A New Scheme for Formation of Ultracold Molecules via Photoassociation and Resonant Coupling

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The formation of cold molecules by photoassociation of cold atoms has recently been observed for Cs_2 [1], K_2 [2], and Rb_2 [3]. The first step is a photoassociation reaction such as

$$Cs(6s) + Cs(6s) + h(\nu_0 + \Delta_L) \rightarrow Cs_2(6s + 6p\ ^2P_{1/2,3/2}, \Omega_{u,g}, v, J) \quad (1)$$

where ν_0 is the frequency of the resonance line and Δ_L the detuning of the laser. This can be understood as a vertical transition at a large distance $R_L = (C_3/\Delta_L)^{1/3}$ from the continuum of the ground state to an excited state with an asymptotic $-C_3/R^3$ potential [see Fig. 1(a)]. The second step, efficient population, by spontaneous emission, of bound levels in the ground state

$$Cs_2(6s + 6p\ ^2P_{1/2,3/2}, \Omega_{u,g}, v, J) \rightarrow Cs_2(6s + 6s, ^{1,3}\Sigma_{g,u}, v', J') + h(\nu_0 + \Delta) \quad (2)$$

requires a favourable Franck-Condon overlap between the two vibrational wave functions [see Fig. 1(b)]. Due to the $-C_6/R^6$ asymptotic behaviour of the ground state potential, reaction (2) necessitates a vibrational wave function in the excited state with a non negligible probability of presence at intermediate distance. An efficient mechanism [1] is offered by the double well structure of the Cs_2 $0_g^-(6s + 6p\ ^2P_{3/2})$ excited potential which provides a favorable probability of presence both at large and intermediate internuclear distances. We now show a new scheme taking advantage of the coupling between two excited channels. For small detunings Δ_L below the $6p\ ^2P_{1/2}$ dissociation limit, photoassociation can proceed from the continuum of the $^1\Sigma_g^+$ ground state to a vibrational level of the $P_{1/2}$ state of $0_u^+(6s + 6p)$. Resonant coupling can then transfer population to a bound level of the $P_{3/2}$ state, allowing a sufficient probability density at intermediate internuclear distances, as shown in Fig. 1. Using a mapped Fourier Grid Representation method [4], we obtain the vibrational wave functions of Cs_2, from which we calculate the probability of forming ground state molecules. We are able to reproduce the experimental molecular ion spectrum, confirming that resonances are indeed responsible for the observed ultracold molecules [see Fig. 2].

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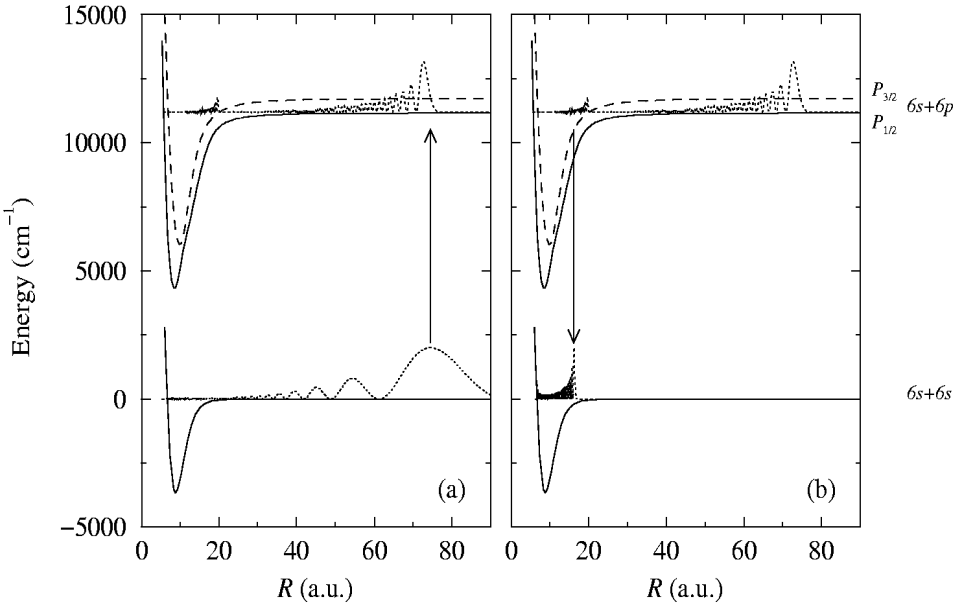

Figure 1: (a) Photoassociation at large internuclear distance; (b) Spontaneous emission at small internuclear distance and formation of an ultracold molecule.

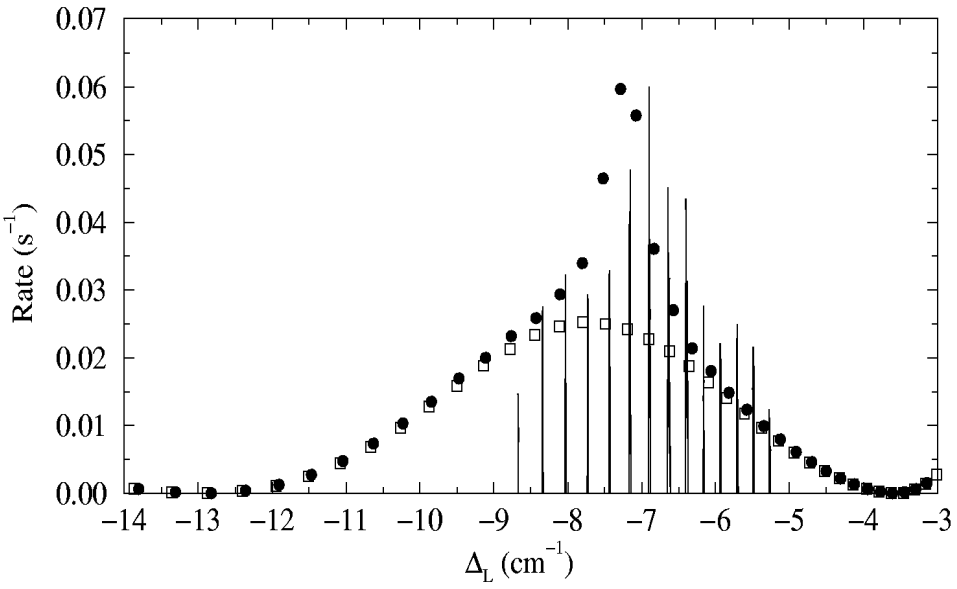

Figure 2: Ultracold Cs_2 formation rate (expressed in molecules per second per atom) through the $0_u^+(6s+6p)$ electronic states, with detuning Δ_L given with respect to the $P_{1/2}$ dissociation limit. Solid line: Experimental molecular ion signal (arbitrary units); Circles: Calculated values, starting from the $^1\Sigma_g^+$ continuum at $T = 140$ μK; Squares: same without the coupling between the 0_u^+ channels.