

- 14 Chen Z Y, Walder, G J and Castleman, Jr, A W, *J Phys Chem*, 1992, **96**, 9581-9582
- 15 Pilgrim J S and Duncan, M A, *J Am Chem Soc*, 1993, **115**, 6958-6961
- 16 Guo, B C, Kerns, K. P. and Castleman, Jr, A W, *J Am Chem Soc*, 1993, **115**, 7415-7418
- 17 Deng, H T, Guo, B C. and Castleman, Jr, A W, *J Phys Chem*, 1994, **98**, 13373-13378
- 18 Yeh, C S, Afzaal, S, Lee, S A, Bynn, Y G and Freiser, B S, *J Am Chem Soc*, 1994, **116**, 8806-8807
- 19 Lin, Z and Hall, M B, *J Am Chem Soc*, 1992, **114**, 10054-10055
- 20 Lin, Z and Hall, M B, *J Am Chem Soc*, 1993, **115**, 11165-11168
- 21 Rohmer, M-M, de Vaal, P. and Benard, M, *J Am Chem Soc*, 1992, **114**, 9696-9697.
- 22 Ceulemans, A and Fowler, P W, *J Chem Soc Faraday Trans*, 1992, **88**, 2797-2798
- 23 Grimes, R W and Gale, J D, *J Phys Chem*, 1993, **97**, 4616-4620
- 24 Grimes, R W and Gale, J D, *J Chem Soc Chem Commun*, 1992, 1222-1224
- 25 Lou, L, Guo, T, Nordlander, P and Smalley, R E, *J Chem Phys*, 1993, **99**, 5301-5305
- 26 Dance, I, *J Chem Soc Chem Commun*, 1992, 1779-1780.
- 27 Hay, P J, *J Phys Chem*, 1993, **97**, 3081-3083
- 28 Methfessel, M, van Schilfgaarde, M and Scheffler, M, *Phys Rev Lett*, 1993, **70**, 29-32
- 29 Methfessel, M, van Schilfgaarde, M and Scheffler, M, *Phys Rev Lett*, 1993
- 30 Reddy, B V, Khanna, S N and Jena, P, *Science*, 1992, **258**, 1640-1643
- 31 Rohmer, M-M, Benard, M, Henriot, C, Bo, C and Poblet, J-M, *J Chem Soc Chem Commun*, 1993, 1152-1885
- 32 Pauling, L, *Proc Natl Acad Sci USA*, 1992, **89**, 8125.
- 33 Khan, A, *J Phys Chem*, 1993, **97**, 10937-10938
- 34 Bowers, M T, *Acc Chem Res*, 1994, **27**, 324-332.
- 35 Reddy, B V and Khanna, S N, *J Phys Chem*, 1994, **98**, 9446-9449.
- 36 Rubio, A, Alonso, J A and Lopez, J M, *An Fis*, 1993, **89**, 174-179
- 37 Naga Srinivas, G, Srinivas, H and Jemmis, E D, *Proc Indian Acad Sci (Chem Sci)*, 1994, **106**, 169-181
- 38 Cartier, S F, Chen, Z. Y, Walder, G J, Sleppy, G R. and Castleman, Jr, A W., *Science*, 1993, **260**, 195-196

Received 20 January 1995, revised accepted 20 March 1995

RESEARCH ARTICLE

Correlating more than two particles in quantum mechanics

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A few years ago, Greenberger, Horne and Zeilinger showed that quantum nonlocality effects, already well-known for systems of two correlated particles, are more spectacular with three particles or more. For observing these effects, one has to initially put the system in an entangled quantum state that is very specific, and not necessarily easy to produce. We discuss some properties of these states and we propose an experimental scheme which allows to create them by successive applications of the postulate of measurement in quantum mechanics. The procedure should, in principle, allow the creation of the desired correlations in systems containing a relatively large number of particles. We propose to apply it to single paramagnetic ions stored in traps, through the measurement of the rotation of the plane of polarization of a laser beam interacting with the ions (quantized Faraday effect).

ONE of the great contributions of Einstein, with Podolsky and Rosen¹, later completed by the celebrated theo-

rem of Bell², was to show that the most surprising feature of quantum mechanics is not indeterminism, as was initially thought, but nonlocality. While Von Neumann's arguments, attempting to prove that the results of quantum mechanics cannot be reproduced by deterministic theories, have been shown by Bell to be not conclusive³, the Bell theorem provides quantitative and general criteria about locality violation. Now, it is a delicate matter to decide in what sense exactly quantum mechanics is nonlocal, or, more generally, to word a precise definition of what is a local theory⁴; this question, at the border of physics and philosophy, is probably still a matter of debate, and we will not discuss it here. What is clear, anyway, is that we do not have an equivalent of the Bell theorem for indeterminism so that, for the moment at least, believing or not believing in the fundamentally nondeterministic character of microscopic processes is more or less a matter of taste. In fact, it is not impossible that the fundamental theory of microscopic processes will, one day, include a (nonlocal) deterministic mechanism behind what

we now call the orthodox quantum theory of measurement.

At the level of the standard mathematical formalism of quantum mechanics, both nonlocality and indeterminism appear explicitly. The former has the status of a postulate: for the results of measurements, one is allowed only to write probabilities, so that the left-hand side of the equations is automatically a nondeterministic quantity. But nonlocality is also visible when, for systems made of two particles located in remote regions of space, one writes the components of the two-particle states of the type considered by Bell: the description of the system is given by an 'entangled' quantum state (see note 1) covering a large region of space; separate descriptions of the two particles are never equivalent to this overall description. To illustrate this point, we make a brief digression from our main subject (the rest of this paragraph may be skipped): we discuss as an example a model which preserves explicitly the nondeterministic features of quantum mechanics – including the use of state vectors, from which probabilities can be calculated – but which at the same time attempts to give separate quantum descriptions for the two remote particles; the latter point, of course, implies that the rules of standard quantum mechanics are altered at some stage. In other words, what we are now trying to do is to keep the usual quantum prescriptions, but for each particle separately, and see if the model can be made compatible with the predictions of standard quantum mechanics. For this purpose, we can obviously ascribe to each of the particles a (one-particle) state vector (or a density matrix); in order to introduce correlations, we have to assume that the two states vectors in question are randomly correlated. Physically, this could happen, for instance, under the effect of fluctuations of the conditions under which the two particles were initially created into quantum states. In this way, we have a rather natural 'semi-quantum' model where the rules of quantum mechanics, including the postulate of measurement, are preserved separately in each region of space; but the correlations arise from correlations of local state vectors, which themselves originate from local fluctuations of a past event. The beauty and the power of the Bell theorem is that we do not have to develop the model any further in order to examine its compatibility with the quantum predictions: the theorem immediately tells us that, whatever correlations we introduce between the individual state vectors (or density operators), any model of this sort will automatically fail to reproduce the full predictions of quantum mechanics. Only a non-separable description of the two particles in one single, nonlocal, ket is therefore permissible. The example illustrates in an explicit manner why locality is harder to incorporate into quantum mechanics than determinism.

Recently, Greenberger *et al.*⁵ showed that considering systems of more than two correlated particles is not a

trivial generalization of the argument, but leads to conflicts between locality and quantum mechanics that are different and more spectacular. Roughly speaking, while the two-particle systems can lead to violations of locality of the order of 25% and require the evaluation of a combination of suitable correlation rates, 100% violations become possible in theory with three particles (or more), in one single experimental result (see note 2); see, for instance, ref. 6 for a discussion of the subject. The quantum state for which these violations should occur is again an entangled state, similar to that usually considered for two particles, but the fact that it now contains more particles makes it even more complex. This does not weaken the basic argument⁵ since, in principle, there is no reason to believe that not all state vectors of a many-particle system are accessible. In some textbooks on quantum mechanics the fact that all states in the linear space are physical states is even an explicit postulate. It is nevertheless interesting to examine the question of how these initial states can be produced.

This is the object of the present article, which originates from several seminars for nonspecialists that the author gave on various occasions during the last few years. The general ideas are not different from those of Greenberger *et al.*⁵; they will just be introduced in a slightly different way. Our main purpose is to discuss methods by which many-particle entangled states of the required sort could be produced: knowing that the state space of a many-particle system contains curious states with exotic quantum properties, it is natural to examine how to produce at least some of them. Several approaches for constructing interesting entangled states are possible and have already been proposed. For instance, in a recent article Hardy⁷ considers a thought experiment where a specific entangled state is produced when particles cancel each other in one of the four paths available to a system crossing two interferometers; this state has interesting nonlocal properties (see also ref. 8). Another even more recent proposal is that of Cirac and Zoller⁹. Our scheme is different and rests in an essential way on the use of the postulate of wave packet reduction in quantum mechanics; it is recursive in the number of particles that it could correlate. At the end of this article we examine how it could be applied to single ions stored in electromagnetic traps, combined with an optical detection method relying on the observation of the 'quantized Faraday effect'. We are not claiming that the proposal is perfectly practical at this stage; we just try to come a little bit closer to realistic experimental schemes than pure thought experiments.

Nonlocality and entangled states

The purpose of this section is to show why certain entangled quantum states, which we call 'states with

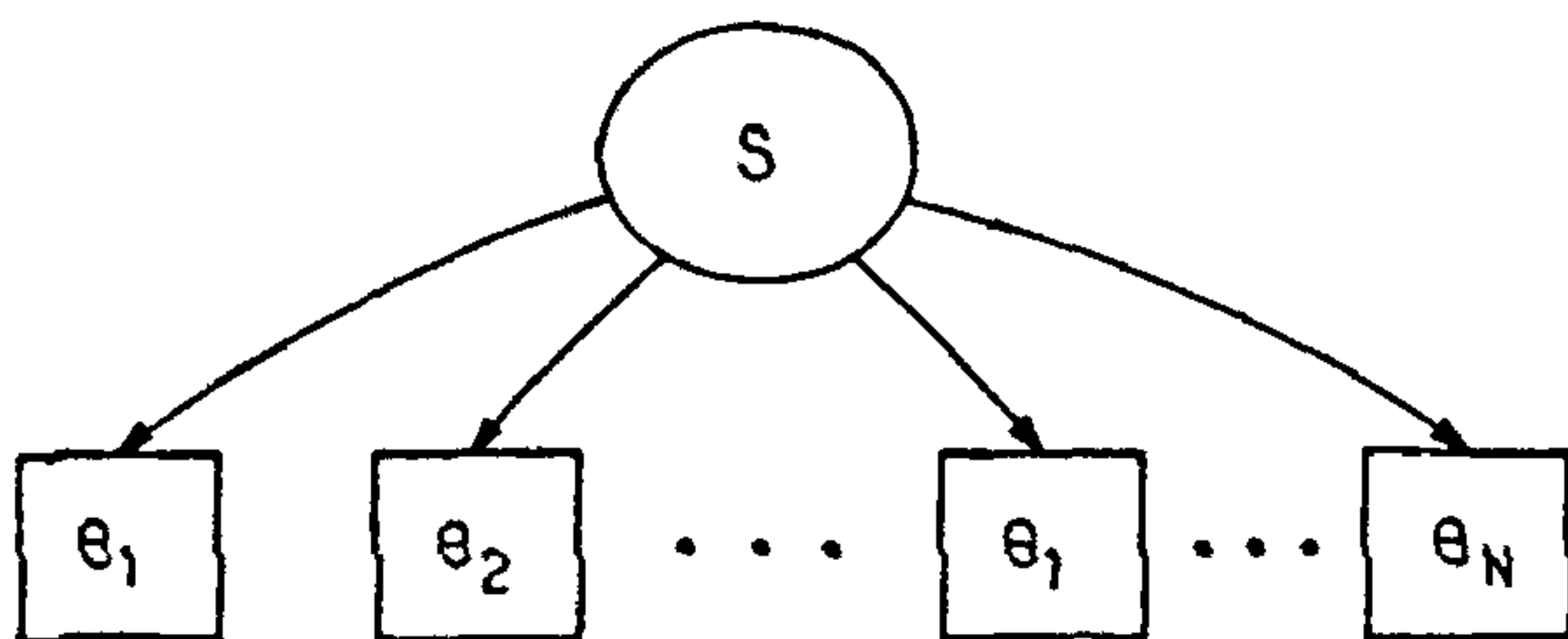


Figure 1. A source S emits groups of N particles, each of which is directed to a region of space $i = 1, 2, 3, \dots, N$ and undergoes a measurement, which depends on the setting θ that is locally chosen by an experimenter

extreme coherence' (they could as well be called the Greenberger–Horne–Zeilinger states), correspond to predictions that strongly violate simple locality concepts. In a first step, it is convenient to forget temporarily about quantum mechanics and to discuss the consequences of locality for systems of several particles with correlations that resemble those contained in the entangled quantum states, leaving the quantum discussion for a second step.

Correlated events in a local theory

We consider the situation schematically shown in Figure 1: in N regions of space, labelled with an index i ($i = 1, 2, \dots, N$), measurements are performed by N experimenters on particles originating simultaneously from a common source S . Each time the source produces a group of N particles, they are distributed among the experimenters, one per region of space, where they undergo a measurement. Actually what we consider is a series of repeated experiments where P successive groups (or bursts) of N particles are emitted by the source and measured, and we label the successive groups by an index K ($K = 1, 2, \dots, P$). The nature of the measurement performed by every experimenter is not fixed: it depends on an apparatus setting which is available on each apparatus and which can be adjusted freely. We symbolize this setting by the letter θ ; if the measurements are, for instance, spin measurements, θ may define the direction along which the experimenter decides to measure a spin component (orientation of the Stern and Gerlach magnet). For the sake of simplicity, in what follows, we call the θ 's 'angles' and treat them as continuous variables. In a series of P measurements, each experimenter chooses P values of θ in succession so that, altogether, $N \times P$ choices are made.

The results and their dependence on the settings. We use the notation A for the results obtained in this experimental procedure: $A_i(K)$ is the result obtained at site i in the run number K . Because the results of an ex-

periment can depend on its nature, the A 's are clearly functions of the θ 's. To proceed in our reasoning, we will need three assumptions:

First assumption. We assume that it makes sense to speak about, in fact to reason mathematically on, the values of the A 's for any possible choice of the θ 's. It is indeed true that, at the end of any given run, every experimenter will have made only one series of choices, so that one could validly argue that the A 's can be defined only for these values of the variable. But, on the other hand, every experimenter acts according to his free will, and we can envisage the different choices that might have been made (or will be made if the experiment is not yet done), and therefore the corresponding results. In other words, whatever 'questions to Nature' will be asked by the experimenters in a given series of measurements, we assume that there is always a well-defined 'reply', and that it makes sense to reason on it (see note 3).

There are actually various ways to justify this first assumption. For instance, if we follow Einstein *et al.*¹, there is no special difficulty here since the A 's are determined by the θ 's and the 'elements of reality' that the particles carry with them; the A 's may actually be identified with these elements of reality, or some function of them. The reader who takes this point of view will probably feel that no special discussion is needed at this stage and should skip this paragraph. Other justifications do not imply at all microscopic properties of systems, but just considerations on the results of measurements and on free will of the experimenters, as sketched at the end of the preceding paragraph. On the other hand, there are also contexts in which the assumption should immediately be rejected: in the orthodox Copenhagen point of view, for instance, saying anything about the (unknown) values of the results for several choices of the θ 's is not allowed.

We touch here the philosophical notion of contrafactuality^{10,11}, a problem which extends beyond pure physics. Instead of giving a long list of references on the discussion of contrafactuality and its philosophical background, we remain in pure physics and just refer the reader to one short article by Peres¹², emphasizing the opposite point of view where 'unperformed experiments have no results'. Indeed, if we may speak only about past experiments or, among future experiments, only of exactly those that will in fact be performed, all the rest of the present article becomes meaningless! We nevertheless assume that this is not the case and continue our reasoning.

Locality. An important issue is then the θ dependence of the A 's. For the group of particles number K , the only values of the settings that matter are the θ 's chosen by the experimenters on that measurement; because the time which separates every experiment may be arbitrar-

ily long, we do not assume any influence of apparatus settings made long ago in the past (or in the future!). So, we can write

$$A_i(\theta) = A_i(\theta_1, \theta_2, \dots, \theta_m, \dots, \theta_N; K) \quad (1)$$

where the θ 's are the settings for the group of particles numbered K ; we do not have to add K indices to these θ values since there is no ambiguity. At this stage, the issue which becomes obvious is locality or nonlocality: if we assume that $A_i(\theta)$ depends only on the local choice made by the experimenter for the apparatus setting, and not on that made by the other experimenters (who can live in different galaxies), we may simplify eq. (1) into

$$A_i(\theta) = A_i(\theta; K). \quad (2)$$

If, on the other hand, we do not assume locality, we must keep the whole θ dependence contained in eq. (1).

In what follows, we discuss the consequences of the local equation (2); assuming the validity of this equation is, therefore, our second assumption.

Correlations. For the sake of simplicity, we limit ourselves to the case where every local measurement can provide only two different results, as would happen with Stern and Gerlach apparatuses; as usual, we take the convention where the two corresponding values are ± 1 . At this stage, we have not yet assumed anything about the A 's, and we need more ingredients to proceed; we have to assume something about the results, at least some kind of correlation. In fact, we assume only one single property, namely that for any combination of the values of the θ 's that has a sum of zero (or any integer multiple of 2π), the product of all the results takes only one of the two possible values ± 1 , $+1$, for instance:

$$A_1(\theta_1; K) \times A_2(\theta_2; K) \times \dots \times A_N(\theta_N; K) = 1 \quad (3)$$

whenever $\theta_1 + \theta_2 + \dots + \theta_N = 0$

(the above relation is valid for any K). The validity of eq. (3) is our third assumption. We note in passing that this equation does not forbid strong fluctuations of every A separately; only the product of all the A 's is fixed provided that the sum of the angles has this particular value. We note that condition (3) is satisfied by the quantum predictions concerning measurements on two spins in a singlet state (see note 4).

Three lemmas. In this section, we first show that all the A 's have the same θ dependence; then that this dependence is necessarily even in θ ; and finally that the θ dependence must disappear altogether.

(i) Assume that for a particular run the experimenters choose a set of angles with a vanishing sum. We then necessarily have

$$A_1(\theta_1; K) \times A_2(\theta_2; K) \times \dots \times A_i(\theta_i; K) \times \dots \times A_N(\theta_N; K) = 1, \quad (4)$$

but also, if the first two angles are interchanged

$$A_1(\theta_2; K) \times A_2(\theta_1; K) \times \dots \times A_i(\theta_i; K) \times \dots \times A_N(\theta_N; K) = 1, \quad (5)$$

so that taking the ratio provides

$$\frac{A_1(\theta_1, K)}{A_1(\theta_2, K)} = \frac{A_2(\theta_1, K)}{A_2(\theta_2, K)}. \quad (6)$$

This relation holds for any value of the angles since it is always possible to find values of $\theta_3, \theta_4, \dots$, for which the sum of all angles is zero (see note 5). Obviously, the same reasoning applies to any other pair of particles than the first two, so that all ratios $A_i(\theta, K)/A_i(\theta', K)$ are necessarily equal for any value of i (but for a given value of K). Taking the value $\theta' = 0$ as a reference, we may set

$$\frac{A_i(\theta, K)}{A_i(0, K)} = X(\theta, K), \quad (7)$$

where $X(\theta, K)$ is a function which describes the common dependence of all the A 's on θ . We then have, for any value of the θ 's,

$$A_1(\theta_1; K) \times A_2(\theta_2; K) \times \dots \times A_N(\theta_N; K) = X(\theta_1, K) \times X(\theta_2, K) \times \dots \times X(\theta_N, K). \quad (8)$$

To write this equation, we have used the fact that, according to eq. (3), the product of all the $A_i(0; K)$'s is always equal to one.

(ii) Assume now that θ_1 and θ_2 have opposite values, all the other θ 's being equal to zero. We then have

$$A_1(\theta; K) \times A_2(-\theta; K) \times A_3(0; K) \times \dots \times A_N(0; K) = 1, \quad (9)$$

which in terms of X 's translates into

$$X(\theta, K) \times X(-\theta, K) [X(0, K)]^{N-2} = 1 \quad (10)$$

for any value of K . Since by their definition [eq. (7)] all X 's are equal to one when $\theta = 0$, the bracket disappears from this equality; moreover, the possible values of the A 's, and therefore of the X 's, being only ± 1 , they are equal to their inverse so that eq. (10) is equivalent to the statement that X is an even function of θ :

$$X(\theta, K) = X(-\theta, K) \quad \text{for any } K. \quad (11)$$

(iii) Let us choose any value of θ for θ_1 , choose $-\theta/2$ for θ_2 , and complete the series of θ 's so that the sum is zero ($\theta_3 + \theta_4 + \dots = -\theta/2$). We then have

$$X(\theta, K) \times X(-\theta/2, K) \times X(\theta_3, K) \times \dots = 1, \quad (12)$$

but also

$$X(\theta/2, K) \times X(0, K) \times X(\theta_1, K) \times \dots = 1, \quad (13)$$

from which we get, by taking the ratio and by using the fact that $X(-\theta/2, K)$ and $X(\theta/2, K)$ are equal,

$$X(\theta, K) = X(0, K) = 1 \text{ for any value of } \theta \text{ and } K. \quad (14)$$

In the end we see that our assumptions lead to a very simple conclusion: the settings used by the experimenters have no influence whatsoever on the results obtained! This may be somewhat unexpected in view of the generality of condition (3), especially of the fact that it relates to a correlation of high order (it says nothing about the correlations observed at one site of measurement, and even nothing about the correlations observed at different sites as long as one of them is ignored). Nevertheless, combined with the condition of locality, condition (3) has dramatic consequences on the θ dependence of the results at each site.

Entangled quantum states with extreme coherence

We now reason in a different context, quantum mechanics, but we will indeed introduce situations where condition (3) is still valid. We consider a system containing N spin-1/2 particles each of which has access to two quantum states, $|+\rangle$ and $|-\rangle$ (orbital variables are ignored). The state vector $|\Psi\rangle$ of the N -particle system belongs to a space of dimension 2^N and it is clear that there is a broad choice on the kind of state we can select as an initial state for a thought experiment. For instance, there are states where all particles are in the same one-particle state $\alpha|+\rangle + \beta|-\rangle$:

$$|\Psi\rangle = [\alpha|1:+\rangle + \beta|1:-\rangle] \otimes [\alpha|2:+\rangle + \beta|2:-\rangle] \otimes \dots, \quad (15)$$

which can be symbolized as

$$|\Psi\rangle = [\alpha|+\rangle + \beta|-\rangle]^N. \quad (16)$$

In this kind of state, every particle is separately in a coherent state; on the other hand, since $|\Psi\rangle$ is an uncorrelated product, there is no overall coherence at the level of many-particles properties. Such states occur frequently in the discussion of coherent spin precession experiments in NMR, or in the theory of Bose-Einstein condensation, where a macroscopic number of particles occupies the same coherent quantum state. When written explicitly on the basis of the kets $|\pm, \pm, \pm, \dots, \pm\rangle$, $|\Psi\rangle$ has in general 2^N components.

Let us now consider the opposite kind of coherence where the number of components of the state is reduced to its minimum: we assume that $|\Psi\rangle$ has only nonzero components on the two 'extreme' states, where all spin components have the value $+$ for the former, or the value $-$ for the latter:

$$|\Psi\rangle = \alpha|+, +, +, \dots, +\rangle + \beta|-, -, -, \dots, -\rangle. \quad (17)$$

This is a coherent superposition between states which are macroscopically different (if N is large): since values for all spin components are different (see note 6), they are actually built to have the maximum possible difference in their single-particle properties. While it was not the case for eq. (16), eq. (17) defines a truly macroscopic quantum state; we call it an 'entangled state with extreme coherence'. We note in passing that, at the level of measurements made on one particle only, the coherence is lost; the density matrix of one particle corresponding to eq. (17) is merely

$$\rho_1 = \begin{pmatrix} |\alpha|^2 & 0 \\ 0 & |\beta|^2 \end{pmatrix}, \quad (18)$$

which describes an incoherent superposition of the two states, $|+\rangle$ and $|-\rangle$. More generally, taking from eq. (17) any partial trace completely destroys the coherence, precisely because all particles change state from one component to the other. So, in this state, quantum coherence can appear only for measurements concerning the ensemble of all N particles.

Correlations between the quantum results. The measurement performed by every experimenter is the component of the spin along a unit vector $u(\theta)$, which we assume to be in the xOy plane:

$$u(\theta) = \cos \theta u_x + \sin \theta u_y, \quad (19)$$

with obvious notation. If S is the spin operator acting in the one-spin space of states (divided by $\hbar/2$ so that its eigenvalues become ± 1), the operator corresponding to the measurement performed in region of space number labelled j is then

$$S_j(\theta) = S_j \cdot u(\theta) = e^{-i\theta} \sigma^+ + e^{i\theta} \sigma^- \quad (20)$$

where, as usual, the σ^\pm operators are defined by $\sigma^\pm|\mp\rangle = |\pm\rangle$. When the experimenters choose the values $\theta_1, \theta_2, \dots, \theta_N$ for the angles, the quantum average (see note 7) of the product of all results is

$$E(\theta_1, \theta_2, \dots, \theta_N) = \langle \Psi | S_1(\theta_1) \times S_2(\theta_2) \times \dots \times S_N(\theta_N) | \Psi \rangle. \quad (21)$$

This expression is easy to calculate when the state vector is given by eq. (17). Let us, for instance, write the result of the action of the operator in the middle of this equation on the component $|-, -, -, \dots, -\rangle$ of $|\Psi\rangle$. Since the effect of σ^- on $|-\rangle$ is to give zero, considering eq. (20) one easily sees that only the term in σ^+ in every $S_j(\theta_j)$ contributes, and one gets

$$S_1(\theta_1) \times S_2(\theta_2) \times \dots \times S_N(\theta_N) |-, -, -, \dots, -\rangle = e^{-i(\theta_1 + \theta_2 + \dots + \theta_N)} |+, +, +, \dots, +\rangle \quad (22)$$

which introduces a first term in $\alpha^* \beta e^{-i(\theta_1 + \theta_2 + \dots + \theta_N)}$ in the average value. The second contribution, corresponding

to the effect of all σ^- operators, is simply the complex conjugate. Therefore,

$$E(\theta_1, \theta_2, \dots, \theta_N) = \alpha^* \beta e^{-i(\theta_1 + \theta_2 + \dots + \theta_N)} + \text{c.c.}, \quad (23)$$

where c.c. is for complex conjugate. In the particular case where

$$\alpha = \beta = \frac{1}{\sqrt{2}}, \quad (24)$$

this results simplifies into

$$E(\theta_1, \theta_2, \dots, \theta_N) = \cos(\theta_1 + \theta_2 + \dots + \theta_N). \quad (25)$$

This shows that, when the initial N -particle state has the form (17), the results of all N measurements are strongly correlated, and that full correlations are obtained each time the sum of all the angles is zero or 180° . We also note that the predictions of quantum mechanics fulfill condition (3).

Conflict with locality. Let us take the simple case where all the angles are equal. Then

$$E(\theta, \theta, \dots, \theta) = E_N(\theta) = \cos(N\theta). \quad (26)$$

If we plot the θ dependence of M in this case, we obtain a sinusoid as shown in Figure 2; the higher the number of particles N , the faster are the oscillations of the curve. They can indeed be called quantum, or non-locality, oscillations, since we have shown that, in any local theory (with, in addition, very general contrafactuality assumptions) the function $E_N(\theta)$ is necessarily a constant, as represented by the broken line of Figure 2. This figure emphasizes, therefore, a particularly strong conflict: for some values of θ the difference is 100% (an opposite sign is obtained), a remarkable fact that was first emphasized by Greenberger *et al.*⁵

Production of entangled states

In this section we propose a scheme which could be used to put a system of two particles or more into a

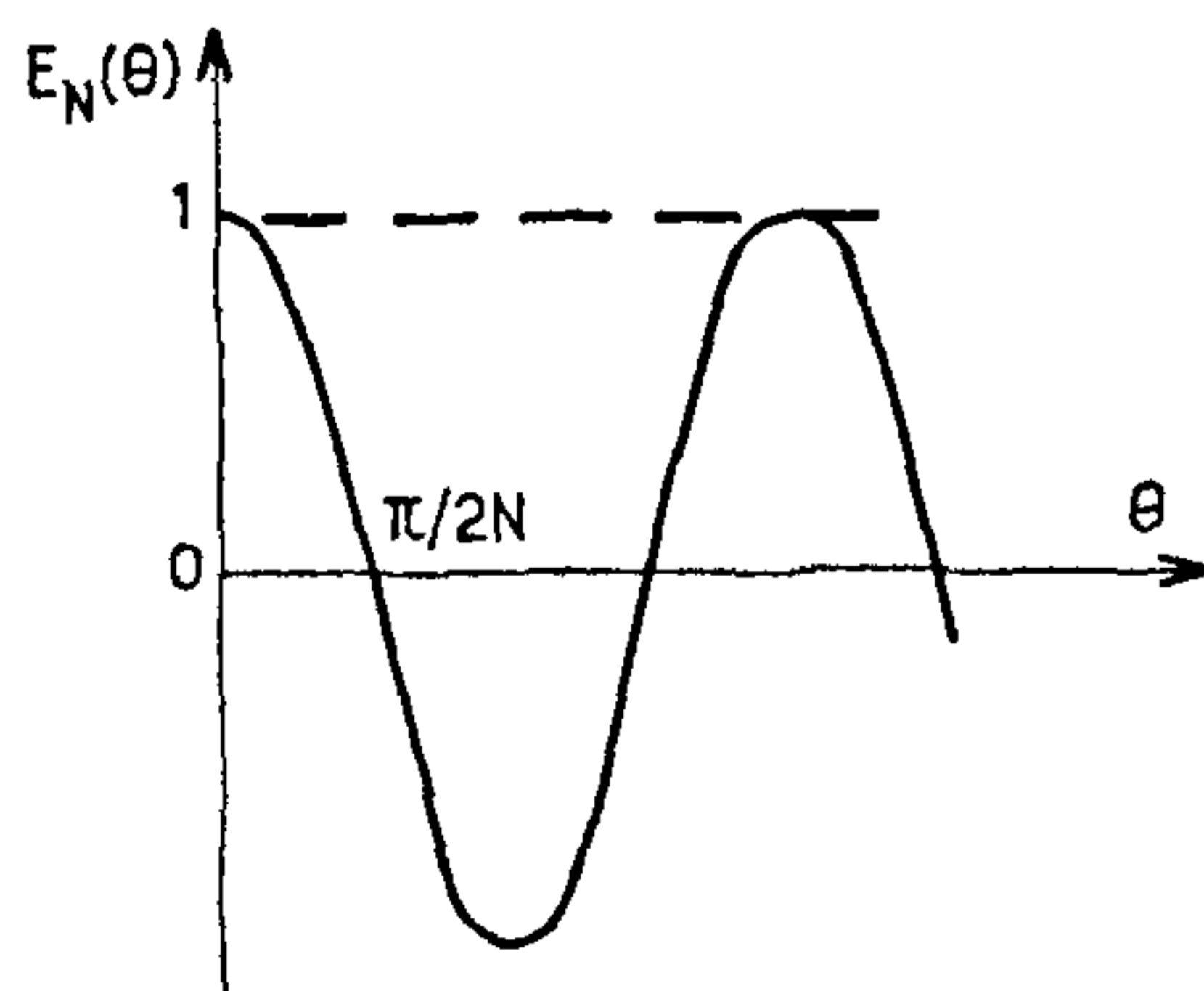


Figure 2. Mean value of the product of all measurements when all settings θ are equal: quantum theory predicts for some initial entangled states an oscillation between $+1$ and -1 , while under the same conditions a local theory allows only the value $+1$.

'state of extreme coherence'. The traditional approach in experiments on locality violation is to make use of particle disintegration, often of atomic cascades, where one system (atom) spontaneously emits a group of correlated particles; in some cases, it turns out that the quantum correlations are precisely those required for locality tests, so that the desired entangled state is produced 'naturally'. Instead of seeking such cases, we use a strategy where we try to create the correlations 'actively' during measurement processes; the choice on the nature of the measurements may introduce more parameters on the production of the initial state. Also, the usual experimental schemes rely on the production use of short-lived particles, photons, for instance, which fly at the velocity of light, so that they are annihilated (either by the detector or by the environment) after a few nanoseconds. Here we will discuss systems which stay almost immobile in the laboratory and with lifetimes at the human scale, minutes or more, making locality violations in a sense more spectacular.

Optical detection of single ions in traps

The trapping and observation of single ions is now a well-known technique^{13,14}; it provides isolated quantum systems which remain stable for long periods. If, moreover, the ion is paramagnetic and has a spin-1/2 in its ground (or metastable) state, it can retain a quantum coherence for a long time (see note 8). The classical technique for detecting the presence of a single ion in a trap is the observation of resonance fluorescence excited by a laser; since an atom or an ion can scatter one photon per lifetime of the excited state, one can obtain photon fluxes that are of the order of 10^8 photons per second – much more than what a human eye can see in the dark. This is an attractive possibility but, for experiments aiming at testing quantum mechanics with single particles, fluorescence detection does not seem too well-suited: every photon absorption, followed by the spontaneous emission of another photon, results in an optical pumping cycle for the ion, and destroys almost completely the ground state coherence¹⁵.

Measuring scattered light is not the only possibility; one can also rely on absorption measurements. Indeed, the absorption of a laser beam produced by a single ion is not negligible when the beam is strongly focused on the ion; one does not need a macroscopic number of ions. The reason is that the resonant photon absorption is of the order of the square of the resonant wavelength (see note 9), while this is precisely the order of magnitude of the minimum area of a 'laser beam waist', a straightforward consequence of the laws of diffraction. Significant changes of the properties of the transmitted laser beam can, therefore, be observed with one single ion. Moreover, instead of measuring photon absorption

(imaginary part of the index of refraction), it is possible to detect dispersive effects (real part of the index); this requires shifting the frequency of the laser beam to outside of the natural line width (the detuning $\delta\omega$ is larger than the inverse lifetime γ of the excited state), while remaining sufficiently close to the ionic resonance frequency. The theory of optical pumping^{15,16} shows that, under these conditions, the effect of the optical field is no longer to relax the ionic ground state but to shift the two sublevels of the ground state in a way which can be described by an effective hamiltonian (virtual transitions). The most convenient way to detect the effects^{16,17} of dispersion is to monitor polarization changes, more precisely rotation of the plane of polarization for spin-1/2 particles (paramagnetic Faraday effect). If a linearly polarized photon beam propagates along the quantization axis, it undergoes a rotation by an angle α if the spin is in state $|+\rangle$, but $-\alpha$ if it is in state $|-\rangle$. Since with a single ion only two rotation angles are possible, the result is quantized and the situation is similar to a Stern and Gerlach experiment. Ideally, the order of magnitude of $|\alpha|$ could be one radian, but this would require the choice of a relatively small value for the ratio $\delta\omega/\gamma$, of the order of one, and automatically allow a high rate of real photon absorption. Instead, one could detune the laser more and assume that $\delta\omega/\gamma \approx 100$, for instance; this would reduce (see note 10) the angle of Faraday rotation by a factor of 100 but, at the same time, the rate of absorption by a factor 10^4 (it decreases as the inverse of square of the detuning factor). See the Appendix for a brief discussion of the effect of the parameter $\delta\omega/\gamma$.

Quantum measurements

We first discuss how the optical techniques discussed in the preceding section can be used to perform quantum measurements on systems containing two particles. A possible scheme is shown in Figure 3: two distinct traps, which can be separated by a macroscopic distance (meters), hold each a single ion; a linearly polarized

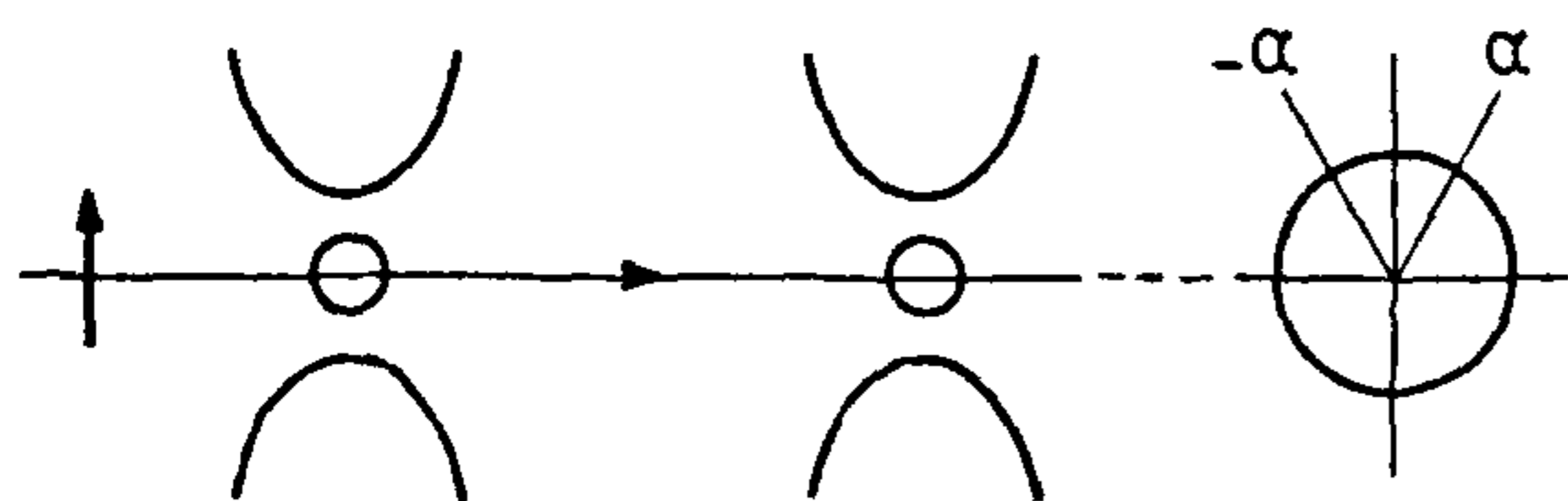


Figure 3. Two ions (circles) are stored at two different traps (schematically represented by hyperbolic electrodes) and a linearly polarized laser beam (arrows) of non-resonant frequency measures the sum of the two spin orientations along its direction. After crossing the two traps, the linear polarization may have three directions, with angles $\pm\alpha$ or 0, these angles are schematically shown in the right part of the figure.

laser beam propagates along the line defined by the two ions, so that the Faraday rotation after interactions in the two traps is proportional to the sum of the two spin components (this is not equivalent to separate information on the orientation of each ion). Three results can be obtained: a rotation of 2α will be observed if the two ions are in their $|+\rangle$ state; a rotation of -2α if they are in their $|-\rangle$ state; and no rotation at all if they are in opposite spin states. In other words, the relevant 'quantum observable' has three eigenvalues, two of which are nondegenerate; the third, corresponding to a zero rotation, is associated with a two-dimensional space of eigenstates:

$$b|+, -\rangle + c|-, +\rangle \tag{27}$$

(where b and c are arbitrary complex numbers obeying $|b|^2 + |c|^2 = 1$). The possible angles of rotation are schematically shown in the right part of Figure 3.

Suppose now, for instance, that, after interacting with the two ions, the beam crosses a polarization analyser which is oriented exactly at 90° to the polarizer inserted before the traps. No photon can then cross the system unless it changes polarization; this means that, if a photon is detected by photomultiplier sitting behind the analyser, the components of the two-spin state $|\Psi\rangle$ corresponding to zero rotation are eliminated by the effect of wave packet reduction. Let us denote by $|\parallel\rangle$ the initial polarization state of the photons and by $|\perp\rangle$ the state of orthogonal polarization. We assume that, before the interaction, the state of two spins plus one photon is described by the ket

$$|\Phi\rangle = |\Psi\rangle_s \otimes |\parallel\rangle, \tag{28}$$

where $|\Psi\rangle_s$ is any spin state:

$$|\Psi\rangle_s = a|+, +\rangle + b|+, -\rangle + c|-, +\rangle + d|-, -\rangle. \tag{29}$$

After interaction $|\Phi\rangle$ is changed into

$$|\Phi'\rangle = a|+, +; 2\alpha\rangle + b|+, -; \parallel\rangle + c|-, +; \parallel\rangle + d|-, -; -2\alpha\rangle \tag{30}$$

(with obvious notation) or

$$|\Phi'\rangle = [|\Psi\rangle_s - 2 \sin^2 \alpha [a|+, +\rangle + d|-, -\rangle]] |\parallel\rangle + \sin 2\alpha [a|+, +\rangle - d|-, -\rangle] |\perp\rangle. \tag{31}$$

This equation shows that, if the photon is detected, the state of the spins is instantaneously projected on to the new state

$$|\Psi'\rangle_s = a|+, +\rangle - d|-, -\rangle, \tag{32}$$

which is precisely of the general form (17). There is even flexibility on the state obtained, since the values of a and b can be adjusted freely. This can be obtained by initially polarizing the two spins along the quantization

axis (which can be obtained, beforehand, by optical pumping, for instance) and using NMR radiofrequency pulses (or any deterministic process rotating the spins) to put them individually into an arbitrary coherent state $\alpha|+\rangle + \beta|-\rangle$, which may be the same or different for the two spins. If they are the same, one gets

$$a = \alpha^2, \quad d = \beta^2, \quad (33)$$

which shows that both the components of eq. (32) may be chosen arbitrarily. For instance, if, initially, both the spins were polarized along Ox , the state $|\Psi\rangle'$ which is reached is the triplet state $|1, 1\rangle$ (with usual notation); if, on the other hand, they start from opposite polarizations along Ox , one gets after measurement the triplet $|1, 0\rangle$ state, and so on. But we must remember that the system goes into state $|\Psi\rangle'$ only if the photon is detected by the photomultiplier; another possibility is that a negative measurement is performed, which projects the state vector on to the ket in brackets in the first line of eq. (31). The desired state is, therefore, not always obtained. Moreover, if a photon is not detected for some experimental reason (limited quantum efficiency of the photomultiplier, for instance), after interaction the state of the spins does not remain pure, as easily seen by partial trace over the photon variables. This is, in fact, not catastrophic since the same experiment can be repeated a large number of times with weak optical pulses (containing about one photon) from fresh-spin initial conditions; one may try until a photon is indeed detected in order to get the desired initial quantum state. If the quantum efficiency of the photomultiplier is low, more attempts before success may be necessary, but the desired state is obtained as soon as a photon is eventually detected (see note 11).

What happens now if, instead of two spins in two traps, we have four on the same line? We can apply the preceding scheme recursively by doing, in a first step, experiments separately on the two spins on the right and the two on the left. The experiment can be initialized as many times as necessary until one detects a photon on each side. This provides a quantum state of the form

$$|\Psi\rangle' = \{ a|+, +\rangle + d|-, -\rangle \} \otimes \{ e|+, +\rangle + h|-, -\rangle \}, \quad (34)$$

where the coefficients a , d , e and h are, as before, fixed by the arbitrary initial orientations of the spins. Now, in a second step, by changing the optical set-up (moving mirrors, for instance), we can perform a measurement on all four spins in the same line, and measure the sum of the four spin components. As for two spins, three results are possible, and actually the predictions are exactly the same except that the Faraday angles are now doubled since all spins are grouped into pairs. The same type of set-up with crossed polarizer and analyser will lead to the same prediction: if a photon is detected after inter-

acting with all the four spins, the wave packet reduction will project $|\Psi\rangle'$ on to a state

$$|\Psi\rangle'' = ae|+, +, +, +\rangle + dh|-, -, -, -\rangle. \quad (35)$$

This, again, provides a 'state with extreme coherence', now for four particles (see note 12). Clearly, the scheme is iterative, and one could polarize 8, 16, ... particles; but, of course, there are practical limitations: since at each stage of the process the production of the desired state assumes that one photon is detected, the overall probability of this occurring at all stages decreases with the number of steps.

Conclusion

There is probably a whole variety of other possible schemes for constructing quantum correlations by iterative application of the reduction of the wave packet. The particular scheme we have discussed allows one to correlate 2^n particles in n successive steps. In theory, n could be arbitrarily large but, since at each step there is a possibility of 'failure' – a possibility that the system will spontaneously be projected onto the states that are orthogonal to the desired subspace – it is clear that the probability of 'success' decreases exponentially for large values of n . In practice, due in particular to limitations arising from the limited quantum efficiency of photomultipliers, n will probably be limited to relatively small numbers (less than 10). In terms of detection efficiency, one of the advantages of detecting a transmitted laser beam, as compared to detecting the fluorescence that is scattered in all directions, is to suppress the losses arising from solid angle limitations; they are indeed the main source of loss in many photon cascade experiments. Generally speaking, it may be interesting to make use of the postulates of quantum mechanics, not only in the final step of the measurement, as usual in quantum mechanics experiments, but also at the initial stage of the preparation of the system; this may offer more flexibility in testing the unexpected predictions of quantum mechanics in critical situations.

Appendix

In this appendix we give a slightly more precise discussion of the change of polarization that a laser beam undergoes when it goes across a single ion sitting in a trap. It is classical, in optical pumping experiments, to monitor the spin orientation of an atomic gas through changes of the polarization of a transmitted beam of light¹⁶. There is, nevertheless, one difference from the case that we study here: in optical pumping the gas of atoms usually extends over a transverse distance that is much larger than the optical wavelength, and the structure of the detection beam can be close to a plane wave. For a

single ion, the light scatterer is localized in a small region of space so that a significant polarization change can be obtained only if the beam is focused strongly. In other words, if a laser beam waist is formed on the ion, the spatial average over the position of the scatterer may become different from that which is usually taken in calculations of detection signals in optical pumping¹⁷.

Let us nevertheless briefly recall what happens in the usual situation where there are many scatterers randomly distributed across the beam. We assume that the transition is electric dipolar. A convenient method to predict the properties of the optical beam after it crosses the sample of atoms is to calculate the electric field as a coherent superposition of the incident field and of the field radiated in the forward direction by the dipoles of the atoms. For simplicity we assume that the ground state level as well as the excited level has a total angular momentum $J = 1/2$ (this simplifies the Grotrian diagram and the angular algebra, but is not essential). In this case, if the atoms are all in the $|+1/2\rangle$ state, whatever the polarization of the incident beam, the field radiated by the atoms in the forward direction has a circular σ^+ polarization. If the incident polarization is also circular, the radiated field is maximum for σ^+ polarization while it vanishes for a σ^- polarization. If the atoms are all in the $|-\rangle$ state, the same results hold except that σ^+ and σ^- are merely interchanged; so we assume that the spin state is $|+1/2\rangle$ and the incident polarization σ^+ . Now, if the laser frequency falls inside the natural linewidth ($\delta\omega \approx \gamma$), it creates resonant excitation, and the phase of the electric field radiated in the forward direction is the same as that of the incident field; on the other hand, if the frequency falls outside the natural linewidth ($|\delta\omega| \gg \gamma$) one has nonresonant excitation and there is a $\pi/2$ dephasing. From this we can predict what will happen when the incident polarization is linear, i.e. a coherent superposition of σ^+ and σ^- :

(i) For resonant excitation, the radiated field will have its direction (which is rotating since it is circularly polarized) parallel to the incident field at times where this field is maximum, while it will be perpendicular a quarter of an optical period later. On an average over many optical periods, and to first order in the radiated field, this results in a reduction of the amplitude of the electrical field, and introduces absorption.

(ii) The situation is just the opposite if the excitation is nonresonant: when the incident linear field is maximum, the direction of the circularly polarized radiated field is now perpendicular, and the net effect (again to first order) is no longer an absorption of energy, but a rotation of the plane of polarization. The angle of rotation is the same in all transverse cross-sections of the beam.

We now come back to the detection of a single particle sitting in a trap. We assume that laser cooling tech-

niques¹⁸ are used so that the orbital variables of the ion are described by the ground state in the trap, which has a small spatial extension. Several situations are possible, depending upon the relative size of this extension and of the laser beam waist. If the laser is focused strongly onto a spot that is smaller than the ground state, the results of the preceding paragraph are basically still valid; the effect of the probability cloud associated with the ground state can be described as that of an index of refraction. If the sizes are comparable, which is probably the best situation in practice, one gets an intermediate case. If the ion is located in a region that is smaller than the laser beam cross-section, the situation is analogous to a small antenna radiating coherently in the middle of a broad electromagnetic beam; it radiates a spherical wave which produces nonuniform interference effects with the incident wave. Since the amplitude of the spherical wave is a slowly varying function of direction, while the incident wave is closer to a plane wave, there is no reason that the polarization of the resulting interference should be uniform; for instance, larger rotations will be observed on the side of the incident wave where the relative effects of the spherical wave are relatively more important. In these conditions one can get a complex polarization (and intensity) pattern after the crossing of the trap, and the situation is far from being as simple as we have assumed in the text. If we assume, nevertheless, that a symmetrical design is used, as shown in Figure 4, where a lens refocuses the beam on the second trap so that the two beam waists have exactly the same geometry, in this case, when the two spins are in opposite spin states, a symmetry argument shows that the polarization of the beam after the two interaction is unchanged (perfectly linear-parallel to the incident polarization), while it undergoes complex changes if the two spin states are the same. Since the aim of the crossed polarizer was precisely to eliminate the components of the state vector on opposite spin states, the detection of a photon keeps the same physical meaning, and the state preparation scheme discussed in the text is preserved.

Another issue is how the value of the parameter $\delta\omega/\gamma$ should be chosen. The difficulty is that, for weak

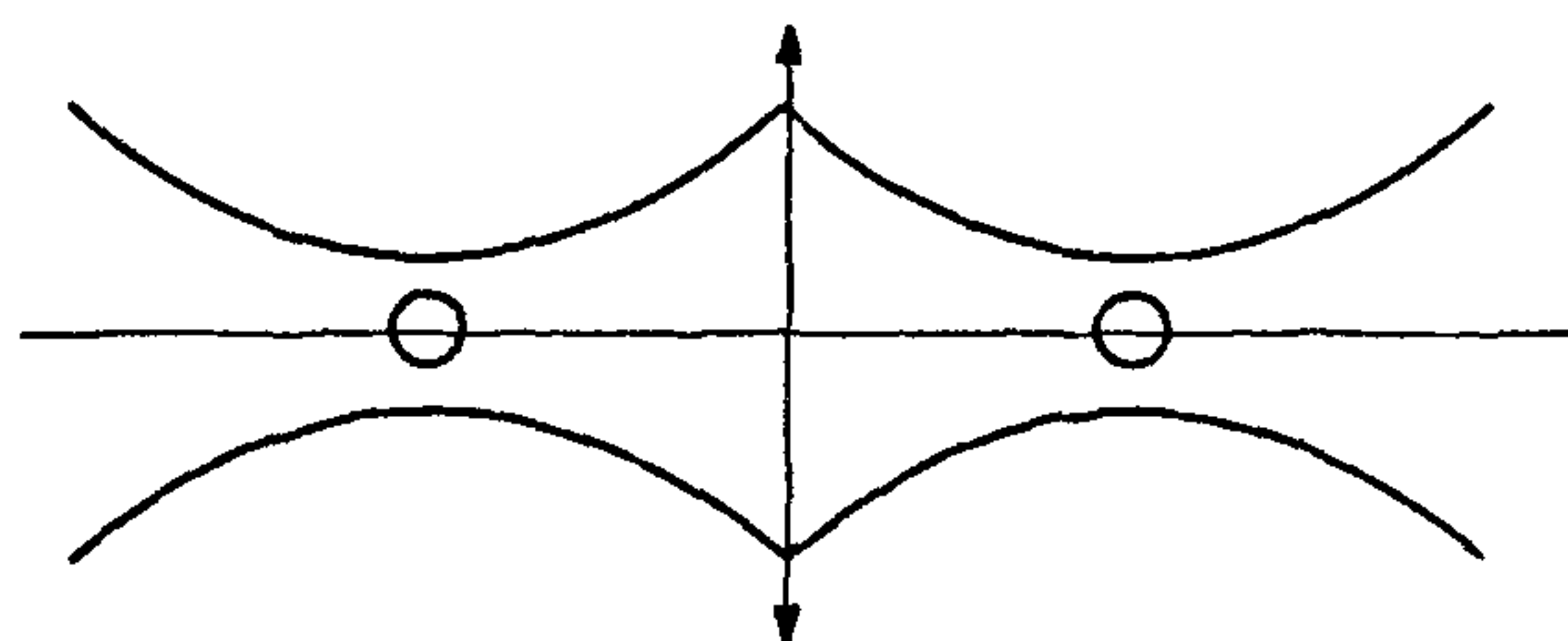


Figure 4. A set-up with a lens in the middle of two identical traps, which, by symmetry, ensures that the initial linear polarization will be pure and unchanged each time the two spins point in opposite directions.

Faraday rotation angles, the probability for a photon to cross the perpendicular analyser is quadratic in the angle, so that both the rate of detecting photons and that of real transitions (which destroy the coherence of the state vector) vary as $(\delta\omega/\gamma)^{-2}$. If we call X the ratio between the area of the laser beam waist and the scattering cross-section λ^2 , and R the quantum efficiency of the photomultiplier, it is easy to convince oneself that the ratio between the rates of photon detection and of real optical excitation is of the order of R/X (when $\delta\omega/\gamma \gg 1$), which is always smaller than one. The strategy to adopt then is similar to that already discussed in the text after eq. (33): rely on rare events and refresh the initial state as often as necessary, until a positive detection result occurs. If, for instance, weak photon pulses are sent on to the ions, one may choose conditions in which the probability of real excitations is, say, 10^{-2} per pulse. If the probability for detecting a photon on the photomultiplier is for instance 10^{-3} , it becomes necessary to repeat the experiment from fresh initial conditions about a thousand times before obtaining a positive result. But, then, the probability of getting the correct initial state will be 99% (assuming, as above, that the intrinsic noise of the photomultiplier is negligible). Another possibility is to use extremely weak pulses each containing one photon only.

Notes

- 1 There are other formalisms for quantum mechanics which are equivalent to that using wave functions or state vectors, e.g. density operators, integrals of action, Wigner transform, etc. In this article we use the words 'quantum state', or 'ket', as well as 'reduction of the wave packet', in a generic sense which contains these different versions of the formalism.
- 2 This is true after the initial full correlations have been established, which may require many experiments, but, once this is done, in principle one single experiment is sufficient.
- 3 Of course, this is not assuming that we know what the reply would be. In mathematics, it is common to reason on unknown quantities (this can be seen as the starting point of algebra).
- 4 With a sign change that is of no importance here, just a matter of definition, we could assume that the product is -1 instead of 1 .
- 5 We see in passing why the reasoning is possible only for more than two particles, the same is true in the reasoning of the third lemma.
- 6 The fact that they are all $+$ in one component and all $-$ in the other does not matter here, although for the sake of simplicity we consider only this case, the only thing that really matters for the argument which follows is that every spin changes to an orthogonal state when one goes from one component of $|\Psi\rangle$ to the other.
- 7 Even if the process we consider is actually a succession of measurements instead of one, it is not necessary to treat it explicitly as a sequence (first considering the effect of one single-particle measurement by applying to $|\Psi\rangle$ the postulate of wave packet reduction to get its new value, second starting from this new state for evaluating the probabilities of the second measurement, etc.) Because all $S_i(\theta)$ commute (they are operators acting in the space of states of different particles), we can directly obtain the desired result by calculating the average of a product as in eq. (21).
- 8 We do not enter technical discussions on the effect of ion motion in magnetic field gradients, etc.; we assume that the spins have a long transverse relaxation time in the ground state.
- 9 We assume that the oscillator strength of the optical transition is one and that the laser intensity is sufficiently weak not to saturate the optical transition.
- 10 Other possible sources of reduction for the angle of rotation are purely geometric; see Appendix for a brief discussion of this question.
- 11 The only real problem arises from false events, due, for instance, to the intrinsic noise of the photomultiplier.
- 12 Alternatively, we could have measured the Faraday rotation of the two spins in the middle; a nonzero rotation would have produced the same four-particle quantum state.

ACKNOWLEDGEMENTS I am grateful to R. Bahian, P. Grangier, B. D'Espagnat, J. M. Raimond and R. Griffiths for useful comments and suggestions on this text. Prof. Griffiths pointed out to the author an error in the initial manuscript, in particular a sign error that he had made in writing eq. (32), and therefore played an essential role in the correctness of this article.

Received 20 December 1994, revised accepted 15 March 1995

- 1 Einstein, A., Podolsky, B. and Rosen, N., *Phys. Rev.*, 1935, **47**, 777.
- 2 Bell, J. S., *Physics*, 1965, **1**, 195.
- 3 Bell, J. S., *Rev. Mod. Phys.*, 1966, **38**, 447.
- 4 d'Espagnat, B., *Le Réel Voilé*, Fayard, Paris and the references therein.
- 5 Greenberger, D. M., Horne, M. A. and Zeilinger, A., in *Bell Theorem, Quantum Theory, and Conceptions of the Universe* (ed. Kafatos, M.), Kluwer, Dordrecht, 1989, p. 74; Greenberger, D. M., Horne, M. A., Shimony, A. and Zeilinger, A., *Am. J. Phys.*, 1990, **58**, 1131.
- 6 Mermin, N. D., *Phys. Today*, June 1990, **9**, *Rev. Mod. Phys.*, 1993, **65**, 803.
- 7 Hardy, L., *Phys. Rev. Lett.*, 1992, **68**, 2981, 1993, **71**, 1665.
- 8 Mermin, N. D., *Phys. Today*, June 1994, **9**.
- 9 Cirac, J. I. and Zoller, P., *Phys. Rev. A*, 1994, **50**, R2799.
- 10 Stapp, H. P., *Nuov. Cim.*, 1977, **40B**, 191; *Found. of Phys.*, 1980, **10**, 767.
- 11 Ref. 4, §9.2.
- 12 Peres, A., *Am. J. Phys.*, 1978, **46**, 745.
- 13 Bergquist, J. C., Diedrich, F., Wayne, M. Itano and Wineland, D. J., *Laser Spectroscopy* (ed. Feld, M. S., Thomas, J. E. and Mooradian, A.), Academic, San Diego, 1989, vol. IX.
- 14 Blatt, R., Gill, P. and Thompson, R. C., *J. Mod. Opt.*, 1992, **39**, 193.
- 15 Cohen Tannoudji, C., *Ann. Physique*, 1962, **7**, 423–469.
- 16 Itapper, W., *Rev. Mod. Phys.*, 1972, **44**, 169.
- 17 Cohen Tannoudji, C. and Laloe, F., *J. Physique*, 1967, **28**, 505, 722.
- 18 Diedrich, F., Bergquist, J. C., Wayne, M. Itano and Wineland, D. J., *Phys. Rev. Lett.*, 1989, **62**, 403.