

## POLARIZATION SPECTROSCOPY FOR INVESTIGATION OF ALKALI-METAL DIATOMIC MOLECULES

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**Abstract.** We introduce the principle of the polarization labelling spectroscopy (PLS) for investigation of alkali-metal diatomic molecules. Signatures of PLS spectrum, which are important for classification of electronic states, for different electronic transition bands are presented. As an illustration for advantages of the technique, show the first rotational resolution for the  $2^1\Pi$  state NaLi.

**Keywords:** diatomic molecules, laser spectroscopy.

### I. INTRODUCTION

Recent years have witnessed a tremendous progress in production and trapping of ultracold molecules culminating in formation of molecular Bose–Einstein condensates [1, 2], with the vast majority of work done on alkali metal dimers. Although not yet much exploited experimentally, the heteronuclear alkali diatomics are of particular interest here, because their permanent dipole moments make them susceptible to manipulate by external electric fields. As several experimental techniques related to cold molecules involve optical excitation either in the formation or detection stages [3], an accurate need of precise spectroscopic characterisation of excited molecular states has arisen.

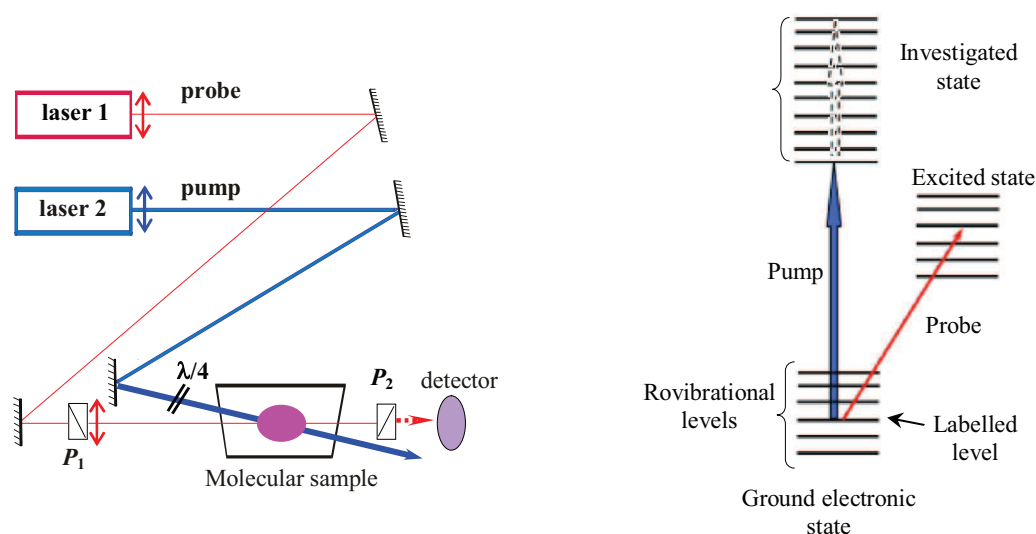
Experimental investigations for the alkali molecules, however, encounter a difficult problem due to a large number of allowed electric dipole transitions between various rovibrational energy levels in electronic states. In order to overcome this problem, the PLS technique is profitable. It based on the combination between the polarization spectroscopy and the optical-optical double resonance [4-6].

### II. BASIC PRINCIPLE OF PLS

The basic idea of the PLS is to monitor changes of polarization of a weak probe laser beam after passing through a molecular sample optically pumped by a strong laser light coming from another laser. A typical experimental scheme is shown in fig 1, where the molecular sample is placed between the two crossed linear polarizers  $P_1$  and  $P_2$ .

Whenever the pump laser is off the molecular sample is optically isotropic because there is no preferential direction of the total angular momentum  $\vec{J}$  of each molecule. Namely, for a given value of quantum number  $J$  there are  $2J + 1$  degenerated sublevels, each has the same statistical weight. Each sublevel corresponds to a value of quantum number  $m_J$  which is a quantum number of the projection of  $\vec{J}$  on a particular quantized axis. The linear polarization of the probe beam is

therefore unchanged after passing through the sample, thus the probe laser beam is blocked at the second polarizer  $P_2$ .



**Fig. 1.** Basic experimental scheme for PLS (left), and the excitation scheme (right).

When the pump beam is on, let's assume that the frequency of the pump laser is tuned to excite a molecular transition  $J'' \rightarrow J'$ . The pump light depopulates the absorbing molecules at the lower level  $J''$  and then populates them to upper level  $J'$ . It is known that the degree of depopulation for the lower level is not the same among the sublevels because of the two following reasons. The first one is due to the selection rules for the transitions between sublevels  $\Delta m_J = 0, +1, \text{ or } -1$  for linear, right circular-, or left circular polarization, respectively. The second reason is due to the dependence of absorption cross-section on value of  $m_J$ . As a result the molecular sample, after absorbing photons, becomes unequal population among the sublevels of the lower or upper level. Therefore, whenever the probe light excites a transition which shares the lower level (so called "labelled" level) with the pumped transition, the polarization of the probe beam is changed, and thus the probe beam passes through the polarizer  $P_2$  and then reaches to the detector. Such transitions possess the so called as V-type optical-optical double resonance scheme.

### III. RELATIVE INTENSITIES OF POLARIZATION SPECTRUM

In the V-type PLS experiments, the lower and upper states of the probe transition is known. On the other hand, the ground electronic states of the alkali-metal diatomic molecules are of the  $^1\Sigma$  symmetry. According to the selection rules for one-photon transitions, therefore, the upper states of the pump and probe transitions in the V-type optical-optical resonance are of the  $^1\Sigma$  or  $^1\Pi$  symmetry.

From the density matrix formalism, relative intensities of PLS spectrum were determined analytically [7]. The main results for PLS spectrum in the case of large  $J$  limit are shown in fig 2. In this figure, the capital letter  $L$  or  $C$  stands for linear or circular polarization of the pump beam, respectively. From the signatures of the PLS spectrum one may classify symmetry of investigated electronic states. For instance, when probe P-line is chosen in the  $^1\Sigma - ^1\Sigma$  band, if the spectrum consists of only P and R lines in both cases of linear and circular polarizations of the pump beam then the investigated state is  $^1\Sigma$ ; if P, Q and R lines appears simultaneously in the case of linear polarization of pump light the investigated state should be  $^1\Pi$ .

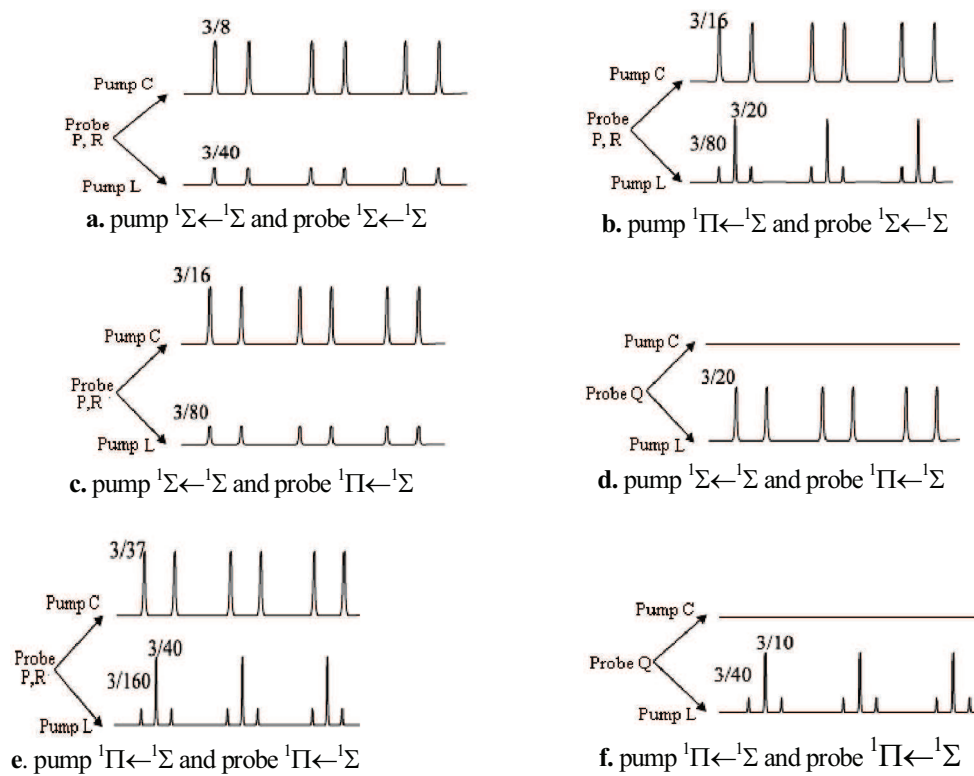


Fig. 2. Relative intensities in PLS spectrum calculated in [7] for different probe bands.

#### IV. EXPERIMENTS

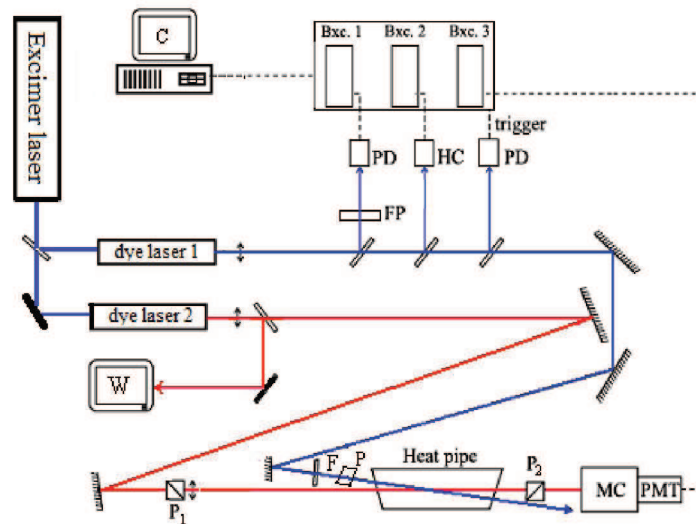


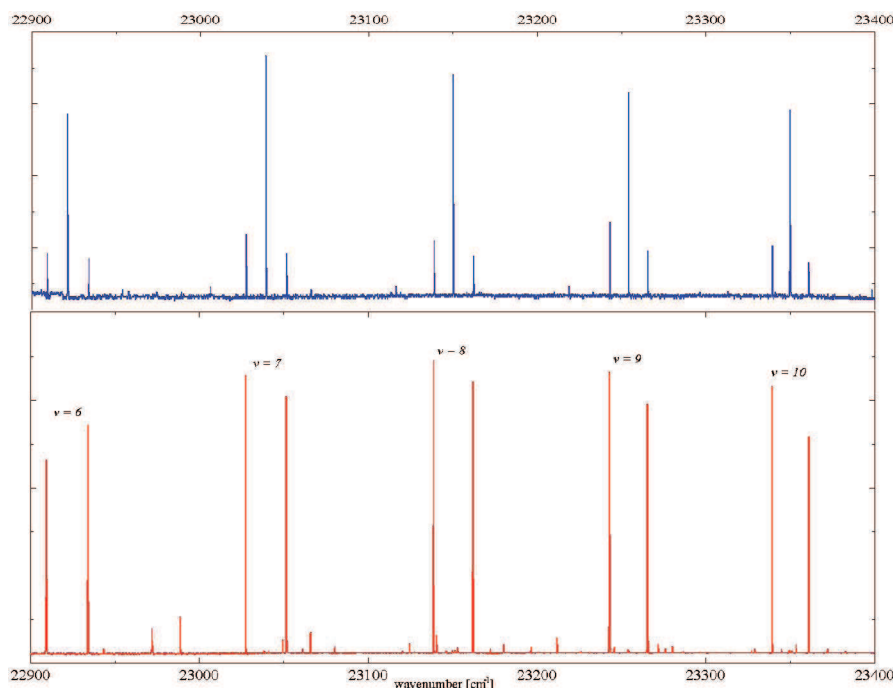
Fig. 3. Experimental setup for V-type PLS: Bxc- boxcar integrator, FP- Fabry-Perot interferometer, HC- hollow-cathode lamp, F-  $\lambda/4$  wave plate, MC- monochromator, P- polarizer, PD- photodiode, PMT- photomultiplier tube.

The arrangement of PLS experiment for NaLi is schemed in fig.3. We use a homebuilt Littman-type dye laser and a Lumonic dye laser as probe and pump sources, respectively. Both lasers are pumped synchronously by an Excimer laser. The Lumonic dye laser works with Styben 3 and Coumarin. It delivers pulse energy of *ca* 5 mJ with  $0.05 \text{ cm}^{-1}$  spectral linewidth.

The homebuilt dye, which works with DCM, provides weak pulse energy (ca 0.05 mJ) with  $0.5 \text{ cm}^{-1}$  spectral linewidth. The probe laser wavelength was tuned to excite resonantly a known transition in the  $2^1\Sigma - 1^1\Sigma$  band, where the probe wavelength is measured with a HighFinesse WS-6 wavemeter. We also use a 496.5 nm  $\text{Ar}^+$  laser to probe a known level (0, 30) in the ground state. The pump laser beam is propagated almost collinearly with the probe beam thus ensuring maximum overlap of the beams inside the sample. The polarization of the pump beam is set to either linear or circular by a removable  $\lambda/4$  wave-plate.

In our experiment, NaLi molecules were produced in a three-section heat-pipe oven [8]. The pump laser wavelength scanned over region  $21500 - 24000 \text{ cm}^{-1}$ . During the scanning a small part of the pump laser beam was sent to a Fabry - Perot interferometer with a free spectral range of  $1 \text{ cm}^{-1}$ , and two neon- and argon-filled hollow-cathode lamps are used to serve the calibration of spectrum. The transmission interference and optogalvanic signals were recorded by two boxcar integrators (Bx1 and Bx2 in fig. 3). The passing probe light through the second polarizer is sent to a monochromator and then detected by a photomultiplier tube. The monochromator was used here to eliminate fluorescence coming from the heat-pipe oven. Finally, the polarization signal was recorded at the boxcar Bx3, simultaneously with the optogalvanic signal and transmission fringes of the interferometer.

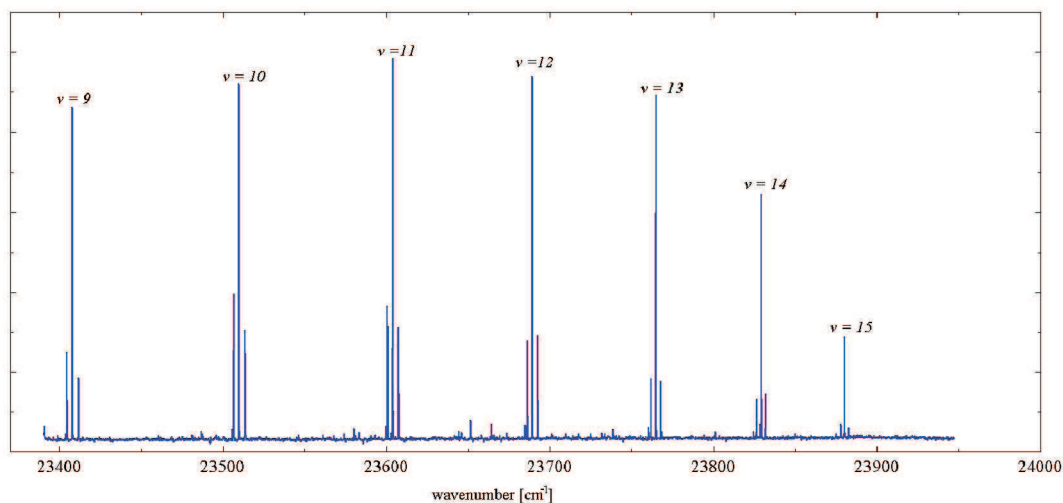
In the scanning spectral range region of the pump laser wavelength, we performed measurements corresponding to 22 labelled levels in linear polarization mode of the pump laser beam and probing via P-branch. We observed spectra of two electronic states in which consist of P, Q and R lines (Fig.4). Such feature showing that the observed spectra are of the  $^1\Pi$  symmetry. By using the atomic asymptotes we attribute the state as  $2^1\Pi$ . It should be noted here that, for the first time, this state is resolved rotational structure.



**Fig. 4.** A fragment of PLS spectrum observed for the case of linear (upper) and circular (down) polarization of the pump beam when probing 496.5 nm, thus labels level (0, 30) in the ground electronic state.

In fig.5 we show part of PLS spectrum observed with  $15594.71 \text{ cm}^{-1}$  probe laser line which labels level  $(v, J) = (0, 9)$  in the ground electronic state. Here, we observed the vibrational progression up to  $v = 15$ . By comparing wave numbers of last triplet in the vibrational

progressions with the atomic asymptotes of this state we see that our observation is close to the dissociation limit. The spectroscopic characterization for the  $2^1\Pi$  state will be reported [9].



**Fig. 5.** A fragment of PLS spectrum near the dissociation limit for the case of linear pump polarization and the  $15594.71\text{ cm}^{-1}$  probe wavenumber, which labels level (0, 9) in the ground state.

## V. CONCLUSION

The polarization labelling spectroscopy has proven to be a simple and efficient technique for investigate spectrum of diatomic molecules. Although the polarization spectra are Doppler-limited, they are greatly simplified in comparison to normal absorption spectra due to labelling process. The technique is therefore useful for the studies of molecules having densely-spaced rovibrational levels.

## VI. ACKNOWLEDGMENTS

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