pi\langle r^3\rangle/3. \quad (9)$$

Here $\langle r^3\rangle$ is the expectation value of r^3 for the near resonant wave function $\phi_{-1}^{\mathrm{cl}}(r) = \exp(-r/a)/(r\sqrt{2\pi a})$ [7,8].

General considerations [9] show that in the presence of open decay channels the resonance enhanced zero energy binary elastic and inelastic collision cross sections can be

described in terms of a complex scattering length $a(B) - ib(B)$, whose imaginary part is related to the loss rate constant by

$$K_2(B) = 16\pi\hbar b(B)/m. \tag{10}$$

Its real part is well approximated by Eq. (8), except that $a^2(B)$ approaches a large but finite value at $B = B_0$, which is determined by the decay width. We shall presuppose in the following that only the bare resonance state ϕ_{res} decays to a set of final channels with a total rate $\gamma_{\text{res}} = 1/\tau_{\text{res}}$, where τ_{res} is the associated lifetime. The imaginary part of the complex scattering length is then given by a Lorentzian function

$$b(B) = a_{bg}\Delta B \frac{\hbar\gamma_{\text{res}}/(4\mu_{\text{res}})}{(B - B_0)^2 + [\hbar\gamma_{\text{res}}/(4\mu_{\text{res}})]^2}. \tag{11}$$

Inserting Eqs. (8), (10), and (11) into Eq. (9) then yields

$$\tau(B) = \tau_{\text{res}} \frac{ma_{bg}^2\mu_{\text{res}}\Delta B}{4\hbar^2} x^2\left(\frac{1}{x} - 1\right)^3 = \tau_{\text{res}}f(B), \tag{12}$$

where we have introduced $x = (B - B_0)/\Delta B$. This formula provides the general functional form of $\tau(B)$ as well as its order of magnitude in terms of the constant factor τ_{res}, which can be obtained either from a measurement of $K_2(B)$, at a single magnetic field strength, or from coupled channels calculations.

We shall demonstrate the predictive power of Eqs. (9) and (12) for the example of the Feshbach molecules associated recently from pairs of ^{85}Rb atoms in a Bose-Einstein condensate [1,2,5] at magnetic field strengths of about 15.5 mT. The nuclear spin $I = 5/2$ of ^{85}Rb gives rise to two energetically relevant hyperfine levels with total spin quantum numbers $f = 2$ and 3. A homogeneous magnetic field B splits the Zeeman sublevels into a nondegenerate set of levels (f, m_f), labeled by the projection quantum number m_f along the axis of the magnetic field and the f value with which it correlates adiabatically at zero field. In the experiments [2,5] the atoms were prepared in the $(f = 2, m_f = -2)$ internal state. The binary asymptotic scattering channels are then characterized by pairs of internal quantum numbers $(f, m_f; f', m'_f)$, as well as the orbital angular momentum quantum number ℓ associated with the relative motion of the atom pair and its projection quantum number m_ℓ. The s wave ($\ell = 0, m_\ell = 0$) entrance channel is thus characterized just by the internal quantum numbers $(2, -2; 2, -2)$. The cylindrical symmetry of the setup implies that the projection quantum number $m_f + m'_f + m_\ell = -4$ of the total angular momentum is conserved in a binary collision.

We have applied the coupled channels approach of Ref. [6] to determine the scattering length $a(B)$ (see Fig. 1), including spin exchange interactions, which couple the entrance channel to the four closed s wave scattering channels $(3, -3; 2, -1)$, $(3, -2; 2, -2)$, $(3, -1; 3, -3)$, and
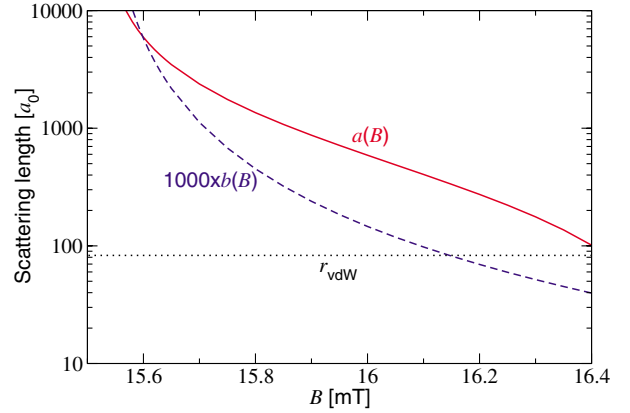

FIG. 1 (color online). Magnetic field dependence of the real (solid curve) and imaginary (dashed curve) parts of the complex scattering length $a(B) - ib(B)$. The dotted line indicates the length scale [13] $r_{\text{vdW}} = (mC_6/\hbar^2)^{1/4}/2$ set by the long-range asymptotic $-C_6/r^6$ van der Waals tail of the background scattering potential. We note that $b(B)$ is multiplied by a factor of 1000.

$(3, -2; 3, -2)$. A fit to Eq. (8) predicts the parameters $B_0 = 15.520$ mT, $\Delta B = 1.065$ mT, and $a_{\text{bg}} = -484.1$ a_0 ($a_0 = 0.0529\,177$ nm) for the experimentally well characterized 15.5 mT zero energy resonance of ^{85}Rb [5]. The decay of the Feshbach molecules is due to spin-dipole interactions, which couple s waves to d waves. Among the 23 d wave channels coupled to the entrance channel only three are open. These d wave decay exit channels are characterized by the internal quantum numbers $(2, -2; 2, -1)$, $(2, -2; 2, 0)$, and $(2, -1; 2, -1)$ in order of increasing energy release. The transition energies are all on the order of several mK (in units of $k_B = 1.3\,806\,505 \times 10^{-23}$ J/K), which implies that the decay products are lost from an atom trap. Including spin-dipole interactions in the coupled channels calculations shifts the predicted position B_0 by less than 1 μT but it provides the small imaginary part $b(B)$ of the scattering length (see Fig. 1), which determines the loss rate constant $K_2(B)$ by Eq. (10).

Given the magnetic moment difference $\mu_{\text{res}}/h = 34.66$ MHz/mT, a fit of Eq. (11) to the curve of $b(B)$ in Fig. 1 determines the lifetime of the bare resonance level to be $\tau_{\text{res}} = 1/\gamma_{\text{res}} = 32$ μs. The solid curve in Fig. 2 shows the associated universal estimates of the lifetimes $\tau(B)$ of the dressed ^{85}Rb$_2$ Feshbach molecules [2,5] determined from Eq. (9), using the exact loss rate constant $K_2(B)$ [cf. Eq. (10) and Fig. 1]. The dashed curve indicates the predictions of Eq. (12). We expect these estimates to be accurate between B_0 and about 16.0 mT, where the scattering length $a(B)$ by far exceeds the length scale $r_{\text{vdW}} = 84a_0$ set by the long-range asymptotic behavior of the van der Waals interaction (see Fig. 1). This trend is confirmed by our direct calculations of the molecular lifetime shown in Fig. 2. The exact lifetime can be obtained from the energy dependence of the inelastic scattering cross section
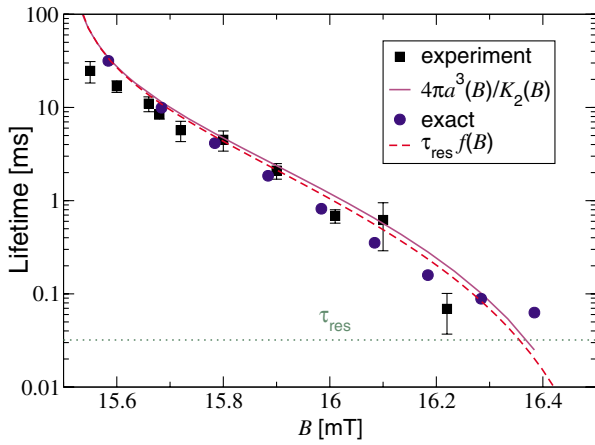
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FIG. 2 (color online). Two-body decay lifetime $\tau(B)$ of the $^{85}\mathrm{Rb}_2$ Feshbach molecules [2,5] versus magnetic field strength B near 15.5 mT. The squares indicate the experimental data of Thompson *et al.* [1]. The circles indicate exact predictions of coupled channels calculations, whereas the solid and dotted curves show the results of the universal formulas (9) and (12), respectively. The magnitude of the bare state lifetime τ_{res} is indicated by the dotted line. For the purpose of comparison, we have slightly corrected the axis of the magnetic field strength in such a way that the theoretical resonance position recovers the measured value of $B_0 = 15.5041$ mT [5].

between any pair of open decay channels. We have chosen transitions between the open d wave channels associated with the internal atomic quantum numbers $(2, -2; 2, -1)$ and $(2, -2; 2, 0)$. Fitting the standard Breit-Wigner formula to the resonance peak at the binding energy $E_b(B)$ [2,5] of the metastable Feshbach molecules then determines the decay rate $\gamma(B) = 1/\tau(B)$ through its width.

The squares in Fig. 2 indicate lifetime measurements by Thompson *et al.* [1] in which the dimer molecules were produced in a dilute vapor of $^{85}\mathrm{Rb}$ atoms at a temperature of 30 nK using a Feshbach resonance crossing technique (see, e.g., Refs. [6,7]). Our theoretical predictions agree with the experimental lifetimes in both their systematic trends and magnitudes. Figure 2 shows the large differences in the order of magnitude of $\tau(B)$ from roughly 30 ms to only 100 μs in the experimentally accessible [1] range of magnetic field strengths from about 15.6 to 16.2 mT. The lifetime correlates with the admixture of the bare resonance level to the dressed molecular state, which, in turn, is related to the spatial extent of the molecules [8]. Equation (9) can indeed be interpreted in the intuitive way that $4\pi\langle r^3 \rangle/3$ sets the scale of the volume confining the bound atom pair, while $K_2(B)/4$ is the event rate constant for molecular decay (which is the same as that for the inelastic collision of a pair of identical Bose condensed

atoms [10]). The large extent of the molecular wave function at near resonant magnetic field strengths thus protects the Feshbach molecules against their spontaneous decay. We caution, however, that purely two-body theory may eventually become inadequate sufficiently close to resonance; for example, the mean distance between atoms at the experimental [1] peak density of 6.6×10^{11} cm^{-3} is only 2×10^4 a_0, which is comparable to the estimated bond length of the molecules [8] of about 3×10^3 a_0 at 15.6 mT.

Our general theory applies to a variety of Feshbach molecules with open decay exit channels [11]. Among these species are the $^{40}\mathrm{K}_2$ [3] and $^6\mathrm{Li}_2$ [12] molecules produced in Fermi gases in the s wave entrance channels associated with the pairs of atomic quantum numbers $(9/2, -9/2; 9/2, -5/2)$ and $(1/2, 1/2; 3/2, -3/2)$, respectively. Measurements of two-body decay lifetimes could allow the characterization of the spin-dipole interaction between various atomic species with an unprecedented precision. Our universal estimate determines the conditions for the stability of Feshbach molecules, which is crucial to all future studies of cold molecular gases in the presence of open decay exit channels.

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[11] In the case of molecules composed of Fermions in different spin states the event rate constant $K_2/4$ in Eq. (9) needs to be replaced by $K_2/2$.
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