

# Spectroscopy of mixed alkali dimers by the polarisation labelling technique: Application to NaLi and NaRb molecules

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## Abstract

The  $4^1\Pi$  state in NaRb and  $5^1\Pi$  state in NaLi molecules are investigated experimentally by polarisation labelling spectroscopy technique. The spectroscopic data are used to construct potential energy curves for both states.

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## 1. Introduction

Recently there has been considerable progress in production of cold diatomic molecules [1,2]. Mixed alkali dimers are of particular interest here because their permanent dipole moments allow to manipulate them by external electric fields. As several experimental techniques related to cold molecules involve optical excitation either in the formation or detection stages [3,4], a need of precise spectroscopic characterisation of molecular states has arisen. An additional motivation for the research on heteronuclear alkali dimers is that they can be treated as the prototypes of diatomic, heteronuclear systems for which quantum mechanical computations are feasible. Therefore, experiments on them may provide valuable tests of the quality of theoretical calculations.

For a number of years we have been involved in investigation of alkali dimers, studying excited electronic states accessible in one photon transitions from the ground states. We have been using the polarisation labelling spectroscopy

(PLS) method [5]. This double resonance technique elegantly surmounts the difficulty of resolving and analysing highly congested molecular spectra: with a proper choice of frequencies and polarisations of two laser beams, interacting with a molecular sample, only transitions from a few known rovibrational levels in the ground state are observed. This selectivity is particularly useful when studying spectra of heteronuclear species, AB, which are often covered by dominating spectra of  $A_2$  and  $B_2$  molecules. Moreover, the rotational structure of the spectra obtained by this technique is fully resolved and relatively easy to understand.

In this paper, we report observation and characterization of two previously unknown molecular states: the  $4^1\Pi$  state in NaRb molecule and the  $5^1\Pi$  state in NaLi. In the following sections we describe principle of the PLS technique and details of two experiments together with discussion of their results.

## 2. The polarisation labelling spectroscopy technique

The goal of polarisation labelling spectroscopy is to obtain simplified, high resolution spectra of the molecule

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under investigation. Two laser beams, a weak, linearly polarised probe beam and a strong pump beam of circular or linear polarisation interact with a molecular sample. When the pump beam is tuned into a molecular resonance, it orients or aligns the levels involved in the transition due to optical pumping and makes the sample optically anisotropic (and birefringent). These levels are said to be ‘labelled’. The probe beam experiences the created anisotropy only if it is resonant with one of the labelled levels and some other molecular level. In such case the linear polarisation of the probe beam becomes slightly rotated and it also acquires a small degree of ellipticity. The signal detected in polarisation spectroscopy arises from analysing these changes in polarisation state of the probe beam.

In our version of the PLS [6] we employ a V-type optical–optical double resonance excitation scheme, with a probe laser of fixed frequency and a tuneable pump laser. The frequency of the probe laser coincides with a set of known transitions in the investigated molecule, and in this variant this is the probe laser which labels the involved rovibrational levels in the ground molecular state. By tuning the pump laser over the investigated spectral range and monitoring changes in polarisation of the probe beam, we obtain excitation spectrum of the molecule simplified by the fact that it originates from a few ground state levels with fixed and known  $(v, J)$  quantum numbers.

### 3. Investigation of the $4^1\Pi$ state in NaRb

NaRb vapour was produced by heating a mixture of sodium and rubidium (1:1 by volume, Rb of natural isotopic composition) in a linear, stainless steel heat pipe. The temperature was maintained at around 370 °C and a pressure of 5 mbar of argon buffer gas was established. The

fixed frequency of the probe laser ( $\text{Ar}^+$  laser working at 514.5, 501.7, 496.5, 488.0 or 476.5 nm, either multimode or in a single mode regime) coincided with a set of known transitions in the  $D^1\Pi \leftarrow X^1\Sigma^+$  system of NaRb, thus labelling the rovibrational levels involved in the ground state. A summary of the identified labelling transitions which give rise to the polarisation spectra is given in Ref. [7]. The pump laser (pulsed dye laser and XeCl excimer laser system, 1 mJ pulse energy,  $0.1 \text{ cm}^{-1}$  spectral line-width) was tuned across the investigated  $4^1\Pi \leftarrow X^1\Sigma^+$  band system, in the spectral range 24,900–26,900  $\text{cm}^{-1}$ . The pump laser frequency was determined absolutely from the optogalvanic spectrum of argon and additionally calibrated by sending part of the laser beam through a Fabry–Pérot interferometer with a free spectral range of  $1 \text{ cm}^{-1}$ . The accuracy in absolute determination of wave numbers over the whole investigated range was better than  $0.1 \text{ cm}^{-1}$ . The pump and probe laser beams were collinearly superimposed in the molecular vapour zone. Crossed polarizers were placed at both sides of the heat pipe in the path of the probe beam. The residual signal transmitted through them was monitored by a photomultiplier and processed with a microcomputer. The rotationally resolved spectra were observed for both  $\text{Na}^{85}\text{Rb}$  and  $\text{Na}^{87}\text{Rb}$  isotopic species (see Fig. 1 for an exemplary fragment of the spectrum).

Altogether 686 transitions in the  $4^1\Pi \leftarrow X^1\Sigma^+$  system were found, with vibrational and rotational quantum numbers of the levels observed in the  $4^1\Pi$  state ranging from 0 to 20 and from 2 to 97, respectively. The absolute vibrational numbering was established using the isotope effect. To convert the experimental line positions to eigenvalues of rovibronic levels in the  $4^1\Pi$  state, the highly accurate molecular constants for the ground state of NaRb by Doc-

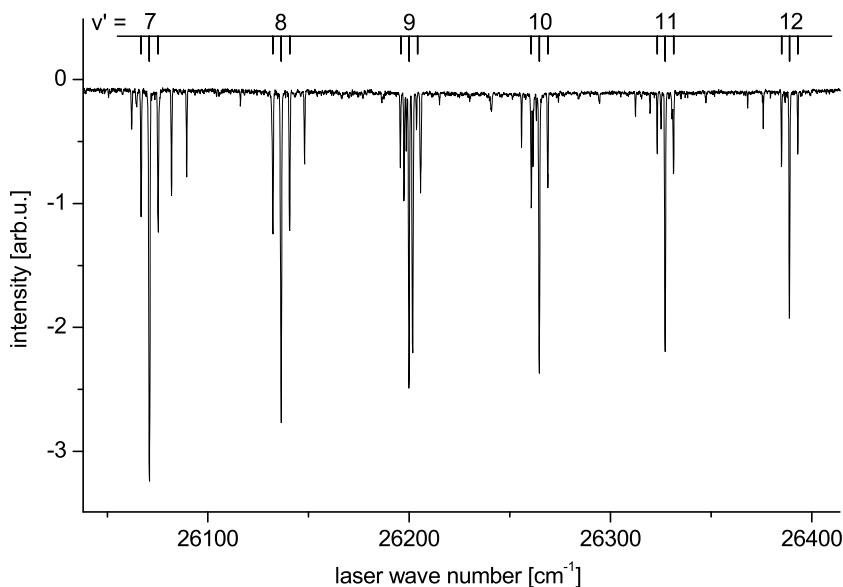


Fig. 1. A portion of the polarisation spectrum of NaRb obtained with 501.7 nm line of the  $\text{Ar}^+$  laser as the probe and linearly polarised pump beam. The assigned progression corresponds to the transitions  $4^1\Pi \leftarrow X^1\Sigma^+$  in  $\text{Na}^{85}\text{Rb}$  from the level  $v'' = 0, J'' = 43$  labelled in the ground state. Note a clear perturbation of the  $v' = 9$  level indicated by doubling of the molecular lines.

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