Multiphoton ionization at AMOLF.

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It was nearly twenty years ago that I first set foot at AMOLF. Being a chemistry student at the Vrije Universiteit I had never heard of this FOM-Institute before Cees de Lange had mentioned it to me as a possible place to look for an opportunity of doing thesis research. I had scribbled the numbers 407 and 946711 on a small piece of paper (yes, phone numbers had only 6 digits in Amsterdam, those days), and phoned to make an appointment with Kistemaker.

"Do you mean professor or doctor Kistemaker?", replied the friendly voice on the other side of the phone. To my embarrasment I was not sure, but I picked the right one. The next embarrasment was that street numbers only are helpful if the majority of buildings carry them, and after traveling up and down the Kruislaan, I was late for the first job interview in my life.

I was thoroughly impressed, and got an offer to choose one of three positions. One of those was in the group of Marnix van der Wiel, and an introduction by Mihai Gavrila into the world of atoms in strong radiation fields convinced me that a chemistry education did not make very good theoretical physicists. So I decided to pick the position in material science, in the group of Frans Saris. "In that case, we have to talk again", was the AMOLF reaction, and I ended up in Marnix' group anyway, but this time only half theorist, half experimentalist.

I have never regretted this course of events. A few years before my entry, AMOLF had started up a major effort in this new and exciting field, that until than had been dominated by the Russians and the French. People understood perturbation theory, but laser technology had progressed to the state where the light could not be considered a small perturbation on the atom. As a consequence, most of what experiments with the new lasers revealed came as a complete surprise and was not understood at all.

The big players were the French, with three groups working on multiphoton ionization in the Centre d'Etude Nucleaire (Saclay) alone, and related theoretical efforts in Orsay, and at 'Pierre et Marie Curie' (Paris). Nano-second YAG lasers were state of the art, and produced horrible multimode pulses. Saclay had the lead where the lasers were concerned, because they did a fair amount of laser development themselves. The French laser company Quantel was practically their neighbor, and every laser they bought there was taken apart immediately, the components installed on an optical table.

AMOLF tried to strike back by concentrating on the development of electron spectrometers. The detection schemes used in Saclay were rather primitive: either time-of-flight measurements of the ions to reveal the charge states produced, or retarding fields to measure electron spectra. With the latter method Agostini and Petite had discovered a tiny peak of photo-electrons with an energy larger than the photon energy of the laser. This absorption of an additional photon, after the electron had already absorbed enough photons from the field to leave the atom, was a hot item good for one of the most cited Phys. Rev. Letters in this field. The Saclay house theoreticians Gontier and Trahin

thought up the name 'above-threshold ionization' (ATI) for this phenomenon.

Progress in studying this process was slow at first. Electron spectroscopy from electrons created in a laser focus was difficult. To reach the required intensities people had to focus their laser as hard as they could, and as few as a thousand ions in such a small volume were able to set up a space-charge potential comparable to the photon energy. Too measure electron energies with enough sensitivity thus required productions of not more than a hundred electrons per shot, and the retarding- voltage analyzers in Saclay intercepted only one in 10^4 of those.

Marnix had realized this problem, and teamed up with Frank Read from Manchester, a recognized expert on electron optics, to develop an electron spectrometer of unprecedented sensitivity. The graduate student working on the project was Pieter Kruit. The spectrometer that came out of this effort was based on the inverse magnetic-bottle principle: a magnetic field guided all electrons to the detector that had a velocity component along the field lines in the right direction (i.e. half of them). The quickly diverging field lines parallellized all electrons so that the time of flight to the detector (which could be positioned at arbitrary distance without loss of sensitivity) only depended on the total energy.

I joined the group when the spectrometer was just producing its first signals, and I remember the excitement of the detection of the first clear ATI signals from xenon ionized by a dye laser. My initial task was to study the theory of such processes, since another graduate student of the group, Jacques Kimman, was still in line before me to use the magnetic bottle for studying resonant multiphoton ionization of molecules. For some reason I drifted away from Mihai, and worked more and more under directions of Adriaan Tip (I guess mostly because we shared the office). I learned to think in terms of operators, rather than differential equations. I kept in close touch with the experiment as well, grabbing the opportunity to use the equipment if Pieter or Jacques were on holiday.

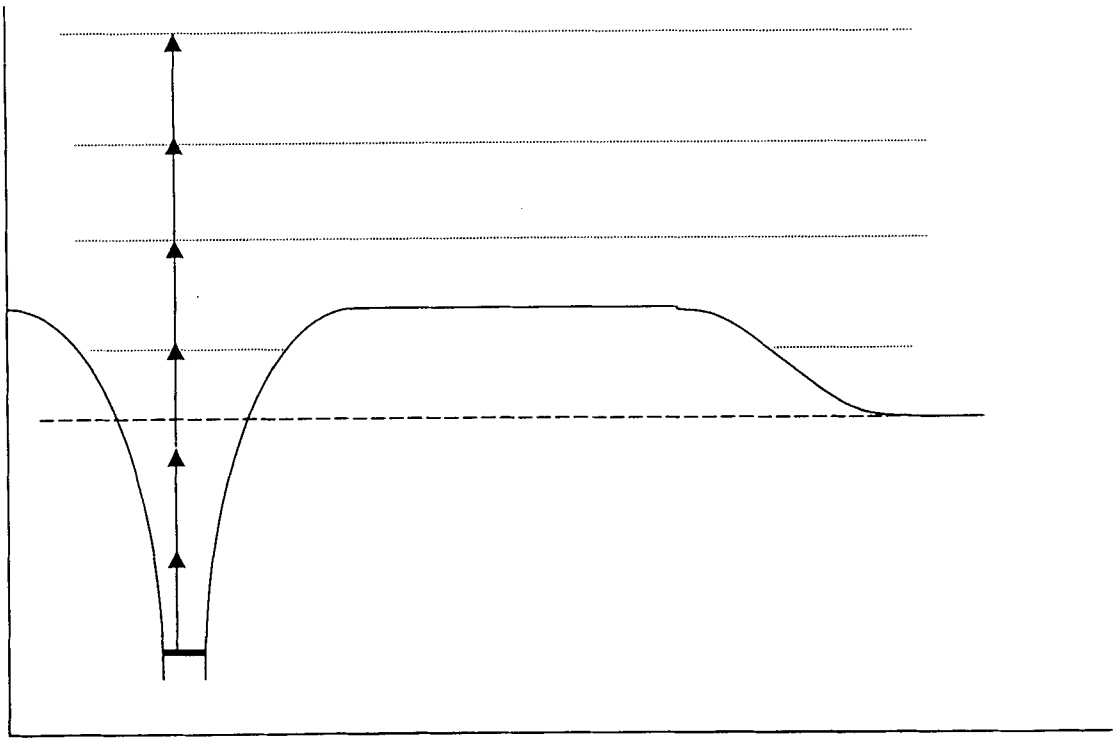
The hot item in the field was 'ponderomotive force', the force that pushes charged particles out of a laser field. The Saclay group had measured ATI spectra with infrared (1.06 um) light, where the ponderomotive force is very big. They saw abundant electrons with kinetic energies of many photons, but could not resolve any peaks. Pieter thought this was a good excersize for our superior spectrometer, and to our astonishment the electron spectrum resolved itself in a sequence of more than ten ATI peaks, all approximately equal in size. This was the most non-perturbative situation ever oberved, and the impact was enormous.

No one had an idea why the ponderomotive force acting on the photo-electrons as they left the focus, did not broaden the observed peaks beyond recognition. A classical calculation of the acceleration due to this effect predicted that it should broaden the peaks about an order of magnitude more than what we saw. In the many group meetings Pieter argued that this acceleration might well be quantized, since after all the photons are responsible for is. The ponderomotive 'acceleration' would then only manifest itself by 'shuffling' electrons between the ATI peaks, moving them on the average to peaks with higher energy. Marnix did not have much faith in classical treatment of this problem, and got a little fed up with Pieters 'simple-man's theory'. Adriaan and I sat down to work on a quantized description of free electrons leaving a laser focus, and this slowly led

to the realization that ponderomotive acceleration is not quantized after all: the force is generated by the scattering of photons to states with slightly different momenta or energies, allowing continuous energy change.

I remember explaining this newly found insight to Marnix, just one week after Pieter had done a precise energy calibration of the spectrometer. This calibration had revealed the embarrasing fact that we had published ATI spectra with an energy scale that was off by one photon: what we thought was the first peak of the spectrum, was actually the second, and at the energy where the first peak was expected there was nothing! Marnix did was not easily convinced by my arguments for continuous ponderomotive acceleration: "we know now exactly with which energy the electrons appear on our detector, and it is the same energy at which the are formed. So they can not have been accelerated." I countered by using the same logic but starting from a different viewpoint: "We know the energy at the detector, and we know that they are accelerated, so the electrons must be formed not at the energy we think, but at a lower one. We do not know for sure what the ionization potential in the field is". A sketch of an energy diagram on the whiteboard, produced to demonstrate the idea (Fig. 1) suddenly showed a remarkable feature: electrons from all ATI channels arrived at the detector at their nominal energy, except that those of the lowest ATI peak were not able to get out of the atom at all!

It was too good to be a coincidence; suddenly everything came together in a coherent picture. The conspicuous absence of the lowest ATI peak, the unobservability of the ponderomotive broadening, the role of Stark shifts, it was all explained by this one assumption that the Stark shift of the ionization potential was equal to the ponderomotive acceleration. And it turned out that Adriaan could prove this equality for a hydrogen atom in a laser field quite easily. The whole process could be understood quite well in classical terms (in which the kinetic energy due to the driven oscillatory motion plays a prominent role), and this new version of the 'Simpleman's theory' became the standard framework for thinking about ATI and related strong-field processes. Even the name caught on internationally.



Ben van Linden van den Heuvell joined the group, and together we performed several experiments to support our ideas. This completed my thesis work, but I remained associated to AMOLF ever since, to participate in the further expansion of strong-field research there. Marnix left to become director of Rijnhuizen, and Ben took over his group. Laser technology progressed at an incredible rate: from a 20ns pulse we moved to 30ps pulse by buying a mode-locked YAG laser. This made it possible to manipulate atoms on a time-scale short compared to the orbit time of the higher Rydberg states. The interest in this was triggered by theoretical considerations on the similarity between ATI (with an intermediate state in the continuum), and multiphoton ionization enhanced by resonance with a high Rydberg state. Arthur ten Wolde and Bart Noordam worked on this for their theses.

The theory group was not idle either. Mihai Gavrila had been developing theory for an interesting limiting case of a super-strong field, where the frequency gets very high, and the intensity even higher to make sure that the light can still overcome the inertia of electrons so that a measurable effect is left. Atoms can be deformed very much away from their spherical shape under such conditions, and this confronted Mihai's student Marcel Pont with many problems during his computations. Cooperation with chemists, who eat non-spherical systems non breakfast, finally overcame all problems: A sabattical stay of Bill McCurdy at AMOLF resulted in accurate predictions of the static behavior of atoms under extreme deformation, where their electron clouds can split up in several parts. Even more exciting was the prediction that above a certain intensity the ionization rate of atoms would start to drop ('stabilization'), so that the calculated structures actually represented a whole new world of exotic but well defined atomic structure, rather than transient structures that would live for just a few optical cycles.

Only a few years after the introduction of picosecond lasers, the pulse duration dropped by another three orders of magnitude. In the framework of a big European network, we imported femtosecond technology from the Laboratoire d'Optique Appliquee in Palaiseau. I went there as a postdoc to learn the tricks of amplifying 80fs pulses with dyes, and together with Ben, Bart and Anton Buiserd we built the FOM-Flits laser system at AMOLF. This laser proved to be an extremely useful 'work-horse' for the later activities in the AMOLF atomic-physics department, mainly because of its flexible design: it was tunable to any wavelength for which we had dyes or doubling crystals, could provide two or three virtually independent (but synchronized) beams, and was exceptionally powerful (for those days), providing pulses upto a mJ.

The exciting experiments done with the FOM-flits laser were so numerous that I will skip most and only mention one. It was the most difficult one in the sense that it required the full capabilities of FOM-flits, overlapping three laser beams in space and time, some of the colors in a hard-to-handle part of the UV. The aim was to demonstrate stabilization for the case of an atomic Rydberg state. It required the effort of the entire department, with students Marc-Paul de Boer, Jan Hoogenraad, Klaasjan van Druten, Raluca Constantinescu, and for three weeks we worked in shifts around the clock to get it done. In the end we obtained incontrovertible evidence for the sought effect.

These were the glory days of strong-field research at AMOLF, in terms of the size of the operation, and it is fair to say that our contributions were paramount to the understanding of this field that we have today. Technology still keeps improving, with pulse

durations now below 20fs, energies of tens or even hundreds of mJ by the introduction of solid-state (Ti:sapph) laser systems, and laser rep-rates increased from 10Hz to 1kHz. Theory of course benefits a lot from technological progress as well. With a single labtop being about 10 times more powerful than the worlds first 'super-computer' (the Cray 1) in 1985, numerical solution of the Schroedinger equation has grown into a very powerful way to study strong-field ionization processes. Surprisingly, considering the dynamics of only a single electron is sufficient to get perfect agreement with experimental results even for multi-electron systems like argon. Through my own efforts, the capability to do such simulations on PC's has expanded into the realm of the truly three-dimensional, allowing us to address questions involving circularly or elliptically polarized laser beams.

Attention nowadays focuses on more subtle effects of ionization, like the things that can happen if a photo-electron re-encounters its parent ion for a second interaction. The probability that this happens is usually well below a percent, but since it can result in final products that would otherwise not occur at all, the effects are very noticeable. Simple elastic scattering from the ion can boost the energy of an electron by a large factor (the laser might accelerate the electron for two consecutive half-cycles in stead of one if back scattering happens just at the time when the field changed polarity). The returning electron can also kick out a second electron in an (e,2e) process, provided it has enough energy. There are many questions about details here, and numerical simulations can reveal details very accurately, producing movies of the process with sub-atomic spatial and temporal resolution.

The future prospects for this field of research are still exciting. Again new technologies will open new windows, and most likely uncover new surprises. Especially free-electron lasers in the EUV or X-ray range might reach unprecedented intensities if we learn how to focus them to the diffraction limit. Relativistic effects will become much more important at these high frequencies. At such frequencies, the response of the various electrons will be much more equal, and many-body effects might dominate the physics, rather than cause secondary effects at the 1happens, there will again be a severe challenge to theory. The trend towards higher frequencies also manifests itself at AMOLF. Through the generation of high harmonics, (in itself is a strong-field effect that until recently enjoyed little attention at AMOLF), intense XUV pulses can be made. The AMOLF XUV group headed by Marc Vrakking now has a very nice setup designed to do exactly that, so we are ready to engage in the new round of exciting physics that will undoubtedly follow.