Lu-Fano plot for the interpretation of photoassociation spectra

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Photoassociation spectrum:

Le Roy-Bernstein analysis

The experimental photoassociation spectrum of Cs₂ molecules obtained in Laboratoire Aimé Cotton (Drag et al., IEEE J. Quantum Electron. 36, 1378 (2000)):

- Two energy regions under the 6S+6P₁/₂ and under the 6S+6P₃/₂ asymptotes (see figures below)
- Several rotational progressions observed
- We are interested in 0⁺(6S+6P₃/₂) progressions
- These progressions are predicted to be strongly perturbed

Interpretation using Lu-Fano plot

The procedure under D₃c:

1. For each experimental energy E, two effective quantum numbers ν₁₂ and ν₂₃ are calculated:
   \[ ν_1 = \frac{(D - E)^{1/2}}{H} \]
2. H, is determined by the long-range coefficient C₃
3. ν₁₂ and ν₂₃ are plotted as abscissa and ordinate on a graph

- Reason for failure of the Le Roy-Bernstein analysis is the presence of one level of the 0⁺(6S+6P₃/₂) progression perturbing the 0⁺(6S+6P₁/₂) progression under the D₃c dissociation limit.

- Lu and Fano (PRA 2, 81 (1970)) proposed a graphical method to deal with perturbed Rydberg series. This method was developed for the Coulomb potential.

- It was shown (Kokoouline et al., PRA 62, 022504 (2000), Ostrovsky et al., J. Phys. B 34, L27 (2001)), that the Lu-Fano method can be applied for molecular potentials with C₃/R₃ behavior.

- The formula well describes the effect of perturbations on two vibrational progressions. They vary slowly with energy and can be used for determination of observables for energies inaccessible in the experiment.

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- Parameters C₃(P₁/₂), C₃(P₃/₂) can be fitted also.

- The probability of transition P is linked to \( \xi \). We obtained P=0.54

- The calculated spectrum for energies above the 6S+6P₃/₂ limit can be fitted also.

Conclusions

1. In presence of perturbations the Le Roy-Bernstein analysis does not work for interpretation of photoassociation spectra.
2. When there are two coupled molecular states, Lu-Fano plot should be used instead of the Le Roy-Bernstein analysis.
3. Lu-Fano analysis allows to determine long-range coefficients with high accuracy.
4. It allows to parameterize perturbations, to predict positions and widths of vibrational levels, cross-section, other observables.
5. Fine structure transition cross section can be determined by extrapolation of parameters fitted on accurate spectroscopic data.