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Proposal of an Einstein-Podolsky-Rosen experiment with molecules

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Abstract. Suppose that a pair of entities which have been generated jointly from a common source are separated by an experimental device so that they cannot interact any more. Then, strictly formulating within the framework of traditional quantum mechanics, the ensemble of all pairs may be represented by *either* a separable *or* a *non*-separable statistical operator. The former stands for the independence of the sub-ensembles whereas the latter admits correlations (EPR correlations) due to the presence of interference or cross terms. The second-order correlation function $\varDelta$, which is also amenable by experiment, adopts different values depending on the choice of the statistical operator. So, by performing an experiment of this kind with molecules, the question could be decided whether EPR correlations appear in the molecular domain too. A detailed elaboration of the following idea is presented: let an achiral precursor molecule M_2 dissociate so that two chiral fragments of different handedness are obtained. After separation R–M (L–M) interacts in a region of space A (B) with a photon I (II) in an already known linear polarization state. The enantiomeric correlation between the fragments is thereby conferred to the photons, and the corresponding changes of the polarization state are detected for different polarizer settings to finally yield $\varDelta$.

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1 Introduction

Since the publication of the study of Einstein, Podolsky, and Rosen ($\equiv$ EPR) [1] on the assumed incompleteness of traditional or pioneer quantum mechanics ($\equiv$ TQM) in 1935 the discussion of the EPR gedankenexperiment did not come to a conclusive result. Two apparently self-evident axioms have been added by EPR to the pure formalism, and a paradox resulted. Up to now the discussion of the consequences has been burdened with an additional problem which emerged from a wide-spread misunderstanding of the first of the axioms, EPR's famous *principle of reality.* At least in their 1935 paper EPR's point of view on the notion of reality was a purely *operationalistic* one where reality appeared as a *consequence* of *predictability.* On the other hand, however, many physicists have equated reality with *existence* in some ontological sense (and still do so) so that the debate on the validity of the principle of reality in TQM got a touch of a question of faith. Depending on the philosophical attitude of the respective physicist numerous attempts to solve the paradox have been made. The first of all dates back to Bohr [2] who refuted the entire EPR argument by pointing out that even this very weak principle of reality is not compatible with his own (and therefore obviously unimpeachable) interpretation of TQM.

The second axiom, the *principle of separability*, is as follows: "If a physical system remains, for a certain time, ... isolated from other systems, then the evolution of its properties during this whole time interval cannot be influenced by operations carried out on other systems" [3]. This axiom can be sacrificed even less light-heartedly because separability is one of the fundamental paradigms of modern science. Only by isolating an entity from the fabric of its interaction with other entities an object of investigation can be created. Separability is the prerequisite of any possibility to perform such an isolation.

It is, however, open to question whether indeed both axioms are necessary to generate a paradox. Without adding any further concepts to TQM, the formalism itself allows for two mutually exclusive possibilities to view the micro-world [4]. Imagine that we separate experimentally a pair of entities which have been generated jointly from a common source, *i.e.*, from a common ancestor. Then, as will be explained later, the ensemble $\{U_i, V_i\}$ of all separated and thereby non-interacting entities U_i and V_i may be represented by *either* a separable (ρ_s) *or* a *non*-separable ($\rho_{\mathrm{n-s}}$) statistical operator depending on the assumed relation between the U-sub-ensemble $\{U_i\}$ and the V-sub-ensemble $\{V_i\}$. ρ_s allows for independent sub-ensembles whereas $\rho_{\mathrm{n-s}}$ admits correlations due to the presence of interference or cross terms. Analyzing a set of suited experiments it can be shown that the second-order

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correlation function $\mathit{\Delta}$ [5–9] adopts different values depending on which ρ has been chosen. Since $\mathit{\Delta}$ is amenable by experiment, a final decision can be made regarding to which statistical operator does predict the experimental result correctly.

Employing ensembles of photon pairs a number of outstanding experiments have been performed (see Refs. [10–12], not to mention the numerous refinements and extensions published since then). Some of these have been severely criticized because of a not well-justified data adjustment technique [13]. However, the recent work of Zeilinger *et al.* [14] has been done without this adjustment, and it confirms without any doubt that these ensembles may *only* be represented by a *non*-separable statistical operator. So, at least in the realm of photons and on the level of pure *description* of phenomena nature behaves non-separable.

The molecules chemists use to explain experimental findings are essentially classical objects due to their internal spatial structure of a certain temporal stability [15]. Nevertheless said objects are micro-entities too (if we leave out of account the case of macro-molecules), and therefore TQM is obviously necessary to make statements about the results of experiments with molecules. But since non-separability is an essential feature of TQM and in fact shows up in photon pairs, the question arises whether also the molecules chemists think of may be correlated in this way. In another paper [16], I have investigated the dissociation reaction $A_2 \rightarrow 2A$, where one internal structural degree of freedom gets lost in favor of two (coupled) translations. In analogy to the formulation of the EPR paradox in the case of two spin-1/2 particles given by d'Espagnat [3] it could be shown that we have the alternative to accept *either* that the two fragments $A^{(1)}$ and $A^{(2)}$ are still correlated even if they do not interact any more *or* to accept the violation of the principle of total energy conservation ($\equiv$ TEC principle). Put it into plain words: we can either sacrifice the principle of separability (in this case we would be obliged automatically to abandon *any* attempt of a *simple* realistic approach to molecules) or admit that (in ultimate conclusion) energy can be created and annihilated arbitrarily. So we are compelled to accept that non-separability even plays a role in the world of our quite classical molecules. This, however, should be detectable by experiment.

2 Non-separability vs. separability

2.1 Basic considerations

Let us perform a gedankenexperiment which dates back to Wheeler [17]: suppose that a coin is sawed up so that one half contains the head of the original coin and the other one the tail. Now each half is put into an envelope. Envelope 1 is sent to an observer named Alice whilst envelope 2 is sent to an other observer named Bob. If Alice opens her envelope she will not only realize which half of the coin she has obtained, but she will also know with *certainty* what Bob will discover if he opens his envelope. Obviously these two processes are correlated. If Alice observes "head" Bob will observe "tail" and *vice versa*. Since Alice and Bob cannot be in possession of the same side of the original coin (unless magic is involved), a combination as $(A_{\mathrm{H}},\ B_{\mathrm{H}})$ will never be found.

Of course this result can also be obtained formally by means of TQM. The ensemble of all coins cut into halves is represented by a statistical operator ρ_{c}, *i.e.*, by a self-adjoint operator with positive spectrum and $\mathrm{Tr}\rho_{\mathrm{c}} = 1$. ρ_{c} acts on the 2×2-dimensional Hilbert space $\mathbf{H} = \mathbf{H}_A \otimes \mathbf{H}_B$. Let $\{|\alpha_i\rangle|\beta_j\rangle\}$ be a basis of $\mathbf{H}$. With P as an operator suited to describe the activities of Alice and Bob the final result of a whole series of single runs is given by

$$O(A, B) = \mathrm{Tr}(\mathsf{P}\rho_{\mathrm{c}}). \tag{1}$$

Let +1 (−1) be the result if "head" ("tail") is observed. Then obviously $O(A, B)$ must be equal to −1. Thereby, and assuming that in a large number of single runs both Alice and Bob will obtain equal shares of "heads" and "tails", ρ_{c} is determined uniquely by

$$\rho_{\mathrm{c}} = 1/2(\mathsf{U}_{\mathrm{hh}} \otimes \mathsf{V}_{\mathrm{tt}} + \mathsf{U}_{\mathrm{tt}} \otimes \mathsf{V}_{\mathrm{hh}}) \tag{2}$$

with $\mathsf{U}_{\mathrm{hh}} = |\alpha_{\mathrm{head}}\rangle\langle\alpha_{\mathrm{head}}|$ and the other operators defined analogously.

> *Definition 1.* A statistical operator ρ on $\mathbf{H} = \mathbf{H}_A \otimes \mathbf{H}_B$, $\mathbf{H}_A \perp \mathbf{H}_B$, is called *separable* iff it can be decomposed into a direct product of two statistical operators ρ_A and ρ_B where ρ_A is confined to $\mathbf{H}_A$ and ρ_B to $\mathbf{H}_B$. Otherwise it is called *non*-separable.

(This definition is a special case of the usual one. For a comparison of the two approaches see Ref. [4].)

It is easy to see that ρ_{c}, which takes care of the correct outcome $O(A, B) = -1$, is non-separable. What does this mean? It does simply mean that the halves are not independent of one another. A half can only be defined with respect to the whole. "Head" makes sense only with respect to a complete coin. So there is no existence of one half independent of the other. Of course they may be sent to the opposite ends of the earth, but nevertheless they remain belonging together. (Note that this experiment serves as an illustration to introduce the notions of separability and non-separability of statistical operators. There is no deeper sense to it.)

2.2 A generalized EPR-type gedankenexperiment

A given quantum entity shall decay at a time t_0 into two parts called U_i and V_i, resp. Suppose that U_i and V_i fly away from one another. An experimental setup shall separate U_i and V_i spatially so that they cannot interact any more by means of any known physical principle. U_i (V_i) shall impinge on an apparatus A (B). A and B are of the same kind. At a time t_1 sufficiently larger than t_0 a property type as spin component or polarization axis, *e.g.*, shall be measured simultaneously on both U_i and V_i. Let us choose this property type E to be dichotomic in such a way that both the property (≡ numerical value of the property type) $E(U_i)$ and $E(V_i)$ shall be equal to ±1 at certain experimental parameters a and b, resp., which are assumed to determine the actual internal structure of the two coupled apparatuses A and B. a and b are, *e.g.*, the unit vectors defined by either the directions of the inhomogeneous magnetic field in two Stern-Gerlach devices or the axes of two polarizers. The outcome of one single run shall then be given by

$$O_i(\mathsf{a},\mathsf{b}) = E(\mathsf{a};U_i)\,E(\mathsf{b};V_i). \tag{3}$$

Suppose further that a sufficient number of single runs has been done. The final result $O(\mathsf{a},\mathsf{b})$ is the mean of all single outcomes.

We associate the ensemble of produced or emitted entity pairs (U_i,V_i) with a statistical operator ρ on a four-dimensional Hilbert space $\mathbf{H}$ which is the direct product of two orthogonal Hilbert spaces $\mathbf{H}_U$ and $\mathbf{H}_V$. $\mathbf{H}_U$ ($\mathbf{H}_V$) is connected to the physical space wherein E is measured on U_i (V_i) using A (B). Let $\{|\alpha_1\rangle,|\alpha_2\rangle\}$ be an orthonormal basis of $\mathbf{H}_U$ and $\{|\beta_1\rangle,|\beta_2\rangle\}$ an orthonormal basis of $\mathbf{H}_V$.

The apparatus to measure E on U_i (V_i) shall be represented by the self-adjoint operator A (B) acting on $\mathbf{H}_U$ ($\mathbf{H}_V$). A and B are determined by the experimental parameters a and b, resp., which are mentioned above. The combination of the two apparatuses to perform simultaneous measurements of E on U_i and V_i yielding $O(\mathsf{a},\mathsf{b})$ has to be represented by the direct product of A and B according to

$$\mathsf{P}(\mathsf{a},\mathsf{b}) = \mathsf{A}\otimes\mathbf{1}_V \times \mathbf{1}_U\otimes\mathsf{B} = \mathsf{A}\otimes\mathsf{B}. \tag{4}$$

What shall be measured? In this place we will *not* refer to a special property type. The only requirements are that

- the single measurements can yield yes-no-decisions, *i.e.*, the eigenvalues of A and B must be equal to ±1, and that
- these decisions depend on rotation.

A is then given in its most simple form by

$$\mathsf{A} = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}. \tag{5}$$

Apparatus B differs from A insofar as $\mathsf{b}\neq\mathsf{a}$. Let a be a principal axis in the laboratory coordinate system. Then B emerges from A by a rotation around the angle χ between the two vectors a and b

$$\Rightarrow \mathsf{B} = \begin{pmatrix}\cos\chi & \sin\chi\\ \sin\chi & -\cos\chi\end{pmatrix}. \tag{6}$$

B must not be diagonal with respect to the basis $\{|\beta_1\rangle,|\beta_2\rangle\}$. Nevertheless the eigenvalues of B are equal to ±1.

In complete analogy we define two further operators, A' and B', where A' represents apparatus A rotated with respect to its first position (determined by a) by an angle ϕ. B' stands for B rotated with respect to a by an angle ψ. Note that A and A' as well as B and B' are in general *non-commuting* operators. The commutators $[\mathsf{A},\mathsf{A}']$ and $[\mathsf{B},\mathsf{B}']$ attain their maximum if $\phi=\pi/2$ and $\psi=\chi+\pi/2$ (χ may be chosen freely).

With the aid of these four operators we can simulate four different experimental arrangements. The corresponding operators of the joint measurements (the coincidence operators) are $\mathsf{P}(\mathsf{a},\mathsf{b})$, $\mathsf{P}(\mathsf{a},\mathsf{b}')$, $\mathsf{P}(\mathsf{a}',\mathsf{b})$, and $\mathsf{P}(\mathsf{a}',\mathsf{b}')$. Each of these experiments, if actually performed, leads to a final result O from which the second-order correlation function Δ can be calculated:

$$\Delta = |O(\mathsf{a},\mathsf{b}) - O(\mathsf{a},\mathsf{b}')| + |O(\mathsf{a}',\mathsf{b}) + O(\mathsf{a}',\mathsf{b}')|. \tag{7}$$

On the other hand we may determine Δ also by means of TQM since there the final outcome of a measurement on an ensemble of entities is given by

$$O(\mathsf{a},\mathsf{b}) = \mathrm{Tr}(\mathsf{P}(\mathsf{a},\mathsf{b})\rho). \tag{8}$$

In this way Δ becomes dependent on the choice of the statistical operator.

It has been shown in reference [4] that the *most general non*-separable statistical operator for the gedankenexperiment considered is

$$\rho_{\mathrm{n-s}} = \sum_{k,l,m,n=1}^{2} c_{klmn}\mathsf{U}_{kl}\otimes\mathsf{V}_{mn} \tag{9}$$

with $c_{klmn}=c^*_{lknm}$, $\mathrm{Tr}\rho_{\mathrm{n-s}}=1$,

$$\mathsf{U}_{kl} = |\alpha_k\rangle\langle\alpha_l|, \tag{10}$$

and V_{mn} defined analogously. Its separable counterpart ρ_{s} is represented by the direct product

$$\rho_{\mathrm{s}} = \rho(U)\otimes\rho(V) \tag{11}$$

where

$$\rho(U) = \sum_{k,l=1}^{2} u_{kl}\mathsf{U}_{kl} \tag{12}$$

and $\rho(V)$ defined analogously. We further have to demand $\mathrm{Tr}\rho(U)=\mathrm{Tr}\rho(V)=1$ so that also $\mathrm{Tr}\rho_{\mathrm{s}}=1$. Based on this preliminary work we are now able to determine Δ in dependence on ρ. Details of the calculation can be found in reference [4]. In the frequently discussed Bohm-Aharonov singlet case, and if the parameters are chosen so that the operators are maximally non-commuting, we obtain $\Delta_{\mathrm{n-s}}\le 2\sqrt{2}$ and $\Delta_{\mathrm{s}}\le\sqrt{2}$.

> *Definition 2.* Two sub-ensembles $\{U_i\}$ and $\{V_i\}$ are said to be *EPR correlated* iff, for a set of four experiments with maximally non-commuting operators, Δ_{exp} can be made equal to $\max(\Delta_{\mathrm{n-s}})$.

As has been mentioned in the introduction, a lot of corresponding experiments have been performed using correlated photon pairs. All Δ_{exp} which have been evaluated so far are very close to $2\sqrt{2}\approx 2.828$ so that we are compelled to conclude that the two photon sub-ensembles employed are EPR correlated. However, what is about larger objects as, *e.g.*, molecules?

3 Non-separability and EPR correlations in the molecular domain

3.1 Summary of previous work [15,16]

Molecules are entities *structured* in an essentially *classical* way (this is the deeper reason why molecules may be denoted as *objects*). "Molecular structure" ($\equiv S_\mathrm{M}$) can be understood as the notion for an arrangement of atomic nuclei in 3-dimensional Euclidic space which is of a certain temporal stability sufficient to obtain data about it. In the framework of TQM, however, molecular structure is *not* an observable property type. There is *no* self-adjoint (and sufficiently time-independent) structure operator S_M, the spectrum of which would contain the different possible molecular structures as eigenvalues. In consequence, S_M had to be introduced to TQM by the so-called Born-Oppenheimer construction [18,19] which makes possible the existence of structure in terms of minima on potential energy surfaces. Its first step, which consists of a series of transformations and neglections, effects the reduction of the full molecular Hamiltonian H^full including all kinds of motions and interactions to an internal Hamiltonian H^int depending on a set of extremely complicated coordinates. In the second step the solution of the corresponding Schrödinger equation is attempted by means of perturbation theory which already presupposes the notion of molecular structure, since otherwise no perturbation could be defined. Furthermore it is important to note that the perturbation expansion

$$\Psi^\mathrm{int} = \Psi^{(0)} + \kappa\Psi^{(1)} + \kappa^2\Psi^{(2)} + \kappa^3\Psi^{(3)} + \cdots \tag{13}$$

with respect to the parameter κ (which depends on the ratio of the electron mass to a mean nuclear mass) *must* be truncated after the κ^2 term. If not, structure in terms of potential energy surfaces gets lost again due to the non-adiabatic coupling of electronic and nuclear motion contained in $\Psi^{(3\cdots)}$. The exact eigenvalue E^int is of course independent of any nuclear coordinates.

From the foregoing it should be clear that molecules attain a zwitterposition in the overlap area of the world of quanta and the world of classical physics. So it is even more challenging to investigate the possibility of non-separability and EPR correlations in the molecular domain.

3.2 Proposal of an EPR-type experiment with molecules

Suppose that a non-chiral molecule M_2 consisting of two identical sub-units M is excited by a laser pulse so that it dissociates at a time t_0 according to $\mathrm{M}_2 \to 2\mathrm{M}$. Suppose further that the emerging fragments are chiral but of opposite handedness. Due to the linear momentum induced by the bond breaking R–M and L–M fly away from one another. An experimental setup shall separate R–M and L–M spatially so that they cannot interact any longer by means of any known physical principle. Now let R–M (L–M) interact at a time t_1 and in a region of space A (B) with a photon I (II) of already known linear polarization (which shall be equal for I and II). This interaction will change the polarization of each photon. Finally photon I (II) will impinge on an apparatus D_A (D_B) suited to detect a change in the orientation of the polarization vector. The readings of D_A and D_B are then combined to yield $O_i(\mathsf{a},\mathsf{b})$, where the vectors a and b give the orientation of the respective detector in relation to the original polarization plane. By N-fold repetition we obtain the final result $O(\mathsf{a},\mathsf{b})$.

What will be the effect of the interaction? The polarization axis of photon I will be rotated from

$$\begin{pmatrix}1\\0\end{pmatrix} \tag{14}$$

in the Hilbert space $\mathbf{H}_\mathrm{I}$ by a M-dependent angle θ to

$$\begin{pmatrix}\cos\theta\\\sin\theta\end{pmatrix}. \tag{15}$$

Analogously the axis of photon II will be rotated starting from the same vector (now being an element of $\mathbf{H}_\mathrm{II}$) by the angle $\theta' = -\theta$ to

$$\begin{pmatrix}\cos\theta\\-\sin\theta\end{pmatrix}. \tag{16}$$

This is due to the fact that, if the fragment interacting with I is right(left)-handed, then the fragment interacting with II must possess the opposite handedness. Recall the frequently discussed Bohm-Aharonov singlet state.

The salient point of the whole argument is as follows: let $\{|\alpha_1\rangle, |\alpha_2\rangle\}$ be an orthonormal basis of $\mathbf{H}_\mathrm{I}$ and $\{|\beta_1\rangle, |\beta_2\rangle\}$ an orthonormal basis of $\mathbf{H}_\mathrm{II}$. Then the *original* statistical operator of the ensemble of photon pairs is of simple product form (see expression (14)):

$$\rho_\mathrm{before} = |\alpha_1\rangle\langle\alpha_1| \otimes |\beta_1\rangle\langle\beta_1|. \tag{17}$$

Comparing this experiment with the example of coin halves discussed in Section 2.1 it becomes evident that the statistical operator of the ensemble of fragments must be non-separable. It is formally equivalent to ρ_c given by equation (2). So we may conclude that the operator T describing the interaction between photons and fragments, *i.e.*, the operator taking care of the proper change of the polarization planes (expression (14) → (15) and (16), resp.), is given by

$$\mathsf{T} = 1/2(\mathsf{T}_\mathrm{I}(\theta) \otimes \mathsf{T}_\mathrm{II}(-\theta) + \mathsf{T}_\mathrm{I}(-\theta) \otimes \mathsf{T}_\mathrm{II}(\theta)). \tag{18}$$

In consequence the ensemble of photon pairs *after* the interaction is represented by

$$\begin{aligned}\rho_\mathrm{after} =& \mathsf{T}\rho_\mathrm{before}\mathsf{T}\\ =& 1/2\{(\cos\theta|\alpha_1\rangle + \sin\theta|\alpha_2\rangle)(\cos\theta\langle\alpha_1| + \sin\theta\langle\alpha_2|)\\ &\otimes(\cos\theta|\beta_1\rangle - \sin\theta|\beta_2\rangle)(\cos\theta\langle\beta_1| - \sin\theta\langle\beta_2|)\\ &+(\cos\theta|\alpha_1\rangle - \sin\theta|\alpha_2\rangle)(\cos\theta\langle\alpha_1| - \sin\theta\langle\alpha_2|)\\ &\otimes(\cos\theta|\beta_1\rangle + \sin\theta|\beta_2\rangle)(\cos\theta\langle\beta_1| + \sin\theta\langle\beta_2|)\}.\end{aligned} \tag{19}$$

ρ_{after} is non-separable, *i.e.*, the non-separability of the fragment sub-ensembles has been transferred to the photon sub-ensembles. (Note that, for further considerations, the molecular components must not be taken into account since the fragments act as preparation devices only. Their future fate is irrelevant.) Equation (19) can be simplified so that we finally arrive at

$$\begin{aligned}\rho_{\text{n-s}} \equiv \rho_{\text{after}} &= (\cos^2\theta\,\mathsf{U}_{11} + \sin^2\theta\,\mathsf{U}_{22}) \otimes (\cos^2\theta\,\mathsf{V}_{11} + \sin^2\theta\,\mathsf{V}_{22}) \\ &\quad - \cos^2\theta\sin^2\theta(\mathsf{U}_{12} + \mathsf{U}_{21}) \otimes (\mathsf{V}_{12} + \mathsf{V}_{21}) \qquad (20)\end{aligned}$$

where we have made use of the already introduced operators U_{kl} and V_{mn}.

$\rho_{\text{n-s}}$ has a separable counterpart ρ_{s} which as well (at least in principle) is a possible candidate to represent the ensemble of photon pairs after the interaction has taken place. We obtain it from $\rho_{\text{n-s}}$ if all cross terms in equation (20) are omitted. It is represented by the direct product

$$\rho_{\text{s}} = \rho_{\text{I}} \otimes \rho_{\text{II}} \qquad (21)$$

where

$$\rho_{\text{I}} = \cos^2\theta\,\mathsf{U}_{11} + \sin^2\theta\,\mathsf{U}_{22} \qquad (22)$$

and ρ_{II} defined analogously. Note that, if $\theta = 0$, $\rho_{\text{s}} = \rho_{\text{n-s}} = \rho_{\text{before}}$.

In this way *any* correlation of the fragments R–M and L–M will be transferred automatically to the photon pair so that a subsequent analysis of the photons will give information about the kind of correlation existing between R–M and L–M.

Let us explain this crucial point from another point of view: the ensemble of fragment pairs may be represented by

$$\rho = 1/2(\mathsf{U}_{\text{RR}} \otimes \mathsf{V}_{\text{LL}} + \mathsf{U}_{\text{LL}} \otimes \mathsf{V}_{\text{RR}}) \qquad (23)$$

with $\mathsf{U}_{\text{RR}} = |\text{R}\rangle\langle\text{R}|$, $\mathsf{V}_{\text{LL}} = |\text{L}\rangle\langle\text{L}|$, and the other operators defined analogously. It is a widespread belief that a superposition of L and R is *not* possible. But nevertheless let us assume for the moment that it *is*. What would happen? The superposition would yield additional operators which are not present in the above formula. Instead of ρ, and in the most general case, we would have to make the ansatz

$$\begin{aligned}\rho' = 1/4(&\mathsf{U}_{\text{RR}} + \mathsf{U}_{\text{RL}} + \mathsf{U}_{\text{LR}} + \mathsf{U}_{\text{LL}}) \\ &\otimes (\mathsf{V}_{\text{RR}} + \mathsf{V}_{\text{RL}} + \mathsf{V}_{\text{LR}} + \mathsf{V}_{\text{LL}}), \qquad (24)\end{aligned}$$

and the new interaction operator T' would be given by

$$\begin{aligned}\mathsf{T}' = 1/4(&\mathsf{T}_{\text{I}}(\theta) \otimes \mathsf{T}_{\text{II}}(\theta) + \mathsf{T}_{\text{I}}(\theta) \otimes \mathsf{T}_{\text{II}}(-\theta) \\ &+ \mathsf{T}_{\text{I}}(-\theta) \otimes \mathsf{T}_{\text{II}}(\theta) + \mathsf{T}_{\text{I}}(-\theta) \otimes \mathsf{T}_{\text{II}}(-\theta)) \qquad (25)\end{aligned}$$

(compare to Eq. (18)). (Note that U_{RL} and analogous (cross-term) operators do not contribute to the change of the polarization. $\mathsf{U}_{\text{RL}} + \mathsf{U}_{\text{LR}} = \mathsf{U}_{\text{RL}} + \mathsf{U}_{\text{RL}}^{\dagger} = 2\mathsf{W}$ is a self-adjoint operator, but $\mathsf{W}^2 \neq \mathsf{W}$.)

T and T' describe what will happen to an ensemble of photon pairs. This ensemble is represented by a *separable* statistical operator (ρ_{before}). Applying T to ρ_{before} results in a *non*-separable operator, *i.e.*, after the interaction the two photon sub-ensembles are linked so that they cannot be separated from one another any more. However, the application of T' to ρ_{before} leads to the *separable* operator

$$\begin{aligned}\rho'_{\text{after}} = (&\cos^2\theta\,\mathsf{U}_{11} + \sin^2\theta\,\mathsf{U}_{22}) \\ &\otimes (\cos^2\theta\,\mathsf{V}_{11} + \sin^2\theta\,\mathsf{V}_{22}) = \rho_{\text{s}}, \qquad (26)\end{aligned}$$

because the additional terms in equation (25) cancel exactly all non-diagonal terms of ρ_{after}! Put it into plain words: the “existence” of said superpositions does not touch on the separability of the ensemble of photon pairs. But if said superpositions are *not* present, *then* the photon pair sub-ensembles become interwoven.

The proposal presupposes that the photon pairs are *not* prepared by an EPR-type setup. If they were already EPR correlated *before* interacting with the fragments, the detection of an EPR correlation *after* the interaction would of course prove nothing. The question whether already a single fragment can evoke a rotation of the polarization plane will be addressed in Section 3.3.

In complete analogy to what has been described in Section 2.2 the experiment has to be performed with four different orientations of D_A and D_B. The corresponding operators of the joint measurements are $\mathsf{P}(\mathsf{a},\mathsf{b})$, $\mathsf{P}(\mathsf{a},\mathsf{b}')$, $\mathsf{P}(\mathsf{a}',\mathsf{b})$, and $\mathsf{P}(\mathsf{a}',\mathsf{b}')$ which already have been defined. With $\rho_{\text{n-s}}$ we obtain after a lengthy but straight-forward calculation

$$\begin{aligned}O_{\text{n-s}}(\mathsf{a},\mathsf{b}) &= \cos\chi(\cos^2\theta - \sin^2\theta)^2, \\ O_{\text{n-s}}(\mathsf{a},\mathsf{b}') &= \cos\psi(\cos^2\theta - \sin^2\theta)^2, \\ O_{\text{n-s}}(\mathsf{a}',\mathsf{b}) &= \cos\phi\cos\chi(\cos^2\theta - \sin^2\theta)^2 \\ &\quad - 4\sin\phi\sin\chi\cos^2\theta\sin^2\theta, \\ O_{\text{n-s}}(\mathsf{a}',\mathsf{b}') &= \cos\phi\cos\psi(\cos^2\theta - \sin^2\theta)^2 \\ &\quad - 4\sin\phi\sin\psi\cos^2\theta\sin^2\theta. \qquad (27)\end{aligned}$$

Inserting these equations into equation (7) yields

$$\begin{aligned}\varDelta_{\text{n-s}} &= |(\cos\chi - \cos\psi)(\cos^2\theta - \sin^2\theta)^2| \\ &\quad + |\cos\phi(\cos\chi + \cos\psi)(\cos^2\theta - \sin^2\theta)^2 \\ &\quad - 4\sin\phi(\sin\chi + \sin\psi)\cos^2\theta\sin^2\theta|. \qquad (28)\end{aligned}$$

By numerical evaluation of equation (28) it can be shown that $\max(\varDelta_{\text{n-s}}) = 2$. This value is reached for $\theta = 0°$, $45°$, or $90°$, and properly chosen (ϕ,χ,ψ). If, for each θ given, the triple of the other angles is adjusted to maximize $\varDelta_{\text{n-s}}$, we obtain $\max(\varDelta_{\text{n-s}})$ as a function of θ. For θ varying from $0°$ to $90°$ the range of $\max(\varDelta_{\text{n-s}})(\theta)$ is the interval $[\sqrt{2}, 2]$ where the lower bound is attained if $\theta = 22.5°$ and $67.5°$, resp.

If the angles ϕ and ψ are chosen to maximize the commutators, equation (28) reduces to

$$\Delta_{\mathrm{n-s}}(\mathrm{n{-}c}) = |(\cos\chi + \sin\chi)(\cos^2\theta - \sin^2\theta)^2| + |4(\cos\chi + \sin\chi)\cos^2\theta\sin^2\theta|. \quad (29)$$

Both θ-dependent terms are positive so that this formula may be rewritten according to

$$\Delta_{\mathrm{n-s}}(\mathrm{n{-}c}) = |\cos\chi + \sin\chi|\{(\cos^2\theta - \sin^2\theta)^2 + 4\cos^2\theta\sin^2\theta\}. \quad (30)$$

It is easy to see that the expression in braces is equal to 1 for *all* θ. So we finally obtain

$$\Delta_{\mathrm{n-s}}(\mathrm{n{-}c}) = |\cos\chi + \sin\chi|, \quad (31)$$

which is quite remarkable because now the correlation function is independent of θ. Equation (31) implies $\max(\Delta_{\mathrm{n-s}}(\mathrm{n{-}c})) = \sqrt{2}$.

We may, on the other hand, employ the *separable* statistical operator ρ_{s} given by equations (21, 22) instead of $\rho_{\mathrm{n-s}}$. Proceeding in the same way as before, Δ_{s} turns out to be

$$\Delta_{\mathrm{s}} = |(\cos\chi - \cos\psi)(\cos^2\theta - \sin^2\theta)^2| + |\cos\phi(\cos\chi + \cos\psi)(\cos^2\theta - \sin^2\theta)^2|. \quad (32)$$

Also $\max(\Delta_{\mathrm{s}}) = 2$, but the corresponding function $\max(\Delta_{\mathrm{s}}(\theta))$ differs *significantly* from its $\Delta_{\mathrm{n-s}}$ analogue: the $\max(\Delta_{\mathrm{s}}(\theta))$ values decrease monotonically from 2 at $\theta = 0°$ to 0 at $\theta = 45°$ and then increase again to reach 2 at $\theta = 90°$. For $\theta \in [5.0°, 85.0°]$ $\Delta_{\mathrm{n-s}} - \Delta_{\mathrm{s}} > 0.04$ (see the discussion in Sect. 3.3), and the difference attains its maximum ($= \sqrt{2}$) at $\theta = 45°$.

Choosing again ϕ and ψ so that $[\mathsf{A}, \mathsf{A}']$ and $[\mathsf{B}, \mathsf{B}']$ are maximized we obtain

$$\Delta_{\mathrm{s}}(\mathrm{n{-}c}) = |(\cos\chi + \sin\chi)(\cos^2\theta - \sin^2\theta)^2|. \quad (33)$$

Obviously also $\max(\Delta_{\mathrm{s}}(\mathrm{n{-}c})) = \sqrt{2}$.

What are the consequences regarding the evaluation of the experiment? A comparison of the *maxima* of the two correlation functions is irrelevant. Instead it is important to realize that for *most* θ a set of angles (ϕ, χ, ψ) can be found so that the difference between the two second-order correlation functions becomes large enough to allow for an experimental decision.

It should be mentioned that, in the case of *commuting* operators (*i.e.*, if $\phi = \pi$ and $\psi = \chi + \pi$), $\rho_{\mathrm{n-s}}$ and ρ_{s} yield *identical* results:

$$\Delta_{\mathrm{n-s}}(\mathrm{c}) = \Delta_{\mathrm{s}}(\mathrm{c}) = 2|\cos\chi(\cos^2\theta - \sin^2\theta)^2|. \quad (34)$$

3.3 Discussion

The proposal outlined in the last section raises a couple of questions.

- Why chirality at all? It is well-known that EPR correlations are subject to decoherence [20]. Based on the Bohm-Aharonov version of the EPR gedankenexperiment [21] Ghosh *et al.* have investigated the case of two EPR correlated spin-1/2 particles coupled to an environment consisting of both a bath of harmonic oscillators (which allows for a Caldeira-Leggett type of dissipative interaction) and a fluctuating magnetic field [22]. They could show that decoherence takes place already within a time frame corresponding to the characteristic time scale of the particles so that the chance to observe non-separable behavior is very small. On the contrary, the *polarization* correlation of photons is scarcely influenced by an environment model of the Ghosh-type. A lot of the property types of molecules, however, react very sensitively to external disturbations so that corresponding EPR correlations would rapidly disappear. Fortunately, handedness is a remarkable exception which is best demonstrated by the enormous enantiomeric stability of naturally occurring compounds as L-amino acids and D-sugars. If, *e.g.*, the amino acids would not remain enantiomerically stable during a tremendous amount of different chemical processes and physical influences, evolution could not have proceeded as it did.

One could, however, argue that chirality is not a suited property type to detect non-separable behavior, because it is not possible to prepare a coherent superposition of "left" and "right". This statement should not be considered an irrefutable dogma [32]. But besides this we have shown in the preceding section that just the *non*-presence of operators representing L ± R is responsible for the non-separability of the photon sub-ensembles after the interaction. So we arrive at a fascinating conclusion: superpositions even may *prevent* EPR correlations.

But with respect to the proposed experiment a technical problem could arise: it is well-known that, if during a chemical reaction a radical intermediate is formed, the racemic mixture of the two possible products is obtained if the reactive center of the intermediate may adopt a planar configuration. Therefore, it is clear that only source molecules which form sufficiently stable *non*-planar radicals may be used in the proposed experiment. The substituted disilane $((CH_3)SiClH)_2$ could be a possible candidate and shall serve as an example. Being itself achiral, the two corresponding fragments $(CH_3)SiClH$ behave as mutual mirror-images. The ground state of the methyl chlorosilane radical $(CH_3)SiClH$ has a dihedral angel C–Si–Cl–H of 120.79°. If the radical is forced into a planar arrangement around the Si atom, the energy rises by 72.2 kcal/mole (more information on the calculations is given in the Appendix). The disilane source molecule prefers a trans conformation which is energetically lower than the cis isomer by 2.4 kcal/mole. The dissociation along the Si–Si bond requires an energy input of 78.5 kcal/mole which, however, will be apportioned equitably to the fragments so that the energy excess will hardly suffice to destroy the correlation by racemization.

- The handedness of a given molecule is an *absolute* property. Since the achiral source molecules M_2 are arbitrarily oriented in space before dissociation takes place, it is to be expected that equal amounts of R–M and L–M will reach the volumina A and B so that we would obtain, after a certain time interval, an essentially racemic mixture preventing any possibility to observe an effect on the photon polarization. Therefore, the fragment production rate must be attenuated to a level low enough to allow for the identification of two entities as belonging together in pairs. This, however, rises a question of fundamental importance: does a *single* molecule (fragment) possess a rotatory power?

In dealing with macroscopic amounts of substance optical rotatory power is normally observed as a *bulk* effect. In the case that ω of the incident (monochromatic) light is close to one of the transition frequencies of the molecule, the rotation angle θ is given by

$$\theta \approx \mathrm{const} N l \omega^2/(\omega_{k0}^2 - \omega^2) R_{k0} \tag{35}$$

with N times l being the number of molecules contained in the optical channel of length l (optical path length in dm) and unit cross-section. R_{k0} is the rotation strength of the respective transition $k \leftarrow 0$,

$$R_{k0} = \mathrm{Im}(\mu_{0k} \mathsf{m}_{k0}) \tag{36}$$

and determined by the eigenfunctions of the rovibronic molecular Hamiltonian. With C being the concentration in mole/liter we obtain

$$\theta \approx \mathrm{const}' N_A l C \omega^2/(\omega_{k0}^2 - \omega^2) R_{k0}. \tag{37}$$

This equation seems to suggest that, for a given molecule, θ is proportional to C. In the case of one individual molecule present in the probe volume its concentration will be roughly comparable to $1/N_A$ so that by sure no rotation could be observed and no change of the photon polarization would occur. But equation (37) does not make sense if we deal with single molecules! The essential step in the derivation of this so-called Rosenfeld equation is the determination of the refractive index difference Δn of a circularly birefringent medium. Δn, however, is a pure *bulk* property. *A single molecule does not have a refractive index.* Similarly, a macroscopic body has a temperature, but the molecules it consists of do *not*. In addition it must be emphasized that the corresponding experiments make use of light, not of single photons. The samples are irradiated with light of *considerable* intensity. From this we conclude that the Rosenfeld equation should be applied to macroscopic systems only. Our question, however, remains open.

A vast amount of experiments have been performed with single ions in a Paul cage. They clearly show that a single entity of molecular dimensions exerts a measurable influence on radiation. Moreover, a new kind of EPR-type experiments with *massive* particles has been proposed already a decade ago [27]. The crucial point is that two initially *independent* atoms, both prepared in circular Rydberg states, are *forced into* an EPR correlation by means of a one by one resonant coupling to a high Q microwave superconducting cavity. This coupling process leads to the exchange of a *single* photon between the atoms. The direct result of said indirect interaction is correlation, *i.e.*, a *single* photon exerts a measurable influence on an entity of molecular dimensions. First experiments [28] demonstrate the feasibility of this procedure. Finally it should be emphasized that molecules *have* a structure already by definition. Therefore, certain molecules also *have* a handedness, since handedness is a possible consequence of structure. Recall the nano-scale realization of Pasteur's experiment [33]: the authors succeeded not only to differentiate but even to *separate* enantiomeric aggregates of 10 (!) single molecules of 1-nitronaphthalene by use of scanning-tunneling-microscopy.

Taking all these facts together it is quite plausible to assume that single molecules indeed possess a rotatory power. But what would be the consequence if *not*? In this case ($\theta = 0$) both $\Delta_{\mathrm{n-s}}$ and Δ_{s} reduce to

$$\Delta_{\mathrm{not}} = |\cos\chi - \cos\psi| + |\cos\phi(\cos\chi + \cos\psi)|. \tag{38}$$

Now, if a triple of angles (ϕ, χ, ψ) could be found so that $\Delta_{\mathrm{not}} \approx 0$ but $\Delta_{\mathrm{n-s}}$ and/or $\Delta_{\mathrm{s}} > 0$ (for any $\theta > 0$), *then* the proposed experiment would yield a definitive answer to our question as well, since then any $\Delta_{\mathrm{exp}} > 0$ would automatically prove that the fragments under consideration possess a rotatory power. Let, for example, $\phi = 90°$, $\chi = 89.5°$, and $\psi = 90.5°$. In this case $\Delta_{\mathrm{not}} = 0.017\,453$. It is seen immediately that $(\cos^2\theta - \sin^2\theta)^2$ is always lower than or equal to 1. Therefore Δ_{s} will never exceed the limit of 0.017 453 irrespective of which value θ attains. On the other hand we obtain

$$\begin{aligned}\Delta_{\mathrm{n-s}}(90°, 89.5°, 90.5°) = |1.7453 \times 10^{-2}(\cos^2\theta - \sin^2\theta)^2| \\ + |7.9997 \cos^2\theta \sin^2\theta|\end{aligned} \tag{39}$$

so that *any* effect larger than about 0.018 would prove two statements at a time, namely (i) that the fragments under consideration possess a rotatory power, and (ii) that the corresponding sub-ensembles behave non-separable.

- The benchmark experiment of Zeilinger *et al.* [14] has been performed using silicon avalanche photodiodes with dark count rates (noise) of a few 100 per second. By comparison to the 10.000–15.000 signal counts per second one obtains a signal-to-noise ratio of about 10^2. The final analyzer had a resolution of 75 ps and an accuracy of 0.5 ns. The visibility amounted to 97%. So Zeilinger and coworkers could determine Δ with an accuracy of ± 0.02. Based on this value a difference $\Delta_{\mathrm{n-s}} - \Delta_{\mathrm{s}} \geq 0.04$ in the proposed experiment is necessary to demonstrate that the fragments are EPR correlated indeed. Using the above equation we can estimate that $\Delta_{\mathrm{n-s}}$ must be greater than or at least equal to about 0.058. Therefore the fragments must provide for a rotation angle $\theta > 4.1°$.

There are only a few other proposals to use molecules in EPR experiments. Lo and Shimony [25] suggested to examine the electronic spin correlation of pairs of Na atoms produced by dissociative excitation of Na_2 $({}^1\Sigma_g^+)$ using the induced Raman effect. Years later Fry, Walther, and

Li [26] described a related experiment: by a similar procedure ^{199}Hg atoms are generated from $^{199}\mathrm{Hg}_2$ dimers, and the *nuclear* spin correlation of the atoms is measured. Both proposals may be considered as heavy-body analogue of the original Bohm-Aharonov idea. Therefore they differ basically from the present work. To the knowledge of the author none of them has been realized until now.

4 Summary

Imagine a pair (U_i, V_i) of entities which have been generated jointly from a common source, *i.e.*, from a common ancestor. Imagine further that they will be separated by an experimental device so that they cannot interact any more by means of any known physical principle. Then the ensemble $\{U_i, V_i\}$ of all pairs may be represented by *either* a separable (ρ_s) *or* a *non*-separable ($\rho_{\mathrm{n-s}}$) statistical operator. ρ_s stands for the independence of the sub-ensembles $\{U_i\}$ and $\{V_i\}$ whereas $\rho_{\mathrm{n-s}}$ admits correlations (EPR correlations) due to the presence of so-called interference or cross terms. A property type E shall be measured simultaneously (*i.e.*, within a time frame $\Delta t < \Delta x/c$ with Δx being the distance of the entities) on both U_i and V_i. The numerical values $E(U_i)$ and $E(V_i)$ shall depend on the setting of two parameters a and b which determine the actual internal structure of the coupled measuring apparatuses A and B, resp. Now choose two additional settings, a' and b', so that a series of four different joint measurements each consisting of a number of single runs sufficient to validate the law of large numbers can be performed. The four results yield the second-order correlation function Δ (see Eq. (8)). It has been shown in Section 2.2 that the value of Δ calculated by means of TQM depends strongly on the choice of the statistical operator. In this way it can be decided by experiment which of the two operators correctly represents the behavior of the ensemble. Thereby also the question is answered whether the sub-ensembles behave separable or not.

If it would be possible to design an analogous experiment with molecules, it could be decided whether EPR correlations appear in the molecular domain too. This question is of enormous relevance for our understanding of the notion of molecule, since a positive answer would mean that separated molecules may not behave separately. None would have an “existence” of its own *despite* the fact that molecules are essentially classical entities.

A detailed elaboration of the following idea has been presented: let an achiral precursor molecule M_2 dissociate so that two chiral fragments of different handedness are obtained. After separation R–M (L–M) interacts in a region of space A (B) with a photon I (II) in an already known linear polarization state. The enantiomeric correlation of the fragments is conferred to the photons, and the corresponding changes of the polarization state are detected for different polarizer settings to finally yield Δ. Comparing the effect of ρ_s and $\rho_{\mathrm{n-s}}$, resp., on the second-order correlation function, the expected values of Δ may differ considerably. *If said settings are chosen properly, any measured value of Δ larger than or equal to 0.058 proves that both*

- EPR correlations “exist” in the molecular domain too, and
- single molecular entities possess a rotatory power.

Appendix

The calculations on 1,2-dichloro-1,2-dimethyl-disilane ($\equiv \mathrm{M}_2$) and on the corresponding chloromethylsilyl radical ($\equiv$ M) have been performed using the software package *Gaussian 98* [29]. The equilibrium geometries have been determined on a DFT level employing Becke’s 1988 exchange [30] and Perdew’s 1986 correlation functional [31] together with the 6–31G** standard basis set. The two possible M_2 isomers (trans–Cl–Cl in C_i symmetry and cis–Cl–Cl in C_s) have been optimized independently. Based on this structures refined energy values have been obtained by means of single point coupled cluster calculations with perturbatively included triple excitations from the Hartree-Fock determinant ($\equiv$ CCSD–T). Instead of 6–31G** the cc–pVDZ basis set has been used for Cl, H, and C, whereas the larger cc–pVTZ basis set has been employed for the silicon atoms.

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