

# Efficient conversion of ultracold Feshbach-resonance-related polar molecules into ultracold ground state ( $X^1\Sigma^+ v = 0, J = 0$ ) molecules

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**Abstract.** It is proposed that bound states of ultracold heteronuclear alkali dimer molecules derived from Feshbach resonances can be efficiently converted into true ground state ultracold molecules ( $X^1\Sigma^+ v = 0, J = 0$ ) by a stimulated Raman process via  $b^3\Pi \sim A^1\Sigma^+$  mixed levels. This is illustrated and discussed for five of the ten polar heteronuclear alkali dimers: LiNa, NaK, NaRb, RbCs, and especially KRb.

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## 1 Homonuclear alkali dimers

One of the current major challenges of molecular physics is the Bose-Einstein condensation (BEC) of molecules in the lowest electronic, vibrational, and rotational level [1]. For the homonuclear alkali metal dimers (and molecular hydrogen), this means BEC in the  $X^1\Sigma_g^+$  electronic state with vibrational quantum number  $v = 0$  and rotational quantum number  $J = 0$ . Note that this lowest level may be highly degenerate (up to  $g_I = 45$  for  $^{40}\text{K}_2$ ), so the total density must be much higher (by a factor of  $g_I$ ) for BEC to occur than for the non-degenerate ( $g_I = 1$ ) case (e.g. for  $\text{H}_2$ ), assuming the degeneracy breaking terms in the molecular Hamiltonian remain small compared to  $kT$  [1]. It has also been suggested that the  $X^1\Sigma_g^+ v = 0, J = 1$  levels (with degeneracy  $(2J + 1)g_I$ , where  $g_I$  varies from 3 to 36) may also undergo BEC [1].

However, the preparation of such molecular BECs has been hindered by the lack of information on the ultracold ( $T < 1$  mK) collisional properties of such molecules. The only molecular systems for which good estimates of the molecule-molecule scattering lengths  $a_{mm}$  are available are para- $\text{H}_2$  + para- $\text{H}_2$ , ortho- $\text{D}_2$  + ortho- $\text{D}_2$ , and para- $\text{T}_2$  + para- $\text{T}_2$  [1,2]. This analysis is based on van der Waals molecule spectra [3–5], low energy  $\text{H}_2 - \text{H}_2$  scattering [6] and accurate ab initio calculations [5,7,8]. The results are  $a_{mm}(\text{p-H}_2) = +11.3a_0$ ,  $a_{mm}(\text{o-D}_2) = -17a_0$ , and  $a_{mm}(\text{p-T}_2) = +17a_0$ . Thus the p- $\text{H}_2$  and p- $\text{T}_2$  BECs have positive scattering lengths and will be stable, while the o- $\text{D}_2$  BEC has a negative scattering length and will be

metastable only for small numbers of molecules in a trap (as for  $^7\text{Li}$ ; see [9–11]). These molecule-molecule scattering lengths are completely unknown for all alkali dimers, however.

During the past year, there has been a great deal of experimental and theoretical activity on highly excited bound states (slightly negative energies with respect to the separated atom asymptote), associated with Feshbach resonances (at positive energies with respect to the separated atom asymptote). The concept of tuning a Feshbach (molecular) resonance into resonance with an atomic asymptote is well-known [12–19]. In the alkali metal systems, the atomic asymptote is often the “stretched” state, with each atom in the  $|F_{max}, m_F| = F_{max}$  state, so the collisions are purely triplet; however, the Feshbach resonance (and corresponding molecular bound state) is of mixed singlet-triplet character (never a pure singlet since the atomic nuclear spin is never zero). The more commonly used broader resonances have only a small amount of singlet character, however.

To quickly review the recent progress, many labs demonstrated rapid homonuclear molecular formation via magnetically tuned Feshbach resonances in  $\text{Cs}_2$  [20,21],  $\text{Rb}_2$  [22,23],  $\text{Na}_2$  [24],  $\text{Li}_2$  [25–29] and  $\text{K}_2$  [30]. The  $^6\text{Li}_2$  and  $^{40}\text{K}_2$  results were remarkable in their long lifetimes and ultimately led to molecular BECs of  $^6\text{Li}_2$  [25,29] and  $^{40}\text{K}_2$  [30]. The long lifetimes (with corresponding molecular BECs) were attributed to Pauli blocking, i.e. the difficulty of reinserting two Fermi fragment atoms in the Fermi distribution [31]. A second factor, true also for boson fragment atoms, is the tendency of molecules near dissociation to thermally dissociate rather than relax to

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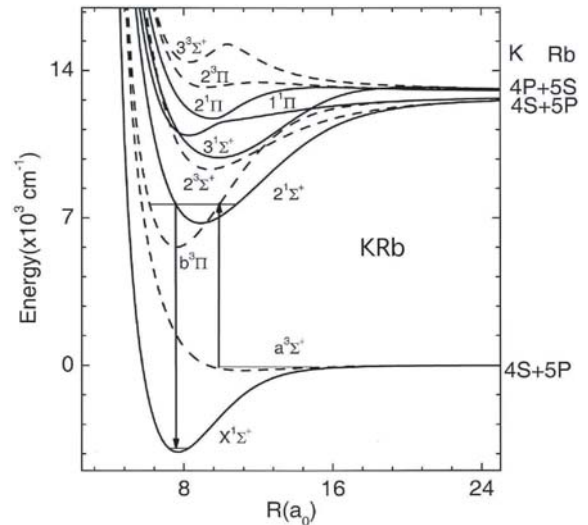
lower levels, where the thermal dissociation can be turned off by ultracold temperatures [2,32]. More recently, attention has also focused on “the BEC-BCS Crossover”, where “Cooper pairs” of resonant unbound Fermi atoms undergo BEC [33–35] and give rise to superfluidity [36]. Very recently, an optically tuned Feshbach resonance in  $^{87}\text{Rb}_2$  has also been observed [37], as proposed in [38] and further discussed in [39,40].

Thus there is great excitement and rapid progress in producing Feshbach-resonance-related ultracold alkali dimer molecules, particularly  $^6\text{Li}_2$  and  $^{40}\text{K}_2$ . However, the conversion of these molecules to  $X\ ^1\Sigma_g^+$   $v=0$ ,  $J=0$  molecules and a “stable” molecule BEC is not straightforward. Ignoring hyperfine, a stimulated Raman process would not work efficiently to convert a predominantly triplet ungerade Feshbach-resonance-related state into a singlet gerade ground state. The hyperfine terms in the Hamiltonian do mix u and g symmetries and would allow some conversion, but the Franck-Condon factors for  $X\ ^1\Sigma_g^+$  ( $v_{max}$ )  $\rightarrow$   $A\ ^1\Sigma_u^+$  (various  $v$ )  $\rightarrow$   $X\ ^1\Sigma_g^+$  ( $v=0$ ) are quite small. Nevertheless, weak transitions to primarily triplet levels very near dissociation have been observed [41]; thus a stimulated Raman (e.g. STIRAP [42]) conversion of such levels to  $X\ ^1\Sigma_g^+$  ( $v=0$ ,  $J=0$ ) remains a challenging possibility. Moreover, there is a large potential barrier if the  $B\ ^1\Pi_u$  state replaces the  $A\ ^1\Sigma_u^+$  state as the intermediate state. There are similar problems for other higher energy  $^1\Sigma_u^+$  and  $^1\Pi_u$  states as well.

## 2 Heteronuclear alkali dimers

The progress on photoassociation [43], formation of ultracold molecules [1,44], determination of Feshbach resonances, etc. for heteronuclear alkali dimer molecules is well behind that for homonuclear alkali dimer molecules. Ultracold polar alkali dimer molecules have been observed for  $\text{NaCs}^+$  [45],  $\text{RbCs}$  [46,47], and  $\text{KRb}$  [48–50]. Photoassociative spectra have only been recently observed for  $\text{RbCs}$  [51] and  $\text{KRb}$  [49,50]. However, accurate ab initio calculations and limited molecular spectra are available for all polar alkali dimers (excluding those containing Fr). The non-polar  $^6\text{Li}^7\text{Li}$  molecule has also been observed by photoassociation [52]. Very recently Feshbach resonances have been experimentally observed for  $\text{LiNa}$  [53] and  $\text{KRb}$  [54], opening the way for copious production of near-dissociation levels of heteronuclear polar molecules and formation of quantum degenerate gases of such molecules. What we wish to emphasize here is that we believe such molecules can be efficiently converted to  $v=0$ ,  $J=0$ , ground  $X\ ^1\Sigma^+$  state molecules by a stimulated Raman process (e.g. STIRAP [42]) via  $b\ ^3\Pi \sim A\ ^1\Sigma^+$  mixed levels.

Five examples of heteronuclear polar alkali dimers are shown in Figures 1–5. In each case, the potential curves plotted are based on high quality ab initio calculations, but experimentally-based RKR potentials are virtually identical, e.g. for  $\text{NaK}$  for which the greatest amount of spectroscopic data is available. In each case, the proposed initial state is a very slightly bound state correlating to a

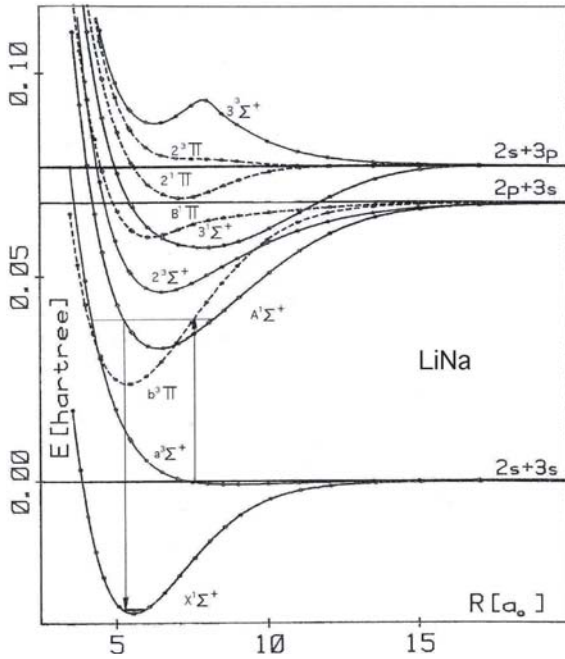


**Fig. 1.** Ab initio potential energy curves of  $\text{KRb}$  [55]. The horizontal lines represent the three levels involved in the proposed stimulated Raman transition: the initial  $a\ ^3\Sigma^+$  level near dissociation created via a Feshbach resonance; the intermediate level of mixed singlet-triplet character ( $b(1)\ ^3\Pi \sim A(2)\ ^1\Sigma^+$ ) with four important turning points (see text); and the final true ground state level,  $X\ ^1\Sigma^+$   $v=0$ ,  $J=0$ . The PUMP “triplet” transition excites the initial level up to the intermediate level and the DUMP “singlet” transition de-excites the intermediate level to the final level.

quasibound Feshbach-resonance-related state of predominantly triplet character (especially near its inner turning  $R_{FR-}$ ). A vertical excitation from  $R_{FR-}$  in accord with the Franck-Condon Principle will efficiently reach an outer turning point of the  $b(1)\ ^3\Pi$  state,  $R_{v_b+}$ . However, especially for the heteronuclear alkali dimers including a Rb or Cs atom, levels of the  $b\ ^3\Pi$  state will be strongly mixed with levels of the  $A(2)\ ^1\Sigma^+$  state. Such mixed upper levels will be sparse in  $\text{LiNa}$  (as in  $\text{Li}_2$ ), dense in  $\text{NaRb}$ ,  $\text{KRb}$  and  $\text{RbCs}$ , and intermediate in  $\text{NaK}$ . Such levels will have *four* turning points, two associated with the  $b\ ^3\Pi$  state ( $R_{v_b\pm}$ ) and two associated with the  $A\ ^1\Sigma^+$  state ( $R_{v_A\pm}$ ). By a somewhat fortunate coincidence, the appropriate  $R_{v_A-}$  turning points are directly above the ground  $X\ ^1\Sigma^+$   $R_e$  region, i.e. between the turning points  $R_{0_X\pm}$ . Such turning point matches imply large transition moment/Franck-Condon factors for both the PUMP step ( $a\ ^3\Sigma^+ \rightarrow b\ ^3\Pi \sim A\ ^1\Sigma^+$ ) and the DUMP step ( $b\ ^3\Pi \sim A\ ^1\Sigma^+ \rightarrow X\ ^1\Sigma^+$ ) of a stimulated Raman transition.

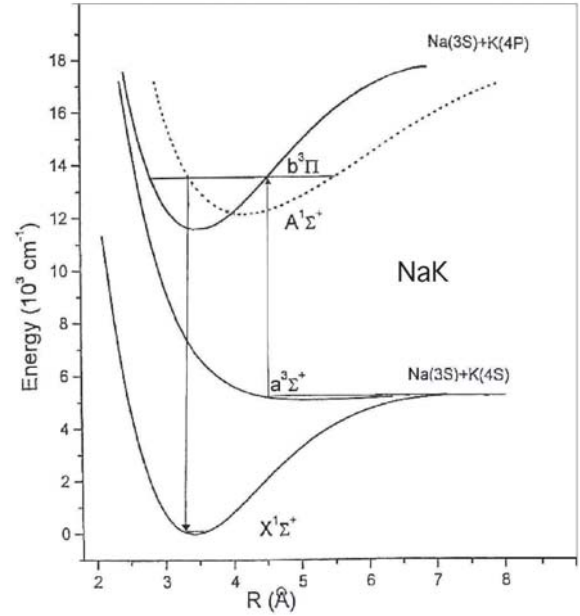
## 3 Specific heteronuclear alkali dimers

Let us begin a more detailed discussion with  $\text{KRb}$ , as shown in Figure 1, a case for which both photoassociative spectra [49,50] and Feshbach resonance data [54] are available. There are three sets of high quality ab initio calculations available [55–57] as well as an accurate description of the long-range portion of the potential in terms



**Fig. 2.** Ab initio potential curves of LiNa [60]. The horizontal and vertical lines are defined as in Figure 1.

of exchange and dispersion interactions calculated from atomic properties only [58]. To indicate the mutual consistency of these calculations at  $16a_0$ , we find the following  $a^3\Sigma^+$  potential binding energies (in  $\text{cm}^{-1}$ ) with respect to separated atoms:  $-77.0$  [55],  $-76.8781$  [56],  $-75.2710$  [57], and  $-75.5010$  [58]. Likewise, at shorter range we have determined by spline interpolation the distances (in Bohr radii,  $a_0$ ) at which the various  $a^3\Sigma^+$  potentials go through a minimum ( $R_e$ ):  $11.15$  [55],  $11.22$  [56],  $11.20$  [57]; and go through zero energy:  $9.28$  [55],  $9.33$  [56], and  $9.16$  [57]. This latter distance is nearly identical to the inner turning point near dissociation,  $R_{FR-}$ , discussed above. Thus the PUMP “triplet” transition will correspond to approximately  $8400 \text{ cm}^{-1}$ . The upper  $b(1)^3\Pi \sim A(2)^1\Sigma^+$  mixed level will be in a dense region of mixed levels and will have four principal components of spontaneous emission corresponding to the four turning points: (a) near  $R_{v_b-}$ , weak (small  $\nu^3$ ) triplet  $b^3\Pi \rightarrow a^3\Sigma^+$  bound-free emission creating very hot atoms; (b) near  $R_{v_A-}$ , strong (large  $\nu^3$ ) singlet  $A^1\Sigma^+ \rightarrow X^1\Sigma^+$  bound-bound emission creating low  $v$  X state levels; (c) near  $R_{v_b+}$ , moderate  $b^3\Pi \rightarrow a^3\Sigma^+$  bound-free emission creating cold atoms as well as bound-bound emission creating high  $v$  a state levels; and (d) near  $R_{v_A+}$ , moderate  $A(2)^1\Sigma^+ \rightarrow X^1\Sigma^+$  bound-bound emission at intermediate  $v$  X state levels. In the proposed stimulated Raman process, the DUMP transition is (b), in particular to  $X^1\Sigma^+ v=0, J=0$  assuming the mixed  $b \sim A$  level is  $J=1$ . In the STIRAP scheme [42], with the counter-intuitive DUMP, then PUMP sequence, one can approach 100% conversion of the near dissociation  $a^3\Sigma^+$  level associated with the Feshbach resonance to the true ground state ( $X^1\Sigma^+ v=0, J=0$ ) with no population of the  $b \sim A$  intermediate state. It is

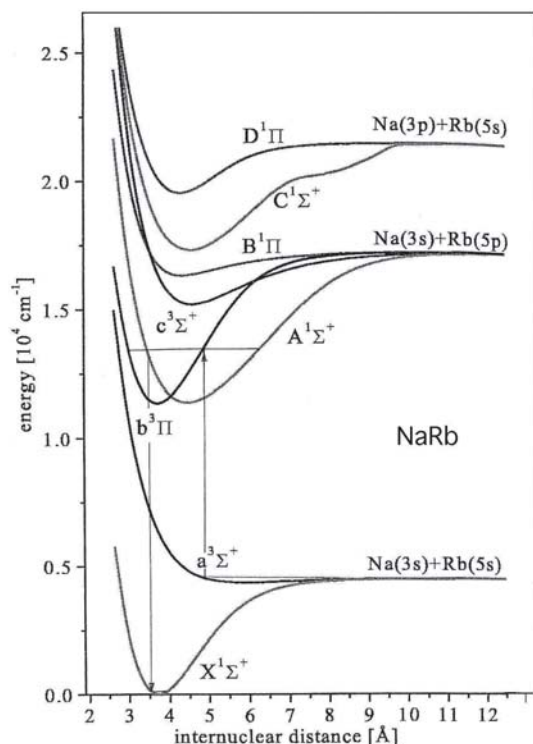


**Fig. 3.** Ab initio potential curves of NaK [59]. The horizontal and vertical lines are defined as in Figure 1.

also possible that an oscillation between the two quantum degenerate gases of selected molecular states (e.g.  $a^3\Sigma^+ v_{max}, N=0$  and  $X^1\Sigma^+ v=0, J=0$ ) could be established, analogous to atom/molecule oscillations (see e.g. [17] and references therein). We do not mean to imply that other schemes for transferring population from a Feshbach-resonance-related  $a^3\Sigma^+$  high  $v$  level to  $X^1\Sigma^+ v=0, J=0$  will not also work efficiently. For example, the  $1^1\Pi \sim 2^3\Sigma^+ \sim b^3\Pi$  mixing in NaK has been studied [59], and, in KRb, the  $2^1\Pi$  and  $3^1\Sigma^+$  states will probably mix in as well! Thus there is much opportunity, but also much spectroscopic complexity, in higher mixed levels. There is an urgent need for conventional short-range spectroscopy, e.g. laser-induced fluorescence at the DUMP “singlet” transition frequency, ideally with a Fourier transform spectrometer to directly observe the four turning point components of the  $b^3\Pi \sim A^1\Sigma^+$  state predicted and described above.

Let us now consider LiNa, the other case for which Feshbach resonance data [53] are available. High quality ab initio calculations [60] are used for Figure 2. The sparsity of mixed  $b \sim A$  levels in  $\text{Li}_2$  [61] suggests such levels will be sparse in LiNa as well, so there is definitely a need for high resolution short-range spectroscopy to identify such levels. One might, for example, tune near the DUMP “singlet” transition frequency in Figure 2 while monitoring the IR emission at the PUMP “triplet” transition frequency in order to identify such mixed levels. Nevertheless, efficient conversion to the  $X^1\Sigma^+ v=0, J=0$  level is predicted once appropriate mixed levels are identified.

For NaK, shown in Figure 3, there are two sets of high quality ab initio calculations of potential energy curves [62,63]. Again we predict efficient conversion of the initial to the final state.

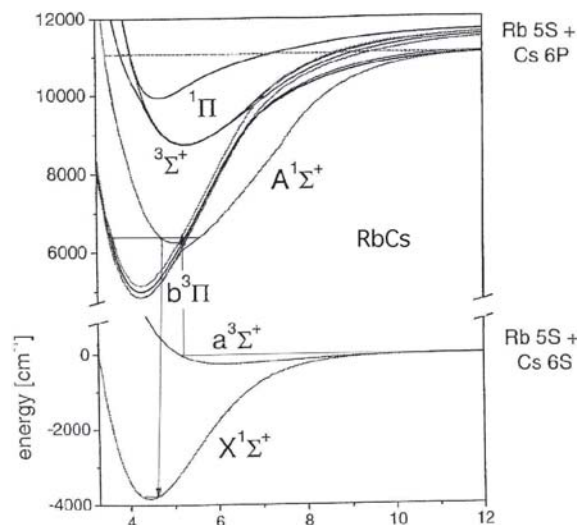


**Fig. 4.** Ab initio potential curves of NaRb [63]. The horizontal and vertical lines are defined as in Figure 1.

For NaRb, shown in Figure 4, there are again two sets of quality calculations [63,64]. We chose [63], where the minima of the  $b^3\Pi$  and  $A^1\Sigma^+$  states are at nearly the same energy, rather than [64], where the  $b^3\Pi$  state minimum is lower (as in Figs. 1, 2, 3 and 5). Experiments to study the  $b^3\Pi \sim A^1\Sigma^+$  mixing are underway in Latvia [65]. Since both  $^{23}\text{Na}$  and  $^{87}\text{Rb}$  have been used to produce high density Bose-Einstein condensates, NaRb Feshbach resonance studies should become available soon.

For RbCs, as shown in Figure 5, high quality ab initio calculations [66] are available. Additional relativistic calculations are also available [67]. The calculations appear to agree well with preliminary photoassociation results [47,51]. Note that the mixing of the  $b^3\Pi$  and  $A^1\Sigma^+$  states is so strong that adiabatic rather than nonadiabatic states are displayed. Nevertheless, efficient conversion again seems likely.

In summary, for the five heteronuclear polar alkali dimers examined (KRb, LiNa, NaK, NaRb, and RbCs), it seems likely that highly efficient stimulated Raman conversion of Feshbach-resonance-related  $a^3\Sigma^+$  states into  $X^1\Sigma^+ v=0, J=0$  ground state ultracold molecules can be achieved, leading, for example, to quantum degeneracy of the corresponding gases. The other five species (LiK, LiRb, LiCs, NaCs and KCs) will likely be equally advantageous for this conversion, although a comparably detailed analysis has not been carried out.



**Fig. 5.** Ab initio potential curves of RbCs [67]. The horizontal and vertical lines are defined as in Figure 1.

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