Experiments with supersonic beam: from molecular rotations towards entanglement of atoms

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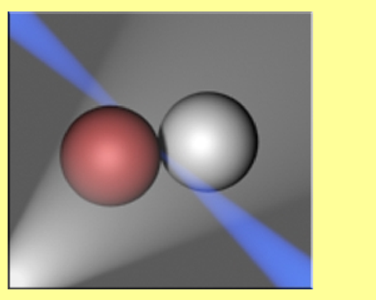

Molecular Spectroscopy and Quantum Information

Narodowe Laboratorium Technologii Kwantowych

Atomic Scale Science for Innovative Economy

INTRODUCTION

Laser spectroscopy of 12-group van der Waals (vdW) molecules produced and ro-vibrationally cooled in a free-jet expansion beam is one of methods for investigation of molecular energy structure [1]. Presently, the method is used in an investigation of vibronic and isotopic structures in the $D^10_u^+(6^1S_0)$ and F^31_u (6^3P_2) electronic energy Rydberg states of Cd_2. Laser induced fluorescence excitation spectra recorded using the $D^10_u^+ \leftarrow X^10_g^+(5^1S_0)$ and $F^31_u \leftarrow X^10_g^1$ transitions in the region of 206-218 nm provided spectroscopic characteristics of the excited states and allowed constructing of their interatomic potentials. Isotopic structures recorded in the (v',v'') bands of the $D^10_u^+ \leftarrow X^10_g^+$ transition were used in determination of the $D^10_u^+$-state vibrational characteristics (ω_e', $\omega_e'x_e'$) and v'-assignment. The frequency $v_{0,0}$ recorded directly in the $F^31_u \leftarrow X^10_g^+$ transition enabled determination of the bottom of the F^31_u-state potential well.

In the past, the method was applied, among others, in experimental studies of the $a^31_u(5^3P_1)$ [2], $b^30_u^+(5^3P_1)$ [3-5], $c^31_u(5^3P_2)$ [6,7], $A^10_u^+(5^1P_1)$ [8,9] and $B^11_u(5^1P_1)$ [10,11] excited as well as the $X^10_g^+$ ground [6,8-10] electronic-energy states of Cd_2 (see **Fig. 2** for reference).

Valence *ab initio* calculations of Cd_2 interatomic potentials were performed with relativistic and spin-orbit effects taken into account (see ***Ab initio* calculations**). The experimental results were compared with the obtained theoretical results.

Finally, a supersonic expansion of Cd_2 as a source of entangled atoms for a test of Bell's inequality is analysed. The experimental set-up is assembled in our laboratory (see **Fig. 5**).

EXPERIMENTAL SET-UP

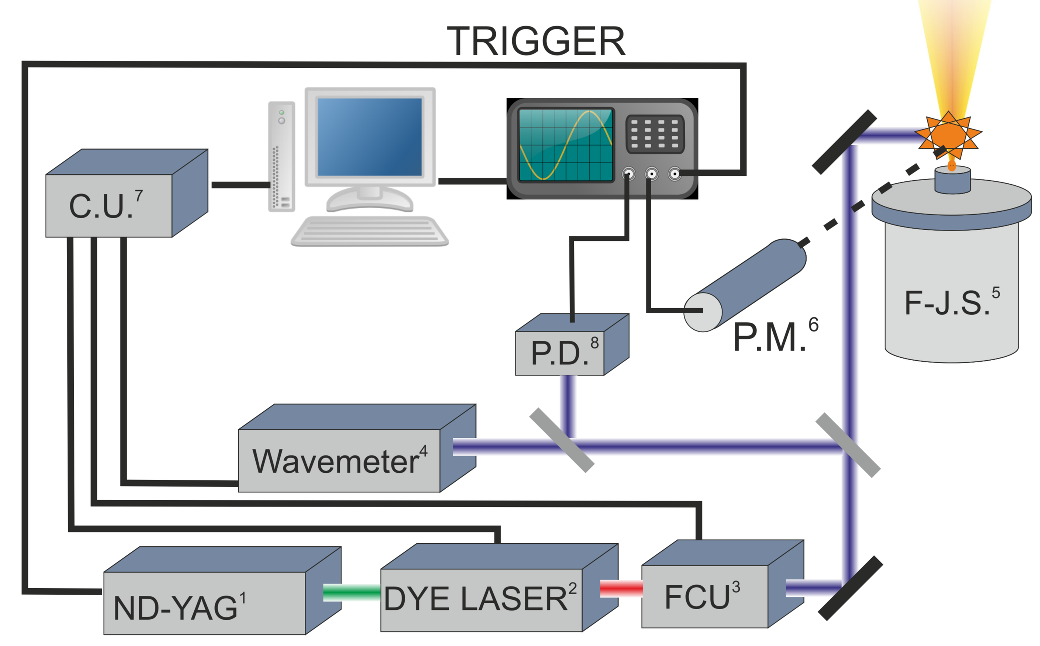

Fig. 1. Experimental set-up
[1] Nd-YAG Laser Powerlite 7010 of Continuum, [2]Dye laser (TDLIII of Quantel), [3]Frequency Conversion Unit, [4]Wavemeter, [5] Free-jet source, [6]Photomultiplier, [7] Controler unit, [8] Photodiode.

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AB INITIO CALCULATIONS

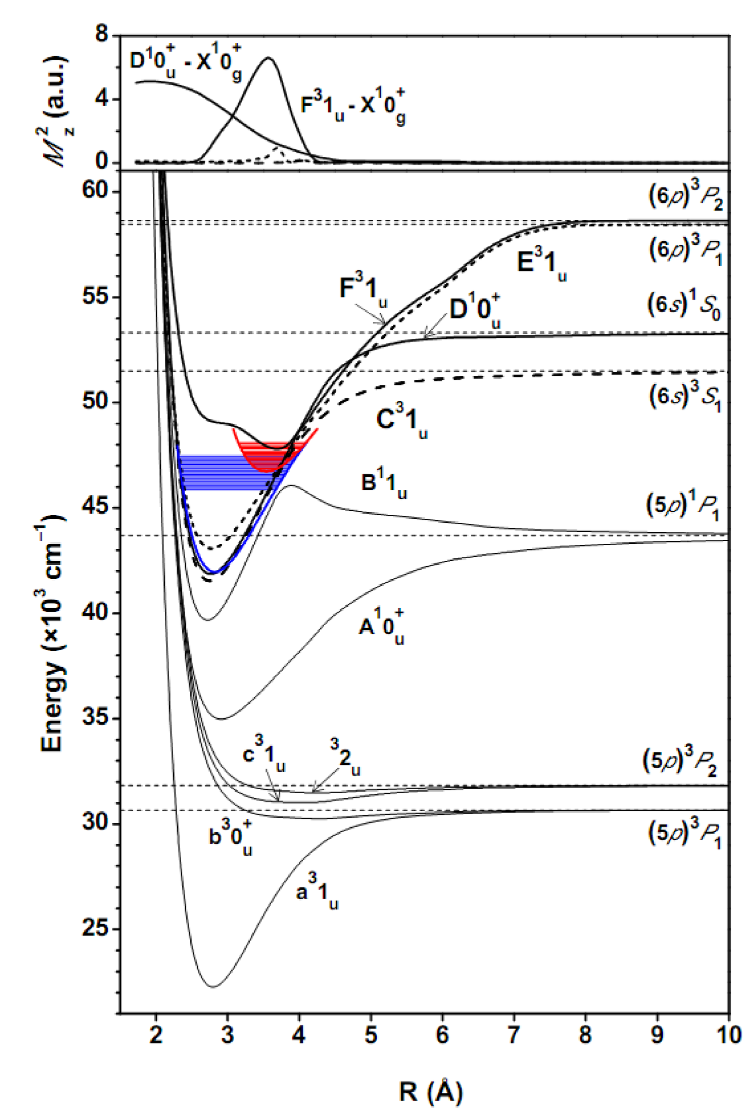
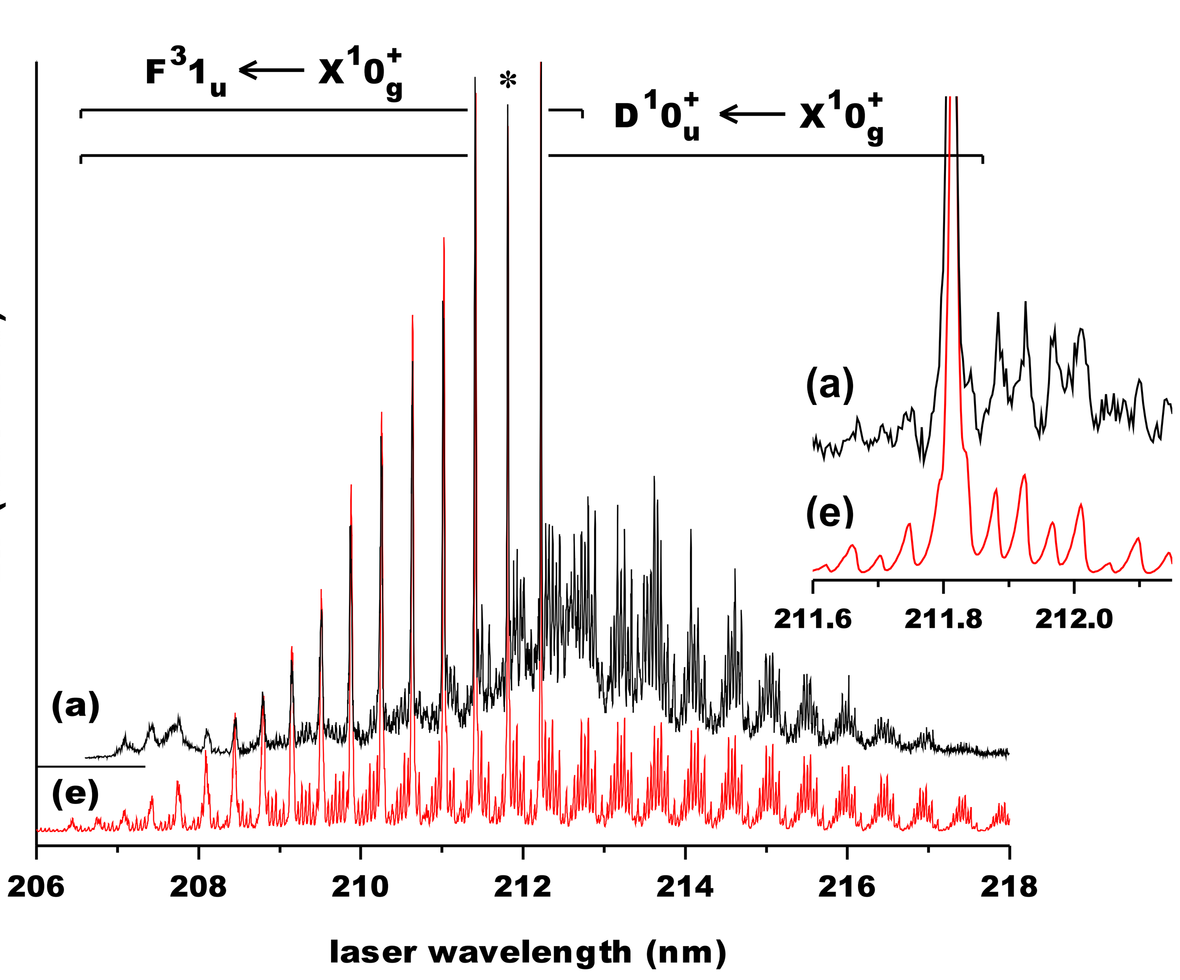


Fig.2. *Ungerade* interatomic potentials of Cd_2 obtained as a result of *ab initio* calculations. An energy range probed in this experiment is represented with positions of the u′ energy levels (horizontal lines) recorded in the excitation spectra. The interatomic potentials of the $D^10_u^+(6^1S_0)$ and $F^31_u(6^3P_2)$ states are plotted with thick solid lines while the $C^31_u(6^3S_1)$ and $E^31_u(6^3P_1)$ potentials are drawn with dashed thick solid lines. Upper part shows M_z^2 dipole transition moments squared calculated for transitions between the ground and C^31_u, $D^10_u^+$, E^31_u and F^31_u excited states. Experimentally determined potentials of the $D^10_u^+$ (blue thick line) and F^31_u (red thick line) states are plotted for comparison.

***Ab initio* calculations.** In *ab initio* calculations of this work, which were performed using a MOLPRO paackage [12], 20 electrons of the Cd atom were treated explicitly while the rest of the core electrons were replaced by the effective core pseudopotential [13]. In the calculations we used an augmented correlation-consistent polarized valence quadruple-zeta (aug-cc-pVQZ) atomic basis set [14] augmented by three *s*, two *p* and one *d* even tempered, diffuse basis set functions. The molecular orbitals used in the calculations of the excited triplet and singlet states were separately optimized for *gerade* and *ungerade* symmetry states in the state averaged complete-active-space multiconfiguration self-consisted field (CASSCF) method [15,16] for all triplet states correlating with the $(5p)^3P$, $(6s)^3S$, $(6p)^3P$ and all singlet states correlating with the $(5s)\,^1S$, $(5p)^1P$, $(6s)^1S$ and $(5d)^1D$ atomic asymptotes, respectively.

SPECTROSCOPY OF Cd_2 - RESULTS

Designation	$D^10_u^+(6^1S_0)$	$F^31_u(6^3P_2)$
ω_e'(cm⁻¹)	117.31±0.64	90.57±0.62
$\omega_e' x_e'$ (cm⁻¹)	0.22±0.02	0.480±0.025
D_o'(cm⁻¹)	11288.72±286.42	11847.5±3.2
D_e'(cm⁻¹)	11347.32±286.74	11892.7±4.4
R_e'(cm⁻¹)	2.82±0.03	3.54±0.01
$v_{0.0}$(cm⁻¹)	42338.84±283.32	47105.6±0.1
T_e(cm⁻¹)	42290.85±283.73	47071.05±0.54



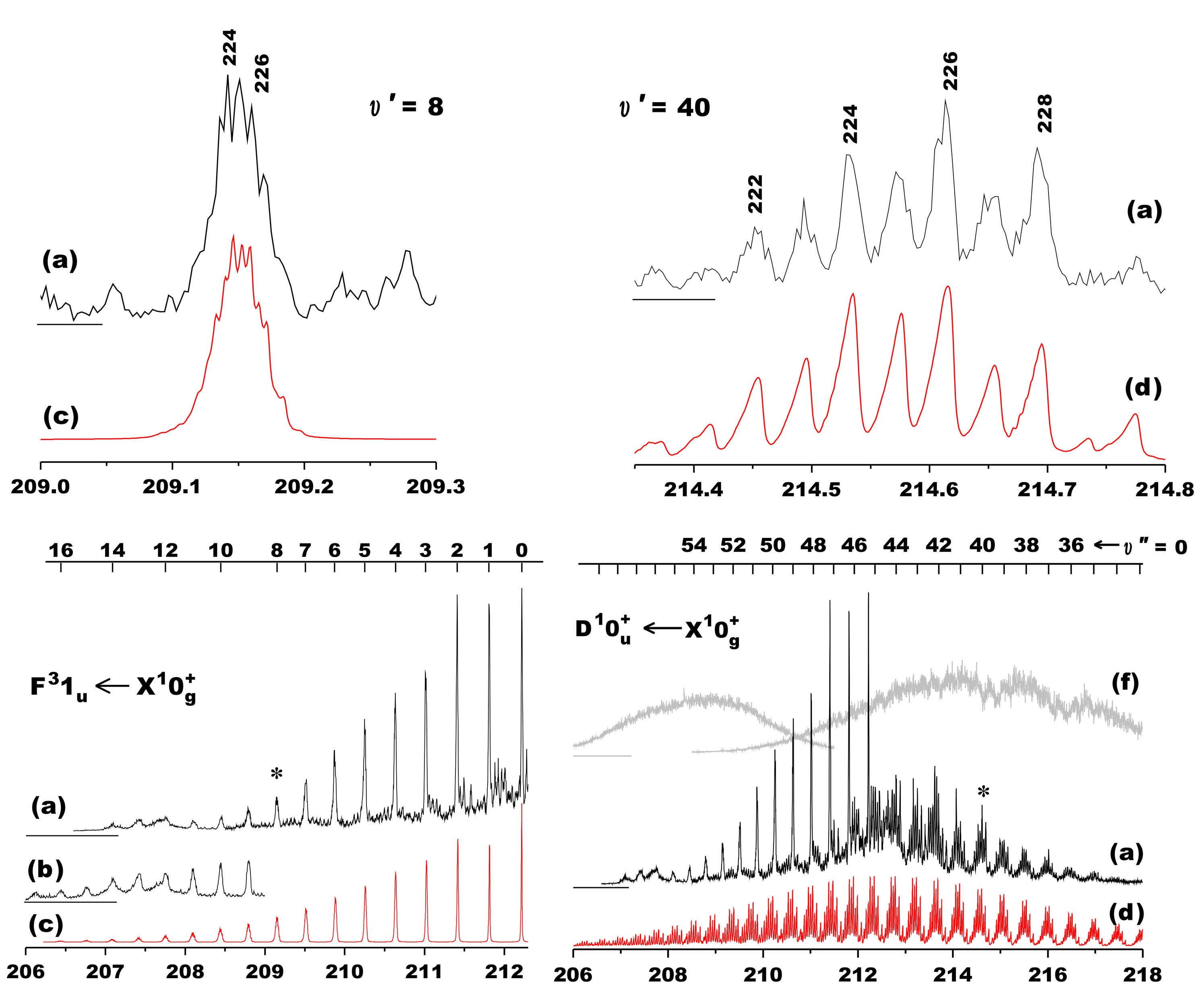


Fig.3. LIF excitation spectra recorded using the $F^31_u(6^3P_2) \leftarrow X^10_g^+$ and $D^10_u^+(6^1S_0) \leftarrow X^10_g^+$ transitions for (a) D=200μm, p_{Ar}=11 bar, X=9 mm, and (b) D=200μm, p_{Ar}=13 bar, X=6 mm. (c) Simulation of the excitation spectrum recorded using the $F^31_u \leftarrow X^10_g^+$ transition; for the simulation, parameters from table below as well as rotational temperature T_{rot}=5 K, maximum rotational quantum number J_{max}=25 and combined Lorentzian (Δ_{las}=0.5 cm⁻¹) and Gaussian (Δ_{dopp}=0.7cm⁻¹) profiles were used. The Δ_{las} and Δ_{Dopp} represent FWHM corresponding to the experimental values responsible for the bandwidth of the excitation laser and residual Doppler broadening associated with a transversal divergence of the molecular beam, respectively. (d) Simulation of the excitation spectrum recorded using the $D^10_u^+ \leftarrow X^10_g^+$ transition; for the simulation, parameters from table below as well as T_{rot}=5K, J_{max}=25 and combined Δ_{las}=0.5 cm⁻¹ and Δ_{Dopp}=0.7cm⁻¹ were used. (e) Two simulations of (c) and (d) combined together to illustrate a complexity of the total excitation spectrum. (f) Laser power curves plotted to show an influence of non-uniform dye-laser intensity. Components marked with asterisks are enlarged.

ENTANGLED ^{111}Cd ATOMS FROM $^{111}Cd_2$ MOLECULES

The knowledge on the Cd_2 interatomic potentials (**Fig.2**) is essential with respect to the planned experiment aiming at a verification of Bell's inequality for a pair of neutral ^{111}Cd atoms "born" in a controlled dissociation of $^{111}Cd_2$ molecules in a free-jet expansion beam [17]. The general scheme for the experiment is shown in **Fig. 4**. The experiment will engage four laser beams: 4th harmonic of an alexandrite pulsed ring laser (257.1 nm, 30 MHz) and three 2nd harmonics of a YAG- laser-pumped-dye-lasers: (305 nm, 2 GHz), (326.2 nm, 4 GHz) and (230.7 nm, 4 GHz).

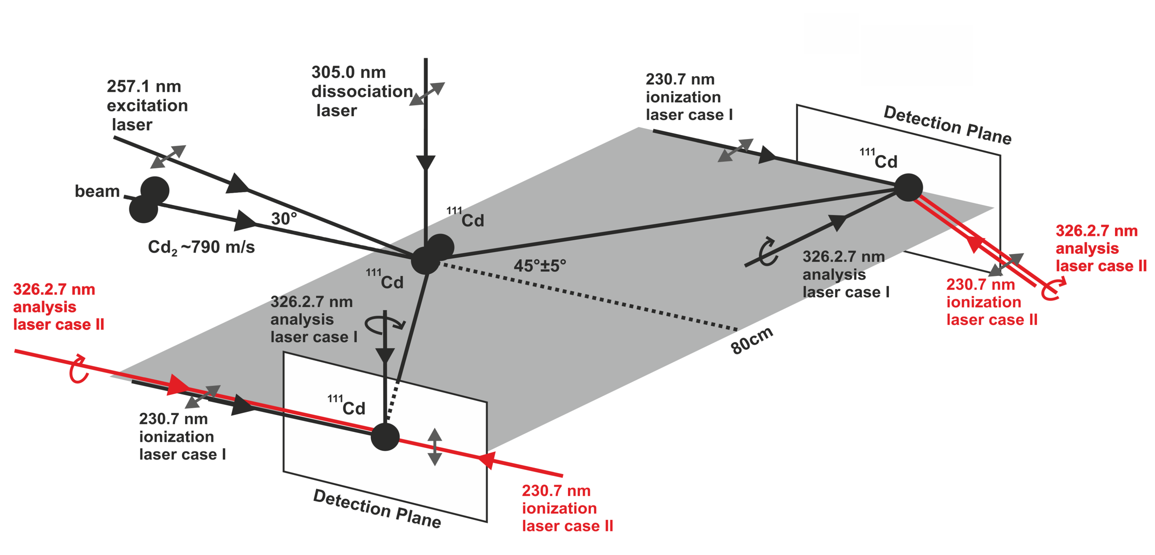


Fig. 4.

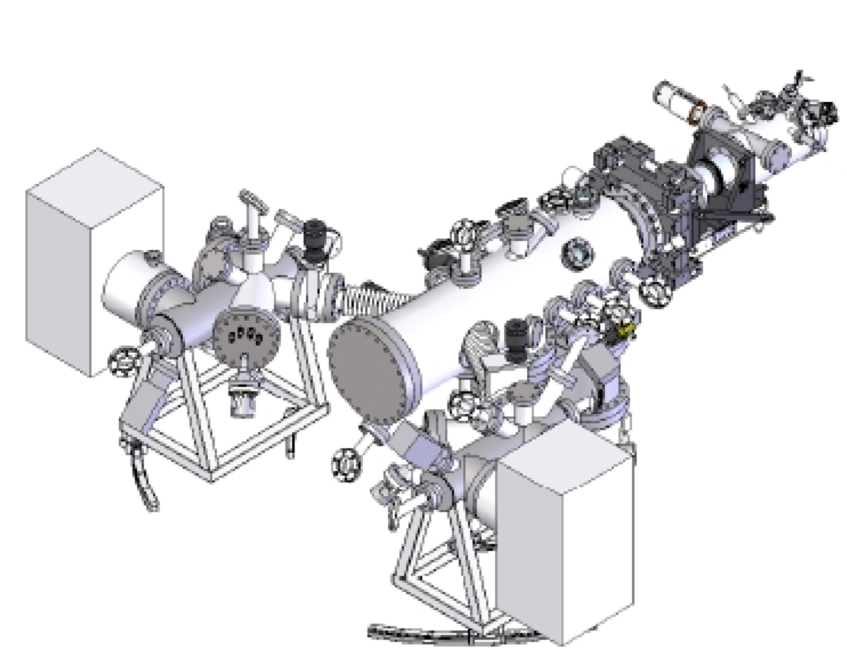


Fig. 5. The experimental apparatus – the main and side vacuum chambers. The latter accommodate detection planes. The apparatus is planned to be used in realization of the scheme shown in **Fig. 4**.

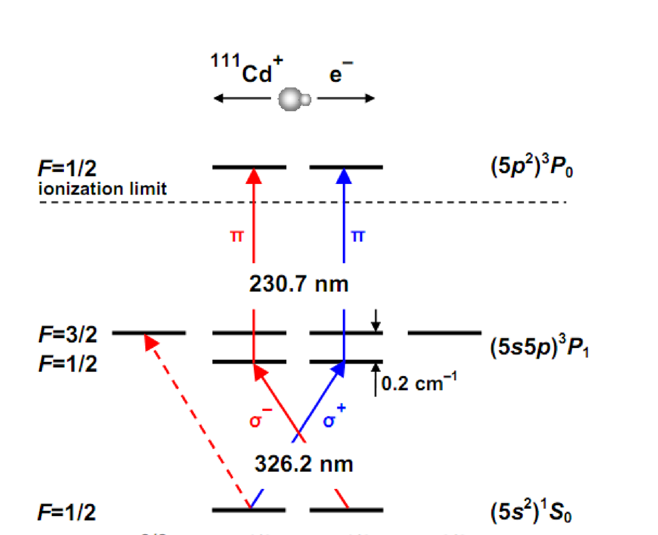


Fig. 6. Spin-state-selective two-photon excitation-ionization (TPEI) scheme allowing detection of the m_F=–1/2 or m_F=+1/2 components of F total atomic angular momentum using two 326.2 nm and 230.7 nm wavelengths. The dashed arrow shows an excitation that is impossible due to the 0.2 cm⁻¹ HFS splitting and sufficiently narrow laser spectral bandwidth.

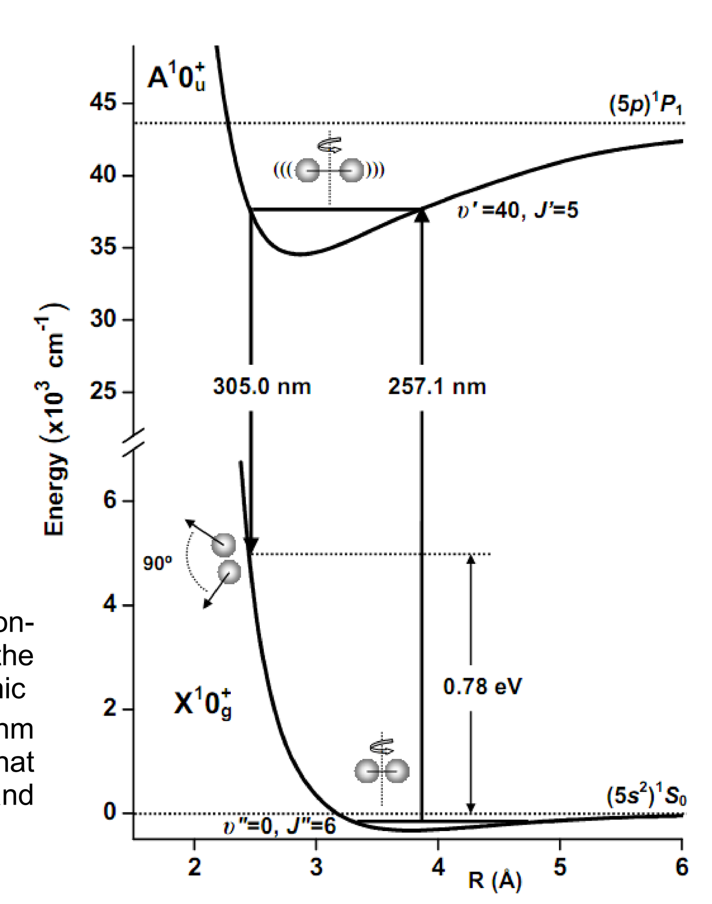


Fig. 7. Creation of a pair of entangled ^{111}Cd atoms. Diagram of the electronic energy states of Cd2 and the relevant stimulated Raman adiabatic passage (STIRAP) scheme between the $A^10_u^+(5^1P_1)$ and $X^10_g^+(5^1S_0)$ states. It consists of the excitation and dissociation of the molecule at 257.1 nm and 305.0 nm, respectively followed by creation of the pair of entangled ^{111}Cd atoms. Diagram of the rotating (υ″=0, J″=6), ro-vibrating (υ′=40, J′=5) and dissociating (with 90º separation angle and 0.78 eV CM kinetic energy) molecules is included.

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