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LETTER TO THE EDITOR

Observational line broadening and the duration of a quantum jump

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Abstract. Experimental observations of 'quantum jumps' suggest that one can associate a timescale shorter than 'lifetime' with quantum transitions; one such 'jump time' is proposed here. We also find that experimental temporal localization of the jump *does* affect it (a lá Bohr), and predict an 'observational line broadening'.

1. Introduction

How quick is a quantum jump? Experiments performed 10 years ago [1] in which jumps were observed permit concrete meaning to be attached to this question. An atom in a (long-lived) metastable state is bathed in light at the frequency of another of its transitions. Departure from the metastable state is marked—*measured*—by the onset of fluorescence at that other frequency. This onset is sudden at the time resolution of the experiments. Presumably, the passage from the metastable state to the ground state occurs on a scale more rapid than the lifetime, just as for quantum tunnelling. There has been a great deal of interest [2] in assigning a 'tunnelling time' to the latter sort of transition. Can any sense be made of the more general case?

By way of confirmation, in the aforementioned experiments the lifetime of the metastable state was checked, and found to agree within experimental error with values previously known. One might ask, according to the views advocated by Bohr, whether the 'observation' of this decay should not affect it in any way.

We show in this letter that observation of the decay *does* affect it, and that this effect is a line broadening (or splitting) that increases as the precision of temporal localization—the quickness of the jump—increases.

In the first part of the letter we discuss the timescale for the quantum jump, then provide corroborative evidence for this result, followed by estimates of minimal jump times for certain transitions. In the second part we calculate the changes in the properties of the transition due to its being under observation, and in particular consider the changes in the line shape—what we call 'observational broadening'—due to this observation. We also discuss points related to an experimental search for this effect.

2. Jump time

The definition of the timescale for a quantum jump is based on the idea that if a system is disturbed at intervals Δt , but those disturbances do not affect the decay, then Δt is longer

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than the jump. On the other hand, if one does manage thereby to affect the decay, then its time scale is reached. As the disturbance to be used in the definition we take a projection onto the original state—a check on whether the system has decayed. In this way we arrive at a formal context similar to that in the so-called quantum Zeno effect (QZE) [3,4].

Let the system begin in a state ψ and let the full Hamiltonian be H. After a time Δt , ψ evolves to $\exp(-iH\Delta t/\hbar)|\psi\rangle$. One checks for decay by applying $\langle \psi |$. The probability that it is still in ψ is $p(\Delta t) = |\langle \psi | \exp(-iH\Delta t/\hbar) |\psi\rangle|^2$. By familiar manipulations [5] this can be written

$$p(\Delta t) = 1 - \left(\frac{\Delta t}{\tau_Z}\right)^2 \mathcal{O}(\Delta t^4) \tag{1}$$

with $E_{\psi} \equiv \langle \psi | H | \psi \rangle$ and

$$\tau_Z^2 \equiv \frac{\hbar^2}{\langle \psi | (H - E_\psi)^2 | \psi \rangle}.$$
(2)

Call τ_Z the 'Zeno time,' notwithstanding the lack of full concurrence with the classical allusion [5].

Suppose many projections are made during a time t, carried out at intervals Δt . Then to leading order at time t the probability of being in ψ is

$$p_{\text{Interrupted}} = \left[p(\Delta t)\right]^{t/\Delta t} = \left[1 - \left(\frac{\Delta t}{t}\frac{t\Delta t}{\tau_Z^2}\right)\right]^{t/\Delta t} \approx \exp(-t\Delta t/\tau_Z^2).$$
(3)

To define a jump time, we want to know whether this differs from standard decay. Without projections the probability for being in ψ is

$$p_{\text{Uninterrupted}} = \exp(-t/\tau_{\text{L}})$$
 (4)

with τ_L the usual lifetime. When the 'Golden rule' can be used τ_L is given by

$$1/\tau_{\rm L} = 2\pi\rho(E_{\psi})|\langle f|H|\psi\rangle|^2/\hbar \tag{5}$$

with $|f\rangle$ the final state, and ρ the density of states (at the energy of the initial state). Comparing equation (3) and (4), we see that the interrupted decay will be slower for $\Delta t < \tau_Z^2/\tau_L$. (Reversing the inequality suggests *faster* decay, but in fact signifies breakdown in the expansion.) We are thus led to define the 'jump time' as the time for which the slow down would begin to be significant, namely

$$\tau_{\rm J} \equiv \tau_{\rm Z}^2 / \tau_{\rm L}. \tag{6}$$

In words, τ_J is the time such that if one inspected a system's integrity at intervals of this duration, the decay would be affected.

We offer two pieces of corroborative evidence for equation (6). First, for barrier penetration one does have an idea of what the tunnelling time is and it ought to be the same scale as the jump time. In [5] there is a calculation of the Zeno time, and it was found to be related to the tunnelling time in the following way: $\tau_Z^2 = \tau_L \tau_T$, with τ_T the semiclassical tunnelling time. Second, in [6] there is an explicit theoretical calculation of a quantum jump. That is, a system ordinarily decays at a certain rate, but when prepared in a special way it can be forced to decay much more quickly. The relation between the various times—the lifetime, the forced-decay time and the inverse of the appropriate matrix elements of the Hamiltonian—are found to satisfy equation (6). (More details are found in [7] and the dramatic 'jump' is illustrated in the figures in [6].)

An interpretation of τ_J in terms of bandwidth and uncertainty relations can be found by combining equation (2), for τ_Z , with (5), for lifetime. After some manipulation one obtains

$$\tau_{\rm J} = 1 \bigg/ \int \frac{\mathrm{d}E}{\hbar} \frac{\rho(E) |\langle E|H - E_{\psi}|\psi\rangle|^2}{\rho(E_{\psi}) |\langle f|H|\psi\rangle|^2}. \tag{7}$$

Because of the orthogonality of the initial and final states, one can insert a $(-E_{\psi})$ into the Golden rule matrix element. Thus the ratio

$$\frac{\rho(E)|\langle E|H - E_{\psi}|\psi\rangle|^2}{\rho(E_{\psi})|\langle f|H|\psi\rangle|^2}$$

takes the value 1 when E passes through E_{ψ} . In effect, this ratio measures the ability of the Hamiltonian H to move the system away from its initial state. One thus has a band of accessible transition states.

Clearly, τ_J is (the inverse of) an integral over energies (or frequencies) of an order unity function describing the modulation of the band of accessible states. It follows that τ_J is the inverse bandwidth for the transition. This is a completely reasonable conclusion: one would like to create a situation where the system's transition is sudden. The success is governed by the frequencies available. The accessibility of those frequencies is the essence of the bandwidth.

It is of interest to see whether τ_J could be of practical significance, whether there are transitions where one could catch them in the act, so to speak. We will see that for atomic transitions this would be rather difficult, but that in other cases, not only is it possible, but that in a sense it is going on all the time.

First, we make a rough estimate of τ_J for atomic transitions. We start from the standard Hamiltonian $H = (p - eA/c)^2/2m + V + H_{\text{EM, free}}$. Noting, from equation (7), that only transition elements are of interest, to a good approximation we need only be concerned with $\langle 0|\mathbf{p} \cdot \mathbf{A}|\psi \rangle$ with 0 the ground state and ψ the decaying state. From (7) we also see that only the *ratio* of densities of states is of interest, introducing a factor $(k/k_0)^2$ [8] (with k_0 the wavenumber for the $\psi \rightarrow 0$ transition). Finally we note that the momentum k portion of the field operator A contains $\exp(ikr)$, and that r can be written $i\partial/\partial p$. (A also contains a $1/\sqrt{k}$ contribution from its normalization.) Combining all these observations, after little manipulation one finds

$$\tau_{\rm J} = \frac{2\pi a}{c} \frac{1}{I} \qquad \text{with } I = a \int dk \frac{k}{k_0} \frac{|\int d^3 p \, \psi_0^*(p) p \psi(p+k)|^2}{|\int d^3 p \, \psi_0^*(p) p \psi(p+k_0)|^2} \tag{8}$$

with I dimensionless and a a (not yet specified) quantity with dimensions of length.

To obtain a number from this it is convenient to take *a* to be the Bohr radius and use momentum space atomic wavefunctions. For the ground state we use $1/(p^2 + 1)^2$ and for the excited state we take a p-state, with dependence $p_z/(p^2 + 1/4)^3$ (see, e.g., [9]). This leads to integrals that can easily be performed numerically. Combining all relevant terms it turns out that $I \approx 10^2$ so that for τ_J we find the remarkably small value, 10^{-20} s. This brevity, 1/100 the time for the electron to circle the nucleus, is reminiscent of the superluminal transmission one encounters in some tunnelling calculations; however, since there is no specific distance to be covered there is no corresponding paradox. What might be puzzling here would be the suggestion that this short time could imply a correspondingly high energy. We will see below that indeed observation of a short time would bring in new energy scales, not as a property of the atom alone, but in conjunction with the 'apparatus' performing the observation.

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We next turn to situations where the value of the jump time is closer to the experimentally accessible regime and experimental consequences of our definition may be noted. For example, a reasonable bandwidth for phonon mediated transitions is 100 meV. For this

$$au_{
m J} \sim rac{\hbar}{100 \ {
m meV}} pprox 10^{-14} \ {
m s}$$

For such transitions one expects that intervention is possible and would indeed influence the transition. In other transitions there are collective, condensed matter effects that narrow the bands; these should present yet richer possibilities for the realization of effects arising from the finiteness of the jump time.

Probably the best known example of successful intervention is the stability of chiral isomers, attributed to environmental monitoring. In this case it has been shown that a QZE-like effect prevents their passage to non-chiral theoretical ground states. We give more details (plus references) in section 4.

3. Observational line broadening

To calculate the effect of monitoring the system's state we introduce a Hamiltonian description. Much of the calculation is as in [4]. There are three levels: state 1 the ground state, 2 the metastable state and 3 the short-lived state. As Hamiltonian we take

$$H = \sum_{j=1,2,3} W_j |j\rangle \langle j| + \sum_k \omega_k a_k^{\dagger} a_k + \sum_k [a_k^{\dagger} (\phi_k |1\rangle \langle 2| + \Phi_k |1\rangle \langle 3|) + \text{adjoint}]$$
(9)

with $|j\rangle$ the atomic states, a_k photon operators, and ϕ and Φ matrix elements for the corresponding transitions.

To study the decay we solve the time-dependent problem. Initially the atom is in state 2. The electromagnetic field is described by photon number. (Use of coherent states is more customary at this point, but for an intense field photon-number spread is not significant.) The mode corresponding to the laser beam is highly excited. We designate its frequency Ω_0 , which equals $W_3 - W_1$ ($\hbar = 1$ in this section), since it stimulates the 1 \leftrightarrow 3 transition. The initial state of the field is N_0 photons in this mode; all other field modes have zero photons. We thus write the initial state $|2, 0, N_0\rangle$, the '0' in the second argument referring to field modes other than Ω_0 , and the '2' the atomic state (which we designated '|2)' above). From this state, with amplitude ϕ_k , the system can go to states of the form $|1, 1_k, N_0\rangle$. From here, the largest amplitude by far is to go to $|3, 1_k, N_0 - 1\rangle$. This is because, first, $|\Phi| \gg |\phi|$ (since the 3 \leftrightarrow 1 transition is much faster than the 2 \leftrightarrow 1 transition), and, second, because of the factor $\sqrt{N_0}$ that a_0^{\dagger} introduces. Once in $|3, 1_k, N_0 - 1\rangle$, the largest amplitude is to drop right back to $|1, 1_k, N_0\rangle$, now because of the factor $\sqrt{N_0}$ alone, since matrix elements for other-directional radiative decays are also large. However, since our interest is primarily in the first step of this process, the decay $2 \rightarrow 1$, these smaller, later effects do not change our conclusions. The general expression for the state vector is therefore

$$\Psi(t) = \sum_{k} x_{k}(t) |3, 1_{k}, N_{0} - 1\rangle + y(t) |2, 0, N_{0}\rangle + \sum_{k} z_{k}(t) |1, 1_{k}, N_{0}\rangle.$$
(10)

The initial conditions are y(0) = 1, all others 0. This yields the equations of motion

$$i\dot{x}_k = E_k x_k + \sqrt{N_0} \Phi_0^* z_k$$
 $i\dot{y} = \sum_k \phi_k^* z_k$ $i\dot{z}_k = E_k z_k + \phi_k y + \sqrt{N_0} \Phi_0 x_k$ (11)

where by convention we take $W_2 + N_0\Omega_0 = 0$ and $E_k \equiv \omega_k - (W_2 - W_1)$. By taking the Laplace transform, making use of the boundary conditions, and defining $B^2 \equiv N_0 |\Phi_0|^2$ we

find

$$y(t) = \frac{1}{2\pi i} \int_{\epsilon-i\infty}^{\epsilon+i\infty} \frac{e^{st}}{s+Q(s)} \,\mathrm{d}s \qquad \text{with} \qquad Q(s) \equiv \sum_{k} |\phi_k|^2 \frac{s+iE_k}{(s+iE_k)^2+B^2}. \tag{12}$$

During intermediate times (which is when the vast majority of decays take place) this is well approximated by the pole in the integrand. This is found by taking the continuum approximation for the photon-mode integration and assuming Re s (which is essentially the decay rate) is small and negative. The continuum approximation encompasses the following assignments and transformations:

$$E_k \to E$$
 $\sum_k |\phi_k|^2 \to \int dE \,\rho(E) |\varphi(E)|^2 \equiv \int dE \,g(E)$

where ρ is the density of states, $\varphi(E)$ are scaled matrix elements $(\phi_k \sim \phi(E)/\sqrt{\mathcal{V}}, \text{ with } \mathcal{V})$ a fiducial volume) and we define $g \equiv \rho |\varphi|^2$. With B = 0, the expression $(s + Q(s))^{-1}$ has a pole near the imaginary axis giving the usual decay rate (as we will see below). With non-zero *B* there is a bit more algebra. One first shows that[†]

$$Q(s) = -iP \int_{-\infty}^{\infty} dE g(E) \frac{E}{E^2 - B^2} + \pi \frac{g(B) + g(-B)}{2}$$
(13)

with 'P' the principal value. Writing

$$s = -\gamma/2 + i\Delta E$$
 $(\gamma, \Delta E \in \mathbb{R})$ (14)

the value of the pole in the Laplace inversion is

$$\Delta E = P \int dE g(E) \frac{E}{E^2 - B^2}$$

$$\gamma = \pi [g(B) + g(-B)] = \pi [\rho(B)|\varphi(B)|^2 + \rho(-B)|\varphi(-B)|^2].$$
(15)

Note that for B = 0 one recovers the usual 'Golden Rule' formula. Furthermore, if g(E) is not a rapidly varying function of E (and in general there is no reason it should be), the decay rate, in the presence of B, is substantially unchanged. This was the experimental observation [1]. (In [4] we propose that for much stronger fields the decay should be halted, basically because of the decline in g(B), but this is another matter.)

Nevertheless, by examining the spectrum of emitted photons, we will see that observation does induce a change. The line shape is essentially

$$I(k) \equiv \lim_{t \to \infty} [|x_k(t)|^2 + |z_k(t)|^2]$$

(as a function of k). This can be calculated in a straightforward way by going back to equation (11) and solving for $x_k(t)$ and $z_k(t)$, taking y(t) from equation (12). If y(t) is dominated by the pole contribution, it is given by $y(t) = \exp[-it(\Delta E - \gamma/2)]$. Inserting this in the two relevant equations in equation (11), gives the x_k s and z_k s as forced oscillators, independent, for different k. It is easy to show that

$$I(k) \sim \frac{1}{(E_k - \Delta E - B)^2 + \gamma^2/4} + \frac{1}{(E_k - \Delta E + B)^2 + \gamma^2/4}.$$
 (16)

This means that the line is split, no longer centred about the original energy (shifted by ΔE), but pushed in both directions by an amount *B*. For small *B* this will show up as a line broadening, only for $B > \gamma/2\sqrt{3}$ will the peaks be separated. This broadening (or splitting)

 $[\]dagger$ Note that to lowest order Q is independent of s. See [4] for details of the derivation.

occurs because the decay episode has been *temporally localized* by a time whose inverse is determined by B (in appropriate units). For this reason we call the effect 'observational broadening'.

The experimental observation of this effect need not be carried out under the demanding circumstances of the first quantum jump experiments, namely with a single atom. Rather, more data could be collected by having many atoms, so long as collisional broadening did not overwhelm the effect to be observed.

4. Discussion and conclusions

This letter takes up two related issues. The first is the duration of a 'quantum jump'. The second concerns changes in the decay that result from observations that give meaning to the first.

Our definition of jump time yields a quantity that is basically inverse bandwidth. It should be considered a *minimal* jump time. Our estimate of this time for an atomic transition gave a small value, about 10^{-20} s. However, we observed that for other transitions the time can be considerably longer, allowing experimental probes. It turns out that such a phenomenon is already known, and our discussion in terms of jump time can be considered simply as a shift of perspective.

Consider the chirality of certain isomers. In [10], Cina and Harris argue that the inversion transition between left- and right-handed versions of some isomers is inhibited by environmental interruptions. The environment provides a 'continuous' check of whether it has flipped or not. In practice, the meaning of 'continuous' is essentially that the timescale should be short on the jump timescale. In other words, the transition suppression due to environmental jostling effectively gives an example of the QZE.

Although observations that are intrusive enough to monitor the jump have little effect on its lifetime, as experimentally established, nevertheless *they do change other properties*. This is a vindication of Bohr's point of view on the effect of observation (but it says nothing about the more elaborate constructs of the Copenhagen interpretation). Moreover, it explains the appearance of the high energies that one might have wanted to associate with the jump times. For example, our 10^{-20} s jump time for an atom could suggest MeV scale energies; where in an atom could there be such an energy? The answer is that an actual experiment that pinned the transition to such times would involve the many degrees of freedom and associated energy contributions of the apparatus. A milder temporal localization, such as that in [1], would involve correspondingly smaller energies. The quantitative measure of this is the split (or broadening), $\pm B$, that is seen in the line shape, equation (16).

The feasibility of observing 'observational line broadening' (or splitting) will depend on (among other things) two important factors. First, there is the value of ' $B' \equiv \sqrt{N_0 |\Phi_0|^2}$, where N_0 is the effective photon number excitation at the location of the atom and Φ_0 the transition matrix element for the short-lived transition. Second, there will be a playoff between the value of increasing the number of atoms studied—for a stronger decay-photon signal—and the resulting problems, such as collisional broadening, that would interfere with the observation.

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