PAPER

The history of laser conditions in semiconductors

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The history of laser conditions in semiconductors

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Abstract

Fifty years ago, summer 1962: in three independent laboratories of the US East Coast, almost simultaneously, IR coherent light was for the first time emitted from semiconductor crystals. No theory was associated with these results. Two years before, Georges Duraffourg and Maurice Bernard had readily proved that for such phenomenon to occur requires that a relation is fulfilled between quasi-Fermi levels and photon energy: $F_n - F_p > h\nu$. This paper presents an overview of this important period of history and the events that occurred around that time.

Today, all over the world, millions of tiny semiconductor lasers emit minute amounts of coherent light, mostly IR. They are the heart of all sorts of electronic devices, computer and communication systems. When laser radiation is emitted, the so called 'laser conditions' are necessarily fulfilled. This is a law of nature.

Fifty years ago I was a physicist, but am no longer. I remain a Professor and I now feel more and more attracted by the history of the last-century science. Thus, I fully appreciate being invited to the marking of the 50th anniversary of the semiconductor laser, a component so widely spread today all over the world.

My contribution to this occasion deals with how Georges Duraffourg and I discovered in around 1960 the condition for a semiconductor to emit stimulated light, that is to say, to display laser action. This work was reported in 1961 [1], and effective laser action was observed during the summer of 1962, half a century ago. The present text also describes the way we, young physicists, were working at that time.

In the late 1950s it had been known for some time that a population inversion is necessary for stimulated emission of light to take place, that is to say, for maser or laser action to occur. In 1958, Schawlow and Townes in USA [2], and Basov and Prokhorov in Russia [3] had given the main ideas to achieve this goal.

As early as 1957, before the paper by Arthur Schawlow and Charles Townes appeared, Pierre Aigrain and I were wondering about the possibility of maser effect at optical frequencies, having obviously in mind semiconductors. In 1958, I was already in friendly relations with Pierre Aigrain, an extremely imaginative physicist in the field of solid state physics. I remember the many discussions, in 1957 or 1958, during lunches we had in small restaurants of the Quartier Latin, somewhere between École Normale Supérieure, rue d'Ulm where Pierre had his laboratory and École Polytechnique where I used to teach. We were intuitively convinced that stimulated emission of light ought to occur also in semiconductors, otherwise it would have been a very strange exception in the field of interaction between light and matter! But how would the process appear and on which transitions would it take place? These were the questions.

At the beginning of 1957, Pierre had a brilliant qualitative idea: in germanium and silicon, recombination of electrons and holes produces photons with simultaneous absorption or emission of phonons, i.e. quanta of lattice vibrations. He knew very well this topic which had received considerable attention in his laboratory at École Normale Supérieure. I clearly remember his argument. It was very simple, too simple in fact, as we shall see later:

Let N_o be the population of ground state, N_e the population of excited states, let n be the number of photons actually present and n' the number of actually present phonons. Then the rate of absorbed photons by such a semiconductor crystal is:

$$W\ N_o\, n\, n',$$

where W is a constant.

The rate of induced photons (simultaneous phonons being stimulated as well as spontaneously emitted) is:

$$W\ N_e\ n(n' + 1)$$

[1] Formerly vice president of teaching and research at the École Polytechnique, France (1983–1990).

Thus, the rate of induced photons is larger than the rate of absorbed photons, if:

$$W\ N_e\ n(n' + 1) > WN_o nn'$$

Or:

$$N_e/N_o > n'/(n' + 1).$$

As $n'/(n'+1)$ is smaller than 1 the condition turns out to be less restrictive than the so-called population inversion. That was Aigrain's approximate idea.

This argument was orally and briefly exposed by Pierre Aigrain in May 1958 at the Brussels Semiconductor Conference, but was never published, nor even written. And of course no experimental work was mentioned.

Pierre Aigrain was a renowned physicist. He made frequent visits to the main US laboratories where he had many friends. Very likely, in 1958 or 1959, on several occasions he expressed his subtle but incomplete idea. At that time there was no web, but exchanges and travel were already commonplace.

However, testimonies on Aigrain's ideas are few and some seem rather doubtful. For instance, in 1965 Ben Lax wrote [4]: 'another possibility of using point contact devices for excitation was entertained by Pierre Aigrain in 1957 during the course of a series of lectures at the Massachusetts Institute of Technology'. Ben Lax continues: 'He (Pierre Aigrain) and his group began active work on this and other schemes for electrical and optical pumping of semiconductors, such as germanium and silicon, without success and reported these ideas at the international conference at Brussels in 1958'.

I was not at MIT at that time but Ben Lax's text is in complete disagreement with my own recollection and with the actual activity of Aigrain's Laboratory. At that time the Aigrain Laboratory was by no means involved in point contact devices.

Pierre Aigrain was a physicist brilliant and generous, 'brillant et généreux': someone had to look carefully at his idea. Georges and I did it in 1959. Now, 50 years later I still do not understand why no other scientist looked at the theory of laser effect in semiconductors?

Let us come back to Aigrain's argument. In a solid, one cannot consider discrete quantum states and label N_0 or N_e the numbers of the occupied states. The only energy states we can consider are delocalized energy states associated with allowed energy bands[2] . In a semiconductor there are a large number of quantum states grouped into allowed energy bands, the conduction band for electrons and the valence band for holes. Their probability of occupancy is given by the Fermi–Dirac statistics.

With Georges we were particularly interested in the application of the so-called detailed balance of electron-hole pairs interacting with photons inside a semiconductor crystal. We had in mind to deduce the radiative lifetime of excess electron-hole pairs in germanium and silicon from the spectra of optical absorption curves, and thus better understand why the radiative efficiency in these semiconductors was so small. In fact the physical reason lies in the fact that in Ge and Si, the optical transitions are indirect while the transitions in III-V semiconductors are direct. With the tool of the detailed balance calculations it was easy, for each optical frequency ν, to sum up all the electron–hole transitions emitting or absorbing a photon of frequency ν, whether the transition is, or is not, accompanied by the emission or absorption of phonons. The results are extremely simple:

Let us assume a semiconductor containing a large number of excess electron–hole pairs. It is a sound hypothesis to consider that electrons in the conduction band and holes in the valence band are in equilibrium with the lattice vibrations of the crystal. Let F_n be the quasi Fermi level of electrons in the conduction band and F_p the quasi Fermi level of holes in the valence band, A straightforward calculation shows that the rate of emission of stimulated photons is larger than the rate of absorption if:

$$F_n - F_p > \text{than } h\nu. \quad (1)$$

These results were presented at a special meeting of the Société Française de Physique in Paris on July 1961 and published in the *Journal de Physique* on December 1961 [5]. On the 12th of July, I submitted a detailed manuscript written in English at *Physical Review Letters*. This mail was addressed to S A Goudsmit, chief-editor of *Physical Review Letters*, with a copy to Elias Burstein, director of the Solid State Physics division of the American Physical Society. The manuscript contained the above argument, also with a reference to the second principle of thermodynamics which has some connections with the formula (1). A few weeks later, Georges and I were deeply disappointed to receive a negative answer from *Physical Review Letters*. Pierre Aigrain, who knew about our paper, was also disappointed. He suggested to me to submit our manuscript to *Physica Status Solidi*, a new international journal just appearing in Berlin, East Germany. It was accepted and was published in their first volume [1].

Why this refusal? In fact the very large amount of papers reporting maser and laser effect in new materials raised problems for editing reviews. This was explained by Jeff Hecht in *Laser Pioneers* [6], quoting the Maiman controversy, page 134, 'Maiman's paper ran afoul of Samuel Goudsmit, an eminent physicist best known for his co-discovery of electron spin, who had founded the letter journal to facilitate rapid publication of new results. Goudsmit had grown impatient with a glut of maser research, and in 1959 announced that the journal would not publish maser papers that did not contain significant contribution to basic physics'.

In France we were not conscious of this. If we had known it, Georges Duraffourg would have been very upset to learn that his work had not been received as basic physics! And a few weeks later, Alfred Kastler who presided over the jury of George's thesis would have been very surprised.

Clearly our work did not receive much attention. Nevertheless experimental groups were successful, and in 1962 three different American laboratories published the first evidence of laser emission in semiconductors. None of them mentioned our paper published in 1961, one year before!

I was invited in 1962 to be assistant professor at Stanford University. In September 1962, as I was driving back to the

[2] Of course all this was perfectly clear in Aigrain's mind; we just had to look more carefully at his original guess.

East Coast, I stopped in Schenectady to visit R N Hall at his General Electric laboratory. In the room close to the office where we chatted, an GaAs diode was emitting stimulated radiation for the first time in the world, but Bob did not mention it. Many years later R N Hall recalled this meeting [6]. But I know that Bob Hall who had visited our group knew our publications.

In 1965 Georges Duraffourg submitted his PhD thesis 'L'emission induite dans les semiconducteurs'. Alfred Astler presided over the jury, Grivet, Bok and myself were examiners. This work included many other considerations, for instance concerning the role of excitons and impurities. There was matter for subsequent work and other papers. Indeed Georges should have published much more, as Pierre Aigrain recalled with sorrow in his last interview [7].

In 1962, I should have led our CNET Laboratories into experimental work on semiconductor laser diodes. Of course optical communications were not ready: optical fibers had too high transmission losses. However, I have no excuse, there was still so much work to achieve but my group was turned toward nonlinear optics.

So is the path of science. Today you have invited me to contribute to this wonderful occasion and you have given me an excellent opportunity to recall memories of the past and, believe me, it has been a great pleasure for me.

The history of 'Laser conditions in Semiconductors' appeared in French in 2009 in REE, N° 3, mars 2009

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