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journal homepage: [www.elsevier.com/locate/jqsrt](http://www.elsevier.com/locate/jqsrt)Spectroscopic study of the  $C(3)^1\Sigma^+ \leftarrow X^1\Sigma^+$  and  $c(2)^3\Sigma^+ \leftarrow X^1\Sigma^+$  transitions in KCs moleculeJacek Szczepkowski<sup>a,\*</sup>, Anna Grochola<sup>a</sup>, Pawel Kowalczyk<sup>b</sup>, Włodzimierz Jastrzebski<sup>a</sup><sup>a</sup>Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, 02-668 Warszawa, Poland<sup>b</sup>Institute of Experimental Physics, Faculty of Physics, University of Warsaw, ul. Pasteura 5, 02-093 Warszawa, Poland

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## ABSTRACT

We present an experimental investigation of the  $C(3)^1\Sigma^+$  and  $c(2)^3\Sigma^+$  states of KCs using two-colour polarisation labelling spectroscopy (PLS) technique. Pointwise potential energy curves are generated for both states using the inverted perturbation approach (IPA) method. The experimental pointwise potentials and molecular constants  $T_e$ ,  $\omega_e$  and  $R_e$  are compared with theoretical calculations.

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## 1. Introduction

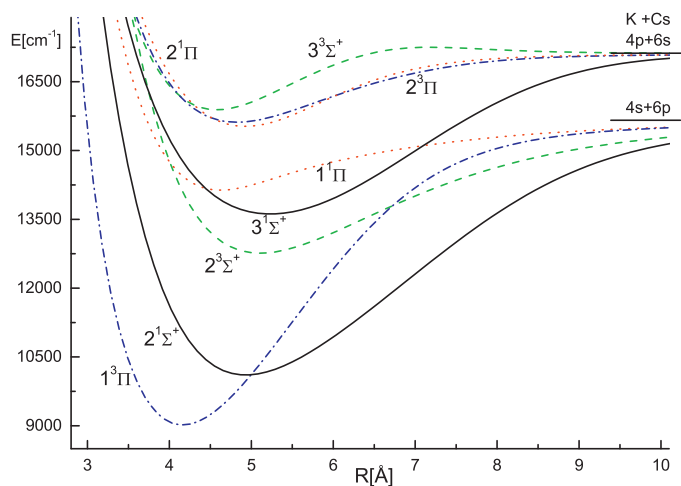
Molecules with permanent electric dipole moments are a basis for experiments from many different areas of physics. Polar molecules give opportunity to control interactions with external fields [1], allow for precision measurements of fundamental constants [2,3], open the way to exploration of quantum phases in dipolar gases [4,5] and testing universality in few-body systems [6,7]. One may also notice a development of so called ultracold chemistry [8]. A particular goal for many groups is to create molecules in their absolute ground state, since it is expected to be the first step in achieving heteronuclear molecular Bose-Einstein condensate. It has been done only for few molecules so far: KRb [9] and RbCs [10,11] using Feshbach resonances and stimulated Raman adiabatic passage (STIRAP) process and for LiCs using photoassociation [12]. KCs with its permanent electric dipole moment of 1.92 D is the next promising candidate. First K-Cs interspecies Feshbach resonances were observed [13] and several optical coherent schemes to create ultracold KCs molecules in their absolute rovibrational ground state were proposed [14]. Described processes base on spectroscopic studies performed so far.

Spectroscopic data are particularly important for all physicists interested in experiments conducted in an ultracold environment, since they give precise information about energies of rovibrational levels, couplings influencing transition probabilities and possible local perturbations, which may become an obstacle as well as an opportunity in process of manipulation of molecules. So far several electronic states of KCs molecule were investigated in high precision spectroscopic experiments: the ground state [15] and a few higher excited states [16–19]. But particular interest is focused on states dissociating to lower lying asymptotes [20–22], since most of excitation schemes lead through these states [14,23]. Couplings between these states were investigated and deperturbation procedures were performed [24] to open a possibility of further study on cooling and manipulating of KCs molecules.

To extend present knowledge of the electronic structure of KCs in the most interesting region we present the first experimental data concerning the  $c(2)^3\Sigma^+$  state dissociating to the first excited asymptote  $K(4^2S_{1/2}) + Cs(6^2P_{3/2})$  and the  $C(3)^1\Sigma^+$  state dissociating to the  $K(4^2P_{1/2}) + Cs(6^2S_{1/2})$  asymptote, but interacting with the states lying below. All potential energy curves correlating to both mentioned asymptotes are presented in Fig. 1.

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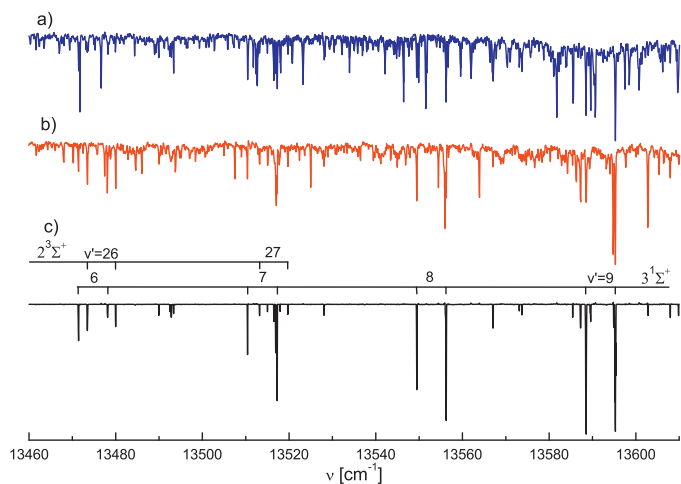
**Fig. 1.** *Ab initio* potential energy curves of KCs [25] for all states correlating to the  $K(4s) + Cs(6p)$  and  $K(4p) + Cs(6s)$  asymptotes. Energies are given with respect to the minimum of the electronic ground state. The horizontal lines with atomic products on them indicate the energies of the adiabatic dissociation limits.

## 2. Experiment

Metallic samples of potassium and caesium (5 g each) were used to generate the vapour of KCs molecules in a stainless steel linear heat-pipe oven, heated to 430 °C. Helium under the pressure of 5 Torr served as a buffer gas to prevent alkali metal deposition on the heat-pipe windows. The polarisation labelling spectroscopy (PLS) technique was applied to record the KCs excitation spectra. Details of the method may be found in our previous papers [26,27]. In general, excitation spectrum was studied using the V-type optical-optical double resonance method with two independent pump and probe lasers. As a pump laser served a parametric oscillator/amplifier system (OPO/OPA, Sunlite EX, Continuum) pumped with the third harmonic of an injection seeded Nd:YAG laser (Powerlite 8000). Either the signal or idler beams of the system were used to scan the region  $11800\text{ cm}^{-1} - 14000\text{ cm}^{-1}$ . The probe beam, of a fixed wavelength controlled with a HighFinesse WS-7 wavemeter, originated from a home-made dye laser operating on Rhodamine 6G and pumped synchronously by the same Nd:YAG laser. Its wavelength was in resonance with selected transitions in the well known KCs  $4^1\Sigma^+ \leftarrow X^1\Sigma^+$  band system [16]. The rotationally resolved  $C(3)^1\Sigma^+ \leftarrow X^1\Sigma^+$  and  $c(2)^3\Sigma^+ \leftarrow X^1\Sigma^+$  excitation spectra were recorded, with the accuracy of observed molecular line positions better than  $0.1\text{ cm}^{-1}$ . Calibration of molecular spectra was based on simultaneously generated optical-van spectra of argon and transmission fringes from a Fabry-Pérot interferometer of  $\text{FSR} = 1\text{ cm}^{-1}$ .

## 3. Analysis and results

In the spectral region examined in the present experiment, we could observe several characteristic patterns. They consisted either of vibrational progressions of P, Q and R lines ('triplets') or of P and R lines only ('doublets'). All recorded P, Q, R 'triplets' were identified as transitions to the  $B(1)^1\Pi$  state, studied before [20]. The remaining P and R lines from doublets were corresponding to transitions to four different electronic states in KCs. In the lower part of the scanned area traces of the coupled  $A(2)^1\Sigma^+$  and  $b(1)^3\Pi$  states were recognized. In the upper part of the examined region transitions to the  $c(2)^3\Sigma^+$  and  $C(3)^1\Sigma^+$  states overlap with strong lines registered due to a presence of the  $B(1)^1\Pi$  state. Observation of five electronic states in the same spectral region resulted in complexity of the spectra. Moreover the probe laser was typically lab-



**Fig. 2.** Exemplary spectrum of KCs after taking numerical product of two experimental polarisation spectra (c). The assigned P,R progressions correspond to transitions to consecutive vibrational levels in the  $c(2)^3\Sigma^+$  and  $C(3)^1\Sigma^+$  states when the ( $\nu' = 3, J' = 87$ ) ground state level is labelled by the probe laser. In the two upper panels spectra before multiplication procedure are presented, recorded with the circularly polarised probe laser set at wavelengths corresponding nominally to transitions  $4^1\Sigma^+(\nu' = 16, J' = 86) \leftarrow X^1\Sigma^+(\nu' = 3, J' = 87)$  (a) and  $4^1\Sigma^+(\nu' = 16, J' = 88) \leftarrow X^1\Sigma^+(\nu' = 3, J' = 87)$  (b). The unassigned, randomly distributed lines in (c) are artefacts resulting from accidental overlap of different transitions in (a) and (b).

elling more than one rovibrational level in the ground state, what additionally increased the number of the recorded spectral lines. In such situation, despite the expected simplification of experimental spectra due to the polarisation labelling method, they remained highly congested. As a solution to this problem we recorded two or more spectra containing progressions originating from the same rovibrational level in the ground state, but labelled with different wavelengths of the probe laser. An example of two spectra originating from the same rovibrational level in the ground state is presented in Fig. 2a and b. Then we looked for common features in the spectra, taking numerical product of two or three of them. Such procedure allowed for unambiguous assignment of spectral lines, as shown in Fig. 2c.

Transitions to the  $C(3)^1\Sigma^+$  state were relatively easy to identify, since observation of several rovibrational levels of this state has been already reported [20]. The remaining spectral lines, much weaker, could correspond to transitions to either upper part of the  $b(1)^3\Pi$  and  $A(2)^1\Sigma^+$  states or to the  $c(2)^3\Sigma^+$  state. The first two states were described with high precision before [21] and elimination of the corresponding spectral lines [24,28] allowed for conclusion that we recorded mostly transitions to the latter, i.e. the  $c(2)^3\Sigma^+$  state. In the case of KCs molecule direct optical excitation from the singlet ground state to the excited triplet states is possible due to strong spin-orbit coupling and mixing of singlet and triplet rovibrational levels [15,21]. Such spin-forbidden transitions have been observed before in KCs [29], as well as in heavier (RbCs [30]) but also lighter (LiCs [31] and NaCs [32]) alkali metal diatomics containing caesium atoms.

### 3.1. $C(3)^1\Sigma^+$ state

Altogether 441 spectral lines were recorded corresponding to transitions to rovibrational levels of the C state in the range  $\nu' = 3 - 18, J' = 38 - 127$ . Distribution of the experimental data is shown in Fig. 3. Knowing energies of the ground state levels [15], we converted measured transition energies into 390  $C(3)^1\Sigma^+$  state term values  $T(\nu', J')$ . A potential energy curve for the  $C(3)^1\Sigma^+$  state was built using the pointwise Inverted Perturbation Approach (IPA)

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