A tale of quantum jumps

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In this year of light, our attention is quickly drawn to the many inroads into our day-to-day lives made, since the early 1960s, by the laser and the various technologies it has spawned: digital technologies (e.g. the optical fibre network), biomedical imaging and applications (e.g. in ophthalmology and dermatology), and applications in precision and 'not-so-precision' engineering – how did engineers ensure that the ends of the Chunnel would meet up in the middle? Likely only a very few of us are turned to a reflection on quantum mechanics by the year of light; but the rise of light, from its rather side-lined position in the late nineteenth century, as a well-understood Maxwell wave, to its position today as a diverse enabling technology, is very much a story of quantum mechanics, particularly in its early days, the days of the Old Quantum Theory where my tale of quantum jumps begins.

Quantum optics studies the quantum mechanics of light. It brings together the understanding of light as a Maxwell wave with light understood as a stream of photons, where the emission or detection of a photon has, from the earliest days, been executed through a quantum jump (Planck 1900, Bohr 1913, Einstein 1916, 1917). The roots of quantum optics are therefore the roots of quantum mechanics and quantum jumps, although, only after the scientific explosion precipitated by the laser do we meet Quantum Optics as a discipline of its own within the science of light. The discipline was brought to New Zealand by Dan Walls in the 1970s. Dan learned his quantum optics from Roy Glauber (his PhD supervisor) – not an unpromising source, since Glauber went on to share the 2005 Nobel Prize in Physics, for which the citation was: 'for his contribution to the quantum theory of optical coherence'.

This paper cannot provide anything like a complete overview of quantum optics in New Zealand. The scope over 40 years is far too broad and the number of players far too large. Nevertheless, the story of quantum jumps, from the days of the Old Quantum Theory up to the present, serves to highlight some small part of the New Zealand experience. It also offers an encounter with the oddities of light as a quantum mechanical 'something', oddities that the gallant proposers of technologies for the future aim to exploit.

Energy comes in lumps and atoms jump

We are no doubt all aware that the core idea of the quantum theory of light, that energy comes in lumps (which we now call photons), traces back to Max Planck at the close of the nineteen century and to his famous formula for the energy density (energy per unit volume per unit frequency) of electromagnetic radiation in thermal equilibrium at temperature T (Planck 1900):

$$U(\nu) = (8\pi h\nu^3/c^3)[\exp(h\nu/k_BT) - 1]^{-1} \quad (1)$$

In this expression, ν is the frequency of the radiation, c is the velocity of light, k_B is Boltzmann's constant, and h is the now-ubiquitous Planck constant. Planck did not speak directly of quantum jumps; but jumps are implicit if his assigning discrete energies – an integer number of lumps of size $h\nu$ – to radiation oscillators is followed to its logical conclusion. Certainly Niels Bohr spoke of jumps directly (Bohr 1913) as he laid out his refinement of the Rutherford planetary model of the atom, with the relationship, $E_e - E_g = h\nu$, connecting the energy change of the orbiting electron to the frequency of the radiation emitted when it 'jumps' from a high (excited) orbit to the lowest, i.e. the ground state. Remarkably, the Balmer series of spectroscopic lines was recovered from such a simple rule.

It was Einstein, however, in his so-called A and B theory (Einstein 1916, 1917), who elevated quantum jumps to the level of a rule for quantum dynamics with real quantitative content. Einstein proposed to build a jump dynamic from the elementary processes illustrated in Figure 1: today we call them

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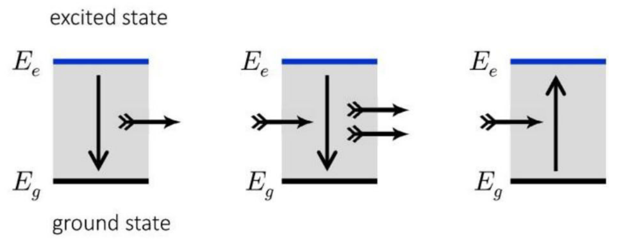

Figure 1.The three jump processes introduced in Einstein A and B theory: spontaneous emission at rate A (left); stimulated emission at rate $BU(\nu)$ (middle); absorption at rate $BU(\nu)$ (right).

spontaneous emission (left), stimulated emission (middle), and absorption (right). The probability that an atom of energy E_e jumps down during a short time, Δt – either spontaneous or stimulated emission – is the sum of

$$p_{\text{spon}} = A\Delta t, \qquad (2a)$$

and

$$p_{\text{stim}} = BU(\nu)\Delta t, \qquad (2b)$$

with A and B constants and $U(\nu)$ the energy density of radiation resonant with the transition. The probability that an atom of energy E_g jumps up is

$$p_{\text{absorb}} = BU(\nu)\Delta t, \qquad (2c)$$

the rate equal, in this simplest case, to the stimulated emission rate. Einstein had a specific job in mind for his little theory, one to put his scheme to a quantitative test. In an average over time, the result of all the jumping up and down would be a steady state. He asked that with $U(\nu)$ given by the Planck formula (1) for radiation in thermal equilibrium, the distribution amongst the energy levels, E_g and E_e, be brought to equilibrium too, i.e. it should accord with the distribution of Maxwell and Boltzmann,

$$N_g(\nu)/\,N_e(\nu) = \exp[(E_e - E_g)/k_BT] = \exp(h\nu/k_BT)\,, \qquad (3)$$

which is, indeed, what Einstein's scheme achieved.

Professor Schrödinger says nothing jumps

In the years 1925 and 1926, quantum mechanics was invented – twice! – as matrix mechanics (Heisenberg 1925) and as wave mechanics (Schrödinger 1926). From here on, the Old Quantum Theory had earned its designation 'old', and the new and true theory raised a challenge to the very idea of a quantum jump: nothing 'jumps' in the continuous evolution over time of the Schrödinger wavefunction, $\psi(\boldsymbol{r},t)$, of the electron in a hydrogen atom, for example. Schrödinger was adamant about the point, as can be read from a comment made during his first meeting with Niels Bohr (Baggot 2011):

> 'If all this damn quantum jumping were really here to stay, I should be sorry I ever got involved with quantum theory.'

His aversion remains with us today, and is stated with equal conviction, e.g., by Heinz-Dieter Zeh, in his paper entitled 'There are no quantum jumps, nor are there particles!' (Zeh 1993). It is most certainly so, that, if a frequency-stabilised laser illuminates an atom, *all else set aside*, Schrödinger's equation does not speak of an atom jumping up and down (Figure 1): the evolution is the continuous Rabi oscillation (Rabi 1937) illustrated in Figure 2 (Scully & Zubairy 1997).

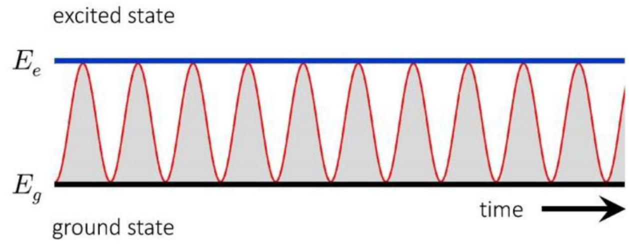

Figure 2. An atom illuminated by a laser does not jump. It executes a Rabi oscillation—a periodic cycling between its ground and excited states, generally in a superposition of the two. The red line tracks the relative weighting of the two states (ground or excited).

But photodetectors 'click'

What, however, of the 'all else set aside'? Spontaneous emission has been set aside to arrive at the continuous Rabi oscillation of Figure 2, and while the Schrödinger equation is convincing about the exchange of energy with a frequency-stabilised laser – no A and B theory-stimulated emission and absorption jumps – jumps in spontaneous emission are another story. Spontaneous emission accounts for the scattering of photons out of the laser beam, where, freely propagating away from the atom, each might be recorded by a photoelectric detector as a sharp 'click'. Surely the 'clicks' speak of quantum jumps: if a photon arrives at a detector (the detector 'clicks') having just travelled a distance $\boldsymbol{r}_{AD}$ from the atom, then, running back the travel time $\boldsymbol{r}_{AD}/c$, the atom must have jumped into its ground state.

This particular slant on quantum jumps – that 'clicks' imply jumps of an emitting atom – was the target of the first work of notoriety produced in New Zealand by the group of Dan Walls. Predicting photon antibunching in resonance fluorescence (Carmichael & Walls 1976a, 1976b), the work explains how jumps of an emitting atom imprint on the emitted photon stream. Figure 3 displays photon streams for so-called bunched, Poisson, and antibunched light, i.e. for thermal light, laser light, and the scattering from an atom illuminated by laser light. The last supports the assertion that 'clicks' imply jumps, since neighbouring 'clicks' repel one another, which produces a

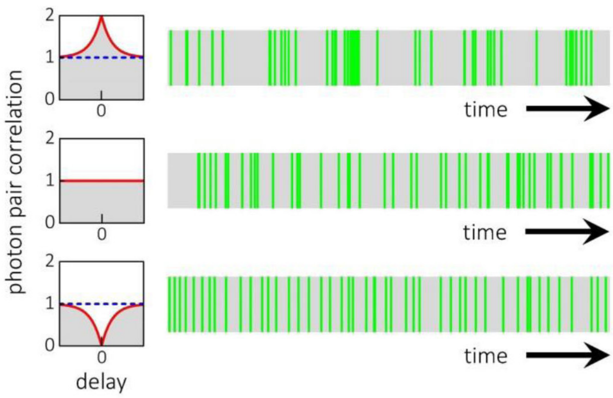

Figure 3. Two-photon correlation functions with sample photon streams for bunched (top), Poisson (middle), and anti-bunched (bottom) light; individual photons are marked by a light vertical line; darker lines indicate the photons are clumped. The two-photon correlation functions present the relative frequency of a given time delay between adjacent photon pairs.

photon pair correlation function that dives to zero at zero delay; there are no simultaneous 'clicks' – as there are for bunched and Poisson light – because the atom becomes temporarily inactive as a source of a second photon when it jumps to its ground state after the emission of a first photon.

Over the almost 40 years that have elapsed since 1976, photon antibunching has been observed in experiments with a wide variety of photon emitters: single atoms, as was originally proposed (Kimble *et al*. 1977, Itano *et al*. 1988), single dye molecules (Basché *et al.* 1992), single quantum dots (Lounis *et al.* 2000), single colour centres in diamond (Kurtsiefer *et al.* 2000, Brouri *et al.* 2000), single protein subunits (Sánchez-Mosteiro *et al*. 2004), for microwave photons in superconducting circuits (Bozyigit *et al*. 2010), in carbon nanotubes (Endo *et al*. 2015), and in a cyclic chemical reaction scheme (Vester *et al*. 2015)…. This is a survey not a thorough review.

Quantum Trajectory Theory (Carmichael 1993) adds the quantum jumps inferred from 'clicks' to Schrödinger's continuous evolution. The composite is not the jumps of Figure 1, nor the Rabi oscillation of Figure 2, but a combination of both – the Rabi oscillation plus jumps (spontaneous emission 'clicks') of Figure 4.

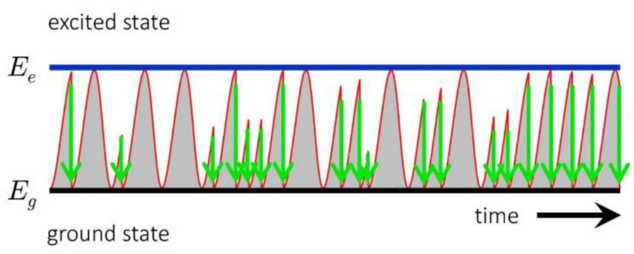

Figure 4. Quantum trajectories combine the Rabi oscillation with jumps for spontaneous emission (scattering out of the laser beam).

And we see the jumps…or do we?

But can quantum jumps be observed in experiments directly? A proposal from the mid-1980s (Cook & Kimble 1985) suggests they can. It builds upon the electron shelving idea of Hans Dehmelt sketched in Figure 5 (Dehmelt 1974). Two transitions – one dipole-allowed and designated 'strong', the other dipole-forbidden and designated 'weak' – share a common ground state. Both are illuminated by resonant laser beams (Ω_s and Ω_w in Figure 5), and the spontaneous emission rate on the strong transition, A_s, is orders of magnitude larger than the rate on the weak transition, A_w. In Figure 5, a stream of photons scattered on the strong transition travels to the left. It acts as a monitor of quantum jumps on the weak transition. For the most part the photon stream is densely packed; but there are four clear gaps. Gaps mark the weak transition jumps. One starts if the atom jumps to the weak excited state, thus 'shelving' the active electron; it is over when the atom jumps back to the ground state – an average wait of one lifetime A_w^{-1}. In short, quantum jumps on the weak transition turn the photon scattering on the strong transition on and off – weak transition jumps, it is proposed (Kimble & Cook 1985), are then observed.

Three independent experiments observed what was proposed in the very next year (Nagourney *et al*. 1986, Sauter *et al*. 1986, Berquist *et al*. 1986), all working with single trapped ions, the technology that would earn Hans Dehmelt a share of

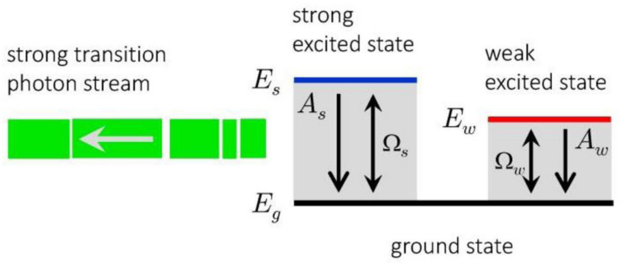

Figure 5. Electron shelving scheme: a monitor of quantum jumps between the ground and weak excited state.

the 1989 physics Nobel Prize. Observations on related systems have followed since (Basché *et al*. 1995, Peil & Gabrielse 1999, Gleyzes *et al.* 2007). Yet the admonition that there are no jumps in absorption and stimulated emission for illumination with laser light remains. How does the evidence from 'gaps' (in the stream of Figure 5) stand up to it?

The answer appears in Figure 6. Two separate plots are shown, each a quantum trajectory (like Figure 4), where the colours coordinate with Figure 5 and a line at 0 (1) means the state is unoccupied (occupied); the photon stream scattered on the strong transition runs down the middle of each plot. The upper plot corresponds to the implemented experimental scenario (e.g. Berquist *et al*. 1986), where the red line moves from 0 to 1 after the photon stream turns off: the electron is 'shelved'. The process is a continuous one, though, not a jump from 0 to 1. The lower plot adds a degree of objectivity to what the upper plot suggests. A well-timed intervention (timely change of phase and amplitude for the weak-transition laser light) reverses the shelving midway through. A quantum jump, being unpredictable and discontinuous, could not be so reversed. Schrödinger's admonition holds true. There is no jump *per se*!

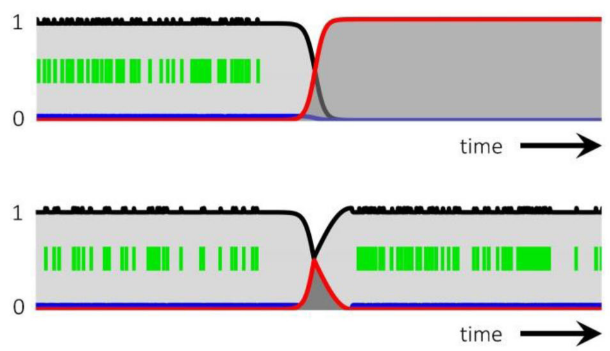

Figure 6. Reversing a 'jump' mid-flight: quantum trajectory without reversal (upper); with reversal (lower).

Epilogue

Little more than a run through history, my tale needs a take-home message. Photons, the quanta of light, are countable and discrete, and one assumes they come and go in jumps. Einstein proposed it so – though only as a pragmatic step – and recent experiments report the jumps observed. Yet the Schrödinger equation is deterministic and nothing within its jurisdiction jumps. What then to make of this unlikely marriage where the *continuous* is to somehow cavort with the *discrete*. 'What to make' is a whole other paper; but philosophy aside, *ways of cavorting is what quantum technologies are all about!*

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