STRUCTURE OF VIBRATIONAL BANDS OF THE $E^3\Sigma^+(6^3S_1) \leftarrow A^3\Pi(5^3P_1)$ ELECTRONIC TRANSITION IN CdRG (RG=Ar,Kr)

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GROUP OF MOLECULAR SPECTROSCOPY AND QUANTUM INFORMATION

INTRODUCTION

Isotopic and rotational structures of the (ν',ν'') vibrational bands in the $E^3\Sigma^+(\nu') \leftarrow A^3\Pi_{0+}(\nu'')$ transition in CdAr and CdKr complexes were investigated using free-jet expansion beam and laser excitation. An optical-optical double resonance (OODR) process was employed starting from the $X^1\Sigma_0^+$ to the $E^3\Sigma^+$ lowest Rydberg *via* the $A^3\Pi_{0+}$ or $B^3\Pi_1$ intermediate electronic state. In the experiment the CdAr and CdKr molecules produced in a free-jet expansion beam were irradiated with two successive laser pulses from two dye lasers pumped simultaneously by the second and third harmonics of the same Nd^+:YAG laser. The first dye-laser pulse, was frequency doubled and excited a maximum number of the CdAr or CdKr molecules from the $X^1\Sigma_0^+$ (ν=0) to the $A^3\Pi_0$ (ν =5 or 9, respectively) or $B^3\Pi_1$ (ν =1) intermediate level. The primarily excited molecules were irradiated with a second dye-laser pulse from the ν to the ν level in the $E^3\Sigma^+$ state. The resulting laser induced fluorescence signal, which was observed perpendicularly to the plane containing the molecular and laser beams, was recorded with photomultiplier tube. The signal from PM was integrated in digital oscilloscope and stored in a computer. The structure of the bands with well defined isotopic structure was analyzed taking into account a complex rotational structure of the triplet ← triplet transition. In the future, a simulation of the bands will provide new values for the ω_e, $\omega_e x_e$, B_u and D_u vibrational and rotational characteristics of the upper and lower states.

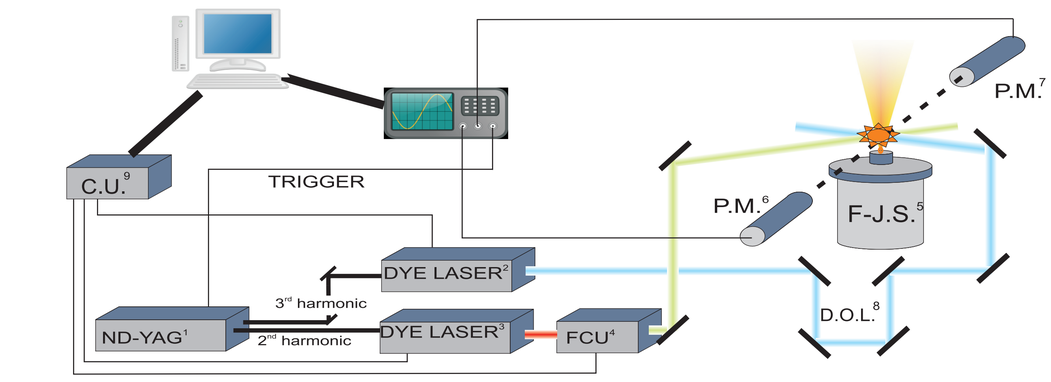

FIG. 1. Experimental set-up used for optical-optical double resonance (OODR) method. [1]Nd⁺YAG laser Powerlite 7010 of Continuum, [2]"Probe" dye laser (NarrowScan of Radiant Dyes Lasers & Accessories) working with Coumarine 480 in Methanol (spectral range 474-488nm), [3]"Pump" dye laser (TDLIII of Quantel) working with DCM in DMSO (spectral range 322.5-332.5 nm), [4]Frequency Conversion Unit, [5]Free-Jet Source, [6]Photomultiplier ("pump"-laser excitation), [7]Photomultiplier ("probe"- laser excitation), [8]Delay Optical Line, [9]Controler Unit.

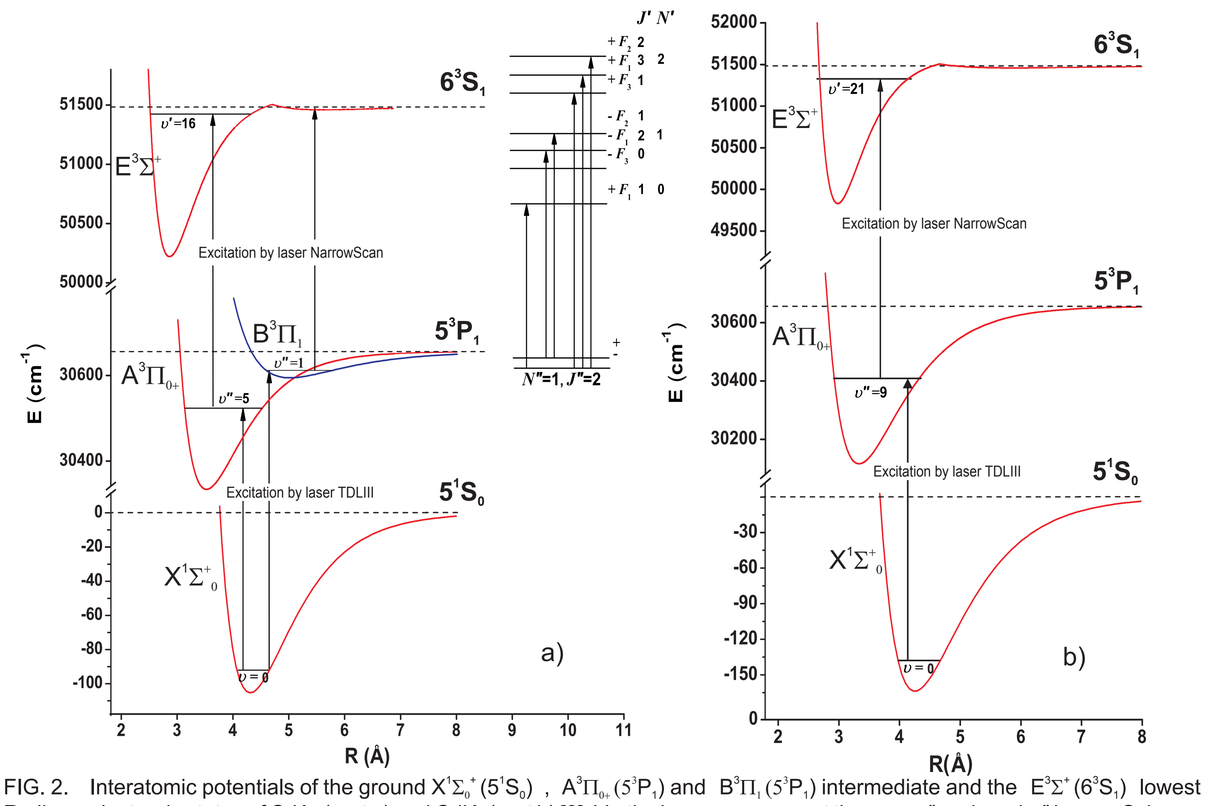

FIG. 2. Interatomic potentials of the ground $X^1\Sigma_0^+(5^1S_0)$, $A^3\Pi_{0+}(5^3P_1)$ and $B^3\Pi_1(5^3P_1)$ intermediate and the $E^3\Sigma^+(6^3S_1)$ lowest Rydberg electronic states of CdAr (part a) and CdKr (part b) [3]. Vertical arrows represent the „pump" and „probe" lasers. Scheme on the right (in part a) illustrates a part of rotational structure of the $E^3\Sigma^+ \leftarrow A^3\Pi_{0+}$ electronic transition (not in scale).

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[4] PGOPHER, a Program for Simulating Rotational Structure, C. M. Western, University of Bristol, http://pgopher.chm.bris.ac.uk.

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CdAr: transition *via* $A^3\Pi_0$ state

FIG.4.

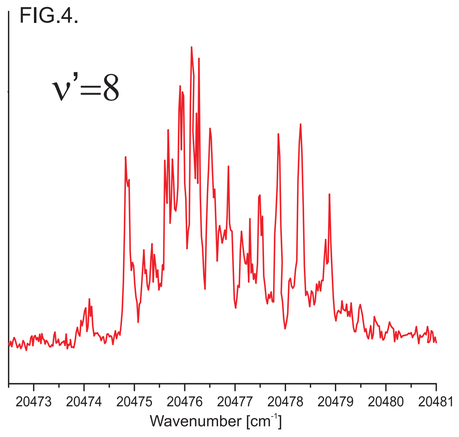


FIG.3.

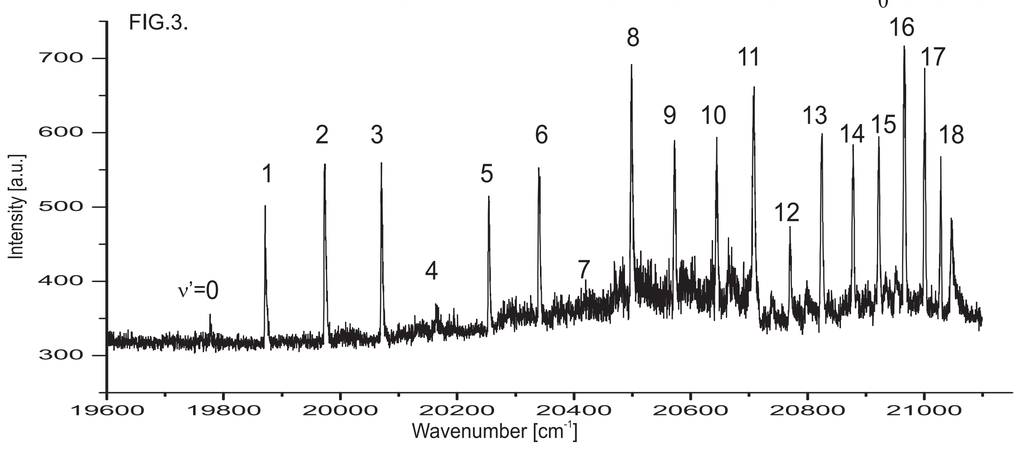


FIG.3. The excitation spectrum recorded using the $E^3\Sigma^+_{(in)}(\nu') \leftarrow A^3\Pi_{0+}(\nu''=5) \leftarrow X^1\Sigma_0^+(\nu=0)$ transitions and OODR method (from [3])

FIG.4-10. The (ν',ν'') vibrational band recorded using the $E^3\Sigma^+_{in}(\nu') \leftarrow A^3\Pi_{0+}(\nu''=5) \leftarrow X^1\Sigma_0^+(\nu=0)$ transitions employing the OODR method (this study).

FIG.10.

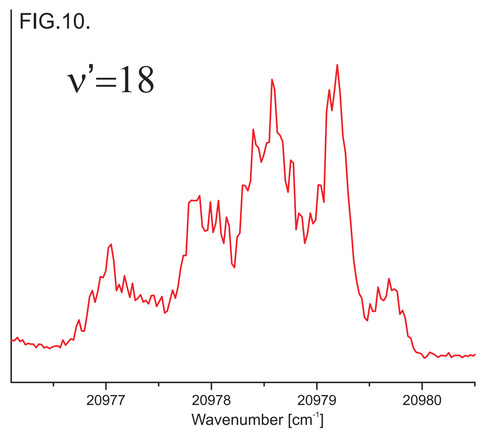


FIG.5.

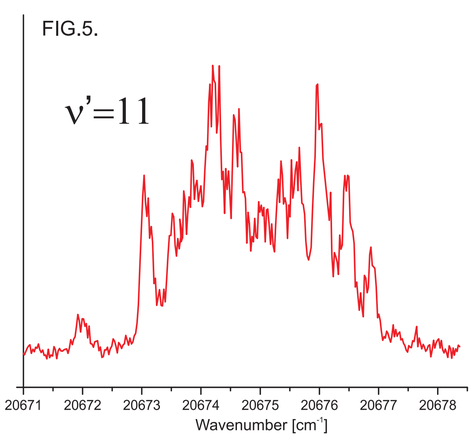


FIG.6.

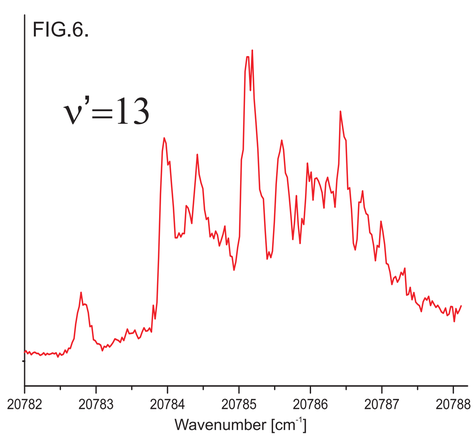


FIG.7.

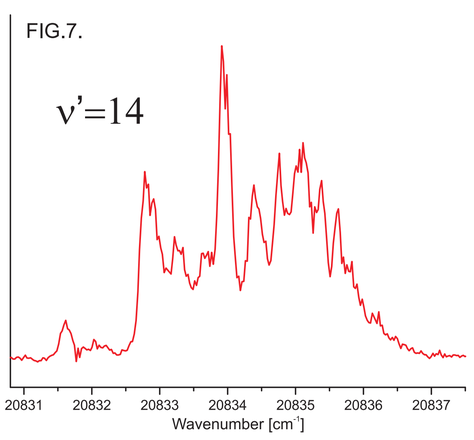


FIG.8.

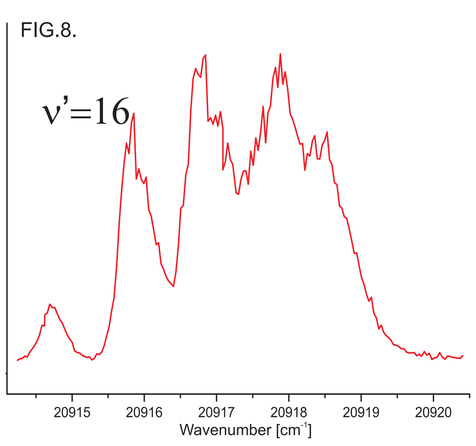
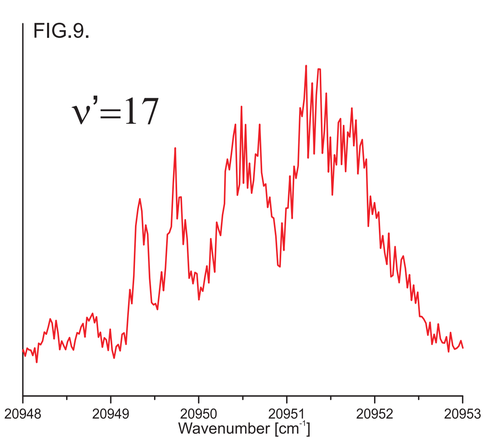


FIG.9.



CdKr: transition *via* $A^3\Pi_0$ state

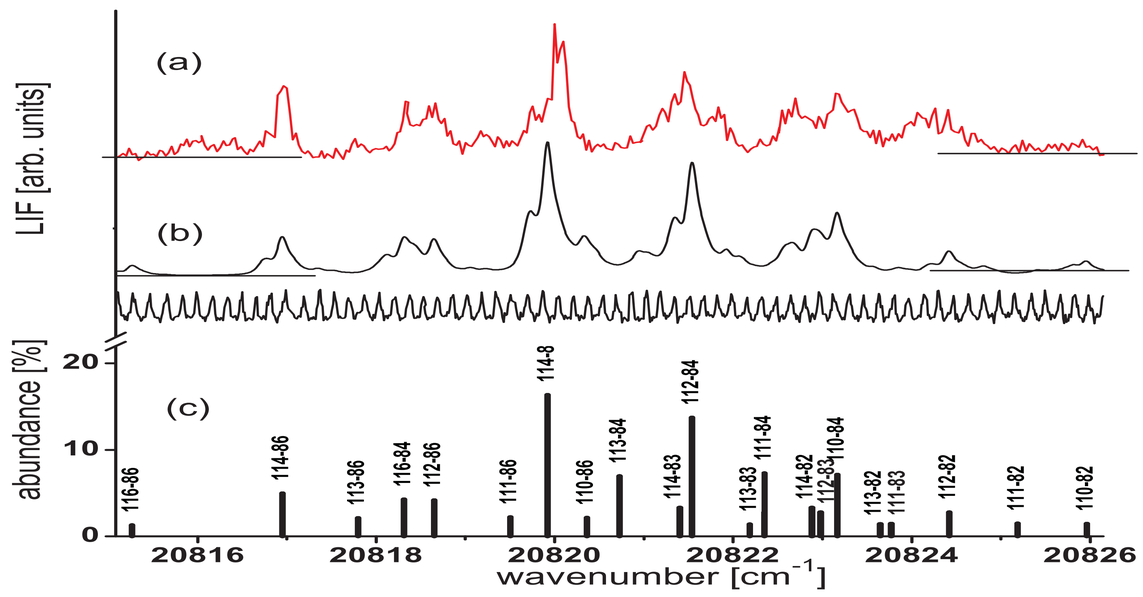


FIG.11. (a) The (ν', ν'')=(21,9) vibrational band recorded using the $E^3\Sigma^+_{(in)}(\nu'=21) \leftarrow A^3\Pi_{0+}(\nu'=9) \leftarrow X^1\Sigma_0^+(\nu=0)$ transitions in CdKr employing the OODR method. (b) Simulation performed using the PGOPHER program [4] with parameters ω'_e=106.5cm^{-1},$\omega_e x_e$' =2.16 cm^{-1},ω'_e=39.2cm^{-1},$\omega_e x_e$' =1.22 cm^{-1} , T_{rot}=2K and combined Lorentzian (Δ_{las}=0.1 cm^{-1}) and Gaussian (Δ_{Dopp}=0.06 cm^{-1}) profiles.

CdAr: transition *via* $B^3\Pi_1$ state

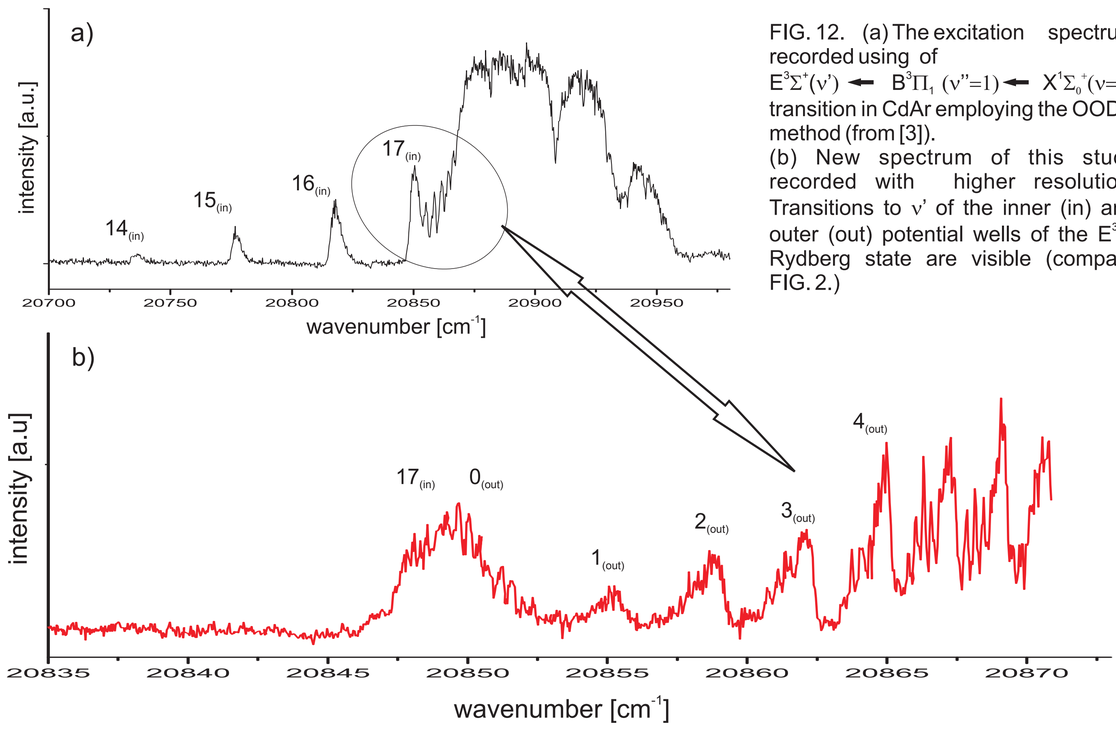


FIG. 12. (a) The excitation spectrum recorded using of $E^3\Sigma^+(\nu') \leftarrow B^3\Pi_1(\nu''=1) \leftarrow X^1\Sigma_0^+(\nu=0)$ transition in CdAr employing the OODR method (from [3]). (b) New spectrum of this study recorded with higher resolution. Transitions to ν' of the inner (in) and outer (out) potential wells of the $E^3\Sigma^+$ Rydberg state are visible (compare FIG. 2.)