

Theoretical treatment of channel mixing in excited Rb₂ and Cs₂ ultracold molecules

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Treatment of the dynamics of ultracold molecules requires new theoretical tools due to the large extension of the vibrational motion in region of weak interaction. The mapped Fourier grid method, using coordinate adapted to the local de Broglie wavelength associated to the relative motion of the two atoms [1], is used to compute the perturbed binding energies and rotational constants of the Cs₂ 0_u⁺ (6s + 6p ²P_{1/2,3/2}) and Rb₂ 0_u⁺ (5s + 5p ²P_{1/2,3/2}) photoassociation spectra below the P_{1/2} dissociation limit. Perturbations due to a strong mixing between the two channels are predicted in a large spectral range (see Fig. 1). The results are very sensitive to the molecular data (potential curves and spin-orbit coupling). They can be interpreted in the framework of generalized Lu-Fano plots (see Fig. 2). This suggests that channel mixing parameters could easily be fitted to experimental spectra, eliminating the problems due to limited accuracy of the short range molecular potentials [2].

The method is generalized to treatment of bound-continuum interaction in a two-channel problem [3]. Two numerical methods (time-dependent and time-independent) are presented: they give equivalent results for the predissociation lifetimes of the vibrational levels in the Rb₂ and Cs₂ spectra. The lifetimes increase with the vibrational quantum number, proportionally to the classical vibration period estimated from Le Roy-Bernstein formula for an asymptotic R⁻³ potential : the predissociation problem is similar to the autoionization of a Rydberg level due to coupling in the core region. A strong isotopic effect is found for the rubidium dimer: the measured width of 8.5 GHz [5] for one predissociated level of ⁸⁷Rb₂ can be reproduced. We show that lifetimes determined by extrapolation of parameters fitted to Lu-Fano plots of bound levels below the P_{1/2} limit are in excellent agreement with the numerical results.

Extrapolation above the P_{3/2} limit would provide accurate determination of fine structure transition cross sections. Channel mixing is creating a favorable situation for the formation of ultracold molecules in the ground electronic state, as described in another abstract [6].

- [1] V. Kokoouline, O. Dulieu, R. Kosloff, and F. Masnou-Seeuws, *J. Chem. Phys.* **110**, 9865 (1999).
- [2] V. Kokoouline, O. Dulieu, and F. Masnou-Seeuws, *Phys. Rev. A* (2000), in press.
- [3] V. Kokoouline, O. Dulieu, R. Kosloff, and F. Masnou-Seeuws, *Phys. Rev. A* (2000), submitted.
- [4] K. T. Lu and U. Fano, *Phys. Rev. A* **2**, 81 (1994).
- [5] R. A. Cline, J. D. Miller, and D. J. Heinzen, *Phys. Rev. Lett.* **73**, 632 (1994).
- [6] C. M. Dion, C. Drag, O. Dulieu, V. Kokoouline, B. Laburthe-Tolra, F. Masnou-Seeuws, and P. Pillet, present conference.

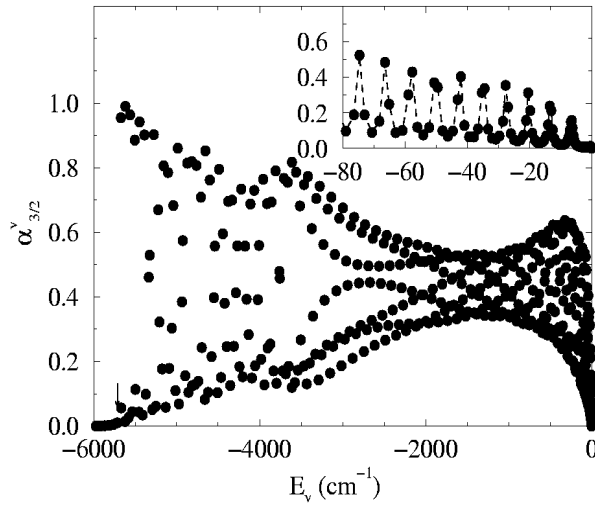


Figure 1: Variation (between 0 and 1) of the relative population of the $P_{3/2}$ component in the two-channel 0_u^+ (${}^2P_{3/2}$) and 0_u^+ (${}^2P_{1/2}$) vibrational function of ${}^{87}\text{Rb}_2$, as a function of the binding energy E_v relative to the $P_{1/2}$ dissociation limit. The inset shows the energy range close to this limit.

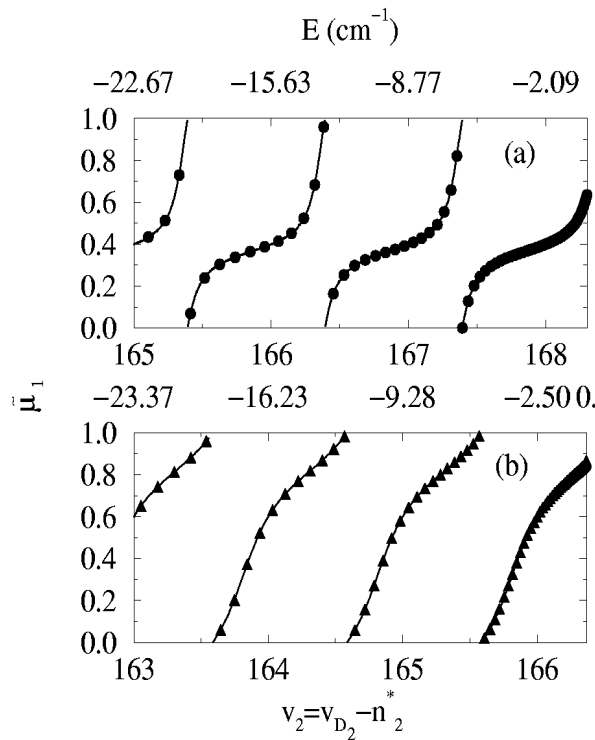


Figure 2: Lu-Fano plot for the quantum numbers of the vibrational levels obtained by two 0_u^+ coupled channel calculations for : black triangles, ${}^{85}\text{Rb}_2$; black circles, ${}^{87}\text{Rb}_2$. lower horizontal scale ; vibrational quantum number for the unperturbed $P_{3/2}$ series. Vertical scale : quantum defect obtained by comparing the perturbed series to the unperturbed $P_{1/2}$ series.