

VIBRATIONAL STRUCTURE OF RESONANCE RAMAN SPECTRA *

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The intensity distribution of vibrational progressions in resonance Raman spectra is studied on a simple model. Expressions are derived for the dependence of this vibrational structure on the wavelength of the incident light and on temperature.

One of the most striking features of resonance Raman (RR) spectra in comparison to ordinary Raman spectra is the presence of prominent vibrational progressions [1]. The first attempts [2,3] to account for the overtones were based solely on the resonance terms in the perturbation description, but this turned out to be quantitatively unsatisfactory [4]. Subsequently Jacon et al. [4] showed that off-resonance terms, in particular the near-resonance terms associated with levels close to the resonance levels, contribute on a par with resonance terms. They demonstrated by direct computation that the inclusion of both terms accurately reproduces the intensities of the first overtones ($\Delta v = 2$) relative to the fundamentals ($\Delta v = 1$) in the RR spectra of I_2 obtained with the 7 suitable lines of the argon ion laser [4].

To understand intensity distributions in RR progressions in a general way it seems useful to start from the analogy with intensity distributions in absorption and emission, which are well-known to be governed by the Franck-Condon principle. In this note we study the RR analog of this principle for simple model potentials. Similar studies applied to absorption and emission spectra have been very helpful in the early days of molecular electronic spectroscopy [5]. Since the vibrational integrals of RR spectra are more complicated than the Franck-Condon

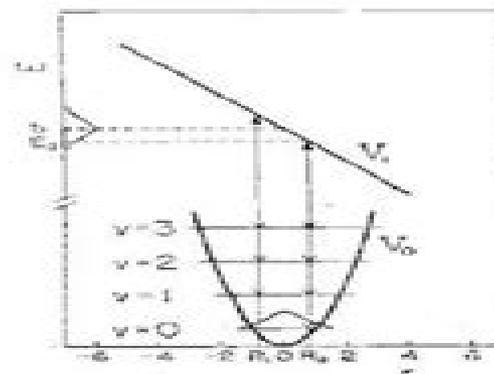


Fig. 1. Model potentials used to derive analytic expressions for resonance Raman intensity (divided by ω^4) distributions.

factors of absorption and emission bands, only the simplest models are expected to be amenable to analytic solution.

The model adopted is illustrated in fig. 1. It consists of an isolated diatomic molecule with a harmonic ground-state potential V_0 and a linear repulsive excited state potential V_1 .

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