Stepwise fluorescence due to level-crossings – A collaboration with S. Pancharatnam

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ANY paper in a learned journal which reports a piece of scientific research inevitably gives the impression that the work has proceeded from beginning to end in a logical and orderly fashion with an inevitable outcome to the interaction between theory and experiment. All the false starts, wrong turnings, difficulties and mishaps which make the work so interesting to the participants are carefully tidied away. I remember reading just one paper which contained the admission by the authors that 'this interesting effect was discovered when one of us mistakenly oriented a polarizer the wrong way'. My collaboration with S. Pancharatnam on 'Interference effects in stepwise fluorescence due to level-crossings' (*Proc. Phys. Soc.*, 1965, **86**, 1351–1363) was, alas, no exception to the convention of presenting a tidy version of reality, and as usual, none of the fascination or excitement of the work as it unfolded comes across. This short note is a belated attempt to describe this other side of my interaction with a most gentle man who was also a great physicist possessing a true and deep insight in his chosen field.

The start of this work was delayed at the outset by some months by his marriage which took place just before he was due to leave India to join us in Oxford. Eventually he did arrive, and was introduced, and then we were left to come to some working arrangement. There was, I tentatively ventured to suggest, a bit of a problem with his name for a start – four syllables was too much for everyday use. So he said his friends shortened it to just Panch, and perhaps I would care to do the same. Next, in total ignorance of his reputation earned by his theoretical and experimental work on the propagation of light in crystals I said that, in view of my interest in experimental work and his in theory, we might have the basis for an interesting and fruitful collaboration. And so it proved.

Both in the old literature on the subject and more recently from other work that we had done in Oxford there was a good deal of experience in exciting resonance fluorescence in mercury vapour to draw upon. Various ways of circumventing the many technical difficulties had already been devised. In our laboratory, for example, we had developed fairly efficient spectral light sources by exciting with microwaves minute quantities of single mercury isotopes sealed into miniature capsules. These capsules were made by glass-blowing fused silica, which transmits the all-important 253.8 nm ultra-violet mercury line.

The microwaves came from medical diathermy units, acquired for the usual reason, that they were relatively cheap. Probably if we were working now, we would use adapted microwave ovens. Matching the output of this device to a discharge in the capsule, which was in a microwave cavity, was difficult. It was achieved by manipulating tuning stubs (that is, transverse cavities provided with adjustable tuning pistons) which were in the coaxial transmission line linking the microwave source to the lamp cavity. Because the incremental impedance of a plasma discharge is negative and changes dramatically as the discharge intensity increases, achieving a sufficiently stable discharge was entirely an empirical matter which often involved an hour or more of pushing the pistons in and out at random like some demented trumpeter. Even then, the quasi-stable source would either be too dim, or paradoxically, too intense. In the latter case, the colder, unexcited atoms near the wall of the capsule would absorb the light from the hotter centre and re-distribute it through other energy level transitions. The result was a broadened spectral line with the useful radiation at its centre entirely missing. The only way to detect this condition was to observe that the lamp failed to excite fluorescence in the target mercury vapour. In that case there followed another session playing the tuning stubs. Now the phenomena Panch and I proposed to investigate needed not one, but two, independent sources and this reduced the likelihood of success with the lamps to near-infinitesimal.

The reward for persisting and obtaining the occasional successful simultaneous operation of both lamps was to be able to perform a striking conjuring trick whereby a ghostly blue glow, dimly visible in the body of the absorbing mercury vapour along the cones of invisible light converging from opposite directions, could be extinguished instantly by blocking off the light from *either* lamp. For one of them, interposing a transparent piece of ordinary glass would suffice.

Meanwhile, whilst helping solve practical problems and combing the literature to devise filters of salt solutions and halogen vapours to suppress unwanted mercury spectral lines, Panch was investigating this phenomenon of stepwise fluorescence theoretically. He once (and only once) asked me to verify some of his calculations. I did my best, but after several fruitless hours I had to ask for help, whereupon he gently pointed out that an effect could not possibly precede its cause, and therefore I had incorrect time limits for a vital integral. A simple matter, but the true geniuses are those who get the crucial fundamentals correct and can build on them. I don't think he appreciated the strength of his own analytical powers because, on another occasion, he asked me to look over some questions he proposed setting for his college undergraduate students, to whom he was teaching the elements of optical crystallography. I felt obliged to say that they might find them a little difficult, whilst privately thinking that those questions would have taxed an Einstein.

Once the problems of lamps and filters had been overcome, we could begin the experimenter's delight of orienting polarizers in the light beams impinging on the mercury vapour, applying a swept magnetic field and seeing the theoretical predictions emerge. In reality, it was still not quite that easy because another severe experimental problem arose from the same self-reversal problem which made the lamps ineffectual, but which now took place in the target mercury vapour. To create 'clean', easily interpreted experimental results it is necessary that incoming photons should interact with just one atom, and not be handed on to another and another before emerging from the containing vessel. This can be avoided by reducing the density of the target atoms by cooling a remote part of the containing vessel to reduce the vapour pressure of mercury. But unfortunately, the amount of light scattered is also reduced and since we were working with conventional light sources before the advent of lasers, there was a limit to the extent to which the intensity of the exciting light beams could be increased to compensate. Even the expedient of collecting more of the solid angle of the light emitted by the lamps (which necessitated scouring the physics department for even bigger and better condenser lenses) meant that the light then entered the scattering vapour from a variety of angles. This spoilt the geometrical cleanliness of the experiment somewhat, and in the limit made interpretation of the results difficult.

The work therefore evolved into a daily struggle for both of us to get convincing, unambiguous results worthy of publication, but eventually we collected enough pen-recorder tracings corresponding to polarizers and analysers being set to a variety of angles. In the end, I do recall being somewhat disappointed that some of the more hard-won and interesting-looking experimental curves obtained with the more exotic geometries – having polarizers oriented at 45° to the magnetic field direction and so on – were not included. But Panch was a purist who held, correctly, that they added nothing extra to the interpretation of the physics. It took me a long time to realize what his physical insight had told him all along, that just as familiar optical interference effects can be seen wherever trains of light waves from a common source are made to traverse different paths before being brought together again, so we were seeing analogous interference effects *in the divided and re-combined wave function of a single atom*, with light acting as a stimulus and final observable. It was a beautiful demonstration of atomic quantum mechanics, and moreover one which could be achieved on a table-top without using particularly exotic or expensive equipment.

The story would not be complete without the inclusion of G. W. Series as the catalyst who brought Panch and I together to work on this problem. He enabled us to have a most enjoyable collaboration which unfortunately ceased after only a few brief months when our ways parted. Nevertheless, it was long enough for me to learn that physics, with its attributes of elegance and symmetrical beauty, is just as much a cultural and artistic activity as, for example, painting or the study of ancient civilizations – a brilliant facet which we tend to let its practical applications obscure. Thank You, Panch.