

DESY Summer Student Program 2010

Photoelectron spectra data analysis

Project report

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Introduction

The XFEL research group, which I joined during the summer student program, has used the possibilities of Free Electron Lasers (FEL) to study the electronic structure of and electron dynamics in Kr atoms. Considering that FELs radiate in the region from extreme ultraviolet to hard x-rays, it is possible to excite the electrons of the inner shells of the atom. Further more, by using two-photon excitations a new kind of Auger resonances are revealed, which are forbidden by parity selection rules for one-photon excitations. Although FELs have a high brilliance, there is some energy jittering and intensity fluctuations, which might blur the fine structure in photoelectron spectrum recorded after excitation of Auger resonances. The purpose of my project was to sort a large number of photoelectron spectra measured each for an individual FEL pulse and to see if it is possible to avoid averaging out this fine structure and extract some clear structures for the decay of the resonance.

Scientific background

The article “Two-photon excitation and Relaxation of the 3d-4d Resonance in Atomic Kr” by M. Meyer et al. was published in Physical Review Letters from this year (PRL**104**, 213001 (2010)). They had used the wavelength 26.9 nm (46 eV) to promote a 3d core electron to the outer 4d shell by two-photon excitation. The subsequent autoionization and Auger decay was studied by means of electron spectroscopy. The experimental data were compared with theoretical predictions, which is shown in figure 1. In the theoretical spectrum, the two-photon resonant excitation from the ground level to the $\text{Kr}^* 3d^9 4d$ states (with total angular momentum $J=0, J=2$) and the subsequent decay to $\text{Kr}^+ 3p^4 4d$ final states was calculated. The $3d^9 4d$ states form two groups of states separated by about 1.2 eV, which according to the spin orbit coupling of the 3d electron to the total angular momentum ($3d_{3/2}$ and $3d_{5/2}$) can be divided into two energetically separated groups. The lower lying group characterized by the $3d_{5/2}$ hole can couple with an outer $4d_{3/2}$ electron to the total angular momentum $J=2$, with an outer $4d_{5/2}$ electron to $J=0,2$, i.e. for the excitation and the decay in total three resonances have to be considered.

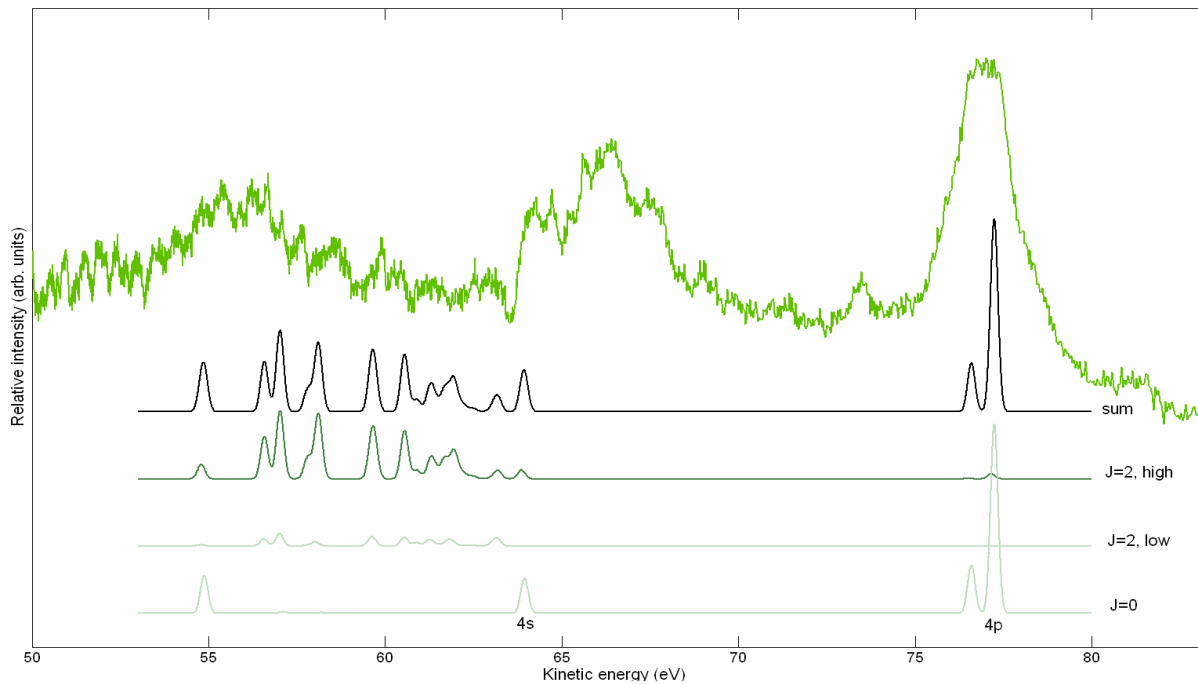


FIG. 1. High resolution electron spectrum of atomic Kr compared to a theoretical spectrum taking into account the relative probabilities for the excitation of different $3d^9 4d$, $J=0,2$ resonances.

In the experimental spectrum three distinct structures can be identified: the peak at 77 eV is formed due to two-photon ionization of neutral Kr atoms, the structure near 65 eV is due to two-photon ionization of Kr^+ ions and the third broad structure between 50 and 60 eV is attributed to the resonant Auger decay, i.e. all structures arise from a non-linear multi-photon process. The two-photon excitation and different ionization pathways are depicted on figure 2.

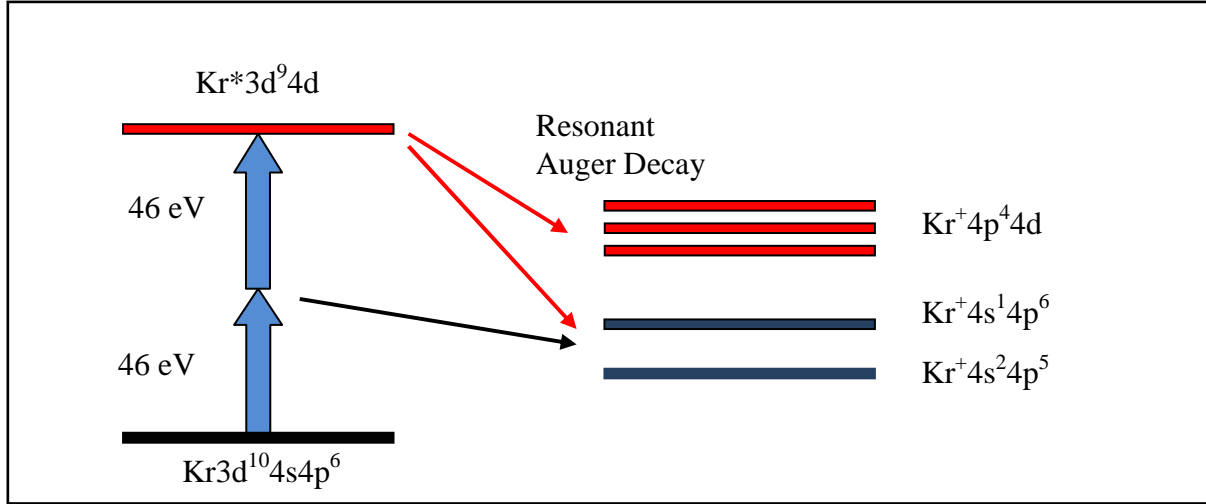


Figure 2. Energy level diagram and excitation scheme for atomic Kr.

The ground state of $\text{Kr} 3d^{10} 4s^2 4p^6$ is taken to $\text{Kr}^* 3d^9 4s^2 4p^6 4d$ excited state by two photons of energy 46 eV. Some femtoseconds after hole creation in the 3d shell, one of the 4p electrons is going to occupy the hole in the 3d shell, the released energy is absorbed by another 4p electron which leaves the Kr atom and ions in the $\text{Kr}^+ 3d^{10} 4s^2 4p^4 4d$ configuration are formed. The other possibility is that the ground state of Kr is taken to the excited state, and autoionizes to $\text{Kr}^+ 4s^1 4p^6$ or $\text{Kr}^+ 4s^2 4p^5$ final states. These last two states can be also formed as a result of one-photon excitation, but in this case the kinetic energy of the photoelectron would be very different (separated by 46 eV).

Fluctuations in FEL beam energy and intensity

To understand why some parameters of the FEL beam are fluctuating, one has to know how the FEL radiation is produced. In the ultraviolet and X-ray region the laser gain is achieved in a single passage of very long undulator. Actually, the FEL radiation is formed in the similar manner as it is formed in the undulators used in synchrotron radiation facilities – electrons are forced to move through oscillating magnetic field and therefore emit photons. The essential advantage of FEL radiation as compared to undulator radiation is its much higher intensity because a large number of electrons radiate coherently. This is a result of so-called self-amplified spontaneous emission (SASE) mechanism. In SASE, electrons, which are losing energy to the light wave, travel on a wavelike trajectory of larger amplitude than electrons, which are gaining energy from the light wave. The result is a modulation of the longitudinal velocity which eventually leads to a concentration of the electrons in slices that are shorter than the wavelength. These micro-bunches are close to the positions where maximum energy transfer to the light wave can happen, and the particles within a micro-bunch radiate like a single particle of high charge. In SASE, electrons produce spontaneous undulator radiation in the first section of the undulator, which then serves as seed radiation in the main part of the undulator. Because of the stochastic nature of spontaneous undulator radiation the SASE process is characterized by fluctuations in wavelength and pulse intensity. If several wavelengths are emitted independently, the wavelength might fluctuate in the order of ± 1 percent. The longitudinal position, where electrons emit radiation varies from electron to electron, but the first emitted wavelength profits the most from the exponential amplification.

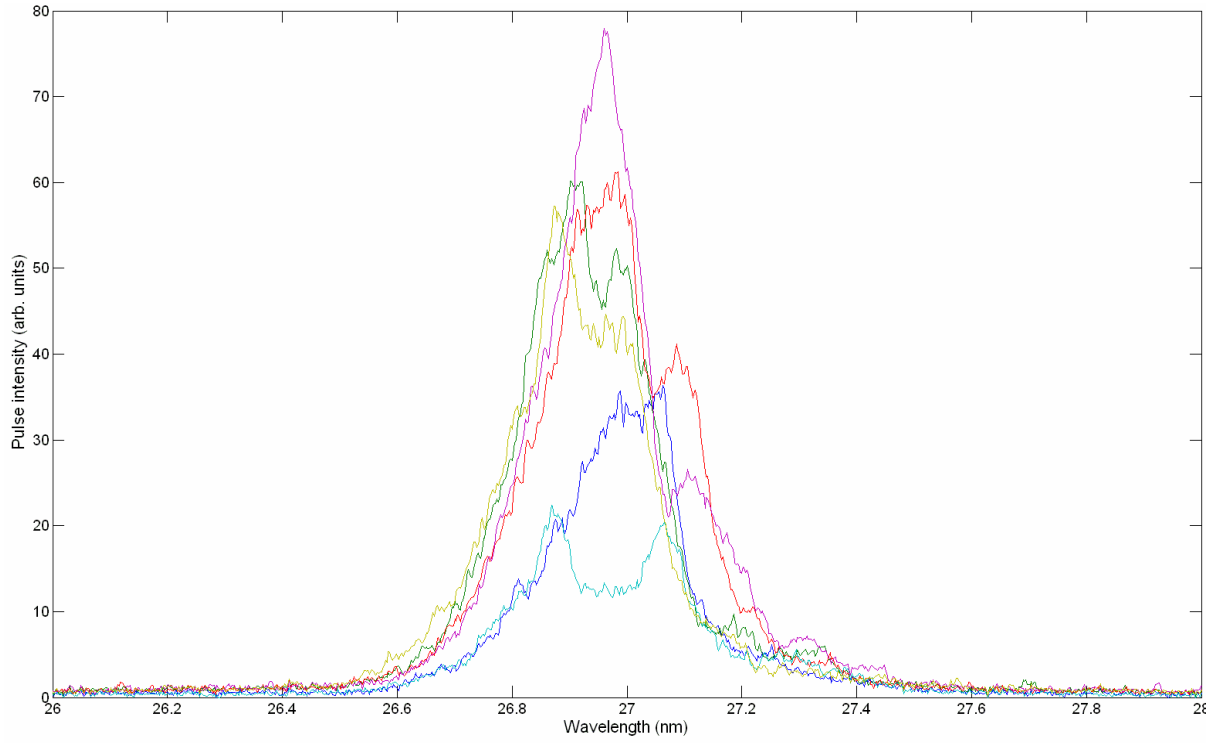


Figure 3. The measured spectra of individual SASE FEL pulses at an average wavelength of 26.95 nm. The single-shot spectra show six peaks fluctuating in size, position and height from shot to shot.

In the end, only few radiation modes will survive, because they absorb most of the energy extracted from the relativistic electron bunch. The spectral distribution of six sequentially measured, individual FEL pulses are shown in figure 3. The intensity fluctuations are clearly seen and also the centre of single pulse fluctuates a little. Because of these fluctuations in the spectral profile, a strong FEL pulse does not always lead to strong resonant excitation and an adequate sorting procedure might be essential for the final data analysis.

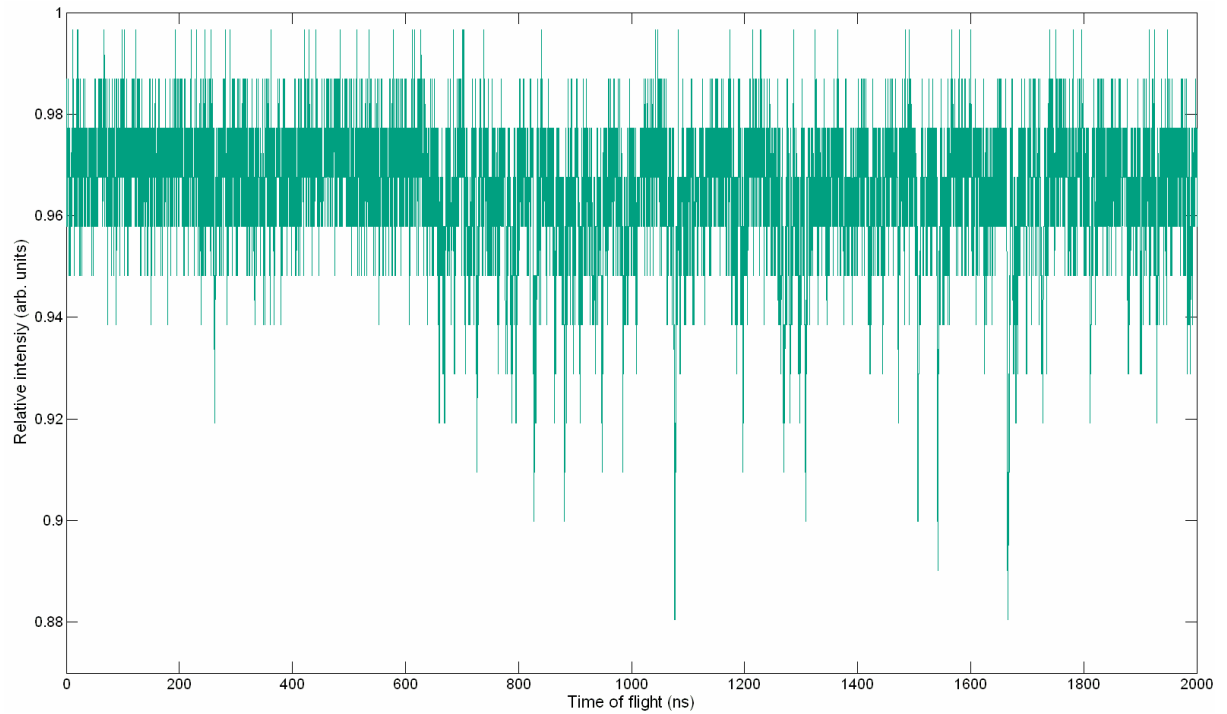


Figure 4. Single-shot photoelectron spectrum of atomic Kr.

Data analysis, results and discussion

In the case of two-photon excitation, which are generally several orders of magnitude weaker than the one-photon processes, it is often difficult to obtain a nice photoelectron spectrum on the bases of one shot, because there is a lot of random noise which stems from thermal noise of the detector and shot noise. One arbitrarily selected single-shot spectrum of atomic Kr is shown in figure 4. With respect to the observed statistics it is clear that averaging over a larger number of single-shot spectra has to be done for a proper analysis.

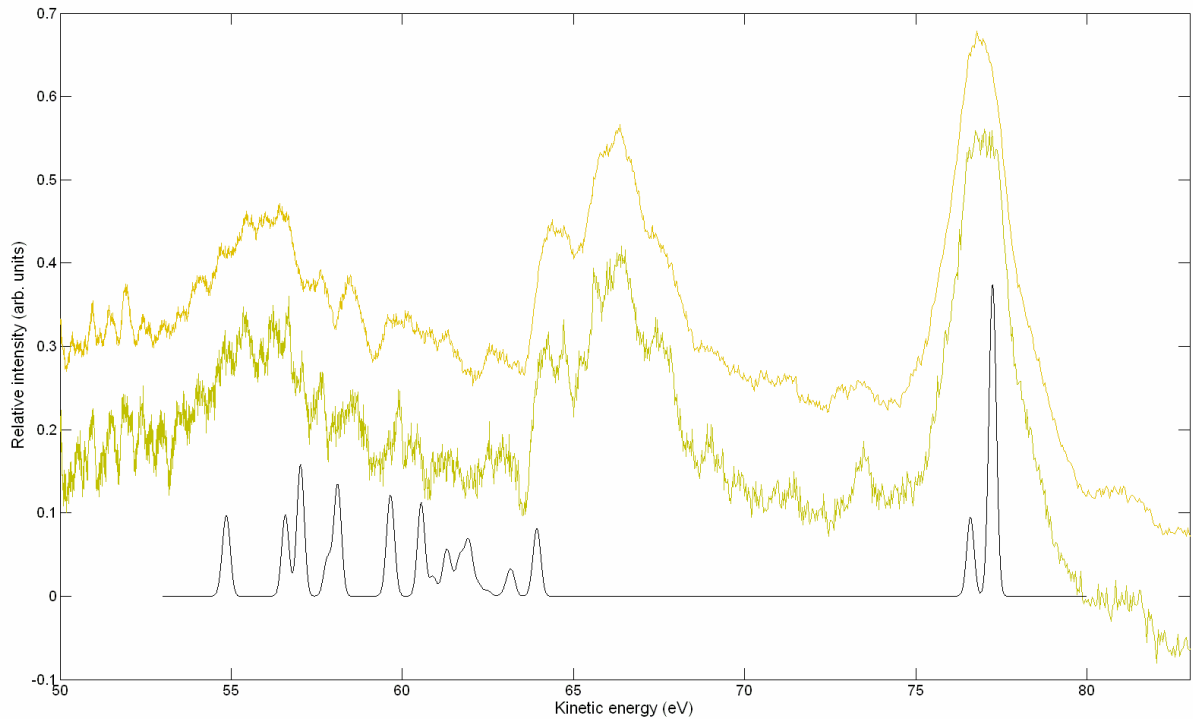


Figure 5. Photoelectron spectra averaged over 500 (green) and 5000 (red) single-shot spectra compared to theory (black).

Indeed, the photoelectron spectrum shown in figure 1 was averaged over 500 single-shots. In order to get better statistics complementary measurements were done and 10 times more single-shot spectra were recorded. For comparison, the photoelectron spectra averaged over respectively 500 and 5000 single-shots are shown in figure 5. The spectrum averaged over the larger number of single-shots looks much smoother and the general shape of the spectrum compared to theoretical calculations is in good agreement. It is necessary to take into account that the energy resolution is about 1.5 eV, therefore it is not possible to resolve all the peaks predicted by the theory. The states separated more than 1.5 eV should appear also in the experimental spectrum. The gap near 59 eV might be observed in the experimental spectrum, but the other bigger gap near 56 eV can not be traced, which might be the result of averaging many different spectra with different intensities or the effect of additional overlapping states (e.g. $3p^4 5d$ shake-up states), which have not been included in the theoretical description up to now. To avoid this kind of averaging over higher and lower intensities, single-shot spectra were divided into 12 intensity bins according to the signal from a gas-monitor detector (GMD), which measures the FEL intensity for each shot. The resulting single-shot histogram is shown in figure 6. The number referenced as GMD intensity is indicating a signal sum over the first peak in the gas monitor detector signal, see figure 7.

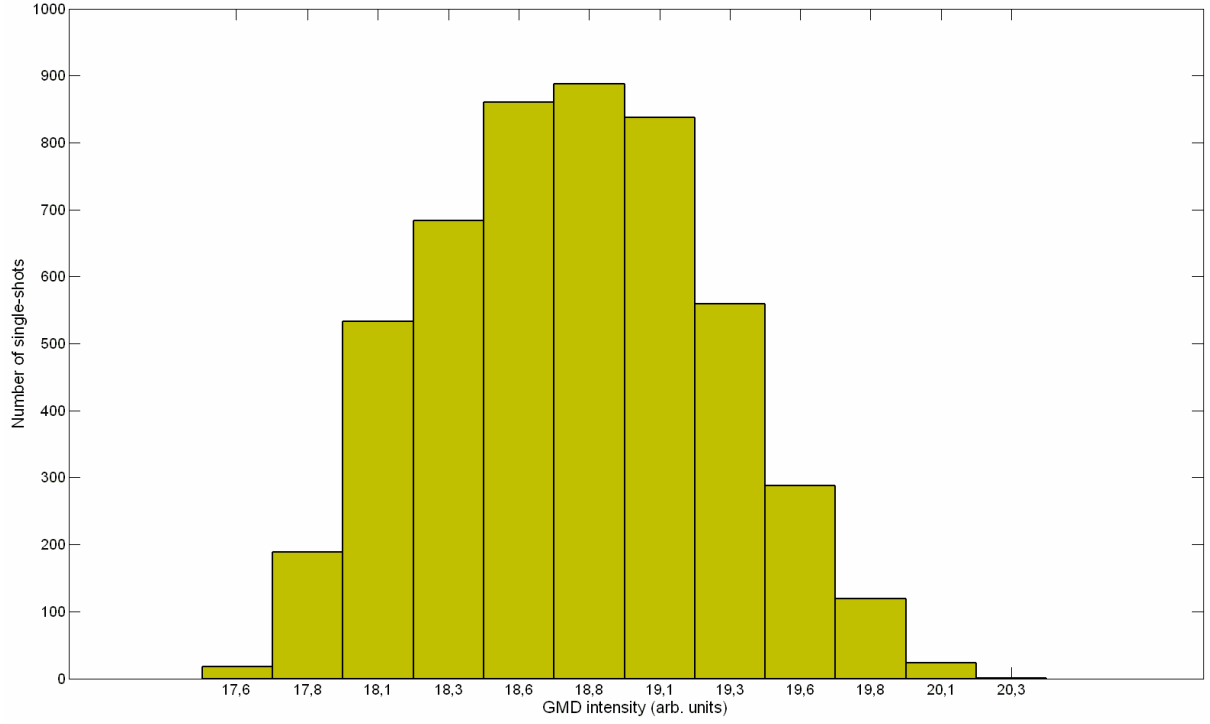


Figure 6. Single-shot spectra binning according to the gas-monitor detector signal.

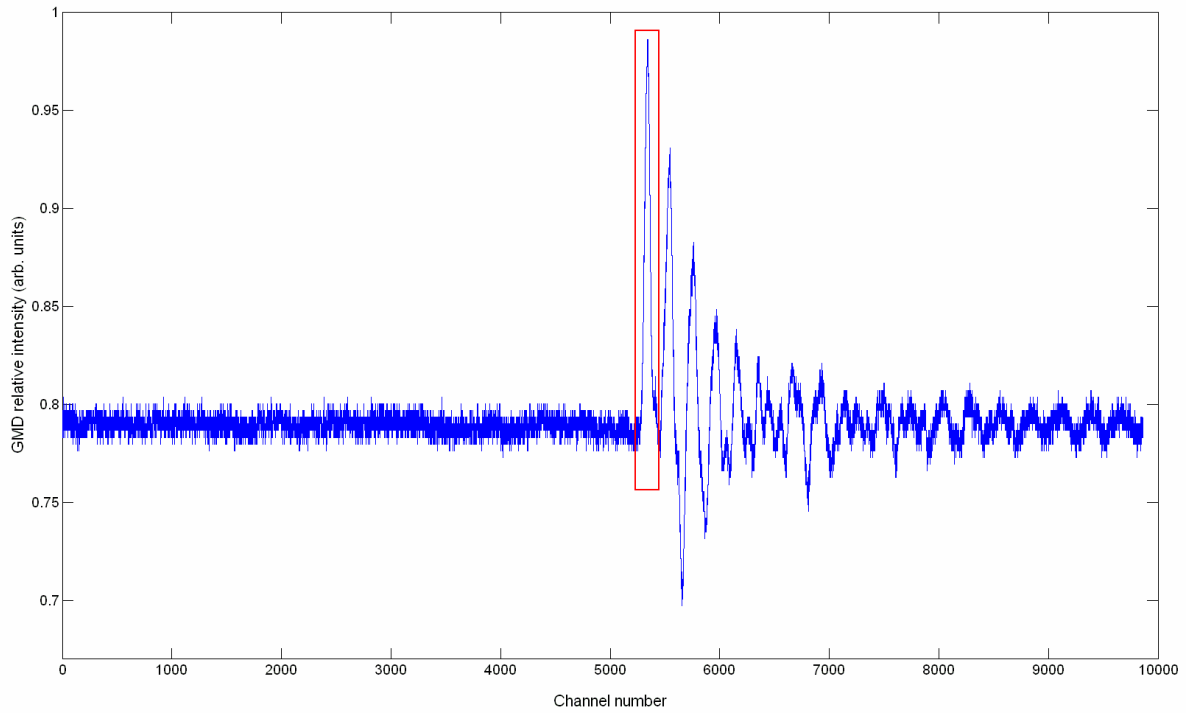


Figure 7. Gas monitor detector (GMD) signal recorded with a digital oscilloscope during one FEL pulse. We took the area of the first peak as a measure of x-ray beam intensity.

For finding the overall width (here the Full Width at Half Maximum FWHM) of the beam intensity distribution we repeated the same kind of binning as before, but with a larger number of bins, see figure 8. From the determination of the FWHM we estimated that the intensity fluctuations are about 6%.

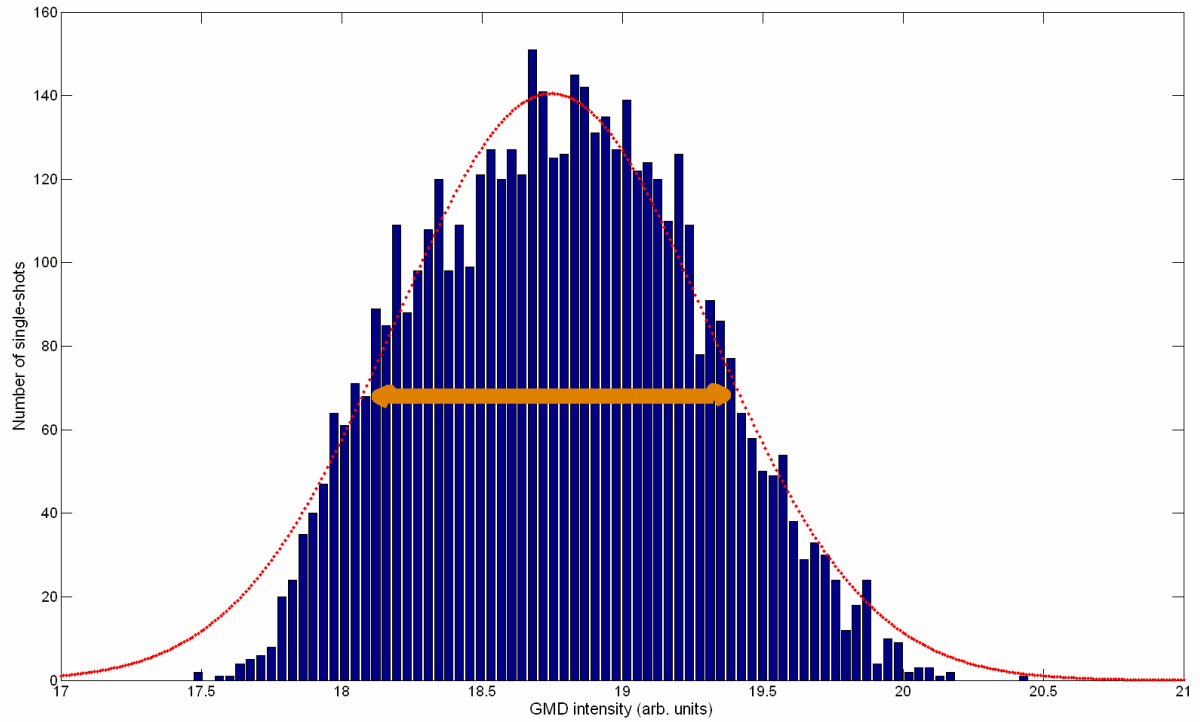


Figure 8. Distribution of the number of single-shot spectra according to GMD intensity. Determination of the FWHM by a Gaussian profile

The averaged spectra received due to binning into 12 GMD intensity bins is shown in figure 9. Binning just by the beam intensity did not give us a more detailed spectrum. For further analysis, we chose one of the twelve spectra that had sufficient statistics and also as high GMD intensity as possible. Therefore intensity bin number 7 was chosen. Inside this bin, the individual spectra were sorted by the Auger intensity into three groups, figure 10.

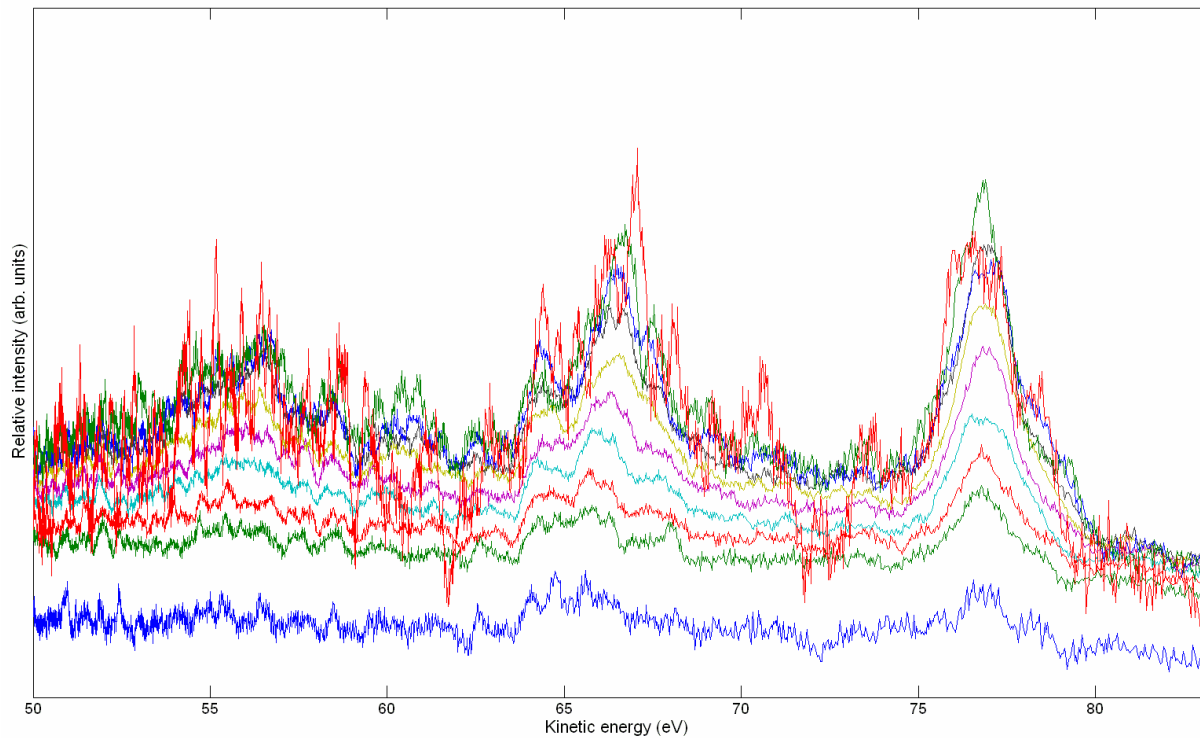


Figure 9. Photoelectron kinetic energy spectra according to GMD signal binning. The spectra of the most and least GMD intensity are not shown because there was too little statistics for them. In this graph individual spectra are shifted relative to each other, the upper ones have higher GMD intensity.

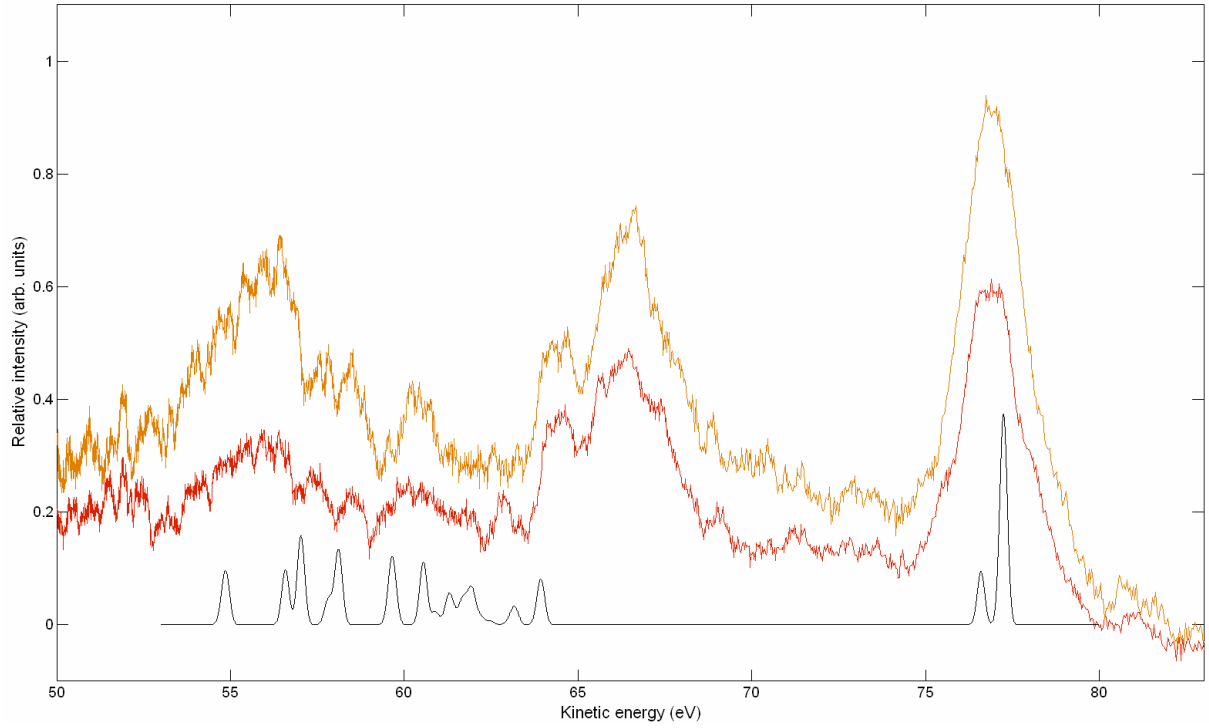


Figure 10. Photoelectron kinetic energy spectra of a chosen range of GMD intensity, which is sorted into three groups according to the Auger peak intensity. The spectrum with most intense Auger structure is not shown because of too low statistics

The aim was to separate those individual single-shot spectra where the Auger structure had a lot of intensity from those, which had less Auger intensity. It is clearly seen in figure 10 that the two spectra are with quite different Auger intensity. In the spectrum with more intense Auger structures also the other peaks are more intense, but less than the Auger structures. This would mean that a higher beam flux increases preferentially the resonant process. In general, the shapes of the two spectra are quite similar actually.

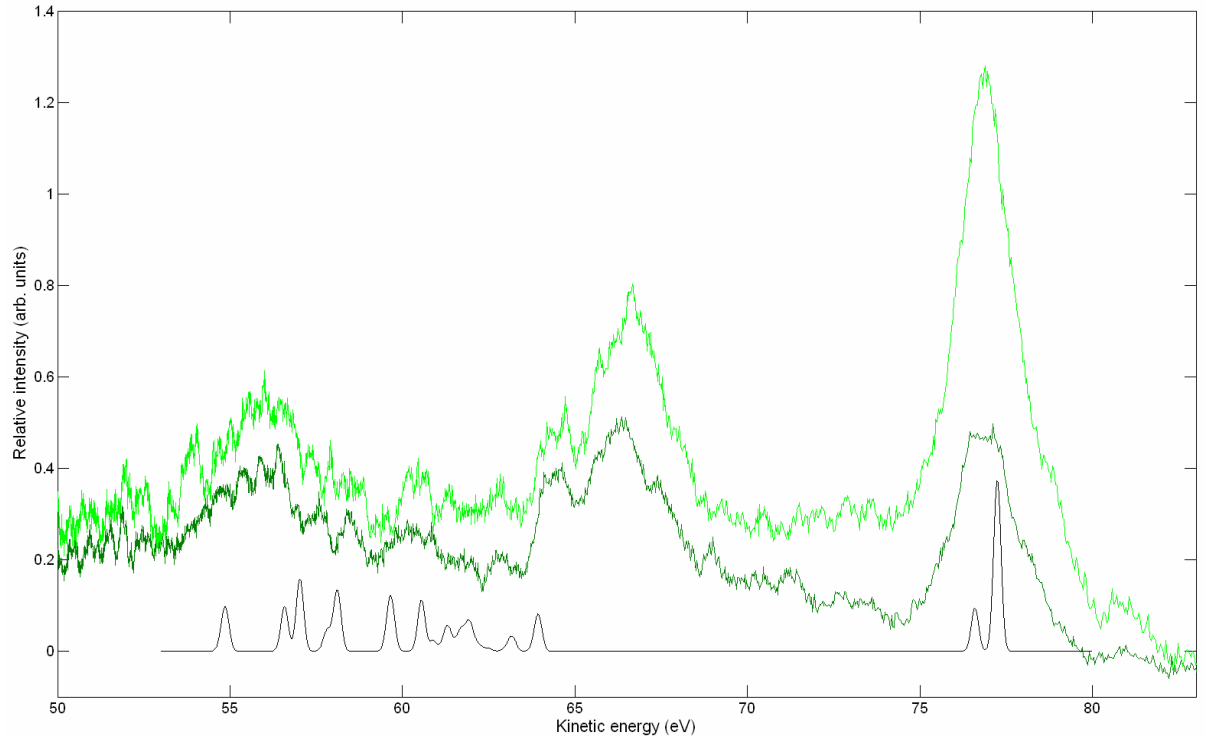


Figure 11. Photoelectron kinetic energy spectra of a chosen range of GMD intensity, which is sorted into three groups according to the 4p peak intensity. The spectrum with most intense 4p peak is not shown because of too low statistics.

Just for interest, we also sorted the intensity bin 7 according to the 4p peak intensity, see figure 11. The result was a bit surprising, because the area of the other peaks seems to stay almost constant, but the area difference of the 4p peak is remarkable. The intensity of the 4p peak depends mainly on the intensity of the incoming beam, therefore the big change in area compared to the other peaks is derived mainly from randomness of single-shot spectra.

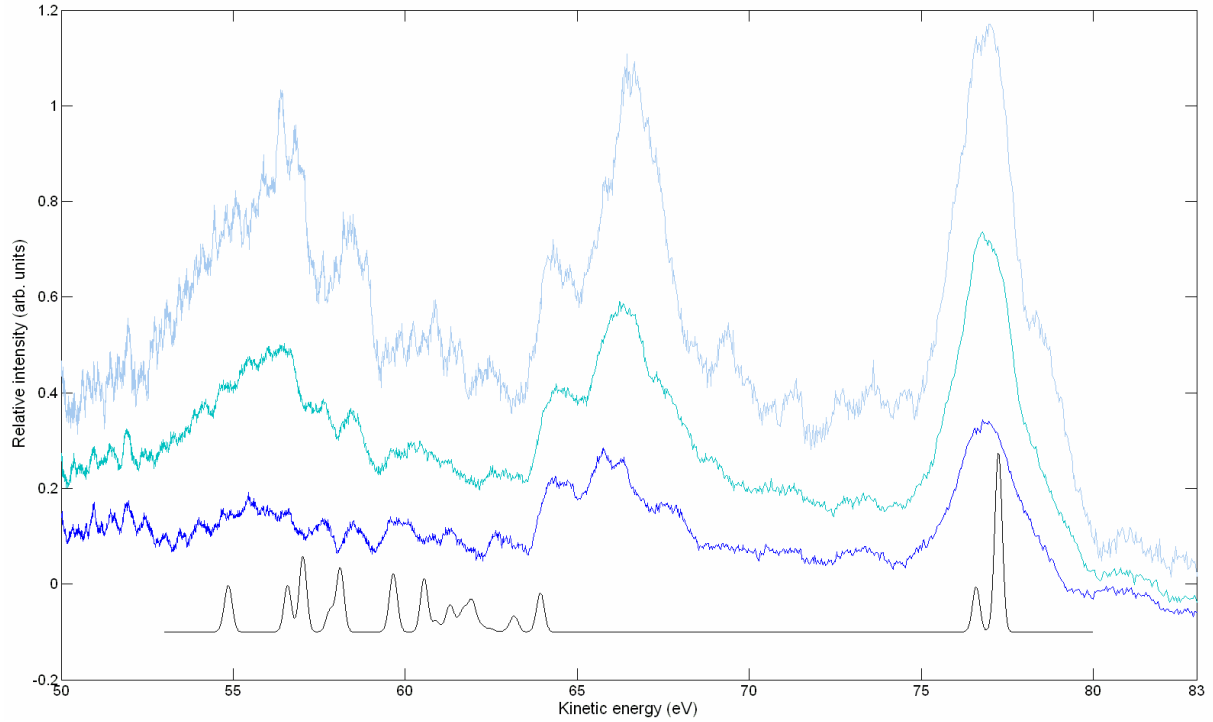


Figure 12. 5000 single-shot measurements sorted only by Auger peak intensity into four bins. The most intense peak is not shown because of too low statistics.

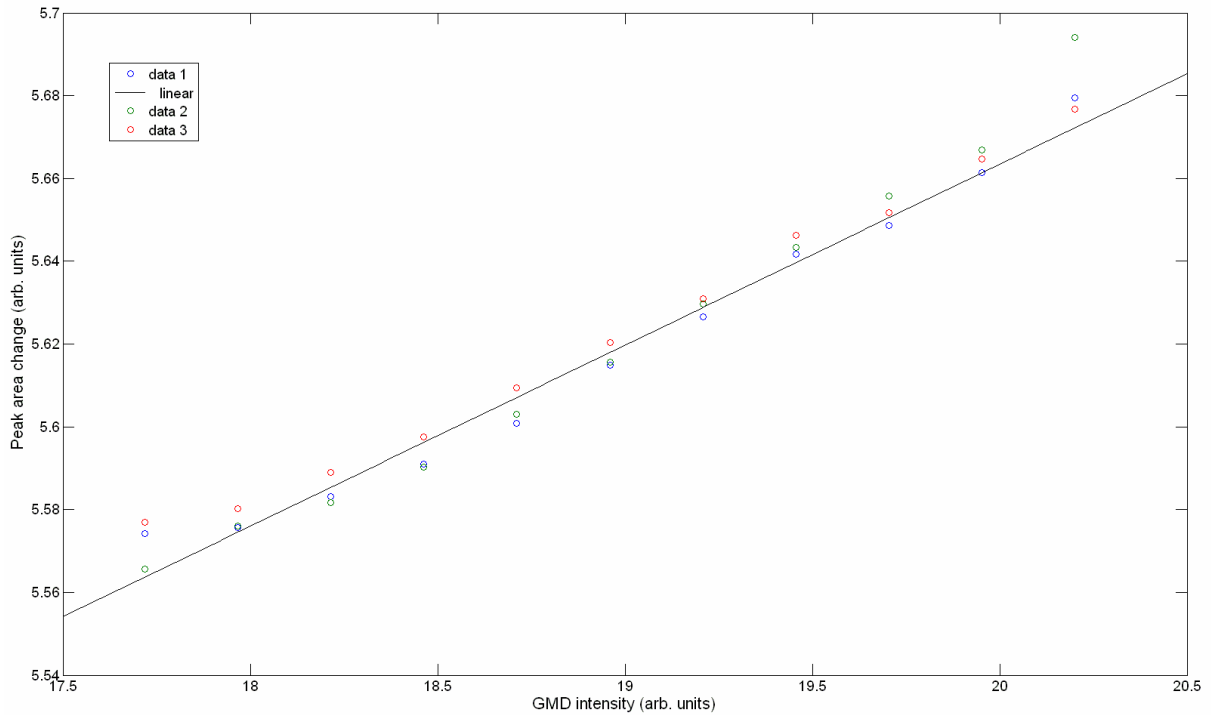


Figure 13. The dependence between incoming beam intensity and peak area change (Auger, Kr^+ and 4p).

As the incoming beam intensity fluctuations were only about 6%, we tried also sorting the single-shot spectra according to the Auger peak intensity without previous GMD intensity binning, figure 12. The two low-intensity spectra look quite similar, but for the upper spectrum with more intense Auger structures it looks like that the position of the three lowest states would be actually closer to each other than predicted by the theory. That could be another reason why there is no gap near 56 eV.

In the next step, we studied how the intensities of the three structures depend on the incoming beam intensity, figure 13. By increasing the exciting beam intensity, the intensities of all three structures are increasing linearly. This is a natural result, because the number of transitions is directly connected to the number of exciting photons.

To find out, if higher incoming beam intensity is affecting the peaks' positions, we observed the position of the 4p peak as this is the narrowest one, see figure 14. At first glance, there does not seem to be any change in the position of the 4p peak with increase of the incoming beam intensity, but when we made a close-up to the region marked with the red rectangle (figure 15) and used the linear regression as an indicator of the inclination, we got that the slope of the regression line was approximately 0.2. The small increase of kinetic energy with increasing FEL intensity is very hard to explain, because every effect that we could think of (the Coulomb interaction between positively charged ions and negatively charged electrons, the non-linear effects of high intensity beam) would decrease the kinetic energy of photoelectrons and not increase it. So, probably the variation of the kinetic energy is within the error bar of our method and might not represent a meaningful result.

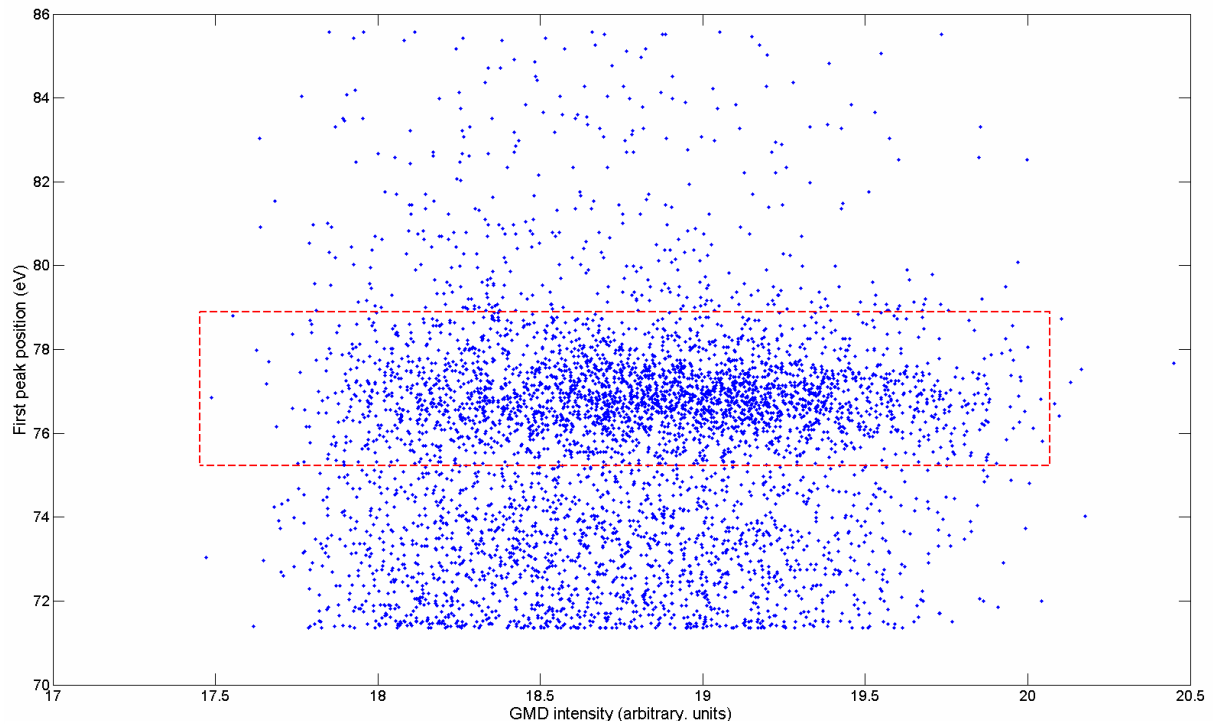


Figure 14. The dependence between incoming beam intensity and 4p peak position. The sharp edge at the bottom of the graph just shows in which region we were looking for the the position change. Although the points on the graph are varying from 71 to 85 eV, it does not mean that the actual peak position would vary that much and these could be just random high peaks in the spectrum.

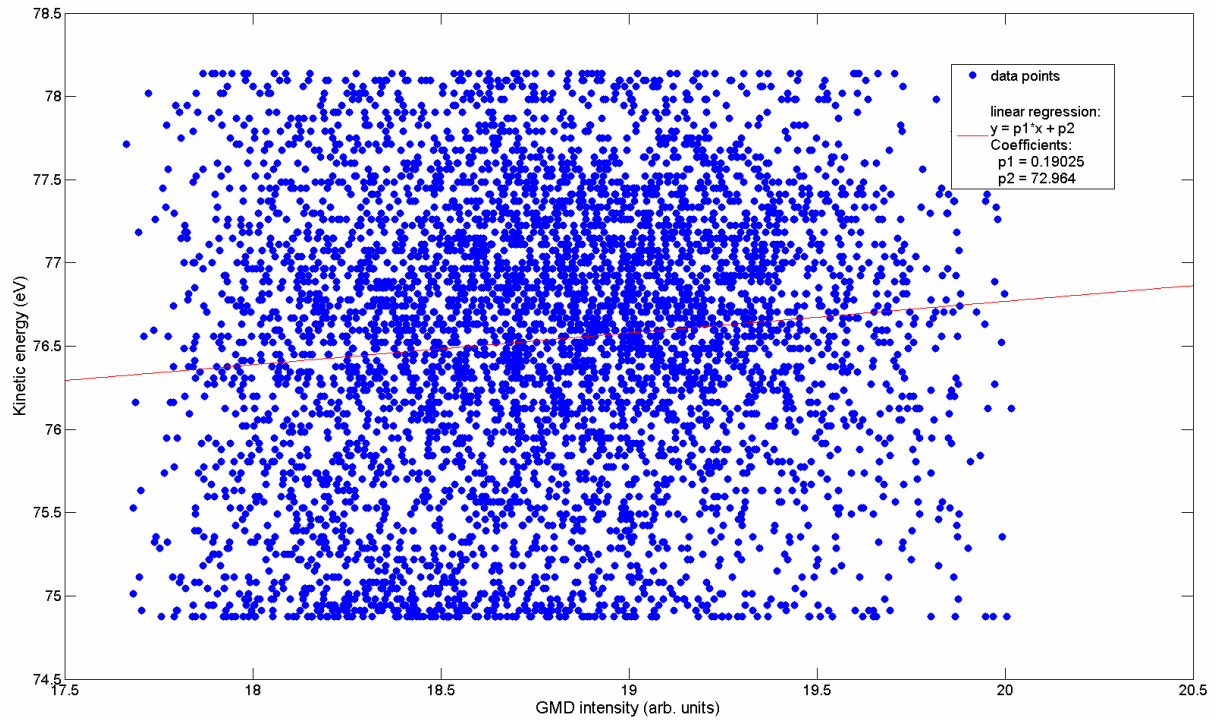


Figure 15. The close-up of the region marked with red rectangle in figure 14. The red line designates the linear regression line calculated by all the points shown in the graph.

Conclusions

- ✓ The incoming beam intensity fluctuations are about 6%.
- ✓ The analysed data had more random peaks than in previous measurements, which might be due to poor incoming beam intensity.
- ✓ Sorting just by the incoming beam intensity is not enough and other additional criteria are needed, like sorting by the Auger peak intensity.
- ✓ The intensity of all three peaks is growing similarly by increasing the incoming beam intensity.
- ✓ The position of the 4p peak might be slightly shifted towards higher kinetic energies by increasing the incoming beam intensity (might be arbitrary).

Summary

My DESY summer student project consisted of analyzing previously measured photoelectron spectra. For the data treatment, I used Matlab software. Different sorting techniques were tested to reveal the fine structure of the Kr photoelectron spectrum. Nevertheless, not all the peaks predicted by the theory could be recognized in experimental spectrum. The reason might lay in still too low intensity of the exciting FEL beam, too little statistics or limited theoretical assumptions. For future analysis, the experiment should be repeated with more intense FEL beam and more statistics should be collected to make any further conclusions. If possible, also the energy resolution of experimental set-up could be improved.

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