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The Problem of Enantiomers: Support for a New Interpretation of Quantum Mechanics

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The problem of enantiomers is a typical case of the difficulties which quantum mechanics encounters concerning the interpretation of molecular structure. Classically, each enantiomer has to be considered as a different molecule. The transformation of one form to the other is interpreted as resulting from a more or less fast oscillation between the two forms. Within strict quantum formalism the two enantiomers are described by the same wave-function, corresponding to a rotatory power equal to zero. The oscillation between the two forms is artificially obtained in considering a non-stationary state, different from the ground state. The difference in behavior between amines (non dedoublable) and arsines (dedoublable) is impossible to explain. A solution is proposed within a new interpretation of quantum mechanics, according to which quantum formalism would give the average value of various properties for a sufficiently long time.

INTRODUCTION

Man does not like changing his habits. He prefers the routine which assures him a certain comfort. That is a consequence of the inertia principle which governs the Universe and which appears in all domains, even in that of the sciences where progress and evolution might be thought to be vital. In fact, when one examines history of the sciences, it is clear that a break-through always arises from an isolated man of genius who does not hesitate to break away from the attitude of mind of his epoch and when the confusion created by the new idea has calmed down and the opposition has been overcome, everyone starts work again in the new way, avoiding any question which raises the risk of the new theory being revised. The history of quantum mechanics is typical in this regard. After a first impetuosity (around 1930), this theory has grown and has accumulated so many successes that, at present, it is presented as being *ultima verba* of science. To criticize this theory is to place oneself beyond the pale of scientific world-community. Nevertheless, all the brilliant successes of this theory must not make us forget that a definitive agreement never could be obtained concerning its physical interpretation. Moreover, in many cases quantum mechanics leads us to rather paradoxical conclusions which are difficult to accept and sometimes even to incoherences which are unconsciously (or hypocritically) passed over in silence. One refuses

to take these into account on the pretext that quantum mechanics holds the truth and that so-called difficulties arise in fact from the pride of our mind which would want the microscopic world to be accessible to our senses. Einstein¹ already said that quantum mechanics, in its orthodox interpretation, is a »tranquilliser philosophy«!

The purpose of this paper is not, of course, to contest the power of quantum mechanics and its validity. By means of an example, the one of enantiomers, we will only show the kind of difficulties against which quantum mechanics comes up. We will explore how one can try to solve these difficulties if willing to go out of the official orthodox interpretations, more precisely, if it is admitted that quantum mechanics is incomplete and cannot give all the informations concerning the behavior of particles in atoms and molecules.

THE CLASSICAL POINT OF VIEW

Classically, a molecule is considered as built up upon nuclei linked by electron pairs (Lewis²). A simple mechanical model consists of comparing the electron pairs with coil springs which allow longitudinal and angular vibrations around the equilibrium positions of the nuclei. This model, in spite of its simplicity, permits, on the one hand, explanation of molecular conformations as revealed by experiment (X-rays, electron diffraction, etc.), and, on the other, interpretation of vibration spectra (infra-red and Raman) in a very satisfying manner.

The oscillations of nuclei around their equilibrium positions being generally of small amplitude, the topology of the molecule is not affected by the vibrations, so that the latter can be neglected in defining the chemical species under consideration. Moreover, it is well known that, following the works of Pasteur and those of Le Bel and Van't Hoff on crystals and molecules, if a molecular structure — defined as a set of points at rest (the nuclei) — cannot be superposed on its image, it exhibits a rotatory power. Two structures symmetrical with respect to a mirror and not superposable on their image possess opposite rotatory powers. They are called *enantiomers*. Two enantiomers (A^+ and A^-) exhibit the same scalar properties (density, melt-point, frequencies and modes of vibration, etc.). Nevertheless, these structures have to be considered as two different molecules, each playing the role of an independent component in the phase rule (existence of an eutectic A^+A^-). Two enantiomers react similarly on inactive molecules to give compounds which are also enantiomers. On the contrary, with an optically active substance (e. g.: B^+), A^+ and A^- give two different compounds. A^+B^+ and A^-B^+ , called *diastereoisomers*, which have different scalar properties, allowing us to separate them.

The case of asymmetric amines $NR_1R_2R_3$ sets a problem. Although these molecules can be conceived in two enantiomeric forms, it is impossible to separate these forms. No optical activity is observable. As far back as the beginning of the twentieth century this impossibility has been interpreted as arising from a very fast oscillation of the nitrogen nucleus with respect to the average plane of the ligands, the life-time of each isomer being too brief to permit reaction on another active molecule or the observation of any optical activity.

The similar case of arsines $AsR_1R_2R_3$, discovered subsequently, supports this interpretation, but, moreover, emphasizes how difficult it is to find a

satisfactory explanation. These molecules can indeed be separated into two enantiomers.^{3,4} Nevertheless, a given enantiomer cannot be indefinitely stored in its pure form. After a certain number of months it becomes racemized, *i. e.* transformed into an equimolecular mixture of both enantiomers. Phosphines present the same phenomenon, but the racemization is faster and only a few weeks are necessary.

Classically, the passing from one form to the other can be conceived only by an intermediate structure in which the three bonds carried by the apical atom (N, As or P) are coplanar. The ease of transformation of one form into the other depends on the height of the potential barrier which divides the two structures, *i. e.* the difference in energy between the planar structure and the enantiomers in their equilibrium position. The higher the barrier, the greater the life-time of each enantiomer. In the case of molecules with an asymmetric atom (*e. g.*: amino-acids) the situation is analogous, but each enantiomer appears to be infinitely stable. All the intermediate cases from amino-acids to amines can occur, so we must conclude that racemization is only a matter of time. In support of this conclusion, we quote the racemization observed for collagen molecules in fossil bones⁵, and for amino-acids in meteorites⁶. However, in the bones this racemization can arise from chemical reactions (F⁻ and OH⁻ ions in soil). In fact, the situation is much more complex. Even in amines where the lowest barrier occurs the corresponding energy is greater than the thermal energy at the ambient temperature, so that the jump from one form to the other appears incomprehensible within the classical interpretation.

THE QUANTAL POINT OF VIEW

The problem of enantiomers in quantum mechanics—raised by Hund⁷ as far back as 1927 — is, in fact, a particular case of the more general problem of the molecular structure. The many works and controversies concerning this question have not exhausted the subject (See the review articles 8—10 *f. ex.*). Usually, the problem of a molecule is treated within the framework of the Born-Oppenheimer approximation which assumes an adiabatic electron-nuclear separation in the total wave-function.

$$\Omega(e, n) = \psi_v(n - n_0) \psi_e(e, n_0) \quad (1)$$

where $\psi_e(e, n_0)$ is the electronic wave-function corresponding to the equilibrium position of the nuclei, n_0 (assumed to be unique), and $\psi_v(n - n_0)$, the vibration function, depending only on the nuclear coordinates, n .

In fact, the correct treatment consists of using the complete Schrödinger equation, *i. e.* including electrons and nuclei as well. The *Generator Coordinate Method*¹¹ is such an attempt. It permits — at least theoretically — the calculation of the eigenvalues and the eigenfunctions of the complete Hamiltonian without assumption of adiabatic electron-nuclear separation. Whatever the procedure may be, if electrons and nuclei are taken into account the symmetry of the Hamiltonian with respect to the coordinates of the particles leads to stationary states (*Complete Molecular Eigenstates*¹²) which exhibit spherical symmetry. This precludes all idea of chemical structure and, consequently, the existence of any optical activity. The Woolley suggestion¹³, according to which the appearance of molecular structure would be essentially due to the effect

of the molecular environment, is a possible mathematical solution of the problem. Nevertheless, such a view seems difficult to be admitted when we consider larger and larger molecular systems, arriving finally at crystals and macroscopic objects.

Given that our purpose is not to study the concept of the molecular structure itself, we can, *formally*, reduce the problem of enantiomers to that of a particle in a symmetric double-well potential. We will neglect the weak neutral current perturbations which, owing to the parity non-conservation, provoke a very small asymmetry for the double well¹⁴ ($\Delta E \sim 10^{-20}$ a. u. for alanine¹⁵).

Consequently, we have to solve the time-independent Schrödinger equation:

$$\hat{H} \psi = E \psi \quad (2)$$

where $\hat{H} = \hat{T} + \hat{V}$, with $V(-x) = V(x)$.

In the general case it is impossible to integrate this equation. Nevertheless, for a qualitative discussion the exact form of V is not very important, so that we can use approximate potentials. Practically, one often uses piecewise continuous potentials, or continuous with discontinuous first derivatives. Let us quote for instance:

$$\begin{cases} V = \infty & \text{if } x < -a \text{ and } x > a \\ V = V_0 & -b < x < b \\ V = 0 & -a < x < -b \text{ and } b < x < a \end{cases} \quad (3-a)$$

or

$$\begin{cases} V = \frac{1}{2} k (x + a)^2 & (x < 0) \\ V = \frac{1}{2} k (x - a)^2 & (x > 0) \end{cases} \quad (3-b)$$

In the first case the solution is obtained by joining the solutions corresponding to the various domains¹⁶. In the second case one builds up the solution as linear combinations of eigenfunctions of the two harmonic oscillators respectively centred at $+a$ and $-a$. These functions are twofold degenerate. The diagonalization of the energy-matrix corresponding to the actual potential splits up the degenerate levels. One obtains pairs of simple levels, alternatively symmetrical and antisymmetrical. Practically, in order to obtain the first two levels, one contents oneself with the following combinations:

$$\begin{cases} \psi_S \sim (\phi_1 + \phi_2) \\ \psi_A \sim (\phi_1 - \phi_2) \end{cases} \quad (4)$$

ϕ_1 and ϕ_2 corresponding respectively to the ground state of each oscillator.

Among the continuous potentials the best known is the one proposed by Manning¹⁷:

$$V = -A \operatorname{Ch}^{-2} \left(\frac{x}{a} \right) + B \operatorname{Ch}^{-4} \left(\frac{x}{a} \right) \quad (5)$$

The solution can be expressed by means of the roots of a continued fraction.

Let us also point out the potential

$$V = \frac{k}{2} \left[\frac{(x+a)^2}{1 + e^{4aax}} + \frac{(x-a)^2}{1 + e^{-4aax}} \right] \quad (6-a)$$

to which corresponds, for the ground state, the following function:

$$\psi(x) \sim e^{-\alpha(x-a)^2} + e^{-\alpha(x+a)^2} \quad (6-b)$$

i. e. the sum of two Gaussian functions.

In any case, whatever the used potential may be, the qualitative results are the same, namely:

- (i) the wave-function corresponding to the ground state is x -symmetrical, consequently the rotatory power is equal to zero,
- (ii) one obtains a spectrum of frequencies connected to the motion of the nuclei,
- (iii) the difference in energy between the first levels, $E_A - E_S$, is directly connected with the height of the potential barrier.

Consequently, on the one hand, the quantal result concerning the optical activity disagrees with experiment, and, on the other, no information is obtained concerning the actual dynamics of the system.

In order to attempt to find again the classical concept of oscillation of the apical nucleus, one reasons as follows: One starts from a distribution strongly concentrated above a well, ϕ_1^2 for instance. Then, using the time-dependent Schrödinger equation, one follows the evolution of the density as a function of time. This density presents a maximum which oscillates between the two wells. Practically, introducing only the first states ψ_S and ψ_A Eq. (4), the wave-function is as follows:

$$\Psi(x, t) = C_S \psi_S(x) \exp(-iE_S t/\hbar) + C_A \psi_A(x) \exp(-iE_A t/\hbar) \quad (7)$$

The coefficients C_S and C_A being practically equal ($E_A - E_S$ is very small), the density is equal to:

$$|\Psi|^2 \sim \psi_S^2 + \psi_A^2 + \psi_S \psi_A \cos[(E_A - E_S) t/\hbar] \quad (8)$$

The most remarkable result is that the oscillation period is precisely equal to $\hbar/(E_A - E_S)$, *i. e.* corresponds to the $\psi_S \rightarrow \psi_A$ transition.

These results are generally considered as justifying the classical concept of oscillation of the apical nucleus between both wells. In connection with this interpretation, it is funny to remark that quantum mechanics considers the planar rigid rotator as being at rest in its ground state, though a wave-packet initially centered at a given point is periodically deformed!¹⁰ In any case, this way of presenting the situation is very questionable. In fact, the actual problem is the one of the ground state of the system. This state, of course, is a stationary state. At a given point its density is constant. It presents no oscillation from one well to the other, even if the nuclei oscillate effectively. A density concentrated above a well does not correspond to such state. The study of its evolution with time is a completely different problem. Moreover, during the oscillations of the density $|\Psi|^2$ the average energy $\langle \Psi \hat{H} \Psi \rangle$ fluctuates, remaining greater than that corresponding to the ground state. In any case, the interpretation of the density variation as a function of time at a given point is not obvious. Eyring *et al.*¹⁸ put the reader on his guard about a formally analogous case, that of the H_2^+ ion. After they had built up a spatio-temporal function from both bonding and antibonding molecular orbitals,

by means of a calculation analogous to the one performed here, these authors showed that the electron density oscillates between the nuclei. From this they concluded that the electron oscillates ceaselessly from one nucleus to the other. But they specified that *this point of view should not be taken too literally!* Quantum mechanics does not give instantaneous information concerning the motion of particles. The motion of oscillation, as also that of trajectory, does not fall under the category of observables. Moreover, this calculation, which gives one frequency only, disagrees with the fact that for arsines or amino-acids one can observe vibration frequencies around the equilibrium position. Certainly, the motion of the nuclei is much more complex.

Another point to which we will draw attention is the paradoxical situation of quantum mechanics when it refers to two enantiomers and their properties. For the ground state it obtains a unique well-determined wave-function. The fact of interpreting the two maximums of the density as corresponding to two well-defined forms is equivalent to admitting that a unique wave-function describes two distinct molecules! Nevertheless, such a procedure can be justified in so far as the splitting of the first two levels, S and A, is small. It is possible, indeed, to consider that these levels are practically independent. Under this condition we can assign a total wave-function Ω of type (1) to each form, ψ_v being one of both functions ϕ_1 or ϕ_2 . But the total energy is greater than that corresponding to the double-well. As the splitting A—S becomes more important, the procedure constitutes a less and less valid approximation, so that, in the general case, the precise signification of enantiomers does not appear clearly.

In conclusion, we see that quantum mechanics not only does not explain the fundamental difference observed for the behavior of various asymmetric compounds, but that it also cannot find the basic notion of enantiomers again, such as experiment shows us. We are in the presence of a new example, after the one of the chemical bond¹⁹ and the one of molecular shape^{8,10,13}, for which the classical concepts and quantum theory not only do not combine but also seem to be mutually exclusive.

A POSSIBLE SOLUTION

In fact, the whole problem turns around the physical significance which we assign to the quantal formalism. Roughly speaking, two chief interpretations have divided the theoreticians for more than 50 years. Does the wave-function completely describe the behavior of a molecule in term of observables, any classical representation being excluded (orthodox interpretation, *i. e.* Copenhagen interpretation), or is there a set of systems of the same nature (statistical interpretation)?

According to the first interpretation, the optical activity which belongs to the category of observables is meaningful, whereas the notion of enantiomers has no significance. Consequently, quantum mechanics would appear to be incomplete given that, in certain cases, we can isolate and observe enantiomers. The statistical interpretation is not more satisfactory. Obviously, it is a question of a phenomenon which concerns each molecule individually. All the properties which quantum mechanics computes are universally considered as corresponding to one isolated molecule and not to the totality of molecules. A chemical reaction is not the result of a global interaction of collections of

molecules, but rather of reactions between the molecules themselves on random collision. Nevertheless, one might think that the oscillation of the density (Eq. (8)) $|\Psi|^2$ militates in favor of the statistical interpretation owing to the fact that starting from a given enantiomer (D for instance) the oscillation corresponds to a periodic variation of the ratio D/L of the two forms. In fact, it is nothing of the kind! Even in the statistical interpretation, the density corresponding to the same quantities of D and L forms is constant for the ground state without giving precise information about the actual mechanism of the phenomenon.

Since neither the orthodox interpretation, nor the statistical interpretation can give a satisfactory solution to our problem, it is necessary to search for another way. However, this entails going outside the traditional framework of quantum mechanics. According to ideas which we have previously developed²⁰, a possible solution consists in admitting that the stability of a system (atom or molecule) arises from the average equilibrium between the electromagnetic energy radiated by the particles of this system in their motion and the energy arising from the other systems which constitute the universe. Under these conditions quantum mechanics should be a formalism which would permit account to be taken of the effect of the rest of the universe on the system, the latter being fictitiously considered as isolated. The most important consequence of this model is that no property remains constant over the course of time. It must necessarily fluctuate. At first sight, this conclusion appears to be completely inconsistent with quantum mechanics which considers that certain properties such as the energy of an isolated system, are constant. In fact, it is well known²⁰⁻²² that if one does not compute the quadratic dispersion for energy from the operator $(H)^2$ as is usual but directly from the operator associated with the square of energy E^2 , using the basic rule $x \rightarrow x$ and $p_x \rightarrow \frac{\hbar}{i} \frac{\partial}{\partial x}$, the expression of E^2 being completely symmetrized with regard

to x and p_x , one obtains a dispersion different from zero. The sharpness of the spectrum lines should not be considered as representing an objection. The absorption spectrum constitutes the response of the system to an exterior electromagnetic excitation. The example of stochastic electrodynamics²³ is typical concerning this point. Although this theory cannot be considered as an acceptable alternative for quantum mechanics because it disagrees with the latter particularly with respect to the anharmonic oscillator²⁴, it does nevertheless show that even with a continuous distribution of energy it is possible to obtain a discrete spectrum (*e. g.* for the harmonic oscillator).

Moreover, a measurement necessitates a finite duration, τ_m , so that the observed result is the average value of the property under consideration during this time. If the duration of the measurement is sufficiently long, the average becomes stable. In other words the phenomenon presents an ergodic character. The minimal duration τ for the observation which allows us to obtain this stabilization appears as the *ergodicity* time for the system.

Consequently, if $\tau \ll \tau_m$, for the property G we must obtain the same value in all cases, the value given by the quantal formalism, $\langle \psi \hat{G} \psi \rangle$. On the contrary, if $\tau \gg \tau_m$, successive measurements must give different results, with a dispersion around an average value which is precisely the one given by quantum mechanics. In purely electronic systems, owing to the very great

speed of electrons, τ always remains very inferior to τ_m , so that quantum mechanics always gives results consistent with experiment. The situation is different when we consider the total wave-function corresponding to the motion of nuclei and electrons. Practically, before the nuclei move in an appreciable manner, the average values corresponding to the various electron properties are stabilized at the quantal values. In other words, the ergodicity time of nuclei is much longer than that of electrons. Thus, the Born-Oppenheimer approximation acquires a physical significance. The ligands carried by an apical atom or by an asymmetric atom (as in amino-acids) can be of very different natures, so that the ergodicity times corresponding to their motion are very different. The ratio τ/τ_m varies practically from zero to infinity (as in the right- and left-handed quartz system).

Moreover, this model allows us to imagine the dynamics of the system. Let us assume that, at a given time, the molecule is in one of the two configurations. Under the effect of interactions with the other systems it oscillates around its equilibrium position. Its energy fluctuates with the oscillation amplitude. When, during these fluctuations, the energy becomes sufficiently high, the system travels above the potential barrier and then comes down into the other well. During a certain time the molecule oscillates in the well, behaving as a new molecule, until it returns into the first well, going back to the first conformation, and so on. If it were possible to draw the histogram corresponding to the position of the apical nucleus of a well determined molecule over a sufficiently long time, we would obtain the quantal density at the limit.

In fact, experiments necessarily relate to a huge number of molecules. If $\tau \ll \tau_m$, as for amines, this necessity has no effect on the result, each molecule, on average, bringing a zero contribution to the rotatory power. On the contrary, if $\tau \gg \tau_m$, as for arsines, although each molecule oscillates unceasingly from a given form to the other, the phenomenon being random for each molecule, a collection of molecules of the same configuration, D for instance, will be progressively changed into a mixture of forms D and L, until the proportions of these two forms become equal. The infinitely small difference in energy arising from the parity non-conservation is not able to affect the ratio D/L in a measurable manner. This mechanism explains why arsines and amino-acids are chemically dedoublable whereas amines are not. The difference arises from the ergodicity times of the systems which are respectively longer or shorter than the reaction times. On the macroscopic scale racemization is the lot of any enantiomer, even when it is chemically isolated! In any case, the passing from a given form to the other arises from a much more complicated mechanism than the one which the subterfuge constituted by the non-stationary function ψ^j seems to indicate. It must be remarked that quantum mechanics gives a spectrum of frequencies so that the actual motion must be a combination of the corresponding vibration modes.

Moreover, we will note that in our model the passing from one configuration to the other is explained without it being necessary to assume that particles possess the mysterious power to pass through potential barriers superior in energy to that of these particles and supposed to be constant, as quantum mechanics claims (tunnelling effect).

A GENERALIZATION OF THE PROBLEM

The case of enantiomers is an example of the difficulty against which quantum mechanics comes up concerning molecular structure. In fact, it is not an isolated case. The problem of enantiomers is only a particular case of the general problem of isomers. Let us give an example. Naphtalene and azulene $C_{10}H_8$, are classically considered as two perfectly distinct molecules, in energy to that of these particles and supposed to be constant, as quantum each possessing well-defined physico-chemical properties and an unlimited life-time in the isolated state. Nevertheless, these molecules correspond to minimums of the same potential hypersurface (which also possesses other minimums). Naphtalene and azulene, as the molecules corresponding to the other minimums, have to be considered as being described by the same wave-function. Formally, the situation is the same as the one encountered for enantiomers. Nevertheless, in the case of isomers, one admits that the very great asymmetry of the potential hypersurface and the height of the barriers which divide the various forms allow us to assign a vibronic wave-function to the structures corresponding to the various minimums, as for independent systems. Nevertheless, the problem of knowing to what extent this kind of approximation is justified remains. More especially since while for enantiomers the vibration functions ϕ_1 and ϕ_2 corresponding to the ground state of each well (considered as being isolated) are in each well very close to the exact solution corresponding to the double well, for azulene the first level of the well corresponds to a highly excited level of the double well. The solution which we have proposed for enantiomers applies to this case. Nevertheless, in the quasi-totality of the cases the spontaneous passing from one form to the other is not observable on the human scale, so that everything occurs as if the isomers were indefinitely stable. But it is possible that pairs of isomers do exist, for which interconversion is observable after a relatively limited time.

The example of isomers, as that of enantiomers, clearly shows that, finally, the notion of molecular structure is very relative and strongly linked to the duration of the observation. Perhaps we have here the solution of the Woolley paradox according to which all molecules would exhibit spherical symmetry.

CONCLUSION

Any theory possesses two complementary but independent aspects, namely, its mathematical formalism and its physical significance. The formalism is based upon a certain number of postulates and well-defined calculation rules. In contrast, the physical interpretation is much more subjective and many interpretations are often possible. Quantum mechanics, in spite of its power, does not escape this fact. The model we present is essentially based upon a physical idea of a general electromagnetic equilibrium between all the systems constituting the universe. It is, consequently, essentially different from the two grand classical interpretations (the statistical interpretation and that of Copenhagen). It would rather have to be connected with the de Broglie interpretation (exchange with the sub-quantal surroundings)²⁵, although in this model no fluctuation in energy is accepted, at least in the stationary states. The analogy with stochastic electrodynamics can seem to be deeper. In fact, this theory effectively introduces an exchange with an *a priori* given vacuum field, but remains silent concerning the origin of this latter. Boyer²⁶

even considers this question as »a teleological question comparable to inquiring after the origin of matter in the universe«! Besides, its failure concerning the anharmonic oscillator, as mentioned above²⁴, clearly shows that the model is too simplistic, even if it constitutes an interesting approach to the problem.

Our model, of course, must be made more precise, but even now, it seems to be acceptable as a general framework for future investigations. In any case, it must be said in its credit that it drops a hint of the possibility of unifying micro- and macrophysics, thereby resolving the thorny question of the classical limit in quantum mechanics.¹⁰⁻²⁷

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SAZETAK**Problem enantiomera: potpora za novu interpretaciju kvantne mehanike***André Julg*

Kvantna mehanika ima stanovitih teškoća u interpretaciji strukture molekula. Jedan od karakterističnih slučajeva jest problem enantiomerâ. S točke gledišta klasične teorije, svaki od enantiomera treba smatrati posebnom molekulom. Transformacija jednoga oblika u drugi tumači se manje više brzim oscilacijama. U formalizmu kvantne mehanike oba enantiomera opisuju se istom valnom funkcijom s optičkom rotacijom koja je jednaka nuli. Oscilacije između dvaju oblika postižu se umjetnim putem, tj. uvođenjem nestacionarnog stanja. U ovom radu predloženo je rješenje u okviru nove interpretacije kvantne mehanike.