Optica Applicata, Vol. XLII, No. 2, 2012
DOI: 10.5277/oa120220

Entangled cadmium atoms – from the method of production to the test of Bell inequalities

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We present recent progress in the implementation of experimental realization of a loophole-free test of Bell inequalities for entangled ^{111}Cd atoms. The experimental approach is a modified version of the proposal of Fry and co-workers (Phys. Rev. A **52**(6), 1995, p. 4381) for the realization of Bohm's 1/2-spin particle version of the Einstein–Podolsky–Rosen (E-P-R) experiment and is based on the production of entangled atoms by photodissociation of the $^{111}Cd_2$ isotopologue in a supersonic expansion beam using the spectroscopically selective stimulated Raman process.

Keywords: entanglement of atoms, Bell inequalities, cadmium dimers, E-P-R experiment, supersonic beam, van der Waals molecules.

1. Theoretical background

1.1. Quantum entanglement

According to quantum mechanics (QM), an entangled state is a specific correlated state of a system of two or more particles, in which the state of the whole system is better described than the states of particular parts (*e.g.*, [2]). From a mathematical point of view, the entangled state exists when the wave function of the composite system cannot be represented as a tensor product of wave functions of individual components:

$$\Psi_{AB} \neq \Psi_A \otimes \Psi_B \tag{1}$$

In other words, if two objects are in an entangled state, it is impossible to precisely describe the state of one of them without taking into consideration the state of the second object (for simplicity, in this article we assume that an entangled state contains only two parts, but in general the number of parts of the entangled system is unlimited). For instance, let us consider two particles with spin 1/2, which can be oriented up or down along the vertical axis of quantization. An example of the entangled state of these two particles can be one of so-called Bell states, which can be defined in the following way [3]:

$$|\Psi^{+}\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\rangle_A |\downarrow\rangle_B - |\downarrow\rangle_A |\uparrow\rangle_B\right) \quad (2)$$

The Copenhagen interpretation constitutes that QM does not describe the physical reality in an exact, deterministic way but only gives the probabilities of the occurrence of certain phenomena [4], *e.g.*, the probability of the occurrence of particle in the point x_0 is proportional to the square of the wave function of that particle in x_0. According to this interpretation, the act of measurement renders that physical quantity assumes – in a quite random way – one value from the whole spectrum of possible values (so-called collapse of the wave function). In this context, the entangled system has an extraordinary feature that the wave functions of entangled objects are in a specific manner correlated and this correlation exists regardless of the distance between them. It means that although initially the results of measurements are undefined, after the measurement process conducted on one of the two particles, we can strictly determine the result of the measurement on the second particle, theoretically even if the particles are at opposite corners of the Universe (so far, it was experimentally confirmed that the entanglement is sustained on a distance of 144 km [5]). For example, in case of a system described with Eq. (2), the spins have opposite orientations and measuring of the orientation of whichever of them gives us also the result of the measurement of the orientation of the second spin.

1.2. Einstein–Podolsky–Rosen (E-P-R) paradox

One of the most basic laws of QM says that there exist pairs of physical quantities, which cannot be measured simultaneously with unlimited precision [6]. Strictly speaking, it is impossible to measure the values of the observables whose operators $\left(\hat{A}, \hat{B}\right)$ do not fulfil the commutation relation:

$$\left[\hat{A}, \hat{B}\right] \neq 0 \quad (3)$$

The most famous example of these quantities is a pair of quantities: position and momentum (Heisenberg uncertainty principle), but there are also other pairs such as two different components of the spin vector (*e.g.*, s_x and s_y). Basing on the above-mentioned principle and the idea of entangled states, in 1935 Albert Einstein, Boris Podolsky and Nathan Rosen (known collectively as E-P-R) proposed a thought experiment [7] which – in intention of its authors – indicated a contradiction between QM and the special theory of relativity (STR) [8] and revealed deficiency of QM.

STR constitutes that any interaction (or any piece of information) cannot propagate with a speed larger than the speed of light in vacuum and also is strictly connected with the principle of locality, according to which any object can be affected only by its surroundings. E-P-R proposed in their thought experiment to measure two non commuting observables at the same time on two entangled objects separated by a distance larger than the distance which light can travel during the time of the measurement. They reasoned that this experiment leads to a paradox (known as the E-P-R paradox). It means that either the entanglement is propagating with the speed greater than the speed of light in vacuum (it violates the principle of locality) or there is a possibility to measure simultaneously two non commuting observables with unlimited precision (it undermines the QM theory). The authors of the E-P-R paradox concluded that QM does not provide the complete description of physical reality and they believed that there should exist a deeper, deterministic theory (so-called hidden variable theory), which should be able to predict any phenomena described by the QM theory.

1.3. Bell inequalities

During next thirty years there had not been any successful attempts on solving the E-P-R paradox. The situation has changed in 1964 when John Stewart Bell formulated and proved so-called Bell theorem: *No physical theory of local hidden variables can reproduce all of the predictions of quantum mechanics* [9]. Consequently, there were formulated the inequalities [10, 11] which have to be satisfied by any hidden variable theory but are violated by QM. One of the simplest case with respect to the experimental testing of Bell inequalities assumes using two entangled 1/2-spin particles (A and B) prepared in the entangled singlet state (it is so-called Bohm's 1/2-spin particle version of the E-P-R experiment [10]):

$$|\Psi\rangle = \frac{1}{\sqrt{2}}\left\{\begin{pmatrix}1\\0\end{pmatrix}_A\begin{pmatrix}0\\1\end{pmatrix}_B - \begin{pmatrix}0\\1\end{pmatrix}_A\begin{pmatrix}1\\0\end{pmatrix}_B\right\} \tag{4}$$

For 1/2-spin particles the probability of detection of $m_F = +1/2$, where m_F is the component of the spin along the quantization axis, at an angle θ with respect to this axis is described by the operator $\mathcal{M}_+(\theta)$ [1]:

$$\mathcal{M}_+(\theta) = d(-\theta)P_+ d(\theta) \tag{5}$$

where $d(\theta)$ is the rotation matrix

$$d(\theta) = \begin{pmatrix} \cos\left(\frac{\theta}{2}\right) & \sin\left(\frac{\theta}{2}\right) \\ -\sin\left(\frac{\theta}{2}\right) & \cos\left(\frac{\theta}{2}\right) \end{pmatrix} \tag{6a}$$

and P_+ is the projection operator

$$P_+ = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \tag{6b}$$

One can also define the variables $R_{\alpha,\beta}(\theta_1, \theta_2)$ and $R_{j,\alpha}(\theta_j)$ as (compare with [1]):

$$R_{\alpha,\beta}(\theta_1, \theta_2) = \eta^2 f g N \langle \Psi | \mathcal{M}_\alpha(\theta_1) \mathcal{M}_\beta(\theta_2) | \Psi \rangle \tag{7}$$

where $R_{\alpha,\beta}(\theta_1, \theta_2)$ is the coincidence rate for simultaneous detection of the particle with $m_F = \alpha 1/2$ at first detector in the direction of θ_1 and the particle with $m_F = \beta 1/2$ at second detector in the direction of θ_2 (α and β are signs plus or minus), N is the total number of entangled pairs produced per unit of time, η is the detector efficiency, f is the detector acceptance solid angle, g is the conditional probability that if one particle reaches the first detector, the other particle reaches the second detector, and

$$R_{j,\alpha}(\theta_j) = \eta f N \langle \Psi | \mathcal{M}_\alpha(\theta_j) | \Psi \rangle \tag{8}$$

where $j = 1$ or 2 and indicates the detector, α is the sign plus or minus, $R_{j,\alpha}(\theta_j)$ is the rate of detection of the particle with $m_F = \alpha 1/2$ in the direction of θ_j at the detector j. After simple calculations one can find:

$$R_{+,+}(\theta_1, \theta_2) = R_{-,-}(\theta_1, \theta_2) = \frac{1}{4}\eta^2 f g N\left[1 - \cos(\theta_1 - \theta_2)\right] \tag{9}$$

$$R_{+,-}(\theta_1, \theta_2) = R_{-,+}(\theta_1, \theta_2) = \frac{1}{4}\eta^2 f g N\left[1 + \cos(\theta_1 - \theta_2)\right] \tag{10}$$

$$R_{1,+}(\theta_1) = R_{1,-}(\theta_1) = R_{2,+}(\theta_2) = R_{2,-}(\theta_2) = \frac{\eta f N}{2} \tag{11}$$

Thus, for the variables mentioned above, the strong Bell–Clauser–Horne (B-C-H) inequality can be formulated in the following way [1, 10]:

$$S(a, b, a', b') = \frac{R_{+,+}(a, b) - R_{+,+}(a, b') + R_{+,+}(a', b) + R_{+,+}(a', b')}{R_{1,+}(a') + R_{2,+}(b)} \leq 1 \tag{12}$$

The maximum violation of the above B-C-H inequality by the QM prediction ($S_{QM} = 1.207\eta g$) occurs when the differences between directions are equal $(a' - a) = 90°$, $(b' - b) = 90°$ and $(a - b) = 135°$.

2. Experiment

The general scheme of our experimental approach is presented in Fig. 1. In general, the $^{111}Cd_2$ molecules are selectively dissociated and consequently two entangled ^{111}Cd atoms are produced in the process and the atoms are investigated towards the correlation between orientations of their nuclear spins components (see the detailed description below). The visualization and the photograph of the vacuum apparatus are presented in Fig. 2. In our experiment, a selected set-up of angles (a, b, a', b'), which ensures the best violation of Bell inequality, is (135°, 0°, 225°, 90°) – details in Fig. 1.

2.1. Production of pairs of entangled atoms

In our experiment [12], the (^{111}Cd–^{111}Cd) pairs of entangled atoms are produced by laser dissociation of $^{111}Cd_2$ dimers produced in a pulsed supersonic beam. The source of Cd_2 which is used in the experiment is shown in Fig. 3. The laser dissociation of $^{111}Cd_2$ takes place with the help of the stimulated Raman transition from the $X^1 0_g^+(5\,^1S_0)$ ground via the $A^1\,0_u^+(5\,^1P_1)$ excited and back to the repulsive part of the $X^1 0_g^+$ ground electronic state (see Fig. 4). Along with the momentum conservation principle it creates a (^{111}Cd–^{111}Cd) pair of entangled atoms, each in $(5s)^1S_0$ ground state. It is crucial that the ^{111}Cd isotope in its ground state possesses only one non-zero angular momentum, *i.e.*, nuclear spin ($I = 1/2$), whereas the other atomic angular

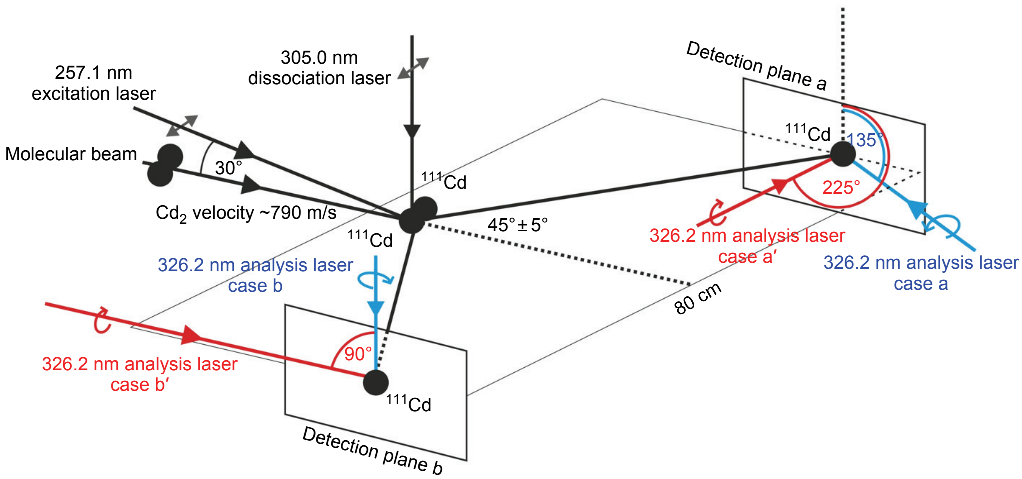

Fig. 1. General scheme of the experiment. The Cd_2 molecule in a supersonic beam is dissociated into two ^{111}Cd atoms (black solid lines). In two parallel detection planes a and b, a spin selective detection of ^{111}Cd atoms is taking place. The detection relies on the two-photon excitation-ionization method (the ionization-laser beams are omitted for simplicity, see Fig. 5 for details). The angles between vertical direction perpendicular to the plane in which the atoms are propagating and analysis beams are selected in the way which ensures the maximum violation of Bell inequality (Eq. (12)).

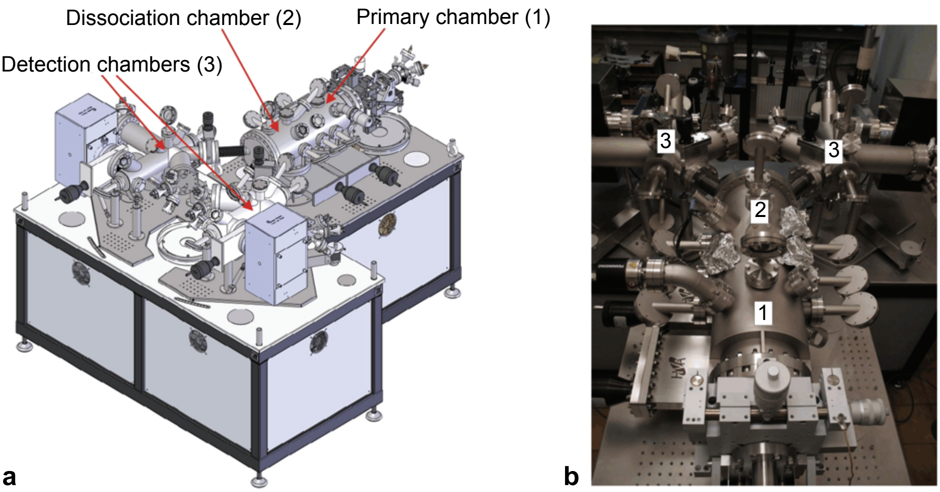

Fig. 2. Vacuum apparatus which will be used in the experiment. Visualization (**a**) and laboratory set-up (**b**) of the apparatus. 1 – Primary chamber in which the supersonic pulsed molecular beam source is located, 2 – dissociation chamber, separated from the primary chamber by a skimer (1 mm in diameter). Both primary and dissociation chambers are pumped down to 10^{-6} mbar of ultimate pressure, 3 – detection chambers in which nuclear spin selective detection of entangled atoms will be performed. Both detection chambers are equipped with ion pumps (ultimate pressure down to 10^{-10} mbar).

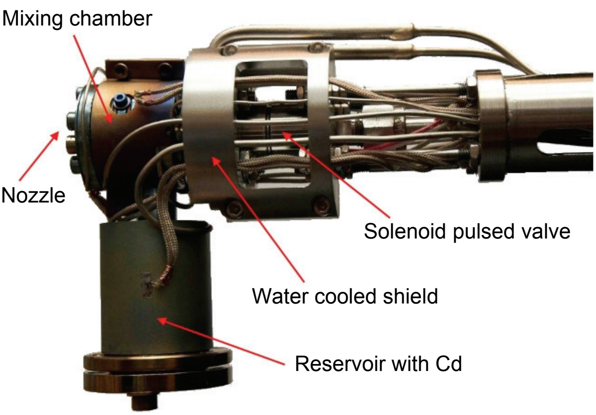

Fig. 3. In the experiment, Cd_2 molecules are produced in a pulsed supersonic expansion beam. Cadmium metal is heated up to 980 K, which is much higher than the cadmium melting point (594 K). Next, cadmium vapours enter the mixing chamber in which they are mixed with carrier gas (noble gas, usually argon). The mixing chamber ends with a nozzle (with orifice of 0.15 mm in diameter) through which the mixture expands to the vacuum chamber forming a supersonic beam. The nozzle is periodically closed and opened with a poppet (made of titanium) with the help of an electrically driven solenoid valve. Adiabatic expansion through a small orifice creates suitable conditions (*i.e.*, internal cooling of vibrational and rotational degrees of freedom) for forming very weakly bound Cd_2 van der Waals molecules.

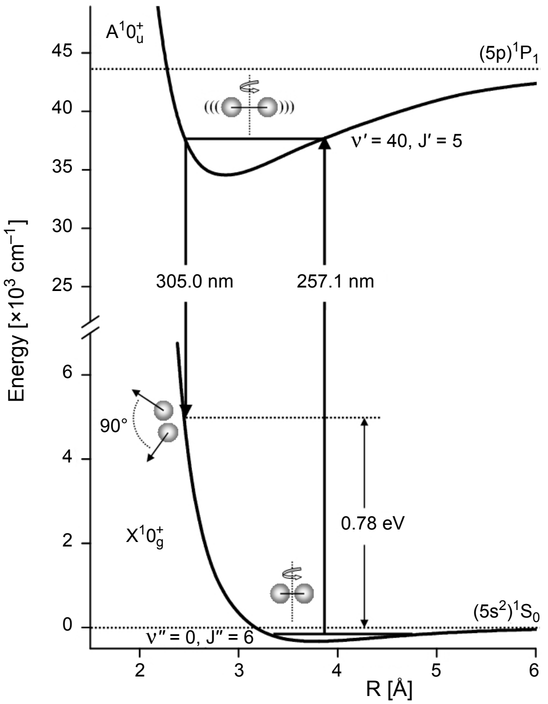

Fig. 4. Scheme of the stimulated Raman transition which is used in the experiment for dissociation of $^{111}Cd_2$ (see text for details).

momenta (*i.e.*, *S* – electronic, *L* – orbital and *J* – total electronic) are zero. Thus, along with the fact that for $^{111}Cd_2$ molecule in the $X^1 0_g^+ ({}^1\Sigma_g^+)$ state all momenta are equal to zero, it leads to the conclusion that the nuclear spins of the two ^{111}Cd atoms (oriented in opposite directions) are entangled. In the process of the stimulated Raman transition it is important to excite the $^{111}Cd_2$ molecule only to one strictly specified J' rotational level in the $A^1 0_u^+$ state. To achieve it, the third harmonic of a pulsed tuneable ring alexandrite laser with 30-MHz spectral bandwidth will be used. It was estimated that the $A^1 0_u^+ (v' = 40, J' = 5) \leftarrow X^1 0_g^+ (v'' = 0, J'' = 6)$ transition, which corresponds to 257.1 nm wavelength, lies in a range of the third harmonic of the alexandrite laser. The dissociating transition (305.0 nm) can be stimulated by the second harmonic of a tuneable Nd:YAG-laser-pumped-dye-laser with 2-GHz spectral bandwidth. It was calculated that the separation angle between the dissociated ^{111}Cd atoms should be equal to $90 \pm 5°$.

2.2. Spin selective detection of atoms

To selectively detect the orientation of a nuclear spin with respect to the quantization axis, we will use the two-photon excitation-ionization method in two parallel detection

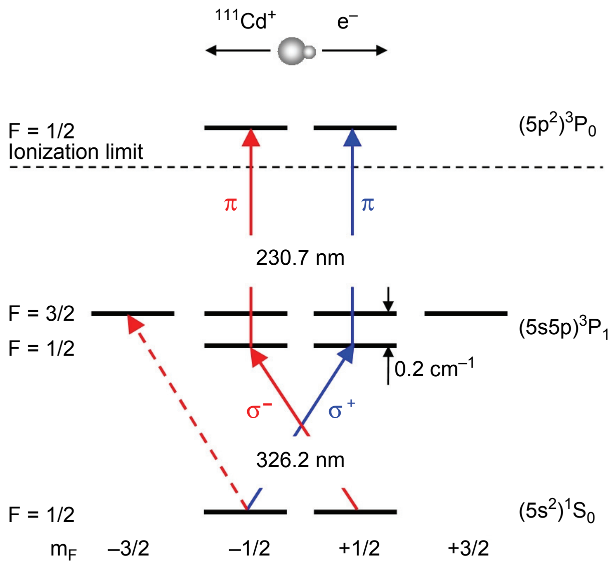

Fig. 5. Scheme of the two-photon excitation-ionization method used for nuclear spin selective detection of ^{111}Cd (see text for details).

planes *a* and *b*. Circularly polarized analysis-laser beam (326.2 nm, second harmonic of the dye laser) is spectrally sufficiently narrow (2 GHz) to selectively excite only one *F* level in the hyperfine structure (HFS) – see Fig. 5. The HFS splitting in the $(5s5p)^3P_1$ excited state of ^{111}Cd is about 6 GHz (0.2 cm^{-1}) [13]. Along with the fact that the only non-zero angular momentum in the $(5s)^1S_0$ state of ^{111}Cd is its nuclear spin, one can anticipate that excitation of the ^{111}Cd atom is possible only if the orientation of the nuclear spin is properly correlated with circular polarization of the analysis-laser beam (σ^+ and σ^- can only excite atoms with $m_F = -1/2$ and $m_F = +1/2$, respectively). The second, ionization-laser beam (230.7 nm, second harmonic of the dye laser) is linearly polarized (π) and ionizes the excited atom to the $(5p^2)^3P_0$ state. After focusing by ion lenses, products of the ionization process (ion and electron) can be detected by channeltrons cooled by liquid nitrogen to minimize dark counts.

3. Conclusions

Recent progress in the implementation of the experimental realization of a loophole--free test of Bell inequalities for entangled ^{111}Cd atoms has been presented. The experimental approach is a modified version of the proposal of [1] for the realization of Bohm's 1/2-spin particle version of the E-P-R experiment. The experiment realized in a newly constructed, dedicated apparatus is based on production of entangled atoms by photodissociation of the $^{111}Cd_2$ isotopologue in a supersonic expansion beam using the spectroscopically selective stimulated Raman process.

The ^{111}Cd atoms are going to be investigated towards the correlation between orientations of their nuclear spins components with the help of the two-photon excitation-ionization method.

Acknowledgements – The research was partially carried out with the equipment purchased thanks to the financial support of the European Regional Development Fund in the framework of the Polish Innovation Economy Operational Program (contract no. POIG.02.01.00-12-023/08 and POIG.02.02.00--00-003/08). The project is financed by the National Science Centre according to the contract no. 2011/01/B/ST2/00495.

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Received March 2, 2012