

## An Experimental Test of Quantum Mechanics in Molecular Predissociation (\*).

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**Summary.** — In this paper we propose a new experimental test of quantum mechanics, dealing with correlation measurements in composite systems. Nonradiative two-body decays in molecular predissociation allow—in the particular case of the NO molecule, that we study in detail—a much more sensitive test compared to the usual two-photon experiments. We discuss a possible way of performing the experiment and verify its real feasibility in all the relevant details.

I. — All the experimental tests of quantum mechanics (QM) that have been made recently <sup>(1)</sup> in connection with those aspects linked with the Einstein, Podolsky and Rosen (EPR) paradox <sup>(2)</sup> dealt with couples of correlated photons. The difficulties of such an experiment are well known. Even the new possi-

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(\* To speed up publication, the author of this paper has agreed to not receive the proofs for correction.

<sup>(1)</sup> S. J. FREEDMAN and J. F. CLAUSER: *Phys. Rev. Lett.*, **28**, 938 (1972); R. A. HOLT and F. M. PIPKIN: unpublished preprint (1974); J. F. CLAUSER: *Phys. Rev. Lett.*, **36**, 1223 (1976); E. S. FRY and R. C. THOMSON: *Phys. Rev. Lett.*, **37**, 465 (1976); G. FARACI, D. GUTKOWSKI, S. NOTARRIGO and A. R. PENNISI: *Lett. Nuovo Cimento*, **9**, 607 (1974); L. R. KASDAY, J. D. ULLMAN and C. S. WU: *Nuovo Cimento*, **25 B**, 633 (1975); M. BRUNO, M. D'AGOSTINO and C. MARONI: Università di Bologna, preprint (1976); M. LAMEHIRACHTI and W. MITTIG: *Phys. Rev. D*, **14**, 2543 (1976).

<sup>(2)</sup> A. EINSTEIN, B. PODOLSKY and N. ROSEN: *Phys. Rev.*, **47**, 777 (1935). The possibility of an experimental test was open by the paper of J. S. BELL: *Physics*, **1**, 195 (1964). For a recent discussion and a list of references see F. SELLERI: *Found. Phys.*, **8**, 103 (1978).

bilities open by the so-called *sensitive observables* <sup>(3)</sup> have been experimentally exploited only in photon experiments <sup>(4)</sup>. Almost all the attention in the foundation of QM was, therefore, limited to dicotomic observables.

For the case of multivalued observables it was shown that an extension of Bell's inequality no longer constitutes a good test of QM <sup>(5)</sup>, while on the basis of the sensitive observables new tests can be devised of increasing sensitivity <sup>(6)</sup>.

In this paper we want to show that a specific test of this kind is really feasible in the realm of molecular physics, using Stern-Gerlach devices as spin detectors. It is worth recalling that the class of tests we are dealing with concerns the correlations between two subsystems coming from the dissociation of a compound system.

We shall briefly discuss in the following first of all the physical process of *predissociation* of diatomic molecules, which is the basis of the experiment we want to propose; we shall then introduce the quantum-mechanical calculation in the specific case of the predissociation of the NO molecule; we shall finally present the detailed project of the experiment and discuss its feasibility.

## 2. - Predissociation of the NO molecule.

Predissociation is a process of dissociation of a molecule into the atoms (we limit ourselves to *diatomic* molecules), due to the overlapping of two different molecular electronic (or rotational) states. Let us consider a pattern of the vibrational levels in the electronic states *A* and *B* of a diatomic molecule like that of fig. 1; while the levels 0, 1, 2, 3 are not significantly perturbed by the levels of the state *A* having usually a different energy, the levels 4, 5, ... have on the contrary energies corresponding to the continuum of *A* and possess consequently a finite probability of a spontaneous and radiationless transition to a level of the continuum of *B* having the same energy, *i.e.* to a state of the two dissociated atoms <sup>(7)</sup>. The mean life of these states is very short ( $10^{-11}$  s) and this causes a broadening of the absorption lines corre-

<sup>(3)</sup> V. CAPASSO, D. FORTUNATO and F. SELLERI: *Int. Journ. Theor. Phys.*, **7**, 319 (1973); N. CUFARO-PETRONI: *Nuovo Cimento*, **40** B, 235, 381 (1977); A. BARACCA, A. CORNIA, A. LUNARDINI and S. RUFFO: Università di Firenze, preprint (1978).

<sup>(4)</sup> J. F. CLAUSER: *Nuovo Cimento*, **33** B, 740 (1976).

<sup>(5)</sup> A. BARACCA, S. BERGIA, R. LIVI and M. RESTIGNOLI: *Int. Journ. Theor. Phys.*, **15**, 473 (1976); A. BARACCA, S. BERGIA, F. CANNATA, S. RUFFO and M. SAVOIA: *Int. Journ. Theor. Phys.*, **16**, 491 (1977).

<sup>(6)</sup> R. LIVI: *Lett. Nuovo Cimento*, **19**, 189 (1977).

<sup>(7)</sup> The process is analogous to the Auger effect for atoms. For a clear and extended discussion about predissociation see G. HERZBERG: *Molecular Spectra and Molecular Structure. - I: Spectra of Diatomic Molecules* (New York, N. Y., 1950).

sponding to such a vibrational state and the disappearance of the corresponding emission lines.

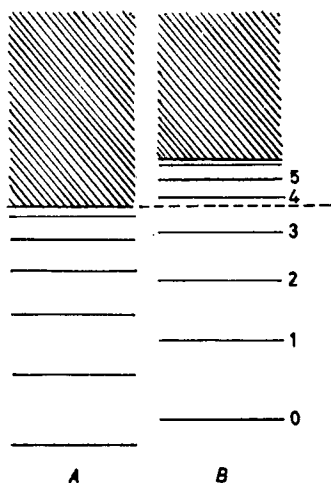


Fig. 1.

The products of predissociation may be normal or excited atoms. The first case is predominant, just because predissociation can be considered as a perturbative phenomenon in which the molecule dissociates adiabatically into normal atoms. The second case is present only when the Wigner-Witmer<sup>(8)</sup> rules do not hold. In what follows we shall take into account only the first case, just because it refers to the specific case we shall deal with.

The various mechanisms of predissociation were discussed by KRÖNIG<sup>(9)</sup>. We simply recall here that for the case of electronic state that we have discussed above the Franck-Condon principle<sup>(10)</sup> requires that the potential curves corresponding to the two electronic states cross like in fig. 2. Moreover, the process satisfies the following selection rules—the same ones of a perturbative decay:

1)  $\Delta J = 0$ , which corresponds to the total angular-momentum conservation;

2)  $s \leftrightarrow a$ ,  $+$   $\leftrightarrow$   $-$ —due to the fact that the initial and the final states must conserve their symmetric structure;

<sup>(8)</sup> E. WIGNER and E. E. WITMER: *Zeits. Phys.*, **51**, 859 (1928).

<sup>(9)</sup> R. DE L. KRÖNIG: *Zeits. Phys.*, **109**, 387 (1938); **11**, 640 (1939).

<sup>(10)</sup> E. U. CONDON: *Phys. Rev.*, **32**, 858 (1928); J. FRANCK: *Trans. Faraday Soc.*, **21**, 536 (1925). For a clear and detailed discussion about this principle in the case of predissociation see also the book mentioned in ref. (7), p. 420-432.

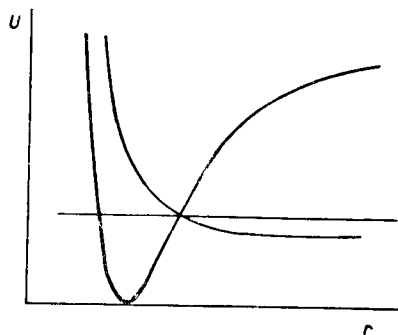


Fig. 2.

3) Hund's cases *a*) and *b*)<sup>(11)</sup>

$$\Delta S = 0, \quad \Delta A = 0, \quad \pm 1,$$

where  $S$  and  $A$  are, respectively, the spin and the component of angular momentum  $L$  along the internuclear axis; and Hund's case *c*)<sup>(11)</sup>

$$\Delta \Omega = 0, \quad \pm 1,$$

where  $\Omega$  is the component of the resultant momentum  $J$  ( $= L + S$ ) along the internuclear axis; moreover, in case *a*)

$$\Delta \Sigma = 0,$$

in *b*)

$$\Delta K = 0,$$

where  $\Sigma$  is the component of the spin  $S$  along the internuclear axis and  $K$  the total angular momentum apart from spin. A specific case of predissociation which is convenient for our purposes is constituted by the NO molecule and was studied by FLORY and JOHNSTON<sup>(12)</sup>. The pattern of the electronic states and the vibrational levels is shown in fig. 3. If a molecule is initially in the electronic state  ${}^2I$  and the fundamental vibrational level absorbs a photon

<sup>(11)</sup> In Hund's case *a*) the interaction of the nuclear rotation with the electronic motion is very weak, whereas the electronic motion itself is coupled very strongly to the line joining the nuclei. In Hund's case *b*)  $\Omega$  is not defined and sometimes, particularly for light molecules,  $S$  may be only very weakly coupled to the internuclear axis. In Hund's case *c*) the interaction between  $L$  and  $S$  is stronger than the interaction with the internuclear axis, so  $A$  and  $\Sigma$  are not defined; this is a peculiar feature of heavy molecules.

<sup>(12)</sup> P. J. FLORY and H. L. JOHNSTON: *Journ. Amer. Chem. Soc.*, **57**, 2641 (1935).

of wave-length between 1.800 and 2.000 Å in the  $\beta$ -band, its final state corresponds to a new electronic state  ${}^2\Pi$  whose fundamental vibrational state has an energy 5.5 eV bigger than the previous one. The potential curve of the final  ${}^2\Pi$ -state crosses the curve of a repulsive  ${}^2\Sigma$ -level, responsible for the predissociation in the region corresponding to an internuclear distance of about 1.7 Å and a potential energy of the molecule of about 6 eV. One has consequently a finite probability for the radiationless transition to the dissociated state  $N({}^4S) + O({}^3P)$ , composed of the two atoms in their normal state.

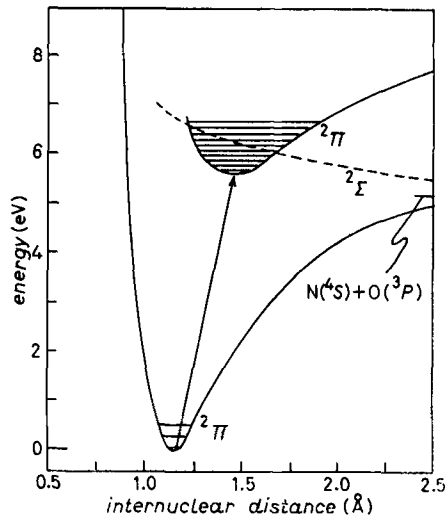


Fig. 3.

### 3. - The state of the NO molecule in terms of the atomic states.

The problem of comparing the predictions of QM with those of a theory based on proper mixtures in the present case requires to express the molecular states in terms of those of the two final atoms: more precisely, in terms of the eigenstates of total angular momentum  $J$  of the dissociated atoms. Such a problem is not a straightforward extension of the usual analysis of a composite system of definite total angular momentum, since the complex structure of atoms and molecules strongly limits the kind of conserved quantities in the dissociation.

The real situation may be resumed as follows:

i) On the one hand, we have the molecular state of predissociation, for which the quantum numbers  $S$  and

$$(1) \quad A = |M_{L_1} + M_{L_2}|$$

are defined, where  $M_{L_i}$  is the component along the internuclear axis of the orbital angular momentum  $L_i$  of the atom  $i$  ( $i = 1, 2$ ).  $A$  may then be interpreted as the component along the internuclear axis of the angular momentum  $L_{\text{mol}}$  coming from the composition of  $L_1$  and  $L_2$ ,

$$(2) \quad L_{\text{mol}} = L_1 + L_2,$$

and allowed obviously to assume all the values between  $L_1 + L_2$  and  $|L_1 - L_2|$ . In the diatomic molecule  $A$ , and not  $L$ , is defined, due to the symmetry property around the internuclear axis.  $S$  is instead obtained as the sum of the spins of the two atoms

$$(3) \quad S_{\text{mol}} = S_1 + S_2$$

and takes the values between  $S_1 + S_2$  and  $|S_1 - S_2|$ .

ii) On the other hand, we have the states of the two dissociated atoms, for which the values of  $L_i$  and  $S_i$  are given.

Nevertheless, a Stern-Gerlach apparatus measures the components of the total angular momentum  $J_i$  of each atom, along the quantization axis, corresponding to the direction of the gradient of the magnetic field. It is obviously

$$(4) \quad J_i = L_i + S_i \quad (i = 1, 2)$$

and the values of  $J_i$  run between  $L_i + S_i$  and  $|L_i - S_i|$ : we have consequently many different values of  $J_i$ , corresponding to given values of  $L_i$  and  $S_i$ . But Hund's rule allows us to determine the value of  $J_i$  in the fundamental state, so that for each atom the value of  $J_i$  is fixed.

In general one has to expand the molecular state

$$(5) \quad |A, S\rangle$$

in terms of the atomic states

$$(6) \quad |J_1, M_{J_1}\rangle, \quad |J_2, M_{J_2}\rangle$$

of total angular momentum  $J_1, J_2$ , respectively of the two atoms, through an intermediate step, which takes into account the values of the other quantum numbers:  $L_1, S_1; L_2, S_2$ . We shall make explicit in the following this expansion in its most general form and we shall take into account, for the specific case of the NO molecule, the conditions discussed above in points i) and ii). Now, let us see the two following steps:

*I step:* one has without difficulties

$$(7) \quad |L_i M_{L_i} S_i M_{S_i}\rangle = \sum_{J_i} \sum_{M_{J_i}} \langle J_i M_{J_i} | L_i M_{L_i} S_i M_{S_i}\rangle |J_i M_{J_i}\rangle \quad (i = 1, 2)$$

with the condition

$$(8) \quad M_{J_i} = M_{L_i} + M_{S_i}.$$

But the summation over  $J_i$  is fictitious, because Hund's rule fixes the value,  $\hat{J}_i$  for the normal state of the atom; as a consequence of this consideration, we stress here that the state (7) must be normalized and expressed as follows:

$$(9) \quad |L_i M_{L_i} S_i M_{S_i}\rangle_{\text{norm}} = \frac{\sum_{M_{J_i}} \langle J_i M_{J_i} | L_i M_{L_i} S_i M_{S_i}\rangle |J_i M_{J_i}\rangle}{\left\{ \sum_{M_{J_i}} |\langle J_i M_{J_i} | L_i M_{L_i} S_i M_{S_i}\rangle|^2 \right\}^{\frac{1}{2}}}.$$

*II step:* now we can expand the molecular state in terms of  $L_i$  and  $S$  with the additional condition that  $S$  and  $\Lambda$  are fixed, but not  $L$ :

$$(10) \quad |(L)\Lambda S\rangle = \sum_{L=|L_1-L_2|}^{L_1+L_2} \sum_{M_{L_1}} \sum_{M_{L_2}} \sum_{M_{S_1}} \sum_{M_{S_2}} \langle L\Lambda | L_1 M_{L_1} L_2 M_{L_2}\rangle \cdot \langle S M_S | S_1 M_{S_1} S_2 M_{S_2}\rangle |L_1 M_{L_1} S_1 M_{S_1}\rangle |L_2 M_{L_2} S_2 M_{S_2}\rangle$$

and also in this case we must normalize this state:

$$(11) \quad |(L)\Lambda S\rangle_{\text{norm}} = \left[ \sum_L \sum_{M_{L_1}} \sum_{M_{L_2}} \sum_{M_{S_1}} \sum_{M_{S_2}} \langle L\Lambda | L_1 M_{L_1} L_2 M_{L_2}\rangle \langle S M_S | S_1 M_{S_1} S_2 M_{S_2}\rangle \cdot |L_1 M_{L_1} S_1 M_{S_1}\rangle |L_2 M_{L_2} S_2 M_{S_2}\rangle \right] / \left[ \sum_L \sum_{M_{L_1}} \sum_{M_{L_2}} \sum_{M_{S_1}} \sum_{M_{S_2}} |\langle L\Lambda | L_1 M_{L_1} L_2 M_{L_2}\rangle|^2 \cdot |\langle S M_S | S_1 M_{S_1} S_2 M_{S_2}\rangle|^2 \right]^{\frac{1}{2}}.$$

Composing now (9) and (11), we get

$$(12) \quad |(L)\Lambda S\rangle_{\text{norm}} = \frac{1}{N} \sum_L \sum_{M_{L_1}} \sum_{M_{L_2}} \sum_{M_S} \sum_{M_{S_1}} \sum_{M_{S_2}} \langle L\Lambda | L_1 M_{L_1} L_2 M_{L_2}\rangle \cdot \langle S M_S | S_1 M_{S_1} S_2 M_{S_2}\rangle \langle L_1 M_{L_1} S_1 M_{S_1} | J_1 M_{J_1}\rangle \langle L_2 M_{L_2} S_2 M_{S_2} | J_2 M_{J_2}\rangle |J_1 M_{J_1}\rangle |J_2 M_{J_2}\rangle,$$

where  $N$  is the normalization coefficient expressed by the relation

$$(13) \quad N = \left[ \sum_L \sum_{M_{L_1}} \sum_{M_{L_2}} \sum_{M_{S_1}} \sum_{M_{S_2}} \sum_{M_{J_1}} \sum_{M_{J_2}} |\langle L\Lambda | L_1 M_{L_1} L_2 M_{L_2}\rangle|^2 \cdot |\langle S M_S | S_1 M_{S_1} S_2 M_{S_2}\rangle|^2 \cdot |\langle L_1 M_{L_1} S_1 M_{S_1} | J_1 M_{J_1}\rangle|^2 \cdot |\langle L_2 M_{L_2} S_2 M_{S_2} | J_2 M_{J_2}\rangle|^2 \right]^{\frac{1}{2}}.$$

We stress here that in any explicit calculation one must take into account the relations from (1) to (4), which relate the summation indices.

Now we may build up the pure state corresponding to the quantum-mechanical state (12); we emphasize that it is sufficient to choose a pure state to perform a comparison between QM and proper-mixture theories; in fact, we shall develop here the calculations to find the maximum value of an appropriate sensitive observable on the most general pure state, just because, as was shown in a previous work <sup>(6)</sup>, this is a superior limit for any proper-mixture theory:

$$(14) \quad |(L)AS\rangle_{\text{pure}} = \sum_{M_{J_1}} \sum_{M_{J_2}} C_{M_{J_1}} C_{M_{J_2}} |J_1 M_{J_1}\rangle |J_2 M_{J_2}\rangle$$

with the normalization conditions

$$(15) \quad \sum_{M_{J_1}} |C_{M_{J_1}}|^2 = \sum_{M_{J_2}} |C_{M_{J_2}}|^2 = 1.$$

Due to the recent results about sensitive observables <sup>(3)</sup> we shall hereafter perform the calculations for the sensitive observable

$$(16) \quad -\mathbf{J}_1 \cdot \mathbf{J}_2 = -[(J_1)_x(J_2)_x + (J_1)_y(J_2)_y + (J_1)_z(J_2)_z].$$

We stress here that from a practical point of view the measurement of the mean value of this observable implies the performance of three measurements, disposing the measuring apparatus opposite along three orthogonal axes.

#### 4. - The case of the NO molecule.

First of all we recall that the state of the NO molecule which gives rise to the dissociated state of the two atoms is characterized by the quantum numbers

$$(17) \quad A = 0, \quad S = \frac{1}{2},$$

while the states of the two composing atoms are

$$(18) \quad \text{for nitrogen} \quad S_1 = \frac{3}{2}, \quad L_1 = 0, \quad J_1 = \frac{3}{2},$$

$$(19) \quad \text{for oxygen} \quad S_2 = 1, \quad L_1 = 1, \quad J_2 = 2,$$

since  $A = 0$  is fixed and, moreover,  $L_1 = 0$ , *i.e.*  $M_{L_1} = 0$ ,  $M_{L_2}$  may assume only the value 0; moreover,  $L = 1$ , so that the summations over  $L$ ,  $M_{L_1}$ ,  $M_{L_2}$  reduce to only one term; the corresponding Clebsch-Gordan coefficient is

$$(20) \quad \langle LA | L_1 M_{L_1} L_2 M_{L_2} \rangle = \langle 10 | 1000 \rangle = 1,$$



as we could have expected on the basis that there is only one way to get a state with  $L = 1$ ,  $\Lambda = 0$  from two states  $L_1 = 0$ ,  $M_{L_1} = 0$ ;  $L_2 = 1$ ,  $M_{L_2} = 0$ .

Now, taking into account expression (8), we have

$$(21) \quad M_{J_1} = M_{S_1} \quad \text{and} \quad M_{J_2} = M_{S_2}.$$

The four remaining summations reduce then only to the two summations over  $M_{J_1}$  and  $M_{J_2}$ ; in conclusion (12) becomes

$$(22) \quad |(1)0\frac{1}{2}\rangle = \frac{1}{N} \sum_{M_{J_1}} \sum_{M_{J_2}} \langle \frac{1}{2} M_S | \frac{3}{2} M_{J_1} 1 M_{J_2} \rangle \langle 00 \frac{3}{2} M_{J_1} | \frac{3}{2} M_{J_1} \rangle \cdot \\ \cdot \langle 101 M_{J_1} | 2 M_{J_2} \rangle | \frac{3}{2} M_{J_1} \rangle | 2 M_{J_2} \rangle,$$

where

$$(23) \quad N = \left\{ \sum_{M_{J_1}} \sum_{M_{J_2}} |\langle \frac{1}{2} M_S | \frac{3}{2} M_{J_1} 1 M_{J_2} \rangle|^2 |\langle 00 \frac{3}{2} M_{J_1} | \frac{3}{2} M_{J_1} \rangle|^2 \cdot \right. \\ \left. \cdot |\langle 101 M_{J_1} | 2 M_{J_2} \rangle|^2 \right\}^{\frac{1}{2}} = \sqrt{10/9}.$$

Now we are able to compare the predictions of quantum-mechanical and propermixture theories in the specific case of the NO molecule, performing the calculation of the mean value of the sensitive observable (16) in both cases.

From eqs. (16) and (22) we get the explicit expression

$$(24) \quad \langle -\mathbf{J}_1 \cdot \mathbf{J}_2 \rangle_{\text{QM}} = -\frac{9}{16} \sum_{M_{J_1}} \sum_{M_{J_2}} \sum_{M'_{J_1}} \sum_{M'_{J_2}} \langle \frac{1}{2} M_S | \frac{3}{2} M_{J_1} 1 M_{J_2} \rangle \langle 00 \frac{3}{2} M_{J_1} | \frac{3}{2} M_{J_1} \rangle \cdot \\ \cdot \langle 101 M_{J_2} | 2 M_{J_2} \rangle \langle \frac{1}{2} M_S | \frac{3}{2} M'_{J_1} 1 M'_{J_2} \rangle \langle 00 \frac{3}{2} M'_{J_1} | \frac{3}{2} M'_{J_1} \rangle \cdot \\ \cdot \langle 101 M'_{J_2} | 2 M'_{J_2} \rangle \langle J_1 M'_{J_1} | \langle J_2 M'_{J_2} | \mathbf{J}_1 \cdot \mathbf{J}_2 | J_2 M_{J_2} \rangle | J_1 M_{J_1} \rangle,$$

where

$$(25) \quad \langle J_1 M'_{J_1} | \langle J_2 M'_{J_2} | -\mathbf{J}_1 \cdot \mathbf{J}_2 | J_2 M_{J_2} \rangle | J_1 M_{J_1} \rangle = \\ = - \left\{ \frac{1}{2} \delta_{M'_{J_1}-1, M_{J_1}} \delta_{M'_{J_2}+1, M_{J_2}} \cdot [(2 - M_{J_2})(2 + M_{J_2} + 1)]^{\frac{1}{2}} \cdot \right. \\ \cdot [(\frac{3}{2} + M_{J_1})(\frac{3}{2} - M_{J_1} + 1)]^{\frac{1}{2}} + \frac{1}{2} \delta_{M'_{J_1}+1, M_{J_1}} \delta_{M'_{J_2}-1, M_{J_2}} [(2 + M_{J_2})(2 - M_{J_2} + 1)]^{\frac{1}{2}} \cdot \\ \left. \cdot [(\frac{3}{2} - M_{J_1})(\frac{3}{2} + M_{J_1} + 1)]^{\frac{1}{2}} + \delta_{M'_{J_1}, M_{J_1}} \delta_{M'_{J_2}, M_{J_2}} M_{J_1} \cdot M_{J_2} \right\}.$$

We get finally the numerical result

$$(26) \quad \langle -\mathbf{J}_1 \cdot \mathbf{J}_2 \rangle_{\text{QM}} = 2.17.$$

From eqs. (16) and (14) we get the other expression

$$(27) \quad \langle -\mathbf{J}_1 \cdot \mathbf{J}_2 \rangle_{\text{pure}} = \\ = - \sum_{M_{J_1}} \sum_{M_{J_2}} \sum_{M'_{J_1}} \sum_{M'_{J_2}} C_{M_{J_1}} C_{M_{J_2}} C_{M'_{J_1}}^* C_{M'_{J_2}}^* \langle J_1 \cdot M'_{J_1} | \langle J_2 M'_{J_2} | \mathbf{J}_1 \cdot \mathbf{J}_2 | J_2 M_{J_2} \rangle | J_1 M_{J_1} \rangle.$$

The maximum value of (27) is

$$(28) \quad \max \{ \langle -\mathbf{J}_1 \cdot \mathbf{J}_2 \rangle_{\text{pure}} \} = 0.38.$$

These latter results, 2.17 to be compared with 0.38, allow a clear-cut discrimination between QM and proper mixtures of about 83%. It is the best experimental test in this field; the actual tests deal, in fact, with the dicotomic case in which the discrimination is about 67%.

### 5. — The experimental apparatus.

In this section we are going to discuss a possible set-up (fig. 4) for an experimental test of the kind proposed in the present paper, *i.e.* for the measurement of the mean value of the correlation  $\langle -\mathbf{J}_{N_1} \cdot \mathbf{J}_O \rangle$ .

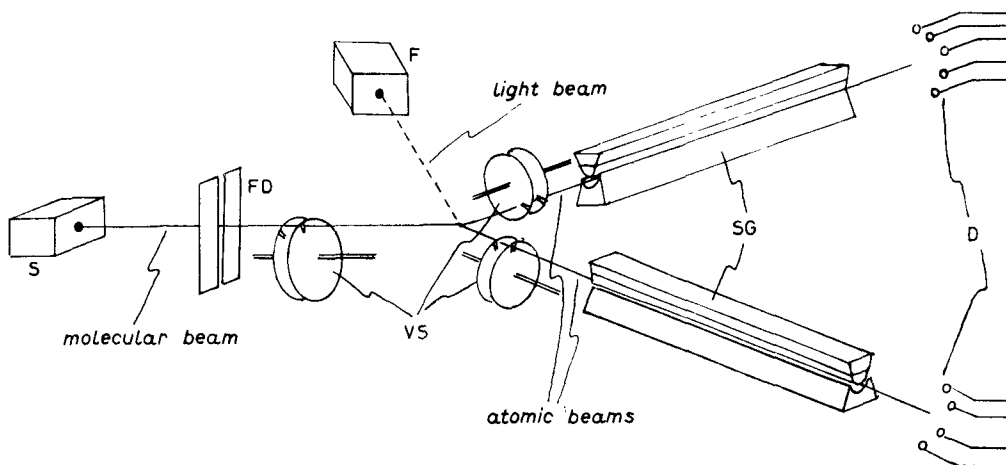


Fig. 4.

One needs a molecular beam source (S) of NO molecules, followed by a velocity selector (VS)—a mechanical one composed of two rotating disks<sup>(13)</sup>. In this way one gets a pulsed beam of NO molecules with approximately uniform velocity: the partial focusing of the velocity selector may be improved by further focusing devices (FD). The importance of getting a uniform velocity of the molecules in the beam is connected to the geometry of the decay, which, in the laboratory system, is fixed by the values of predissociation, rotation and centre-of-mass kinetic energies of the molecules (sect. 6'1).

The pulsed beam then interacts with the predissociation radiation coming from the source F: the frequency of the events may be quite high (sect. 6'2).

<sup>(13)</sup> If we suppose a Maxwell distribution of the molecular velocity, we may select the mean velocity to get a high number of favourable events.

After the predissociation the atoms enter the Stern-Gerlach apparatus (SG); it is important to remark here that, due to the isotropy of the decay, there are oxygen and nitrogen atoms decaying along the same direction, but with different velocities; in order to select only the events in which the oxygen atoms enter always the same Stern-Gerlach apparatus, one can simply add a proper velocity selector to stop the nitrogen atoms (the same holds obviously for the nitrogen atoms and the other apparatus).

In the literature there are several types of Stern-Gerlach apparatus, whose efficiencies are mainly linked to the value of the magnetic gradient and to the dimensions of the region in which it is sufficiently uniform. The maximum value one can reach for the magnetic gradient depends on the nature of the ferromagnetic material used for the magnets—more precisely on its magnetic saturation. For what concerns the second question, it is possible to get a quite uniform region by working on the geometry of the polar expansions; in 1938 RABI, MILLMANN and ZACHARIAS<sup>(14)</sup> built up a Stern-Gerlach apparatus which could reach a uniform gradient of  $10^5$  G/cm in an extended region. With such apparatus it is possible to get deflections of the order of some centimetres in correspondence of the different values of  $J$  components, if the length of the apparatus and the distance of the detectors from them are of the order of 1 meter (sect. 6'3).

In such conditions the detection of the atoms may be performed, for instance, by hot-wire detectors (D), developed by TAYLOR<sup>(15)</sup> on the basis of studies by LANGMUIR and KINGDON<sup>(16)</sup>. The atoms strike against the hot tungsten wire and get ionized; then they are collected by the first dinode of an electromultiplier tube, emitting one or more electrons which give rise to the electric signal; this must be analysed in coincidence with the signal due to the other atom coming from the same molecule—if we take into account the possible delay.

We conclude this section observing that one needs free paths of the molecules and of the dissociated atoms comparable with the linear dimensions of the whole experimental apparatus; for this reason it is necessary to work with pressures below  $10^{-6}$  mm Hg, that may be obtained by diffusion pumps or similar devices.

## 6. — Details of the experiment.

6'1. *Geometry of the experimental set-up.* — We choose the centre-of-mass reference frame. An energy gap of about 0.7 eV exists between the dissociation

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<sup>(14)</sup> For details about the features of this kind of apparatus see the original paper by J. M. B. KELLOGG and S. MILLMAN: *Rev. Mod. Phys.*, **18**, 323 (1946); a specific and wide list of references may be found in N. F. RAMSEY: *Molecular Beams* (Oxford, 1956).

<sup>(15)</sup> J. B. TAYLOR: *Zeits. Phys.*, **57**, 242 (1929); **35**, 375 (1930).

<sup>(16)</sup> E. LANGMUIR and K. H. KINGDON: *Proc. Roy. Soc.*, **107 A**, 61 (1925).

vibrational state in the electronic  ${}^2\Pi$ -state and the dissociated state  $\text{N}({}^4S) + \text{O}({}^3P)$  (fig. 3). After the decay this binding energy appears as kinetic energy of the dissociated atoms.

Moreover, before predissociation the two atoms have a rotational energy, linked to the value of the total angular-momentum number  $J$  of the molecule,

$$(29) \quad 2I\omega = \hbar J(J + 1),$$

where  $I$  is the moment of inertia of the molecule.

As we said in sect. 3, the value of  $J_{\text{mol}}$  is not determined in the molecular state; in fact,  $J$  can assume all the values

$$(30) \quad J = K + S, K + S - 1, \dots, |K - S|,$$

where

$$K = A, A + 1, A + 2, \dots$$

For the NO case—in accordance with the Krönig selection rules—we have

$$J = \frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \dots,$$

corresponding to states with different multiplicity.

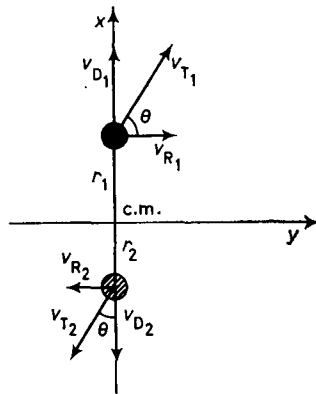


Fig. 5.

It is immediate to conclude that different decay angles correspond to different  $J$  values. The trajectories of the two atoms after the decay are straight lines with constant velocities  $v_{T_1}$  and  $v_{T_2}$  (fig. 5). We stress here that the decay is isotropic and consequently the direction  $x$  in fig. 5 is not fixed. We can calculate  $v_{T_1}$ ,  $v_{T_2}$  and  $\theta_1$ ,  $\theta_2$  in correspondence to the different values of  $J$  by taking into account energy, momentum and angular-momentum conservation laws.

With simple calculations we get

$$(31) \quad \begin{cases} v_{D_N} = 2270 \text{ m/s}, & v_{D_O} = 1980 \text{ m/s}, \\ v_{R_N} = r_N \omega(J) = r_N \frac{\hbar J(J+1)}{2I}, \\ v_{R_O} = r_O \omega(J) = r_O \frac{\hbar J(J+1)}{2I}, \end{cases}$$

and

$$(32) \quad \begin{cases} \theta_N = \arcsin(v_{D_N}/v_{R_N}), \\ \theta_O = \arcsin(v_{D_O}/v_{R_O}), \end{cases}$$

where  $v_D$  and  $v_R$  are, respectively, the dissociation and rotation velocities, and  $\theta_N$ ,  $\theta_O$  the decay angles of the two atoms in the centre-of-mass reference frame.

We want to remark only that, when the orientation  $\varphi$  of one of the measurement apparatus with respect to the beam direction is chosen, the other apparatus must be oriented at an angle  $90 + \theta_N + \theta_O + \varphi$ .

We recall that these calculations are related to the centre-of-mass reference frame; it is trivial to transform to the laboratory reference frame by adding to both  $v_T$ 's the centre-of-mass velocity fixed by the velocity selector (VS).

**6.2. Frequency of the events.** — The number of molecules that dissociate per unit time by fotoactivation is expressed by the following relation as a consequence of Beer's law:

$$(33) \quad \frac{d \ln n}{dt} = \frac{3}{2} KI_0 \left( \frac{\exp[-\alpha N_0] - 1}{N_0} \right),$$

where  $n$  is the number of dissociated molecules,  $K$  is a constant,  $I_0$  the intensity of the radiation,  $\alpha$  the absorption coefficient and  $N_0$  the total number of irradiated molecules.

We can express  $N_0$  as follows:

$$(34) \quad N_0 = \Delta Q \cdot \frac{l}{v},$$

where  $l$  is the length of the beam submitted to the radiation and  $v$  the molecular velocity;  $\Delta Q$  is the number of molecules emitted per unit time in a small solid angle  $\Delta\omega$  in the direction of the beam (fig. 6) and may be determined by the following relation:

$$(35) \quad \Delta Q = \frac{\Delta\omega}{4\pi} N \bar{v} \cos\theta A_s,$$

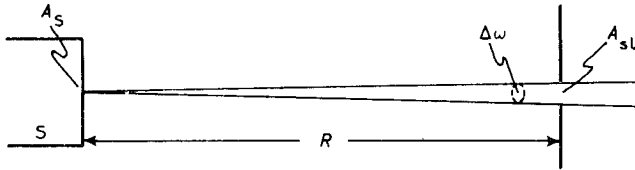


Fig. 6.

where  $A_s$  is the area of the source slit,  $\bar{v}$  the mean velocity of the molecules,  $\theta$  the angle relative to the normal to the plane of the slit and  $N$  the density of the molecules into the source. Moreover,  $\Delta\omega$  can be expressed as follows:

$$(36) \quad \Delta\omega = A_{s1}/R^2,$$

where  $A_{s1}$  is the area of the equivalent slit of the whole focusing and velocity selection apparatus. (The centre of  $A_{s1}$  is on the straight line coming from the centre of  $A_s$  and normal to the plane of  $A_s$ .)

Just to have an order of magnitude we have calculated that for a molecular beam having velocity of about 500 m/s and striken over a length of 2 mm by the radiation emitted by the source used by FLORY and JOHNSTON in their experiment<sup>(17)</sup> (see ref. (12)) the number of molecules which predissociate per unit time is  $10^5$  molecules/s. The above-mentioned source was a weak one compared with the intensity we can actually get, for instance, using synchrotron radiation, by which one could considerably increase the number of molecules dissociated per unit time of a  $10^2$  factor.

**6.3. Deflection and detectability of the atoms.** — For the case of the Stern-Gerlach apparatus considered in sect. 4 the mean deflecting force acting on one atom is

$$(37) \quad \bar{F}_z = g\mu_B J_z \frac{\partial B_z}{\partial z},$$

where  $z$  is the direction of the  $\mathbf{B}$  vector,  $J_z$  the component of  $J$  along this direction,  $g$  Lande's factor and  $\mu_B$  the Bohr magneton.

If  $v$  is the velocity of the atoms which get into the Stern-Gerlach apparatus, then the deflection is expressed by the relation

$$(38) \quad \Delta z = \bar{F}_z l(s + l/2)/Mv^2,$$

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(17) We stress here that the decay time of predissociation is of the order of  $10^{-11}$  s and consequently with a velocity of 500 m/s every molecule runs for a space of  $10^{-7}$  cm before it predissociates: this is perfectly negligible in the framework of the decay's geometry.

where  $M$  is the atom mass,  $l$  the Stern-Gerlach length and  $s$  the distance of the detector from the end of the SG (see fig. 7).

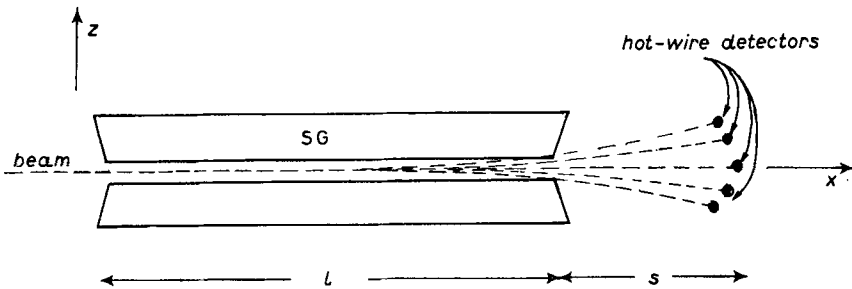


Fig. 7.

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### ● RIASSUNTO

In questo lavoro si propone un nuovo test sperimentale della meccanica quantistica, relativo a misure di correlazione per sistemi composti. Il decadimento in due corpi non radiativo nella predissociazione molecolare permette — nel caso particolare della molecola NO che qui è trattata in dettaglio — un test di gran lunga più sensibile degli usuali esperimenti a due fotoni. Si discute anche un possibile modo di realizzare l'esperimento, verificandone la fattibilità negli aspetti più importanti.

### Экспериментальная проверка квантовой механики в молекулярной преддиссоциации.

**Резюме (\*).** — В этой статье мы предлагаем новую экспериментальную проверку квантовой механики, которая относится к корреляционным измерениям в составных системах. Нерадиационные двухчастичные распады в молекулярной преддиссоциации представляют — в частном случае для молекулы NO, который мы исследуем подробно — более чувствительный тест, по сравнению с обычными двухфотонными экспериментами. Мы обсуждаем возможный путь проведения эксперимента и анализируем реализацию такого эксперимента.

(\* ) *Переведено редакцией.*