Creating Feshbach resonances for ultracold molecule formation with radio-frequency fields

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(Received 13 May 2016; published 15 August 2016)

We show that radio-frequency (rf) radiation may be used to create Feshbach resonances in ultracold gases of alkali-metal atoms at desired magnetic fields that are convenient for atomic cooling and degeneracy. For the case of ^{39}K + ^{133}Cs, where there are no rf-free resonances in regions where Cs may be cooled to degeneracy, we show that a resonance may be created near 21 G with 69.2 MHz rf radiation. This resonance is almost lossless with circularly polarized rf, and the molecules created are long-lived even with plane-polarized rf.

DOI: 10.1103/PhysRevA.94.023619

I. INTRODUCTION

Polar molecules formed from ultracold atoms are opening up new possibilities for quantum-controlled chemistry [1], precision measurement [2–4], quantum computation [5], quantum phase transitions [6], and quantum simulation [7,8]. The last few years have seen major success, with the formation of ultracold ^{40}K ^{87}Rb [9], ^{87}Rb ^{133}Cs [10,11], ^{23}Na ^{40}K [12], and most recently ^{23}Na ^{87}Rb [13] molecules in their absolute ground states. Molecules are first formed by magnetoassociation, in which atom pairs are converted into weakly bound molecules by ramping a magnetic field across a magnetically tunable Feshbach resonance. The resulting "Feshbach molecules" are then transferred to the polar ground state by stimulated Raman adiabatic passage (STIRAP). The ground-state molecules have been confined in one-dimensional [14] and three-dimensional [15] optical lattices and used to study atom-molecule and molecule-molecule collision processes [10,16].

A major problem in this field is that the magnetoassociation step is possible only if there is a Feshbach resonance of suitable width at a magnetic field where there is a lucky combination of intraspecies and interspecies scattering lengths. Ideally, all three scattering lengths have moderate positive values to allow cooling, condensate formation, and mixing of the two atomic clouds. For the intraspecies scattering lengths, negative values cause condensate collapse, whereas excessively positive values cause loss through fast three-body recombination. For the interspecies scattering length, a large negative value can cause collapse of the mixed condensate, while a large positive value can make the condensates of the two species immiscible. Although magnetoassociation can be carried out in low-temperature thermal gases that are not subject to condensate collapse, it is much less efficient than in condensates and does not produce high densities of molecules. This is the so-called one-field problem, because a single field must be chosen to satisfy several different criteria, and such a field may not (often does not) exist.

The purpose of this paper is to show that radio-frequency (rf) fields can be used to produce new Feshbach resonances that offer additional possibilities for magnetoassociation. In particular, they may be used to produce resonances at magnetic fields where the scattering lengths have the desired properties. Formally similar resonances have been considered previously in homonuclear systems [17–19], and molecules have been formed by direct rf association [20,21]. We propose here that rf-induced resonances may provide a solution to the one-field problem in heteronuclear systems.

We recently considered the possibilities for magnetoassociation to form molecules in mixtures of ^{39}K, ^{40}K, and ^{41}K with ^{133}Cs [22] by performing coupled-channel calculations of the Feshbach resonance positions and widths, using interaction potentials obtained from extensive spectroscopic studies [23]. In all three systems, we found Feshbach resonances with widths suitable for magnetoassociation. However, the background intraspecies and interspecies scattering lengths around the resonances present problems. In particular, the intraspecies scattering length for ^{133}Cs is very large and positive except in relatively narrow windows around 21, 559, and 894 G [24], and for ^{39}KCs and ^{40}KCs there are no suitable interspecies Feshbach resonances that lie in these regions. In the present work, we show for the case of ^{39}KCs that a suitable rf field can be used to create a new Feshbach resonance in the magnetic field region near 21 G, where Cs can be cooled to condensation.

II. METHODS

For the present work, we have generalized the MOLSCAT [25], BOUND [26] and FIELD [27] programs to handle interactions of two alkali-metal atoms in the presence of simultaneous magnetic and rf fields. MOLSCAT performs scattering calculations to extract S matrices and scattering lengths, and locate and characterize Feshbach resonances. BOUND locates near-threshold bound states as a function of energy at constant applied magnetic and rf field. The extended version of FIELD is capable of locating bound states at fixed energy as a function of magnetic field, rf field strength, or rf frequency. Both scattering and bound-state calculations use propagation methods that do not rely on basis sets in the interatomic distance coordinate R. Apart from the inclusion of rf fields, which is added in the present work, the coupled-channel methodology is the same as described for Cs in Sec. IV of Ref. [24], so only a brief summary will be given here.

We use a basis set of photon-dressed products of atomic functions in a fully decoupled representation,

$$|s_a m_{sa}\rangle|i_a m_{ia}\rangle|s_b m_{sb}\rangle|i_b m_{ib}\rangle|L M_L\rangle|N M_N\rangle,$$

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where s_a and s_b are the electron spins of the two atoms, i_a and i_b are their nuclear spins, L is the angular momentum of their relative motion, and N is the photon number with respect to the average photon number N_0. The quantities m and M are the corresponding projections onto the magnetic field axis Z. The Hamiltonian and its matrix elements in this basis set have been given in the Appendix of Ref. [28], except for the rf terms, which are described below.

The calculation may be done for a variety of different polarizations of the rf radiation. For radiation polarized along Z (π polarization), $M_N = 0$ for all N and $M_\mathcal{F} = M_F + M_L$ is conserved, where $M_F = m_{sa} + m_{ia} + m_{sb} + m_{ib}$. For radiation polarized in the XY plane, the simplest calculation is for circularly polarized light, with either $M_N = N$ (right-circularly polarized, σ_+) or $M_N = -N$ (left-circularly polarized, σ_-). For radiation linearly polarized along X (σ_X), M_N runs from $-N$ to N in steps of 2 and a correspondingly larger basis set is required. In all these cases, $M_{\rm tot} = M_\mathcal{F} + M_N$ is conserved. In the present work we restrict the basis set to functions with $|N| \leqslant 2$ and the required $M_{\rm tot}$.

The rf terms in the Hamiltonian for each atom are given for σ_+ polarization by

$$H_{\rm rf} = \frac{\mu_{\rm B} B_{\rm rf}}{2\sqrt{N}}[(g_S \hat{s}_+ + g_i \hat{i}_+)\hat{a}_+ + (g_S \hat{s}_- + g_i \hat{i}_-)\hat{a}_+^\dagger] + h\nu(\hat{a}_+\hat{a}_+^\dagger - N_0), \quad (1)$$

where $B_{\rm rf}$ is the oscillating magnetic field, ν is the rf frequency, $\hat{s}_+$ and $\hat{s}_-$ are raising and lowering operators for the electron spin, $\hat{i}_+$ and $\hat{i}_-$ are the corresponding operators for the nuclear spin, and g_S and g_i are electron and nuclear spin g-factors with the sign convention of Arimondo *et al.* [29]. $\hat{a}_+$ and $\hat{a}_+^\dagger$ are photon annihilation and creation operators for σ_+ photons. For σ_- polarization, $\hat{a}_-$ replaces $\hat{a}_+^\dagger$ and $\hat{a}_-^\dagger$ replaces $\hat{a}_+$. For σ_X polarization, both σ_+ and σ_- coupling terms are present, with $B_{\rm rf}$ renormalized by $1/\sqrt{2}$.

III. RESULTS

Figure 1 shows the near-threshold $L = 0$ bound states of ^{39}KCs, in the absence of rf radiation, for both $M_F = +4$, corresponding to ^{39}K and ^{133}Cs atoms in their absolute ground states, and $M_F = +3$. All levels are shown relative to the lowest $M_F = +4$ threshold, and the two $M_F = +3$ thresholds are shown as dotted orange lines. At fields near 21 G, where the scattering length of Cs allows cooling to condensation, it may be seen that there are $M_F = +3$ bound states that lie about -57 and -69 MHz below the $M_F = +4$ threshold.

We choose an rf frequency of 69.2 MHz to bring one of the $M_F = +3$ states into resonance with the $M_F = +4$ threshold near 21 G and carry out scattering calculations in the field-dressed basis set for $M_{\rm tot} = +4$ to identify Feshbach resonances. Figure 2 shows the calculated inter-species scattering length for ^{39}K + ^{133}Cs collisions in the region around 21 G for a variety of strengths $B_{\rm rf}$ of the rf field, with σ_+ polarization and $L_{\rm max} = 0$. It may be seen that a new resonance is induced, with a width that varies approximately quadratically with the rf field. To a good approximation the width Δ is 1.6×10^{-5} $B_{\rm rf}^2$/G. The rf-induced resonance is also shifted significantly from its rf-free position, again nearly quadratically with field.

The rf fields considered in this paper are large, but comparable to those considered previously [17,18]. Rf fields up to 6 G have been applied in experiments to produce ^{87}Rb$_2$

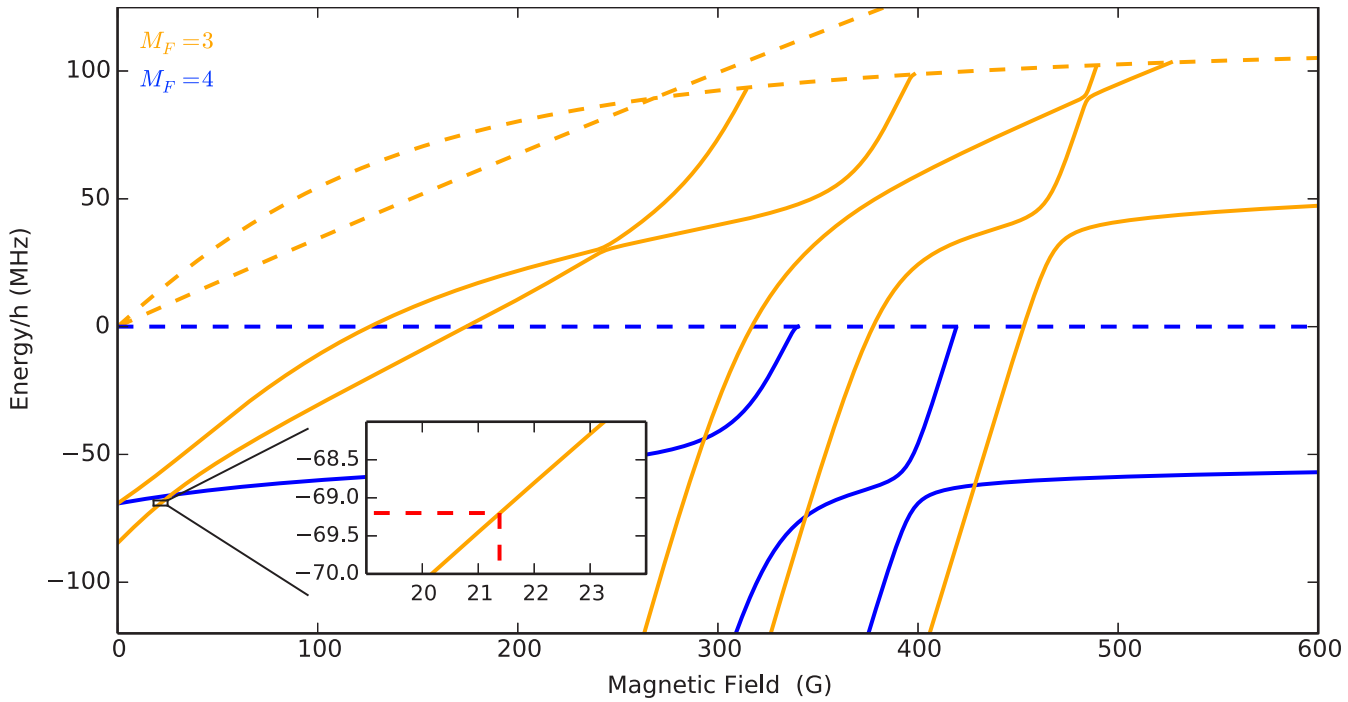

FIG. 1. Thresholds (dashed lines) and near-threshold bound states (solid lines) for ^{39}KCs in the absence of rf radiation for $M_F = +4$ (blue) and $M_F = +3$ (orange). The inset shows an expanded view of the region we consider in detail. All energies are relative to the lowest $M_F = +4$ threshold.

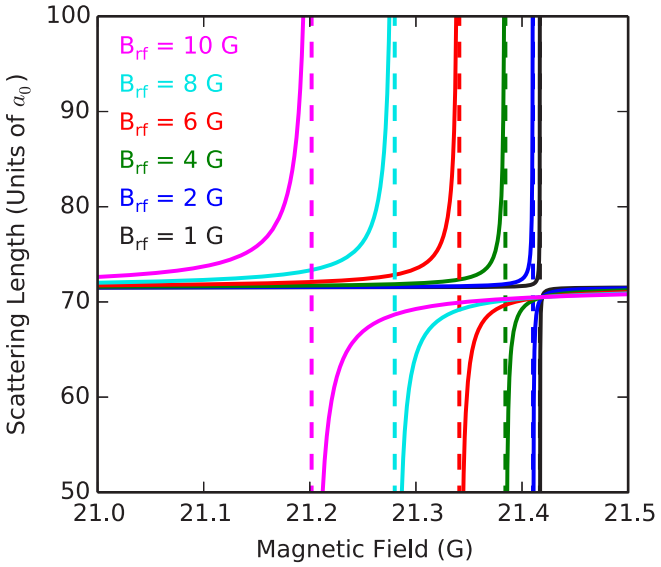

FIG. 2. Calculated scattering length for $^{39}\mathrm{K}+^{133}\mathrm{Cs}$, in the presence of a σ_+ rf field at a frequency of 69.2 MHz, with differing strengths B_{rf} (increasing from right to left).

on atom chips, and higher fields are achievable [30]. The fields currently achievable in conventional atom traps are rather lower, but fields of up to 0.7 G have been achieved [31].

The resonances shown in Fig. 2 are lossless, so they appear as true poles in the scattering length. This is because the incoming channel is the lowest that exists for $M_{\mathrm{tot}} = 4$ and the molecular state that is coupled to it by rf radiation is a true bound state, below the lowest threshold. However, there are two decay mechanisms that can actually exist. First, if the rf radiation has σ_X rather than σ_+ polarization, it can couple to an $M_{\mathrm{tot}} = 4$ channel with $M_F = 3$, $L = 0$, $N = -1$, and $M_N = +1$. Because $N = -1$, this lies below the incoming channel. The resonance is then characterized by a resonant scattering length a_{res} in addition to the width Δ: the real part of the scattering length exhibits an oscillation of amplitude $\pm a_{\mathrm{res}}/2$ instead of a pole, and the imaginary part exhibits a narrow peak of magnitude $\pm a_{\mathrm{res}}$ [32]. We have repeated the calculations of Fig. 2 for σ_X polarization, and find $a_{\mathrm{res}} = 1.5 \times 10^7 (\mathrm{G}/B_{\mathrm{rf}})^2$ bohr. These very large values of a_{res} correspond to very weakly decayed resonances, and should not cause problems in magnetoassociation. Secondly, even for σ_+ polarization, channels with $L > 0$ and $M_L \neq 0$ can cause collisionally assisted one-photon decay, mediated by the atomic spin dipolar (or second-order spin-orbit) interaction. In the present case, for example, there is a channel $M_F = 3$, $L = 2$, $M_L = +2$, $N = -1$, and $M_N = -1$, and thus $M_{\mathcal{F}} = 5$ and $M_{\mathrm{tot}} = 4$, that lies below the incoming channel. Such d-wave participation can in principle cause loss. However, this is a very weak process because of the weakness of the spin-dipolar coupling. We have repeated the calculations of Fig. 2 with all $L = 2$ channels for $M_{\mathrm{tot}} = 4$ included; in this case the resonance is close to pole-like with $a_{\mathrm{res}} = 1.2 \times 10^8$ bohr for $B_{\mathrm{rf}} = 10$ G. Once again, therefore, this loss process should not cause problems in magnetoassociation.

The resonant scattering length a_{res} is given by [32]

$$a_{\mathrm{res}} = -2a_{\mathrm{bg}}\Delta/\Gamma^B_{\mathrm{inel}}, \tag{2}$$

where Γ^B_{inel} is a Breit-Wigner width that describes decay of the field-dressed bound state to atoms. This may be converted into a lifetime for the field-dressed molecules,

$$\tau = \left|\frac{\hbar}{\Gamma^B_{\mathrm{inel}}\Delta\mu}\right| = \left|\frac{-\hbar a_{\mathrm{res}}}{2\Delta\mu a_{\mathrm{bg}}\Delta}\right|, \tag{3}$$

where $\Delta\mu$ is the difference in magnetic moments between the molecular state and the incoming channel, $\Delta\mu = \mu_{\mathrm{molecule}} - \mu_{\mathrm{atoms}}$. The value $a_{\mathrm{res}} = 1.5 \times 10^5$ bohr obtained for σ_X polarization with $B_{\mathrm{rf}} = 10$ G corresponds to a molecular lifetime of 166 ms for photon-assisted decay to the lower field-dressed threshold; the lifetime is approximately proportional to B_{rf}^{-4}, as expected for a 2-photon decay pathway, so increases fast as the rf field is decreased. This decay of course persists only as long as the rf field is switched on.

Different type of decaying rf-induced resonance may be observed if the rf radiation couples the incoming state to a molecular state that is itself above a threshold to which it can decay. At least two such cases may be identified. Tscherbul *et al.* [17] and Hanna *et al.* [18] both considered rf-induced resonances due to bound states of $^{87}\mathrm{Rb}_2$ near the a+e $|1,1\rangle + |2,-1\rangle$ excited hyperfine threshold of ^{87}Rb; these bound states can decay to lower open channels with the same M_F through rf-independent mechanisms, so the resonances are strongly decayed and the molecules have a finite lifetime even after the rf field is switched off. Hanna *et al.* [18] also considered resonances due to bound states of $^6\mathrm{Li}_2$ that lie above the lowest open channel, but have different M_F; these can decay to $L = 2$ open channels by rf-free spin-dipolar coupling, or through 2-photon rf coupling for σ_X polarization.

The coupled-channel approach that we use includes the effect of the rf field nonperturbatively. However, for the rf fields considered here, the resonance widths are clearly dominated by direct couplings from the incoming channel to the resonant bound state. Under these circumstances, the width of the resonance is proportional to the square of a bound-continuum matrix element I of the rf perturbation $\hat{H}_{\mathrm{rf}}$,

$$\Delta = \frac{\pi I^2}{k\Delta\mu a_{\mathrm{bg}}}, \tag{4}$$

where

$$I = \langle\psi_{\mathrm{bound}}|\hat{H}_{\mathrm{rf}}|\psi_{\mathrm{incoming}}\rangle. \tag{5}$$

The incoming wave function is essentially a product of field-dressed atomic functions $|\alpha_{\mathrm{K}} m_{f,\mathrm{K}}\rangle$ and $|\alpha_{\mathrm{Cs}} m_{f,\mathrm{Cs}}\rangle$ and a radial function $\chi_k(r)$. At the low magnetic fields considered here, the atomic functions are approximately $|f, m_f\rangle = |1,1\rangle$ for ^{39}K and $|3,3\rangle$ for ^{133}Cs, where f is the resultant of s and i for each atom. The molecular wave functions are more complicated, and a general treatment is beyond the scope of this paper. However, for the specific case of $^{39}\mathrm{K}\,^{133}\mathrm{Cs}$, Fig. 1 shows that the near-threshold bound states are mostly nearly parallel to the thresholds, indicating that they have similar spin character to the thresholds where this is true. If the scattering lengths for the $M_F = +3$ and $+4$ thresholds were identical, the incoming and bound-state radial functions would be orthogonal to one

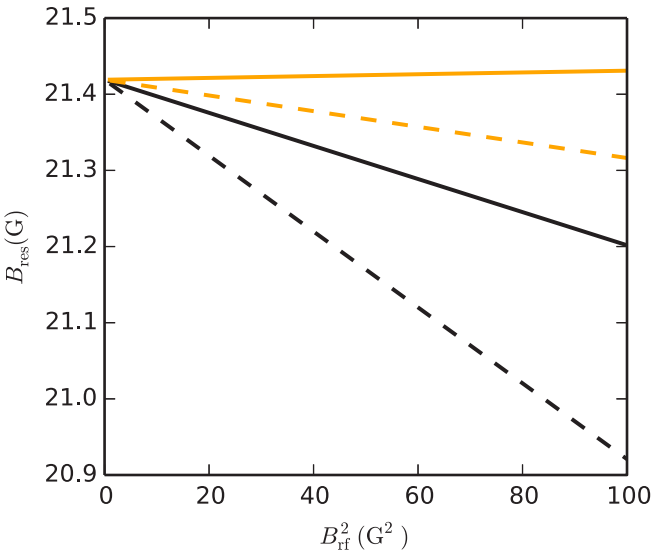

FIG. 3. Calculated resonance positions as a function of $B_{\rm rf}^2$ for σ_+ (black lines) and σ_X polarization (orange lines), for basis sets with $|N| \leqslant 1$ (dashed lines) and $|N| \leqslant 2$ (solid lines).

another, which would produce a very small matrix element I because the spin part of the rf coupling is almost independent of r. In general terms, therefore, the rf coupling is strongest for systems where the scattering lengths for the incoming and bound-state channels differ most, and thus where the singlet and triplet scattering lengths are very different. It is reasonably straightforward to construct a complete map of the near-threshold bound states for any specific system using BOUND and FIELD, but some experimentation is needed to establish which bound states produce rf-induced resonances with useful widths.

Although the resonance widths are dominated by direct couplings between the incoming channel and the resonant bound state, the shifts are not. Figure 3 shows the resonance positions as a function of $B_{\rm rf}^2$ for both σ_+ and σ_X polarization, for basis sets with both $|N| \leqslant 2$ (essentially converged) and $|N| \leqslant 1$ (unconverged). The smaller basis sets give widths that are unchanged to 1 part in 10^3 compared to the larger ones, but the resonance positions shift substantially; they are still close to quadratic in $B_{\rm rf}$, but with different coefficients. This arises because the $M_F = +3$, $N = 1$ bound state that causes the resonances is shifted by ac-Zeeman couplings to both $N = 0$ and $N = 2$ states, but the latter couplings are omitted for the smaller basis sets. The shifts are also significantly different for the two polarizations. Our coupled-channel approach provides a straightforward way to capture such effects properly.

Resonances of the type described here will exist for all the alkali-metal dimers. For all such dimers except those containing ^{40}K, the lowest threshold in a magnetic field has $M_{F,\rm ground} = i_a + i_b - 1$. For those containing ^{40}K, which has inverted hyperfine structure, $M_{F,\rm ground} = i_a + i_b$. In both cases, there are Zeeman-excited thresholds with $M_F < M_{F,\rm ground}$. However, the lowest thresholds with $M_F > M_{F,\rm ground}$ always correlate with excited hyperfine states and are substantially higher in energy. As for ^{39}K ^{133}Cs, resonances due to bound states with $M_F = M_{F,\rm ground} - 1$ are likely to be pole-like, with only weak decay as described above.

IV. CONCLUSIONS

We have shown that radio-frequency fields can be used to engineer magnetically tunable Feshbach resonances in regions of magnetic field where they did not previously exist. This capability may allow the creation of resonances at magnetic fields where the intraspecies and interspecies scattering lengths have values that are favorable for evaporative or sympathetic cooling, and where stable mixed condensates may be created. This in turn may allow magnetoassociation to form molecules from otherwise intractable pairs of ultracold atoms. The resonances we consider are different from those of Refs. [17,18] both because the molecules that can be created at them are heteronuclear and because they are truly bound, so cannot decay to lower atomic thresholds after the rf radiation is switched off.

The present work has used an rf field to bring bring bound states into resonance with threshold and create new Feshbach resonances. This is conceptually the simplest approach, but a similar effect could be achieved with the difference between two laser frequencies, with different (and potentially more versatile) selection rules governing which bound states can cause resonances.

ACKNOWLEDGMENTS

The authors are grateful to Caroline Blackley for initial work on adapting the computer programs to handle rf fields. This work has been supported by the UK Engineering and Physical Sciences Research Council (Grants No. ER/I012044/1 and No. EP/N007085/1 and a Doctoral Training Partnership with Durham University).

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