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Our Personal and Scientific Encounter and Collaboration with Delone (or a Story of the Development of Multiphoton and Tunnel Ionization)[1]

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INTRODUCTION

Each of us, (SLC) and (FI), met N.B. Delone under different circumstances. Nevertheless his scientific work and personal demeanor had profound effect on both of us. Thanks to Delone we were able to work together for a number of years, and that impelled us to form a duet in relating memories of our encounter with Delone's work and life.

In what follows we are stepping in front of each other rushing to convey our stories, mixing first and third person narrative modes, trying to reach through years and continents in an attempt to do the impossible-recreate one's personal live. We admit our failure before hand to stress the fact that such happening as the life of Nikolay Delone is unprecedented and any slice and projection that we claim to remember are just flimsy rendering of the phenomenon.

1. SLC: THE FIRST SCIENTIFIC VIRTUAL ENCOUNTER WITH DELONE (1965–1975)

It dates back to the second part of the 1960's when one of us (SLC) was still a graduate student studying at the University of Waterloo under the supervision of Prof. Neil R. Isenor. Without knowing what lied ahead, SLC "naively" took up the challenge proposed by Isenor and tried the experiment on multiphoton ionization (MPI) of an atom after reading the papers by Delone et al. [1, 2] and the theoretical papers by Bebb and Gold [3] as well as that of Keldysh [4]. When SLC told some senior scientists at some conferences about his multiphoton ionization project, the latter wished him good luck as if this were an impossible subject. At that time, there was at least another group in France, the CEN Saclay group led by G. Mainfray and C. Manus, doing the same experiment and had already obtained similar results as Delone's group. But it was then fun to be able to "play around" in the hitherto under-explored field. That meant adventure and risk but it was "exciting." An intriguing outcome of Delone's first experimental results was the fact that the observed nonlinearity of ionization rate was considerably smaller than the number of photons required by energy conservation law for MPI of the atoms under investigation.

After overcoming some major technical difficulties, SLC's experimental results showed that one could obtain the theoretically predicted nonlinearity during multiphoton ionization (MPI); i.e. the proper slope in the log-log plot of ion yield versus peak laser intensity [5]. It was Isenor who came up with this idea of satu-

Fig. 1. Picture taken by SLC of Nikolai Borisovich Delone and his wife, Tatjana Markovna, in their house in Moscow (September 29, 1991.)

[1] The article is published in the original.

ration (depletion of the neutrals) during MPI and the calculation fitted very well the experimental result [5]. Thus, the proposition by the first results of Delone et al. that there was some Stark shift etc. could not be confirmed. Instead, by comparison, we were led to believe that the reason why Delone et al. obtained a smaller value of the slope was because their results were partially in the saturation region. Saturation or depletion of the neutral is the consequence of an ionization probability equal to unity at the center of the focal volume. It was almost unimaginable at that time because MPI was thought to be very difficult if not impossible. A few years later, Isenor and his student (M. Cervenan) discovered the law of the slope of 3/2 in the saturation region [6].

2. WHAT URGED DELONE DO THAT FIRST MPI EXPERIMENT?

Let us have a pause and ask why Nikolai Delone wanted to do that first difficult unknown and uncertain experiment. We inquired about this among his early students. The answer was that Delone was intrigued by the two-photon absorption theory first proposed by Goeppert-Mayer in the 1931 [7] and would like to do that experiment. Then came the Keldysh theory [4] together with the discovery and advancement of the laser science and technology that originated partially in the USSR (Basov and Prokhorov). We could conclude that it was his imagination, curiosity and adventurous spirit that guided him to do what he became interested to do for the rest of his life. No one, we believe, could have imagined that MPI would eventually lead to high order harmonic and attosecond laser pulse generations. But in between these two events, tunnel ionization seems to be the key that opened up the door towards modern day high laser field physics, high harmonic generation and attosecond laser science.

3. THE FLOURISHING PERIOD OF MPI LED BY THE FRENCH SACLAY GROUP AND MULTIPHOTON MULTIPLE IONIZATION

Staggered political and personal events in the late 1960's and the beginning of the 1970's led to the closing of Delone's laboratory and dissipating the group working with him on those first experiments. In the second half of the 1970's he became an experimentalist without a laboratory of his own and hence was not able to compete in the MPI experiments for quite a while. Meanwhile, it was G. Mainfray and C. Manus in the CEN Saclay (Commissariat à l'Énergie Atomique de Saclay), France, who led a team of dynamic theorists and experimentalists to systematically investigate all the aspects of single electron MPI successfully including the discovery of the above threshold ionization (ATI) and high order harmonic generation (HHG) (except tunnel ionization which they attempted several times but did not obtain any conclusive result [8]). The Saclay Nuclear laboratory became THE dominant laboratory on MPI. Any one working in this field would feel highly honored if he was invited to visit the laboratory and discussed with the many dynamic leaders in the field. At least this was how one of us (SLC) felt. (See also "note added by SLC" at the end of this paper giving a historical glimpse of the initial action on MPI by G. Mainfray.)

Meanwhile, Delone, with an unbeatable spirit and perseverance, scientifically directed two experimental groups outside the Russian Federation. One in Tashkent, Uzbekistan and another in Uzgorod, Ukrain of the former USSR. In Uzgorod, V.V. Suran, I.S. Aleksakhin, and I.P. Zapesochnyi obtained new experimental results, extending single electron MPI to multiphoton double ionization of two electron alkaline earth atoms (Strontium, Barium, etc.) [9]. The experimental results were reported for the first time in the Western world in 1979 in Benodet, France by Mikhail Fedorov on behalf of Delone and co-authors who could not get a permit to visit the West. One of us (SLC) was in the audience. To SLC, the new experimental results of Delone et al. were as surprising as the first MPI experiment. Very soon, he collaborated with K.H. Welge and D. Feldmann et al. in the University of Bielefeld in Germany and performed some experiments on double ionization of Strotium [10]. Later, the Bielefeld group reported many significant results in the field of double ionization of alkaline earth atoms by measuring the electron spectra. However, it was the Saclay group again led by Mainfray and Manus who later observed quadruple ionization of Xe atoms with the signature of nonsequential double ionization [11]. It turns out many years later that nonsequential ionization is explained by the re-scattering process which in turn hinges on tunnel ionization of an electron which later re-scattered back to the parent ion with a certain probability [12–14].

4. TUNNEL IONIZATION (TI)

Before the 1980's, tunnel ionization was the ultimate mystery in laser ionization of an atom that was not totally accepted before an experimental unambiguous observation was made. Keldysh's theory [4] predicted this event in the beginning of the 1960's but was not yet observed in the laboratory till the beginning of the 1980's. Because some attempts to observe this phenomenon was not successful, it has created a heated controversy among the practitioners of strong field ionization including the theories advanced by Faisal Subsection 4.1 and Reiss Subsection 4.2. Later, the theories of Keldysh, Faisal and Reiss were grouped together and called KFR theory. We try to tell the story of Delone's action to do an experiment on tunnel ionization in this direction and that of the successful experimental observation by one of us (SLC).

4.1. An Insider's View, Narrated by FI in the First Person Mode: FI Became a Student of Delone

After completion of my graduate study in 1978, I've joined an experimental group of former Delone's student, T.U. Arslanbekov, in Tashkent. In 1979 Delone gave series of scientific seminars on non-linear ionization of atoms at the Nuclear Physics Institute, where the group was located at the time. Delone's charisma, level of energy he seemed to emanate on every discussed subject and every person he talked to,—were captivating. Remember thinking while listening to Delone's presentation: "if I want to run through my life on "doing" science, this is how I want to feel about it."

In 1980 I became a postgraduate student of Delone in FIAN (Lebedev Physics Institute of the USSR Academy of Sciences). It turned out that Delone didn't have his own experimental lab and staff. In fact, I was the only student Delone had in FIAN at the time and the first one for the last few years. Now, looking back, I recognize that it was an awkward situation for a postgraduate student to start a career in experimental research with no laboratory and support of fellow researchers. But then I felt blissful and eager to 'move scientific mountains'. That is how I felt in the presence of Delone from then on.

At the end of the 1970s and the beginning of the 1980s, one of the 'hottest' topics in experimental physics of nonlinear ionization in laser field, was the so called 'tunnel regime' of ionization. Delone, who pioneered MPI research decade before, felt, with his natural gift of competitive edge, that this should be the focus of our effort. Right at our first meeting he had thrown at me: "Fedja, we are going to design the most important experiment in our field- the tunnel ionization experiment."

The excitement and desire to start were there, but reality set it's own pace on the progress. It turned out that requirements of experimental observation of ionization in the tunneling regime put us at the limit of capabilities of the state of art experimental techniques.

After the work of Keldysh [4], the tunneling regime of ionization was generally accepted to be conditioned by the value of adiabaticity parameter $\gamma = (\omega/F)\sqrt{(2E)}$ to be much less than 1. To get $\gamma \ll 1$ one has to juggle with three experimental parameters: laser frequency (ω), field strength (F) and atom ionization potential (E). Obviously finding an atom with the smallest ionization potential, like potassium, eases requirements on laser parameters. The longer the wavelength, the smaller laser intensity is needed. Only pulsed CO_2 lasers available in FIAN at the time were with pulse duration of $\geq$100 ns. That put hard limit on attainable field strength and reachable ionization rates($<10^{-7}$ s^{-1}). To our anxiety it meant that the experiment had to be done at relatively high pressure of alkaline metal vapor and standard methods of detection using Time of Flight mass-spectrometer wouldn't work since the collective effects in the created plasma preclude accurate measurement of the produced ions and electrons. Working with Delone through problems breaking up one after another I was learning my life long lesson witnessing how seemingly unsurpassable difficulties could be viewed as opportunities to do something new and unconventional. The chosen detection method was monitoring of optical discharge created by focused CO_2 laser pulse in alkaline metal vapor. Initial electron created by TI process is multiplied through electron avalanche sustained by the laser field accompanied by emission of recombination light. This bright flash of light is easily detectable.

After the methods were chosen I started preparing the experiment in the basement of FIAN in G.P. Kuzmin's group (Karlov's laboratory) that had the most powerful CO_2 laser available to us. I enthusiastically worked on solving technical problems associated with focusing of powerful CO_2 laser into a chamber with alkaline metal vapors through NaCl windows without breaking them. In parallel, we prepared theoretical calculation on ionization rates and avalanche conditions favorable to our experiment conditions [15]. By that time a second Delone's student, Vadim Tugushev, helping me with the experiment preparation, started to theoretically explore a possibility of using negative ions for TI observation. We were going full speed ahead aiming to prove the existence of tunnel ionization "à la Keldysh" when two events stopped us unexpectedly.

One was the success of the experiment carried out by SLC and co-workers from Canada. SLC sent Delone his experimental results before publishing them. Delone's group would not be anymore the first to experimentally demonstrate the existence of tunnel ionization. The other more serious difficulty was that the time to use CO_2 laser in Kuzmin's group was up and apparently, no one else believed in the possibility of tunnel ionization in the laboratory.

Being overplayed by "Canadians" we started to look for new scientific challenges. On the wave of energy created by enthusiasm of preparation for the very first TI experiment a new student of Delone, Maxim Ammosov together with Prof. V.P. Krainov, finished the work on an analytical tunneling formula based on the simplification of the Keldysh's [4] and the ppt theory [25]. This workable ADK formula was published later and became one of the most cited in the community of strong field laser physics [18].

Soon after the end of the "Tunnel epopee," Delone and I identified a new scientific problem to solve: Muliphoton Spin-forbidden transitions in 2-electron atoms. I moved to Tashkent and successfully finished experiments in laboratory of another former Delone's student,—D.T. Alimov.

4.2. View from Outside, SLC: First Convincing TI Experiment- Meeting Delone in Person

Right from the beginning, all practitioners of MPI would like to know whether tunnel ionization of an atom could be observed experimentally. In particular, the paper of Keldysh predicted this through the value of the parameter γ which should be much smaller than unity (i.e. $\gamma \ll 1$) for tunnel ionization to be observable. It was not until the technology of the CO_2 laser (λ = 9–10 μm) became mature that such a condition could be fulfilled.

Even in the summers of 1975 and 1976, when SLC visited Mainfray and his group in CEN-Saclay, France, tunnel ionization was often discussed in the laboratory and in particular, between Mainfray and SLC. They discussed the use of the TEA-CO_2 laser which was invented by Jacques Beaulieu in the Canadian Defense Laboratory in Valcartier near Quebec City. Mainfray would even move his high vacuum system to Laval University to make use of the existing high power CO_2 laser in SLC's colleague's laboratory to test the idea of tunnel ionization. Unfortunately, the laser was still not powerful enough to justify the cross-Atlantic move.

Meanwhile, Gy. Farkas (Hungary) visited SLC in Laval University in 1982. They performed experiments on photo-electron emission from a gold surface using a TEA-CO_2 laser emitting a train of mode-locked 2 ns pulses at the wavelength of 10.6 μm. The results indicated that tunneling emission of the photoelectrons could be the main mechanism [16]. They also observed ion signals from Xe and Kr using the same laser [17]. However, because the laser intensity in the laboratory in Laval University was not sufficiently high, the statistics of the experimental points did not allow them to conclude beyond doubt that tunnel ionization of atoms did take place. Some of these results were communicated to Delone, principally by Farkas.

Without any knowledge of the activities of Delone's group described by FI above, SLC continued on pushing the tunnel ionization experiment using a much more powerful CO_2 laser system developed by P. Lavigne and co-workers in INRS-Énergie in Varennes (near Montreal), Canada. Finally, tunnel ionization of Xe atoms was confirmed [18] with a new surprise. That is, multiple ionization of Xe up to Xe^{3+} was easily observed. Later, even up to Xe^{6+} was observed [19]. But the fit of the experimental data with the Keldysh theory was far from satisfactory [20]. The real fit of the CO_2 laser experimental data with an experimentalist-friendly tunneling "theory" (the ADK formula [21]) was not realized until many years later after FI joined SLC's laboratory through the recommendation of Delone [22]. With FI, it was discovered in SLC's laboratory using a new CO_2 laser that the Keldysh γ parameter need not be much smaller than unity for tunnel ionization to take place. It is sufficient that γ be smaller than 1/4 [23, 24]. All atoms and molecules tested so far for tunnel ionization obeyed the corrected ADK formula [23] without any arbitrary shift of the intensity scale in the experimental data. These days, people would say that tunnel ionization is valid when $\gamma < 1$ roughly. A review of the evolution from mutliphoton and tunnel ionization can be found in [27].

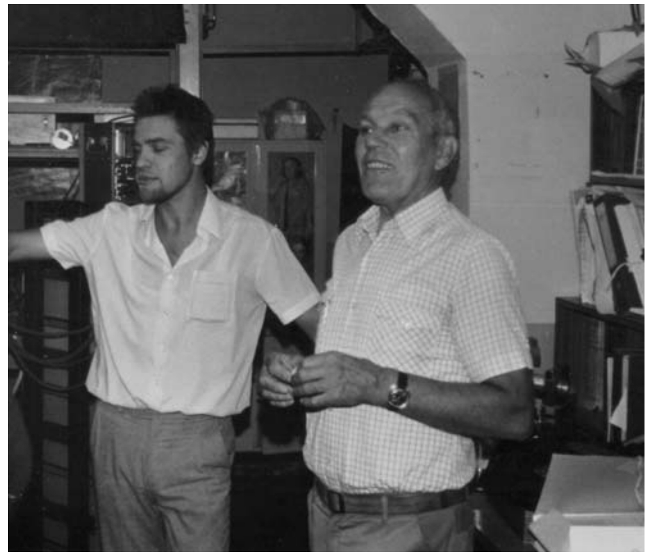

Fig. 2. Delone (right) and FI in FI's Lab. of Department led by D.T. Alimov (Tashkent, 1986).

SLC was invited by Delone to visit Moscow in 1985 at the ICONO conference in Moscow State University. This was his first visit to USSR and it marked the beginning of the real human contact between SLC and Delone together with many of his colleagues. SLC was surprised that Delone spoke French only as his foreign language. (Later, SLC realized that Delone was a descendant of Napoleon's army left behind in Russia.) Luckily, SLC could also speak French after having worked in Laval University (a French speaking university) for many years. The first real face-to-face discussion on science between Delone and SLC took place in a quiet corner outside the main building of Moscow State University with two other Russian scientists. During the more than 30 min conversation between Delone and SLC, the two other Russian scientists did not speak a word. SLC was initially introduced to them but as usual, he could not retain new names immediately in his memory. But he believes that one of them was V.P. Krainov. A very interesting behavior of Delone was observed by SLC. Delone, while speaking, would turn his head and move his body to look around almost constantly. Nevertheless, SLC was impressed by Delone's humble attitude and sincerity in science. Since then, he visited the USSR several times to attend different conferences. Collaborative agreement was signed between Laval University and the General Physical Institute on strong field physics. The political

Fig. 3. First visit to Laval University, Delone (left) and FI (right) (December, 1990; bus station—Quebec).

Fig. 4. Second visit to Laval University Delone (right), Mainfray (center), SLC (left) (June 1991 in SLC's office.[1]

change in the USSR accelerated the direct contact and collaboration between SLC and Delone and his colleagues.

5. POST-USSR AND INTERNATIONAL LINKAGES

After 1991, there was no more USSR. From the political point of view, the Western countries would like to see that those highly trained scientists would be kept employed either inside the former USSR or in the Western countries. It was around this period when Delone recommended SLC to invite FI to work in his laboratory. This turned out to be an excellent recommendation. FI started working in Laval University since January of 1991. For FI it became a perfect loop in fate—ten years after he started a race for observing TI regime in a basement of FIAN (Moscow) he continued to explore peculiarities of TI ionization in a basement of COPL (Center for Optics, Photonics and Laser, Laval University, Quebec). Delone has visited Quebec a few times and took an active part in discussions of experimental results, shaping interpretation of the previously collected data and focusing on new experiments.

Delone also recommended Ammosov to visit SLC's laboratory through an international post-doctoral fellowship from the Natural Sciences and Engineering Research Council of Canada in 1993–1995. Meanwhile, NATO announced the creation of the NATO-linkage grant. Any scientist in a Western country belonging to NATO together with a lead scientist in the former USSR could apply for such a two-year grant in collaboration with one or more former USSR scientists. SLC and Delone were successful in winning this grant several times. The initial participating scientists were V.P. Krainov, S. Goreslavski, V.P. Bykov, A. Popov, and O. Tikhonova all from Moscow. They visited Laval University at different periods of time. All their travelling expenses were paid for by the NATO-Linkage grant. They were also introduced to many Canadian colleagues of SLC such as A D Bandrauk, Paul Corkum etc. We initially discussed strong field stabilization, Stark atoms and ideas of carrying out such experiments in our laboratory. This experiment turned out to be too difficult and was not conclusive although we did observed interference stabilization (population trapping) similar to what Fedorov has proposed in 1996 [26].

Later, Delone recommended V.P. Kandidov and O.G. Kosareva to link up with SLC's new direction of research in filamentation. This latter fruitful collaboration goes on until now. Meanwhile, SLC still keeps in touch with the other visitors/collaborators who have since established their own contacts and collaborators in various different Western countries.

6. CONCLUSIONS

For almost 30 years of our acquaintance with Delone as a scientist, colleague, ally, we always felt being enriched by his presence in our lives. We miss him profoundly as a dear friend. We miss him being reachable- just a call away. We regret missing so many opportunities to share with him our ideas, feelings and simply time being together.

Time to time we caught ourselves in recognizing his qualities in people we are meeting. It is a subconscious act of compensating for what is missing in our lives and sure indication of what he meant for us. We will always cherish our memories of him.

[1]More pictures are in web-album: http://picasaweb.google.com/fedor.ilkov/ProfDeloneNB#).

NOTE ADDED BY SLC

Dr. G. Mainfray told SLC on March 17, 2009 through a trans-Atlantic telephone conversation about the reason why he did MPI experiments in the beginning of the 1960s. It was because a focused Q-switched laser pulse could breakdown atmospheric air which they have observed right from the beginning of the development of the Q-switched laser. They have published some papers on this which unfortunately, he could not remember the exact references. They liked to understand the physical mechanism of laser induced breakdown (R.G. Meyerand, Jr. and A.F. Haught, Phys. Rev. Lett. **11**, 401 (1963)). After much analysis, they arrived at the conclusion that MPI of an atom/molecule generating the first electron would be the initiating process of breakdown. They thus started to investigate the probability of multiphoton ionization of atoms. Unknowingly, this activity converged with that of Delone who tried to understand Keldysh's theory. This route of events of Mainfray coincided with that of SLC since he began his graduate study with Isenor on laser induced breakdown which later branched out into MPI.

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