Abstract

Availability of short, femtosecond laser pulses has recently made feasible the probing of phases in an atomic or molecular wave-packet (superposition of energy eigenstates). With short duration excitations the initial form of the wave-packet is an essentially real "doorway state", and this develops phases for each of its component amplitudes as it evolves. It is suggested that these phases are hallmarks of a time arrow and irreversibility that are inherent in the quantum mechanical processes of preparation and evolution. To display the non-triviality of the result, we show under what conditions it would not hold; to discuss its truth, we consider some apparent contradictions. We propose that (in time-reversal invariant systems) the preparation of "initially" complex wave-packets needs finite times to complete, i.e., is not instantaneous.

1 Introduction

A quantum mechanical system developing in ordinary Hilbert Space according to the time dependent Schrödinger Equation does not exhibit the time arrow. (Formal description of time-reversal invariance is found in almost every text on quantum mechanics, e.g. [1], and is essentially implied by the equality of the absolute magnitude of the wave function developing from zero time at positive and at negative times.) On the other hand, the
"time arrow" is manifest both in the subjective feeling of the passage of time and in statistical mechanics involving macroscopic systems [2-4]. In some recent studies it was therefore proposed to adopt a Rigged Hilbert Space, in which unidirectional evolution, decay of an isolated elementary excitation and evolution of the universe are all possible [5-6]. We wish to focus on "wave-packets", "doorway-states" and "observable phases" and to show that, under circumstances that these are applicable, the quantum mechanical evolution exhibits an experimentally verifiable time arrow from the prepared state. The verifiability is made possible by recently developed or perfected optical techniques for wave-packets of atomic Rydberg states [7-8] and of molecular wave-packets [9]. (The quoted works are recent samples of a multitude of relevant references over the past few years.) In turn, these techniques are based on the possibility of experimental observation of phases by use of coherent radiation (lasers) pulses of femtosecond duration, which is the range appropriate to Rydberg-state and molecular vibrational spectroscopies.

Let us examine the relationship between phase and time-direction in the light of conventional treatments: The main sources for time-reversal invariance (or its violation) are collisions between elementary particles [10]. In that (quantum field theoretical) context, creation and destruction of particles is allowed and time reversal (denoted by T) is normally considered in conjunction with space inversion (P) and charge conjugation (C). The invariance of the combination CPT is assumed in all interactions [11] while based on experiments, CP or T is currently believed to be conserved in strong and electromagnetic interactions, but slightly violated in weak interactions [12, section 3.1]. In this work we shall use a single-particle (not field-theoretical) description with a scalar wave-function that obeys a time dependent Schrödinger equation, and we assume that the Hamiltonian of the system is time-reversal invariant. It then follows from the Schrödinger equation (or from the non-relativistic Lagrangian) that time reversal is equivalent to an inversion of the phase [1]. This equivalence makes clear the relevance of recent phase determinations to the question of time arrow.

The essential features of the physical systems on which our result depends are described in the next section. The whole issue is rephrased more cautiously in the following section. Problems and questions that remain are treated in the remainder.
2 Short time preparation of the wave packet

We start by considering (for the sake of simplicity) a time independent Hamiltonian without a magnetic field. We add a sharp perturbation $\Delta H_{opt}$ that causes a jump ("the preparation") from an initial stable state $|0\rangle$, which can be considered to be real. (For a dipole transition $\Delta H_{opt} = e{\mathcal{E}} \cdot \vec{r}$, where $\mathcal{E}$ is a real electric field and $\vec{r}$ is the electron coordinate.)

The following is a typical scenario, due originally to Feshbach et al. for scattering in nuclear matter [13], but later adopted and extended for excitations into electron-vibrational states in molecules [14-18] Under circumstances of moderately dense bunching of the discrete excited states (such as would exist in excitation of Rydberg states [7] or of vibrational states [9]) and a weak coupling to an adjacent continuum [14], the excitation is into a doorway-state given by

$$|D \rangle \propto \Delta H_{opt} |0\rangle \quad (1)$$

When expanded into real energy-eigenstates, this can be written as

$$|D \rangle = \sum_r a_r |r\rangle \quad (2)$$

where (at $t = 0$) all $a_r$ are real (except possibly for a common phase), since $\Delta H_{opt}$ arises from a real field. The sum is over those energy eigenstates that are included in the spectral width of the excitation. The physical condition for the validity of the doorway description is that the duration of the excitation is shorter than all relevant decay times. In this work, we shall call (for brevity) preparation processes achieved under this condition "instantaneous" or "occurring at $t = 0" (though, of course, the time-energy uncertainty principle forbids state-selective excitations to be made instantaneously). Rhodes [16] and Nitzan and Jortner [17] have derived equations (1) and (2) for excitation times that are of the order of the inverse spectral width, which is the shortest excitation time compatible with the time-energy uncertainty relation.

In contrast to the situation at $t = 0$ when the $a_r$ are real, for $t > 0$ the coefficients become complex, with phases that differ for each component. The precise formulae for $a_r(t)$ are given in [16-17] (and show these to be complex), but for the simplest case, when the energy eigenstates are uncoupled to any other state (discrete or in a continuum), the phases grow in time proportionally to the energies of the r-states. We now come to an essential point in our argument, namely, that this acquisition of phase is irreversible in the sense that it is not possible to prepare with $\Delta H_{opt}$ a wave-packet...
whose coefficients are complex at $t = 0$ and become real at $t > 0$. (We shall show this in more detail in the sequel.) Moreover, the phases are observable (through coupling the system to a phase-interferometer device, as was done, e.g., in [7]).

Also for $t > 0$, when the $r$-states in the sum in (2) interact with states outside the summation, certain coefficients decrease in an analogous way to the behavior of a quasi-stable particle [6]. (For a system in which the states outside the summation form a dense energy level scheme the "decrease" takes a time-exponential form, but the decrease, subject to Poincare recurrences only [18], exists also for microscopic systems in which the $r$-states interact with discrete states outside the summation.) The decrease is time-symmetric, since the absolute value of $a_r$ is even in $t$, but the phase change of the coefficients is time asymmetric.

In summary, we have used a schematic description to argue that wave packets prepared in a physical way (and moving in the full Hilbert space) are by quantum mechanics time-asymmetric and irreversible.

3 A Reformulation

In the light of the foregoing we can restate our result, while phrasing our argument somewhat more precisely.

We start with a suggested definition of reversibility.

A description may be said to be reversible, if the following state of affairs holds: Whenever a "picture" (being a formal description of the physical situation) $P_1$ at $t_1$ is followed by pictures $P_2$ at $t_2$, $P_3$ at $t_3$, ..., $P_N$ at $t_N$, then the description also allows that $P_N$ at $t_1$ will be followed by $P_{N-1}$ at $t_1 + t_{NN-1}$, ..., $P_2$ at $t_1 + t_{N2}$, $P_1$ at $t_1 + t_{N1}$ ($t_{rs} = t_r - t_s$), i.e. at the same time intervals with the order of events taken in the reverse sense. The times $t_r$ are arbitrary and not just some times selected in a way that depends on the properties of the system.

[Two illustrations of the definition:
(a) (A time-reversal invariant case) A classical particle, having coordinate $q$, that moves under a constant force $f$ between times $t = t_1 = 0$ and $t_2$ is subject to the equation of motion: in suitable mass units $\frac{d^2q}{dt^2} = a$. This has a solution $q = \frac{1}{2}at^2$, taking at $t_1$ the values: $q = 0, v = 0$ (velocity), and at $t_2$: $q = \frac{1}{2}at_2^2, v = at_2$. Starting at $t_2$ with a reverse velocity, the solution is $q = \frac{1}{2}a(t - 2t_2)^2$, which has the following values at a time $t$ which is $t_2$ later (namely, $t = 2t_2$): $q = 0, v = 0$. This confirms reversibility for all choices of...
(b) (No time-reversal invariance). A particle moving in the \(x - y\) plane subject to a magnetic field \(B\) perpendicular to the plane. The velocity components satisfy 
\[
\frac{dv_x}{dt} = v_y B, \quad \frac{dv_y}{dt} = -v_x B,
\]
with solutions \(v_x = \sin B t\), \(v_y = \cos B t\). The values are at \(t = 0\): \(v_x = 0, v_y = 1\) and at \(t = t_2\):
\[
v_x = \sin B t_2, \quad v_y = \cos B t_2.
\]
The other solution commencing at \(t = t_2\) is again \(v_x = \sin B t\), \(v_y = \cos B t\), whose values at \(t = t_2\) are identical to those of the previous solution. However, at a time which is \(t_2\) later \((t = 2t_2)\):
\[
v_x = \sin 2B t_2, \quad v_y = \cos 2B t_2
\]
and this can be made to agree with the values of the first solution at \(t = 0\) only for special choices of \(t_2\). This system does not therefore satisfy our criterion for time-reversal invariance. By changing the sign of \(B\), as is done in a spin echo experiment [2], one gets the solution \(v_x = \sin B (2t_2 - t), v_y = \cos B (2t_2 - t)\) and this indeed takes the values of the original state at both times, but this involves a change in the system.

We now return to the wave-packet. For definiteness, we take the case that \(P_1\) is the physical state (as represented by the many-component wave-function in (2)) immediately after preparation of the wave-packet at \(t_1 = 0\) by a short-duration excitation, and for that later pictures we take just one of the several \(P_1\)’s, and call it \(P_{>}\), the picture of the physical state at some arbitrary later time \(t_{>}\). Then we hope that the argument in the previous section has shown that the picture reversal cannot hold, namely, \(P_{>}\) cannot be followed by \(P_1\) at \(t_{>}\), Thus the description in question (quantum mechanics, including preparation and observation) is not reversible. The essential difference is of course the absence of phases in \(P_1\) and their presence in \(P_{>}\). The former occurs at \(t = 0\) and then only (under the mode of preparation here envisaged), and the latter at \(t > 0\).

Have we proven a time arrow, or only a non-equivalence between the time of preparation and time of observation? If the sign of the phase is observable (a proposition which we shall later put forward), then, given the fact of lower-boundedness of energies, there is no question that measurement of the phase establishes the time-arrow. However, even if the sign is unknown, it is clear that the time direction of going from real-to-complex states is uniquely aligned with earlier-to-later. This follows, since the observation of a prepared state must take place at a time posterior to its preparation. Under doorway-excitation conditions phases will be observed at later times than their preparation-time and, indeed, one will get successively larger phases (at least initially) as one carries out the observation at successively later times.
4 Is the result trivial?

We now raise the question, which ingredients in the description would have to be removed, so as not to have a time arrow (or, to have "time-reversal symmetry")? (We are now trying to show that our result is not trivial.)

(1) If the phases were not observable, only the populations of the components, then the pictures $P_1$ and $P_2$ would be identical and our demonstration would fail. Though strictly this is true only for a finite number of component states (energy eigenstates) that are not in overlap with a continuum (recall our earlier remarks about the decrease of the absolute value of $a_r$ when the r-states are coupled), the crucial significance of phase is already made clear.

(2) In classical mechanics, one can start at $t = 0$ with picture $P_1$ which could (as an example) be the state that two particles which repel each other are placed at some distance to each other and then get a picture $P_2$ at $t_2$, in which the particles are at a farther distance from each other. However, one could also start at $t = 0$ with the picture $P_2$, impart to the particles velocities that are opposites to those originally possessed by them at $t_2$ and then observe $P_1$ at $t_2$. Thus in classical, phase-less mechanics our claim does not hold. (Note also that, in the short-duration wave-packet preparation process from $|0 >$, one does not have the luxury of starting the process with negative phases: At $t = 0$ there are just no relative phases of any sign. In the sequel we shall consider a different preparation process, one which appears to be starting with non-zero phases, but in fact does not do so.)

(3) The previous parenthetical remark is invalid in a long-duration preparation process or in preparations with one or more time delays. Then phases can be introduced in any desired manner ("wave-function tailoring", [11]). If this situation could be assumed to take place at $t=0$, then one could argue that one has started with $P_2$ and ended up at $t_2$ with $P_1$. (The bold faced phrase "one could argue" is an expression of the possibility that the signs of the phases cannot be measured. The bulk of present-day literature treats the experimental determination of only the magnitude of phase-differences and not their sign. The "observability" of phase is discussed by [20] and [21]. Recently, it has been shown that in certain circumstances the analytic properties of the time-dependent wave-function can lead to the establishment of a unique and unambiguously signed phase [22]. Based on a discussion with Y. Aharonov, it appears that experimental determination of the sign of phases is, in principle, possible.) It seems, however, that this situation ($P_2$ preceding $P_1$) cannot be regarded as the time-reversed counterpart of
the doorway-state excitation, since the preparation modes are essentially different. Thus, the doorway-state preparation is "instantaneous", while the creation of relative phases requires a finite time.

(4) An alternative definition of time reversal invariance (one that, in our view, would not do justice to the common understanding of the term), different from that given at the beginning of this section, would not suffice to establish our result. Thus, if time reversal invariance were defined as the possibility of the reversal of pictures at some (rather than at any) later time, than the revival time of a wave-packet [18], or a Poincare recurrence time, would constitute a time starting from which phase growth could occur backwards.

5 Apparent exceptions

There are, however, cases of non-zero phases in a prepared state and these should be noted. (We argue that they essentially differ from the "initial state", as employed in the previous sections.)

(a) On its face, it might have been possible to achieve a "phased" doorway state by applying $\Delta H_{opt}$ onto the state $|0> + A|1>$, rather than as in (1) (Such a "phase control, using a coherent laser source, was suggested some time ago [23-24] and elaborated on in several works more recently [7-8].) The coefficient $A$ can have any desired magnitude and phase, through choices of the laser intensity and of the time delay between the applications of the laser on $|0>$ and the later application of $\Delta H_{opt}$ on $|0> + A|1>$. However, the "phased doorway state" obtained in the aforesaid manner is not a "prepared" state in our sense, since it does no arise at $t = 0$ (which is defined as the moment of preparation) but only after a time delay. The "time-arrow test by exploration of the phases" is to be conducted at the moment of preparation ($t = 0$) and at a moment of observation, which is different from $t = 0$, rather than at two times different from zero. Moreover, (not only the preparation of a complex wave-packet, but also) the observational check that a wave-packet state has complex coefficients requires a prior procedure (through the employment of appropriate time delays) that takes place some time before the moment of observation on the wave packet. In other words, no instantaneous checking of phases is possible, either.

(b) A broad class of wave packets that have complex coefficients have been given by a large number of workers.(E.g., [25,26]) These states have a non-zero initial momentum. In the next section ("An application") it will be
shown that to prepare such a complex wave packet, one needs an excitation that has a finite time duration, or a time dependent coupling mechanism.

(c) A free particle state of the form $e^{ikx}$ is necessarily complex. (In other words, the amplitudes of position eigenstates are complex in a momentum eigenstate.) Its special status may be due to its not being a bound state, and indeed if the free particle is confined to a box with reflecting (or absorbing) boundaries, then the amplitudes become real.

(d) Complex states $e^{im\theta}$ (where $2m$ is an integer and $\theta$ an angle) can arise with magnetic excitation. The preparation involves a Hamiltonian that is not time-reversal invariant and is excluded from our present considerations.

6 An application

In the field of molecular wave-packet dynamics, complex "initial" wave-packet states are frequently employed [25,26]. A preparation process for a diatomic molecule has been recently described with great clarity and with provision of considerable formal details [9]. Here we give a simplified and condensed description, using a wave-function rather than a density-matrix formalism. The essential point for the present purpose is the non-instantaneous preparation of a complex state and this is clearly noted also in [9]. The accompanying drawing (Figure 1) is a schematic version of Figure 2 in [9].
We consider a wave packet, which is prepared in the form:

\[ \Psi(q) = N e^{ikq} e^{-M\frac{(q-q_o)^2}{2}} \]  

Here \( q \) is the coordinate of a particle (which could be, say, an atom of mass \( M \) constrained to move inside a parabolic potential wells centered in \( q_o \) in the electronic ground state, and inside a parabolic potential centered at some other point in an excited electronic state, to which it is raised by an electromagnetic field), \( \omega \) is the frequency of harmonic motion, \( N \) is a normalizing factor and \( k \) is the initial momentum of the particle. (\( \hbar = 1 \).)

The complex form of the initial state is apparent in (3). In the sudden approximation for the excitation process, its representation by the (real) eigenstates \( \phi_n(q) \) of the upper potential is

\[ \Psi(q) = \sum_n (a_n + ib_n) \phi_n(q) \]  

where \( a_n = \langle \text{Re}\Psi(q)|\phi_n(q)\rangle \), \( b_n = \langle \text{Im}\Psi(q)|\phi_n(q)\rangle \)

A physical process which ensures the creation of the wave packet (3) or (4) is the application in the ground (electronic and vibrational) state of several coherent (real) electric fields. It was shown in [24] that when a sharply pulsed laser light is applied, each coefficient \( a_n \) or \( b_n \), though initially time varying, will settle down to its long-time value after a time exceeding the pulse duration (which may be as short as several femtoseconds). We represent the real field (during the pulse) by

\[ E(\Omega) = E_0 \cos(\Omega t) \]  

whose oscillatory frequency is \( \Omega \). The field creates a nonzero transition dipole between the ground and the excited electronic state. To obtain the wave packet (4) at, say, \( t_0 \) we need to apply a set of fields some of which, it turns out, have to be advanced in time. First, to construct the \( a_n \) part of the wave packet, we apply at \( t = t_0 \) the superposition

\[ E_1 = \sum_n \left( \frac{a_n}{F_n} \right) E(n\omega + \Delta) \]  

Next, to obtain the imaginary \( b_n \) terms we apply at \( t = t_0 - \delta t_n \), a further set of fields consisting of the sum of fields

\[ E_2 = \sum_n \left( \frac{b_n}{F_n} \right) E(n\omega + \Delta) \]
In the above $\Delta$ is the energy gap between the two electronic states, $F_n$ is the Franck-Condon amplitude (=square-root of the F-C factor) for transitions between the 0 and a displaced n’th vibrational level. (For F-C factors see [15, 27].) The time-advance $\delta t_n$ for the excitation time in (7) is such that later, at $t_0$, the phase factor of the excited level at the height $n\omega + \Delta$ is just

$$e^{i\pi/2} = i$$

and this is achieved by so choosing $\delta t_n$ that

$$(n\omega + \Delta)\delta t_n = \frac{\pi}{2} \mod(2\pi)$$

The required time advances need to be longer than the pulse width, and this is feasible with femtosecond pulses. Also, there is no problem, in principle, of letting light from a coherent source arrive at different times and intensities, employing path deflection into dielectric and partly absorbing channels.

It is now elementary to check that the "initial" state (i.e., the state at $t_0$) in (4) is reconstructed, but only by starting the preparation process earlier than $t_0$. This example illustrates our earlier statement that the preparation of a complex doorway state cannot be achieved instantaneously.

7 A paradox

A nutshell summary of our results might be: One-time preparation means no time reversal; multiple-time preparation can exhibit time reversed development.

It may seem odd that touching the system just once gives a time-direction, whereas manipulating it at subsequent times restores the time reversal invariance.

However, classical systems show just this behavior, as we shall now illustrate. Suppose a (classical) particle is set into motion at $t = 0$ in a Universe that is such that the masses of particles decrease by a fixed fraction after each second. Obviously this Universe contains a time-arrow (just as our Quantum Universe does). The position of the "kicked" particle is shown in Fig. 2 as function of time up to 5 seconds with the upward curve. (This is a sum of five straight line fragments, with successively increasing slopes.) Trying to obtain a time-reversed motion, we cannot just change the sign of the velocity which was present at 5 seconds and let go, as is shown by the downward thin line. Rather, we must interfere at the tick of each second
Figure 2: Position (in velocity-seconds unit) as function of time (in seconds), of a classical particle whose mass decreases by a fixed fraction \((\frac{1}{4})\) after each second, plotted for the first five seconds. The upward curve represents free motion with an initial positive velocity. The thin downward curve shows the position of the same particle if this starts from its position after five seconds and is given a velocity which is the opposite to that which the particle had at that instant, and is then left moving. The motion is not the time-reversed one of the previous. The thick downward curve shows the position, if the particle velocity is adjusted after each second so as to have the opposite velocity to that in the upward curve. The motion is now the time-reversed one to that in the upward curve.

to adjust the speed (downward thick lines in Fig. 2). Thus, in a classical system, too, time reversal can be got artificially, so to speak, by repeated manipulation.

8 Future directions

The following points need further consideration: Is time-asymmetry a special phenomenon that occurs only in the doorway-state spreading, or do asymmetry and irreversibility occur more generally, under any state-preparation procedure. If the latter is the case, how can it be demonstrated? In particular, the relation to time arrows in particle physics and involving basic forces (but exclusive of the gravitational one) need to be discussed. As already noted, our result depends on the practical feasibility of short time pulsed preparation of the initial wave-packet state (also known as the sudden approximation). In collision or scattering of nucleons and elementary particles the preparation of the incoming state normally starts at \(t = -\infty\) and takes
place adiabatically. The difference in the two situations for fundamental processes was emphasized recently [28]. A suggestion to apply at \( t = 0 \) a ‘pulse’ of particles to bombard a stable nucleus was made some time ago by Khalfin [29]. We are not aware of any phase or other measurements that arose from the suggested experiments. More recently, the theory of T.D. Lee [30] about spontaneous T-violations through phase interference has received renewed attention with a view to experimental tests [31,32]. These ideas are clearly beyond the scope of the present paper, which deals with established technology.

How is the above “molecular” irreversibility, occurring after preparation, connected with either the statistical-mechanical time arrow, or with the irreversibility entailed by a measurement process, or with the irreversible processes considered in [5-6] (decay of elementary excitations and evolution of the Universe)?

The foregoing theory has to be fitted in with the description, originally due to Aharonov and coworkers [33], of retrodiction from a performed measurement.

9 Experiments

Recent experiments on Rydberg-states appear to confirm the existence of a ”doorway state”, but one of the wave-packets in the experiments of [7] appears to have one non-zero phase (of \( .15\pi \)), in apparent opposition to the theory. The seven other components have real coefficients as required by the foregoing theory. It appears that the complex coefficient is due either to a time-advanced mode of preparation of the wave packet or to the phase determination being made at some later time than the creation of the wave packet.

It has also been remarked to us that if the preparation and (subsequent) observation procedures are ”put in”, then the time arrow must necessarily ”come out”, since the ”extended” Hamiltonian is time-dependent (rather than time-reversal invariant, as is the Hamiltonian of the measured system). This, however, only means that the formalism is consistent upon extension of what is meant by a ”system”. The essential point remains that the wave function of the measured system is observably different at an initial time (the moment of preparation) and at a later time (the moment of observation), with the difference showing up through the relative phases in the component amplitudes.
10 Conclusion

It has been shown that when a wave-packet is prepared in currently practiced ways, such as described in [7-9], its "properly initial" and its later-time states differ by the acquisition of phases, which are odd in \( t \) (time). Recently developed techniques for atomic and molecular wave-packets can test for phases and, by consequence, for time direction. Even if it is doubted that phase signs are observable, in wave-packets that are prepared via "doorway-state" excitation the weaker, but certainly verifiable reality - complexity property of amplitudes is uni-directionally aligned with a time-arrow implied by the preparation-observation procedure [6].

A further proposition of this work, supported with examples, is that, in systems with time-reversal invariant and spatially bound Hamiltonians, complex (=phased) wave-functions cannot be prepared (or measured) instantaneously.

Several issues have been relegated for future work. This ought to show how the results of this paper will appear when translated from the specific setting of atomic and molecular wave-packets to more general ones.

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state could be produced that would be odd under both P and CP. Once you excited such states, you could observe parity odd correlations of the product particles. At least one of the RHIC detector groups, STAR, will search for P and T violations” (Quote from p. 23.)

[33] Y. Aharonov, P.G. Bergmann and J.L. Leibowitz, Phys. Rev. 134B (1964) 1410