INVESTIGATION OF THE $E^3\Sigma^+(6^3S_1)$ RYDBERG ELECTRONIC ENERGY STATE IN CdRG (RG=Ar,Kr) COMPLEXES USING THE OPTICAL-OPTICAL DOUBLE RESONANCE

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ATOMIN

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INTRODUCTION

Isotopic and rotational structures of the (υ',υ") vibrational bands in the $E^3\Sigma^+(\upsilon') \leftarrow A^3\Pi_{0+}(\upsilon'')$ and $E^3\Sigma^+(\upsilon') \leftarrow B^3\Sigma_1^+(\upsilon'')$ 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 $B^3\Sigma_1^+$ 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 $X^1\Sigma^+_0$ (υ=0) to the $A^3\Pi_{0+}$ (υ =5 or 9, respectively) or $B^3\Sigma_1^+$ (υ =1) intermediate level. The primarily excited molecules were irradiated with a second dye-laser pulse from the υ to the υ 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. Simulation of the bands provides new values for the ω_e, $\omega_e x_e$, B_υ and D_υ vibrational and rotational characteristics of the upper and lower states.

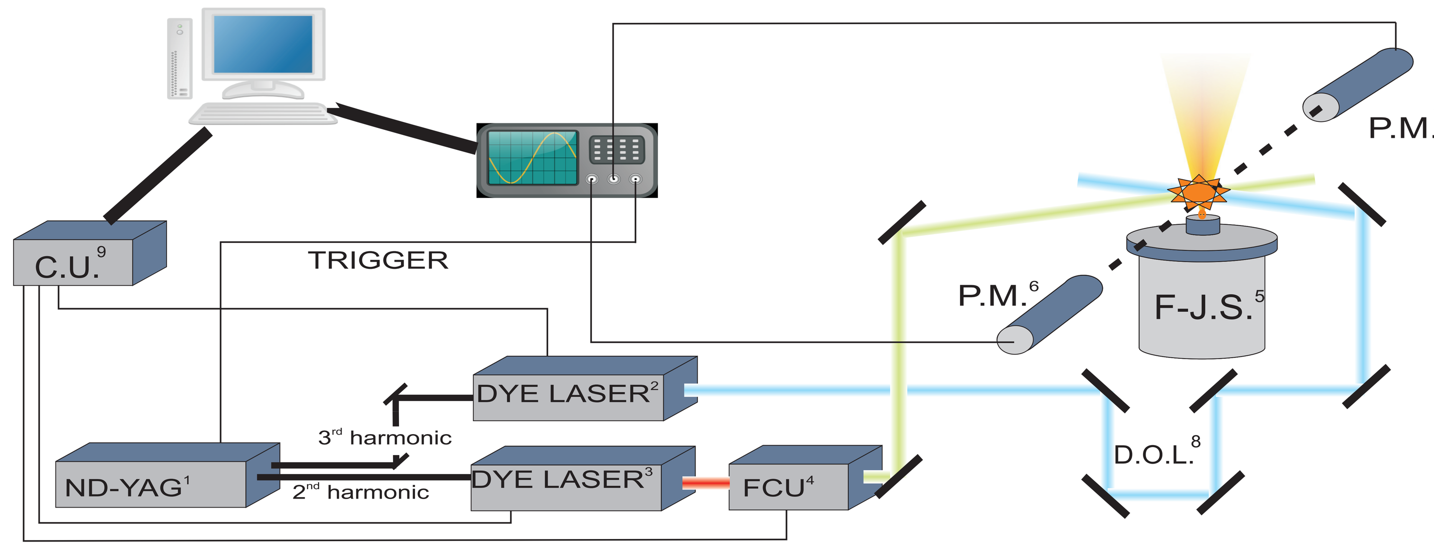

FIG. 1. Experimental set-up used for optical-optical double resonance (OODR) method. [1]Nd^+YAG laser Powerlite 7010 of Continuum, [2]"Probe" dye laser (NarrowScan of Radiant Dyes Lasers & Accessories) working with Coumarine 480 in Methanol (spectral range 474-488nm), [3]"Pump" dye laser (TDLIII of Quantel) 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.

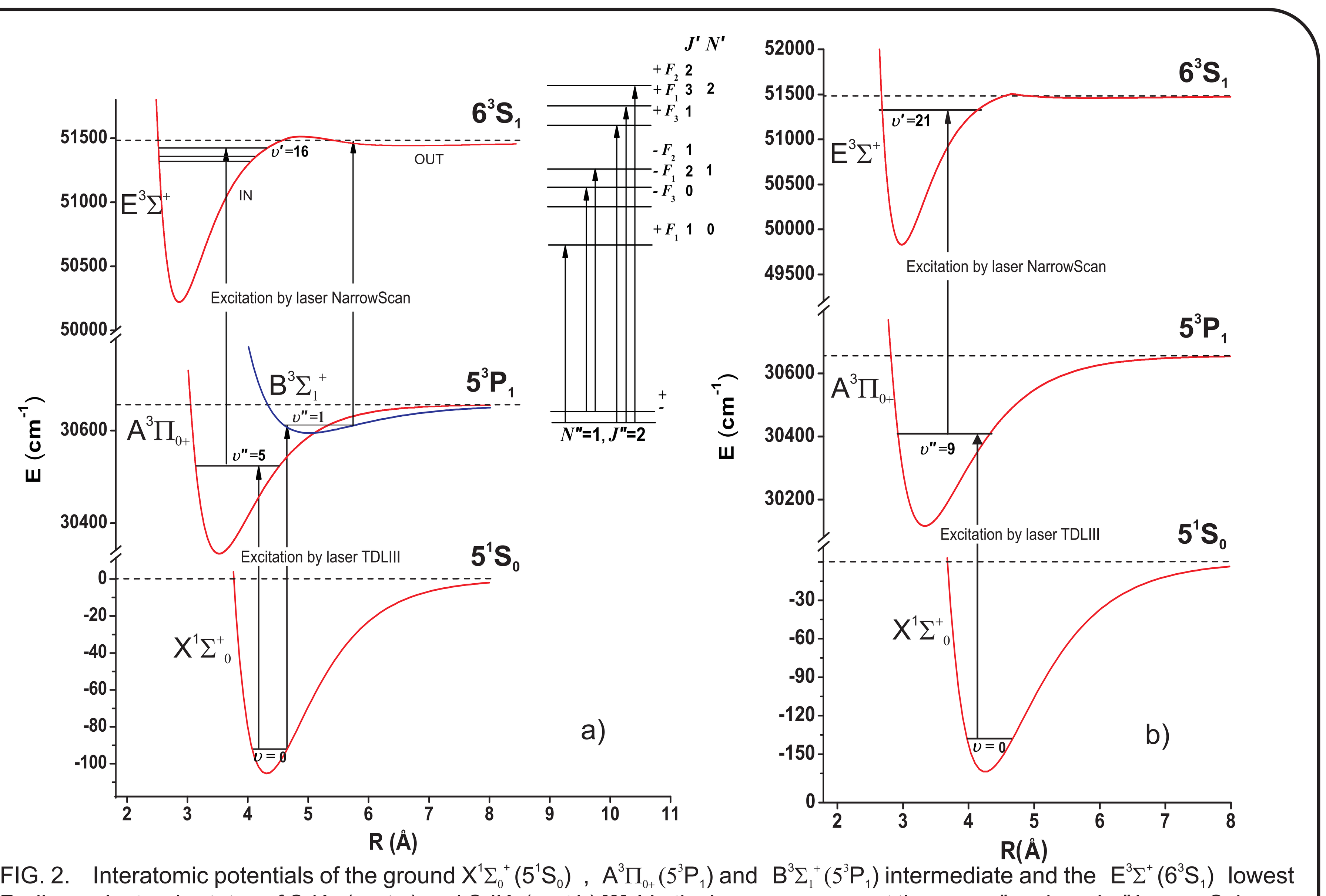

FIG. 2. Interatomic potentials of the ground $X^1\Sigma_0^+(5^1S_0)$, $A^3\Pi_{0+}(5^3P_1)$ and $B^3\Sigma_1^+(5^3P_1)$ intermediate and the $E^3\Sigma^+(6^3S_1)$ lowest Rydberg electronic states of CdAr (part a) and CdKr (part b) [3]. Vertical arrows represent the „pump" and „probe" lasers. Scheme on 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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[3] J. Koperski, M. Czajkowski, Spectrochim. Acta A 59 (2003) 2435.
[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.3 a.

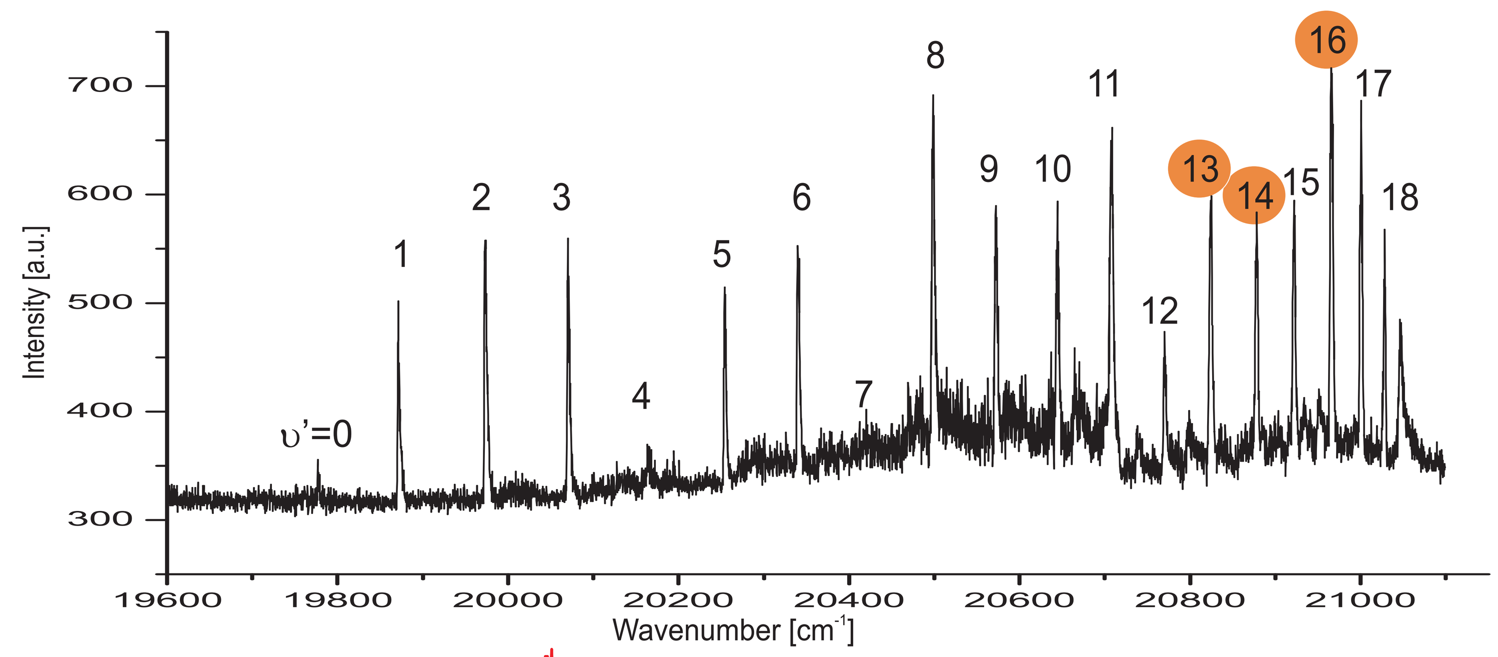
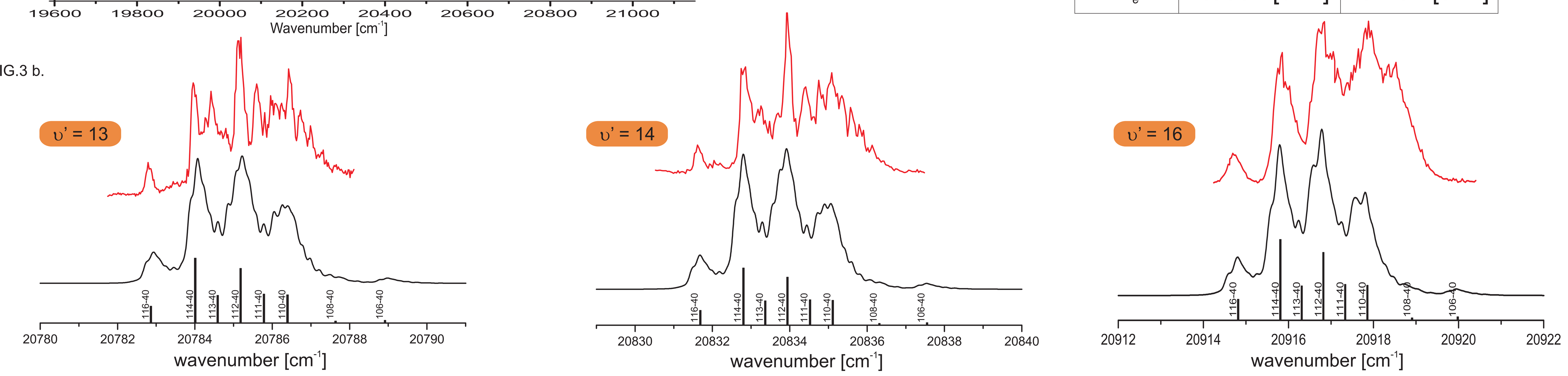


FIG.3 a) The excitation spectrum recorded using the $E^3\Sigma^+_{(in)}(\upsilon') \leftarrow A^3\Pi_{0+}(\upsilon''=5) \leftarrow X^1\Sigma_0^+(\upsilon=0)$ transitions and OODR method (from [3])

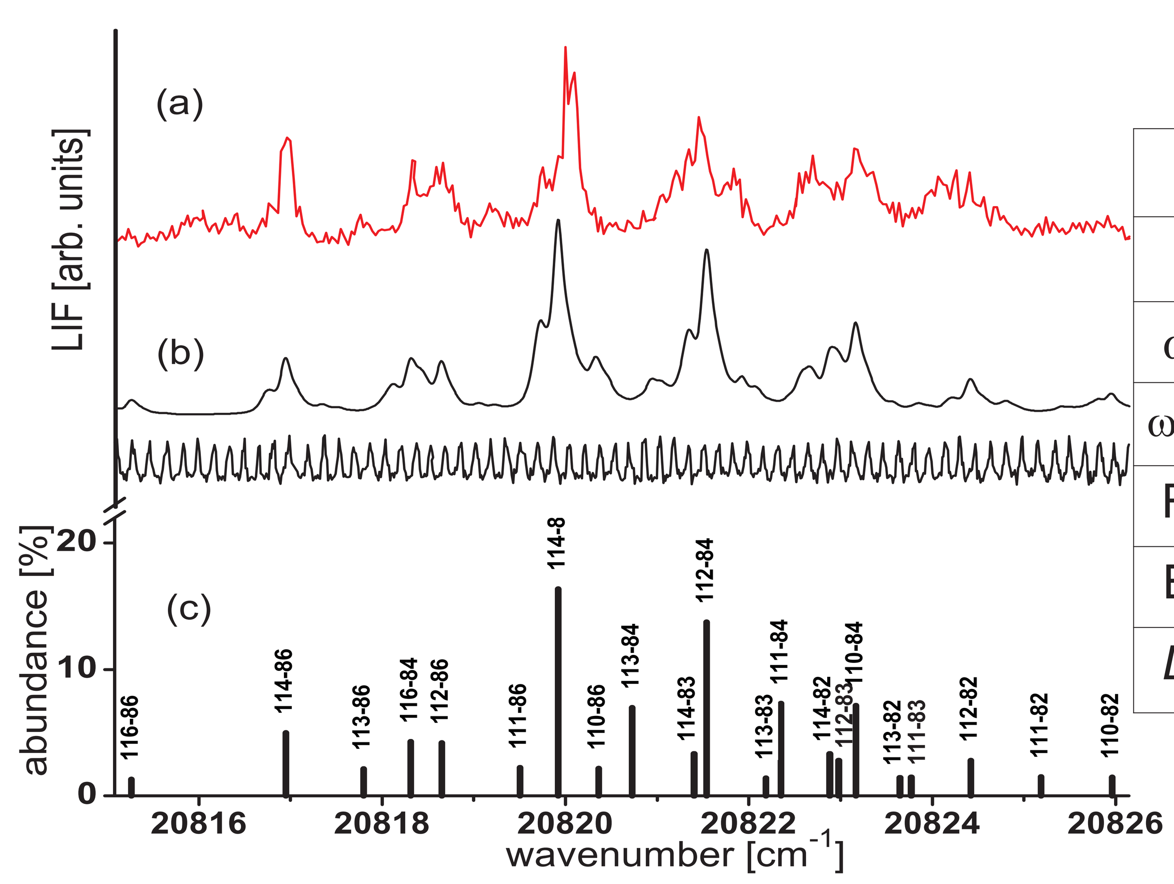
FIG.3 b) The (υ',υ") vibrational band recorded using the $E^3\Sigma^+_{in}(\upsilon') \leftarrow A^3\Pi_{0+}(\upsilon''=5) \leftarrow X^1\Sigma_0^+(\upsilon=0)$ transitions employing the OODR method (this study). Red curves present experimental results, black simulations performed in PGOPHER[4] program.

OUR RESULTS		
	$A^3\Pi_{0+}$	$E^3\Sigma_1^+$ (in)
ω_e	39.2[cm^{-1}]	106.5[cm^{-1}]
$\omega_e x_e$	1.22[cm^{-1}]	2.16[cm^{-1}]
R_e	3.51[A]	2.85[A]
B_e	0.0463[cm^{-1}]	0.0702[cm^{-1}]
D_e	2.58e-7[cm^{-1}]	1.22e-7[cm^{-1}]

FIG.3 b.



CdKr: transition *via* A $^3\Pi_{0+}$ state



OUR RESULTS		
	$A^3\Pi_{0+}$	$E^3\Sigma^+$(in)
ω_e	36.95[cm^{-1}]	90.97[cm^{-1}]
$\omega_e x_e$	0.615[cm^{-1}]	1.374[cm^{-1}]
R_e	3.34[A]	2.99[A]
B_e	0.0309[cm^{-1}]	0.0385[cm^{-1}]
D_e	8.61e-8[cm^{-1}]	2.76e-8[cm^{-1}]

FIG.11. (a) The (υ', υ'')=(21,9) vibrational band recorded using the $E^3\Sigma^+_{(in)}(\upsilon'=21) \leftarrow A^3\Pi_{0+}(\upsilon'=9) \leftarrow X^1\Sigma_0^+(\upsilon=0)$ transitions in CdKr employing the OODR method. (b) Simulation performed using the PGOPHER program [4] with parameters: T_{rot}=2K combined Lorentzian (Δ_{las}=0.1 cm^{-1}) and Gaussian (Δ_{Dopp}=0.06 cm^{-1})
(c) All m_1+m_2 mass combinations that constitute the (21,9) band.
(d) Fringes recorded using a Fabry–Perot etalon (FSR=0.2 cm^{-1}) to monitor the tuning process of the probe laser

CdAr: transition *via* $B^3\Sigma_1^+$ state

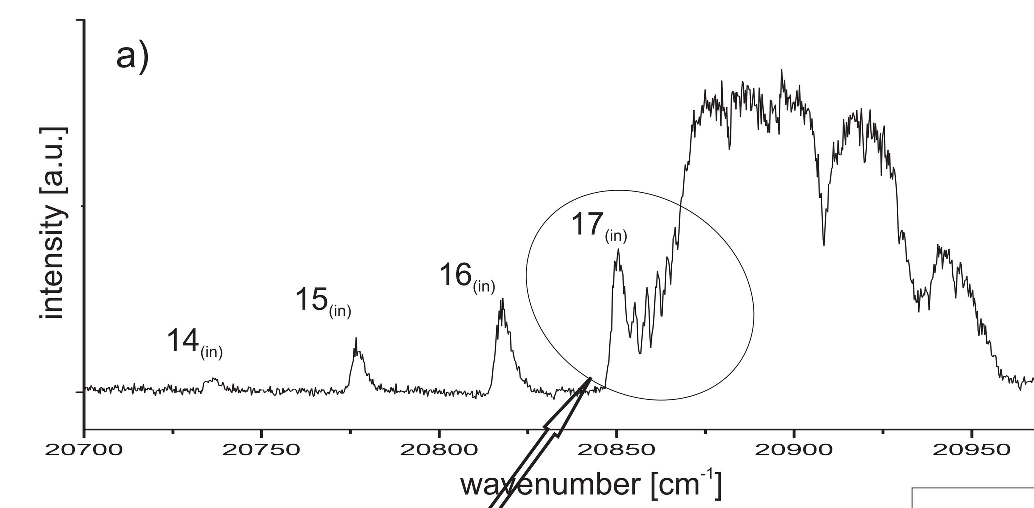
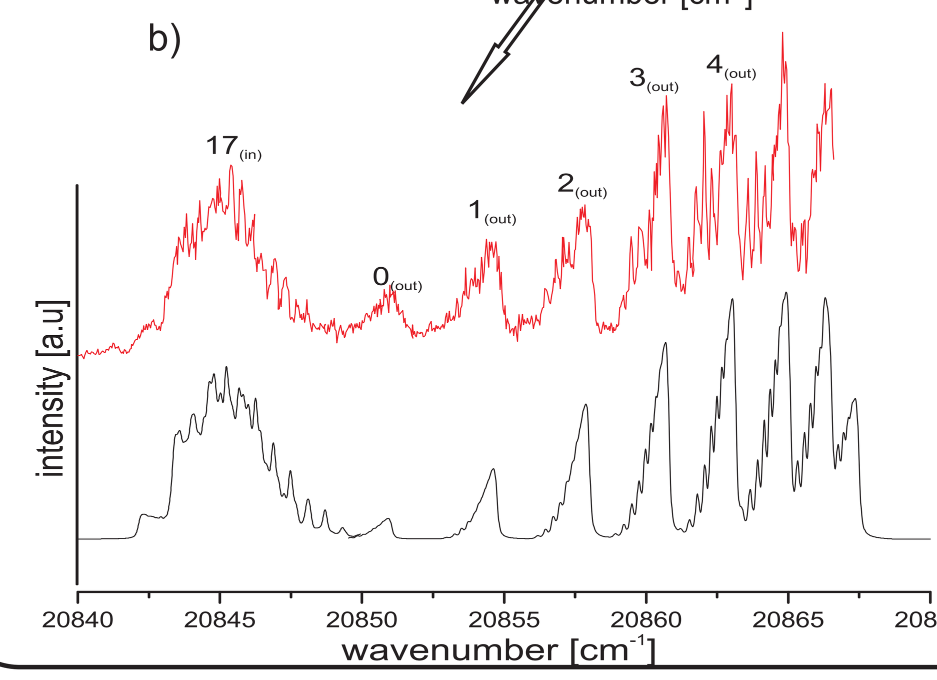


FIG. 5. (a) The excitation spectrum recorded using of $E^3\Sigma^+(\upsilon') \leftarrow B^3\Sigma_1^+(\upsilon''=1) \leftarrow X^1\Sigma_0^+(\upsilon=0)$ transition in CdAr employing the OODR method (from [3]).
(b) New spectrum of this study recorded with higher resolution (red - experiment, black - simulation). Transitions to ν' of the inner (in) and outer (out) potential wells of the $E^3\Sigma^+$ Rydberg state are visible (compare FIG. 2.)



OUR RESULTS		
	B $^3\Sigma_1^+$	$E^3\Sigma^+$(out)
ω_e	11.3[cm^{-1}]	4.15[cm^{-1}]
$\omega_e x_e$	0.56[cm^{-1}]	0.225[cm^{-1}]
R_e	5.07[A]	7.63[A]
B_e	0.0222[cm^{-1}]	0.00979[cm^{-1}]
D_e	3.41e-8[cm^{-1}]	2.18e-7[cm^{-1}]