Commissioning of a collimated VUV Fourier transform spectrometer for inelastic scattering measurements

Julian Kühnert[1]

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[1]Julian.Kuehnert@uni-konstanz.de

Abstract

A parabolic collection mirror is installed in front of a VUV Fourier transform spectrometer for higher efficiency. First tests with visible light are presented and analysed. Then also measurements with light of vacuum wavelengths are described, showing interference patterns and energy spectra. Conclusions are pointed out and possible further improvements are suggested.

Contents

1 Introduction

The aim of this project work is to further develop an instrument for Fourier transform spectroscopy, working in the wavelength range of soft X-ray and vacuum ultraviolet (VUV). The instrument was developed at Uppsala University by the division of molecular and condensed matter physics, including testings at the MAX-III synchrotron in Lund, Sweden [1][2].
A Fourier transform spectrometer includes an interferometer, where light is split into two branches. After a certain path length difference, which can be adjusted, they are merged together again and interfere. The interference pattern is detected and gives with the known path length difference a frequency spectrum by using a Fourier transformation.
Several utile circumstances of the Fourier transform spectroscopy in contrast to other techniques make this kind of spectroscopy attractive to develop for soft X-ray and VUV. The IR-spectroscopy shows advantages of Fourier spectrometers to grating spectrometers, as one gets information over the whole energy range of a sample's spectrum in a small detection area. This information is spatially distributed in grating spectrometers. Besides this, a Fourier transform spectrometer working with very short wavelengths promises a high resolution of the spectrum, since the path length difference of the two branches can be divided into smaller parts and thus more steps can be scanned.
That Fourier transform spectroscopy in the vacuum wavelength range is not yet highly developed is due to the difficulty to manufacture beam splitters for dividing the light into to branches. Amplitude-dividing beam splitters such as semi-transparent mirrors are not feasible here, since light in this wavelength range is easily absorbed. To solve this problem a wave-front-dividing beam splitter was developed, which consists of a highly reflective mirror with periodically etched slits. Here the challenge in manufacturing is up to the fact that the period of the slits must be smaller than the transverse coherence length of the light which shall be analysed. Only then the split light in the two branches does interfere when merged together again [3].
The Fourier transform spectrometer, which is described in this work, uses beam splitters with $100\,\mu$m wide slits in $200\,\mu$m periodicity. The above mentioned testings at the MAX-III laboratory showed that the peak of a primary beam of energies between 10 eV and 55 eV could be resolved even with low light intensities [2]. The aim is now to make the instrument work as a spectrometer, i.e. to be able to detect secondary photons from a sample. As the vacuum wavelength energies make inelastic scattering measurements possible this kind of spectroscopy is very promising to investigate not only solids but also fluids and gases. In this purpose a parabolic collection mirror is installed in front of the spectrometer which allows to capture photons from the sample in an enlarged solid angle.

2 The collection mirror

To be able to get a higher amount of photons from the sample into the spectrometer the idea is to install a collection mirror in front of the instrument. As a mirror a parabolic mirror is used. Its reflecting surface is part of a circular paraboloid. This kind of mirror transforms a plane wave travelling in direction of the rotational axis into a spherical wave converging into one focal point. The other way round it can convert spherical light diverging from a irradiated sample into parallel light which can then be directed into the spectrometer. Thus, one can make use of two effects. On the one hand one observes a larger solid angle, on the other hand parallel light is guided through the interferometer, which also increases the collection angle.
Besides an expensive manufactured parabolic mirror, a hand-polished dummy-mirror was available in the laboratory for first testings.

2.1 Focal point

The focal point of the available parabolic mirrors is given by the distance $f = 450\,\text{mm}$ towards the middle of the mirror surface and the angle $\Theta = 10\,^\circ$ between incoming and reflected beam likewise at the mirror's center.
This was verified at the very beginning of the project work using a collimated light source. The smallest spot of the reflected light was located in the expected focal point. Apparently the light source was not collimated properly enough what could easily be checked by measuring its diameter in different distances. That caused a larger focal spot as estimated before.

2.2 Collection angle

In this section an estimate of the gained intensity due to the collection mirror compared to the set-up without mirror shall be presented. Note that the demonstrated calculations correspond to a simplified light path. The calculation of a more detailed and realistic model would be beyond the scope of this project work. In figure 1 a scheme of the spectrometer without (i) and with (ii) collection mirror from a side-view is shown.
To compare the two set-ups we assume the light path from the source S to the second beam splitter BS2 in (i) being the same as in (ii) with collection mirror, which is approximately $l = 970\,\text{mm}$.
As we have divergent light through the whole interferometer in (i) the second beam splitter is the device which limits the detected light. The dimension of the beam splitters is 35 mm along and 25 mm perpendicular to the beam and the slits. They are positioned in an angle of $10\,^\circ$ towards the horizontally incoming beam. This gives a vertical angle $\alpha_v = 0.35\,^\circ$ and a horizontal angle $\alpha_h = 0.74\,^\circ$ of the collected light from the source.
The solid angle for rectangular optical apertures is calculated by

$$\Omega_\alpha = 4 \arcsin\left(\sin\frac{\alpha_v}{2}\sin\frac{\alpha_h}{2}\right). \tag{1}$$

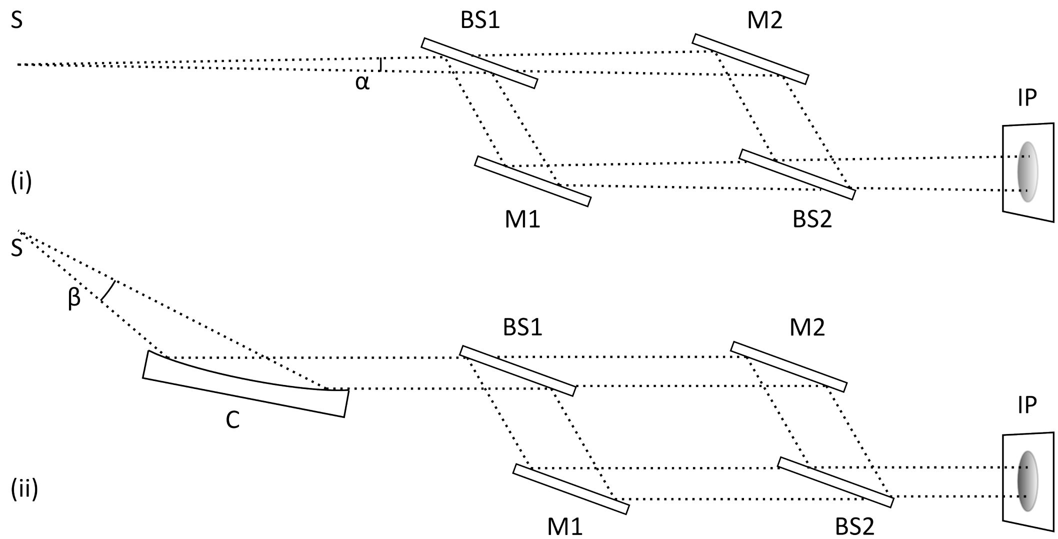

Figure 1: Interferometer without (i) and with (ii) parabolic collection mirror and schematic light-path. S - source, C - collection mirror, BS - beam splitter, M - mirror, IP - interference pattern.

This gives a solid angle $\Omega_\alpha = 0.318 \cdot 10^{-3}\,\mathrm{sr}$ of collected light in (i).
For the second set-up with collecting mirror the light which is directed towards the interferometer is not divergent anymore. Thus, the first beam splitter BS1 is already limiting the the collected light in the same way as the second does. Knowing this, we can calculate the dimension of the actual used area on the mirror, whereas the surface of the mirror is approximated to be flat and positioned in an $5\,°$-angle towards the incoming beam and the reflected beam, respectively. This is only completely true in the center of the mirror. Then, having the focal distance one trigonometrically gets a vertical angle $\beta_v = 0.83\,°$ and a horizontally angle $\beta_h = 1.59\,°$ of the collected light from the source.
In analogy to equation 1 the solid angle of collected light in set-up (ii) is calculated to be $\Omega_\beta = 1.606 \cdot 10^{-3}\,\mathrm{sr}$. Yet the additional reflection decreasing the light intensity by absorption is not taken into account. The mirror's surface is layered with gold for which the reflectivity in this arrangement is about 0.75 [4].
Now calculating the ratio $0.75\,\Omega_\beta/\Omega_\alpha$ minding the reflectivity tells that the collection mirror gives a 4 times larger detected solid angle and hence higher light intensity. This result is less than expected and quite unsatisfying. But till now an additional effect of the divergence in (i) of the light inside the interferometer has not yet been considered. Namely, the collection angle is not only limited by the second beam splitter's total dimension but also by the structure of the periodic slits. The flat position of the beam splitters forces the beam to nearly pass the whole length of the slits. As the light is divergent only part of it passing through the first beam splitter, BM1, is also passing through the second, BM2, which decreases the horizontal angle β_h of the collected light. In figure 2 you can see the spectrometer from a top view,

illustrating the beam from the source through the beam splitters.

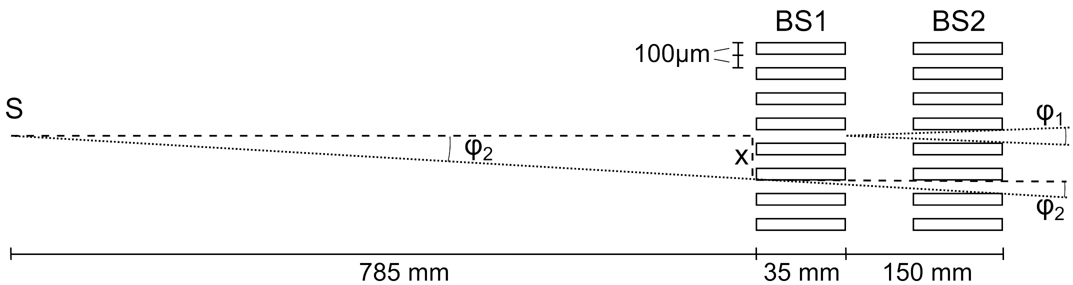

Figure 2: The collected light from the source is limited by the slit-structure of the beam splitters due to its divergence. S - source, BM - beam splitter.

To calculate how much of the light which diverges from the first beam splitter (BM1) passes also the second beam splitter (BM2) we consider a point source at one slit's end in BM1. Knowing the distance to the second beam splitter's end (150 mm) and the width of the slit (100 μm), allows calculating the horizontal angle φ_1 of the light cone getting through:

$$\varphi_1 = 2 \arctan \left(\frac{0.1\,\text{mm}/2}{150\,\text{mm}} \right) = 0.019\,^\circ$$

This angle is to be multiplied by the number slits of the beam splitter to get the whole collection angle. In total the beam splitters hold 125 slits. However, this would give an even larger horizontal angle ($\alpha_{h,slits} = 2.39\,^\circ$) of collected light as before ($\alpha_h = 0.74\,^\circ$) without considering the blocking of the divergent light due to the single slits. Here the view angle of the source must be taken into account which limits the number of the slits actually being used.
Thus, we calculate the angle φ_2 which limits the light getting from the source S through the second beam splitter, BM2, using the distance from the beginning of BM1 to the end of BM2 and the width of the slits:

$$\varphi_2 = \arctan \left(\frac{0.1\,\text{mm}}{(35 + 150)\,\text{mm}} \right) = 0.015\,^\circ$$

Having φ_1 one gets the length x which corresponds to half of the illuminated slits through which light can be detected:

$$x = 785\,\text{mm} \cdot \sin \varphi_2 = 0.42\,\text{mm}$$

The periodicity of the beam splitter (200 μm) gives the total number of slits being used:

$$(2 \cdot 0.42\,\text{mm})/0.2\,\text{mm} \approx 4$$

Thus approximately 4 slits out of the 125 slits of the first beam splitter can be used. Multiplying the amount of slits with the angle φ_1 of the light getting through one

slit gives a reduced horizontal angle $\alpha_{h,new} = 0.08\,^{\circ}$.
Together with the vertical angle α_v from before and using equation 1 the solid angle of collected light in (i) turns out to be $\Omega_{\alpha,new} = 0.035 \cdot 10^{-3}\,\mathrm{sr}$.
The new ratio $0.75\,\Omega_\beta/\Omega_{\alpha,new}$ gives a 35 times larger detected solid angle and higher light intensity accordingly.
This shows that the main property of the parabolic mirror is to collimate the sample's spherical light. The collection angle of the mirror is about 4 times larger than the solid angle without the mirror. The collimation enlarges the angle further by an additional factor of about 9. Note again that the calculated results are approximations. For instance also the dimension of the illuminated spot on the sample has to be considered for further preciseness. Till now the spot has been treated as point source.

2.3 The mirror holder

To be able to install the parabolic collection mirror in front of the interferometer a mirror holder was constructed which allows to even adjust the position of the mirror from the outside of the vacuum chamber. Figure 3 shows the drawing of the interferometer with the mounted mirror in the front.

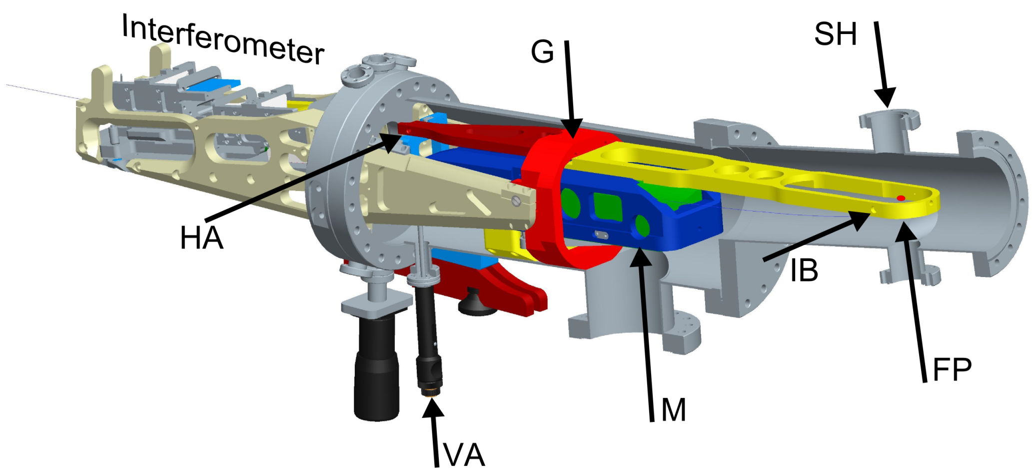

Figure 3: Interferometer with the parabolic collection mirror **M** mounted in a holder, the gimbal **G**, device for horizontal and vertical adjustment (**HA** and **VA**, respectively) of the mirror, its focal point **FP**, the flange for the sample holder **SH**, and the direction of the incoming beam **IB**.

The parabolic mirror **M** sits in a framework, held in place by several steel balls. The balls are sitting on reference surfaces on one side. The mirror is pressed against those by spring-loaded steel balls from the other side. The framework itself is connected by a gimbal set-up (**G**) to holders which are fixed to the old instrument. The gimbal gives freedom for a horizontal and a vertical movement of the mirror around its

center. Here, two flexure pivot bearings were used in each axis. The mounting of the gimbal ring turned out to be problematic as it couldn't pass previously fixed parts and thus couldn't be put into place. Several modifications of the framework were needed until the mounting finally worked.
The position of the mirror can later be adjusted from outside the vacuum chamber by a screw from the side (vertical adjustment **VA**) and a pin from the bottom (horizontal adjustment **HA**). The sample holder **SH** will sit in the very front of the instrument above the focal point **FP** of the parabolic mirror. For the purpose of hitting the sample with the incoming beam **IB** in the focal point an additional bar is mounted onto the framework of the mirror. There are holes in the side of the bar marking the position of the focal point, i.e. if the incoming beam passes the holes on both sides it should also pass the focal point. By this the bar serves as an orientation when aligning the incoming beam.

3 Equipment and principle of operation

For this project available vacuum wavelength light source is a high harmonic generator (HHG). It can provide photon energies in the range of 15 eV to 70 eV. The beam of the HHG is coming sideways into the instrument, i.e. perpendicular to the light path inside the Fourier transform spectrometer. It is focused onto the sample and is meant to hit it in the focal point of the parabolic collection mirror. For this purpose the sample is positioned on a holder which can be adjusted in all three spatial directions and rotated around the z-axis. Photons from the sample shall be collected within the solid angle discussed in section 2.2 and directed into the interferometer, which is of Mach-Zehnder type and in which one of the two light path lengths can be changed by a piezo stage. The interference pattern is amplified by a multi-channel plate (MCP) and observed by a CCD camera on a fluorescence screen. To analyse the pattern, the intensity of several regions of interest are plotted against the amount of changed path length differences. The final frequency-, and accordingly energy-spectrum is received by Fourier transformation, whereas the observed interference pattern of a laser beam with known wavelength, which is also guided through the interferometer, serves as an optical ruler.

4 First testing

To find out about the alignment and the mode of operation of the collection mirror first testings were made in normal air conditions, using a halogen light and a red laser source, respectively. Here the MCP and the fluorescence screen must be removed.

4.1 Transmission and diffuse scattering measurements

First the light source was placed in front of the spectrometer facing it. In order to get light which is enough transverse coherent and hence does interfere after passing

the wave-front-dividing beam splitters, a slit of 25 μm width was located vertically in the focal point of the mirror. The mirror was adjusted looking at the recorded image of the interference pattern. Here, also the advice at the interferometer to block one of the two light paths is very useful to make sure that the two branches interfere in their centers.
After aligning the light source and the collection mirror several spectra were recorded using the instrument as a transmission spectrometer. Besides different colours of safety glasses, fluids of red ink, blue ink and tea with corresponding reference spectra were examined.
In a second set-up the light source was positioned in a 90 °-angle to the spectrometer. A red laser pointer was used hitting on the mat side of aluminium foil in the focal point of the collection mirror. The in such a way diffused scattered light was detected with the spectrometer.

4.2 Results

In figure 4 the Fourier transformations of the transmission patterns with differently coloured safety glasses are illustrated. Figure 5 shows the absorption of the glasses by subtracting the transmission spectra from the reference spectrum without any glasses (black curve in figure 4). As a feedback, we compare the eye-protection specifications on the glasses with the measured data.
The orange glasses are supposed to block wavelengths smaller than 532 nm which correspond to energies above 2.33 eV. This can be verified by both the transmission and the absorption spectrum. The fact, that they already absorb lower energies is due to intensity losses because of scattering or refraction processes within and on the surface of the glasses. This is also true for the next transmission measurements. One could normalize the intensity of the spectra to the level of the reference spectrum and should then get the specified absorption. The green glasses are supposed to block wavelengths larger than 690 nm which are photon energies below 1.80 eV. In the transmission spectrum one sees that all energies up to 2.10 eV are even filtered out. This might be explained by the specification manner. The producer does guarantee the protection of the safety glasses against the specified wavelengths. This doesn't mean that they don't filter out any other wavelengths. The blue glasses are specified to filter out a wavelength of 633 nm which is correlated to 1.96 eV. In the transmission spectrum one can nicely see an totally absorbed window around this energy from approximately 1.8 eV to 2.1 eV. Here again more energies are absorbed in reality than specified.
The transmission and absorption spectra of the measurements with liquids are displayed in figure 6 and 7. Due to absorption, scattering, or refraction processes, the light intensity can also be changed by the transparent container in which the liquids were stored. Thus a reference spectrum with the empty container was recorded and is displayed in figure 6. Subtracting from this reference spectrum gives the absorption spectra.
This time we cannot compare the measurements to any specifications as before. Yet

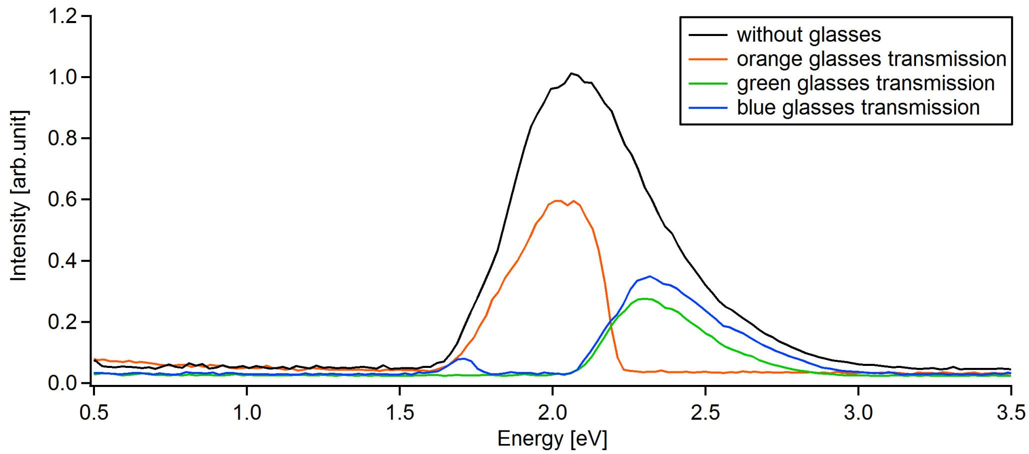

Figure 4: Transmission spectrum of different coloured safety glasses.

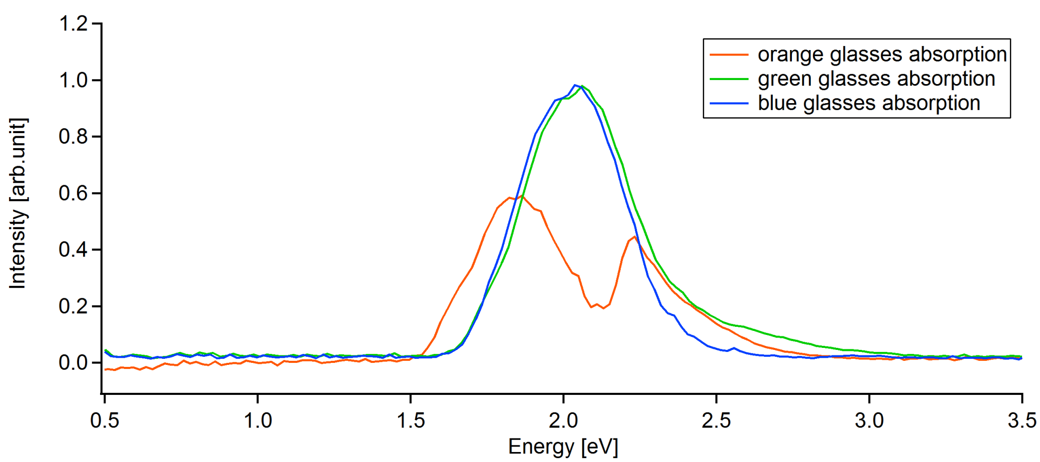

Figure 5: Absorption spectrum of different coloured safety glasses.

they give feedback about the working properties of the spectrometer. In the transmission spectrum of the blue ink one can observe an absorption of light intensity over almost the whole range of the light energy. The absorption spectrum shows, that mostly light up to an energy of 2.3 eV is absorbed. This energy range corresponds to red, orange, and yellow light. Green, blue, and violet light are consequently transmitted what gives the blue ink its colour. The spectra of the red ink show an edge at around 2.2 eV. One can clearly observe how light with lower energies (red, orange, yellow) is transmitted and light with higher energies (green, blue, violet) is absorbed. The spectra of the earl grey tea show a continuous distribution of transmission and absorption. The light intensity of all present energies is decreased. The yellowish colour of the tea causes a small shift of the envelope in the transmission to lower energies and accordingly in the absorption to higher energies.

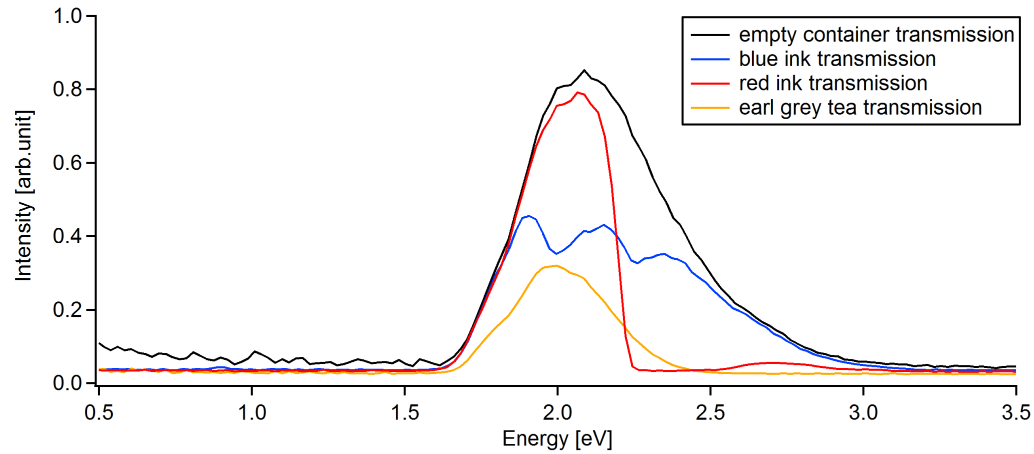

Figure 6: Transmission spectrum of different liquids in a transparent container.

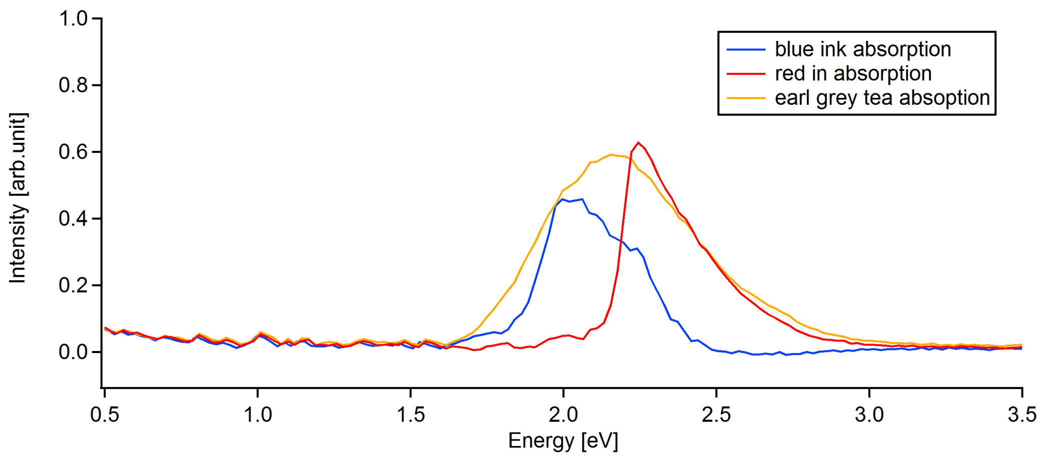

Figure 7: Absorption spectrum of liquids in a transparent container.

In the second set-up of diffused scattering measurement with the light source on the side a clear interference pattern could be detected. The Fourier transformation spectrum showed a high resolved laser peak.

4.3 Conclusion

The results show successful first measurements of the spectrometer including the collection mirror. Even low intensities emerging from the slit in the first set-up, which could hardly be seen on a sheet of paper behind, are detected and moreover lead to well resolved spectra. The second set-up reveals, that an installation with a sideways incoming beam also leads to analysable interference patterns. These two conclusions give green light for the next step to move the instrument to the HHG-lab

to do measurements with a vacuum wavelength light source.
To hold in mind is the high sensitivity of the set-up to the position of the point where the light which shall be detected is diverging from. It should sit exactly in the collection mirror's focal point to get maximal intensity. This was highly noticeable in the interference patterns.

5 Beam time in the HHG-lab

After moving the instrument into the HHG-lab, it could quite easily be attached to the HHG installation using the height adjustable feet. More problematically is the alignment of the whole system, including HHG-beam, sample and spectrometer with collection mirror. A fluorescence powder helped positioning the beam on the sample, but as the focal point of the collection mirror is tiny one has to look on the interference pattern for further alignment.

5.1 Reflection measurements

First measurements were made on a highly reflective sample. Using the zero order of the HHG-beam provides several peaks of different frequencies. The sample was turned in a 45°-angle, in order to directly reflect the incoming beam towards the spectrometer. The so detected interference pattern was intensive enough to observe the arising intensity oscillations and analyse them.

5.2 Inelastic scattering measurements

Next step was to try measurements on non-reflective samples to detect secondary photons. As samples the fluorescence powder and NaCl salt was used. By this only single events could be detected, i.e. individual photons from the sample are amplified by the MCP and can be recorded by the camera. After analysing those images it turned out that the intensity is too small to observe any oscillations due to interference. It was also tried to accumulate several frames of the pattern for each path length difference to get more interfering photons. However it was still too less intensity to observe any oscillations.

5.3 Threshold measurements

After the inelastic resonance measurements failed since too less photons could be detected, again reflection measurements were done to figure out the threshold of how much intensity is needed to get oscillations in the interference pattern in order to resolve photon energies. For this purpose a single energy peak of the HHG was isolated at about 39 eV and targeted on the highly reflective sample. The intensity of the beam was then successively decreased until the peak could barely be resolved anymore.

6 Results

In figure 8 the Fourier transformation of the reflection measurement described in section 5.1 is shown. There are six highly resolved energy peaks of the HHG beam, whereas one more peak at an energy around 42 eV and 43 eV can be assumed. Also the periodicity of the generated light frequencies can be observed (around 3 eV from peak to peak) which is based on the operation mode of the HHG producing multiples of the originating laser. Furthermore an envelope is assumable expressing the varying probabilities of different higher harmonics. The background noise originates from vibrations and other artifacts.

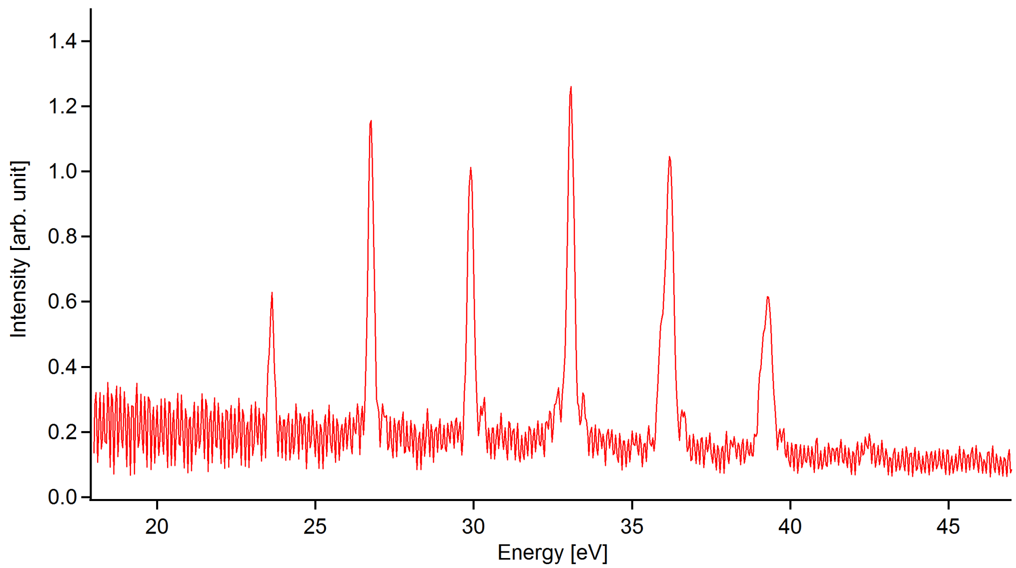

Figure 8: Fourier transformation of the measured interference pattern from a highly reflective sample using the zero order of the HHG.

In figure 9 the results of the threshold measurements described in section 5.3 are presented. On the left hand side one of the recorded interference pattern images during the measurements is shown, on the right hand side you can find the correlating Fourier transformation. The uppermost presented measurement corresponds to the highest recorded HHG beam intensity which was decreased successively towards the lowermost displayed measurement. The more uneven background noise in the first spectra at low energies originates from parts of the interference pattern which are influenced by several spots of interfering light. Those oscillate in a confused way giving raise to lower frequencies and accordingly artificial energies. In measurements with less intensity the interfering light spots are separated more properly, hence they overlap less. Note that while decreasing the beam intensity the amplifying voltage on the MCP was increased to gain the most observable interference pattern for each measurement. In figure 9 this procedure is visible in both the illustration of the patterns and the spectra. If one compares the images of the third and the fourth interference pattern, the last mentioned seems to have a larger illuminated spot. However the crucial interference pattern in the middle is blurred by a higher background intensity. Same in the Fourier transformations. While the peak's intensity is decreasing, the level of the intensity beside is increasing.

7 Conclusion

The first testings showed that the combination of the Fourier transform spectrometer with the parabolic collection mirror works in a successful way. Having a visible light source, transmission and diffuse scattering measurements with even low light intensity led to highly resolved energy spectra. The same is true for the reflection measurements with the HHG source. However, due to too less intensity no inelastic scattering measurements could be done. Only single events were detectable, which didn't lead to analysable interference patterns. More promising patterns could be reached with some improvements of the installation.

First to name is a better adjustability of the whole spectrometer. Till now, the collection mirror can only be aligned in respect to the interferometer but not to the incoming light. The spectrometer including the collection mirror should stand on a $x - z-$table with which one can adjust its height relative to the sample and its distance towards the sample. By this one can ensure that the focal point of the mirror is placed in the point where the incoming beam hits the sample.

Another improvement concerns the detector. To not lose any signal, a more sensitive detector could be of utile use.

Finally one might need a higher source intensity to be able to detect enough photons in the observed solid angle. Using the beam of a synchrotron should give better results.

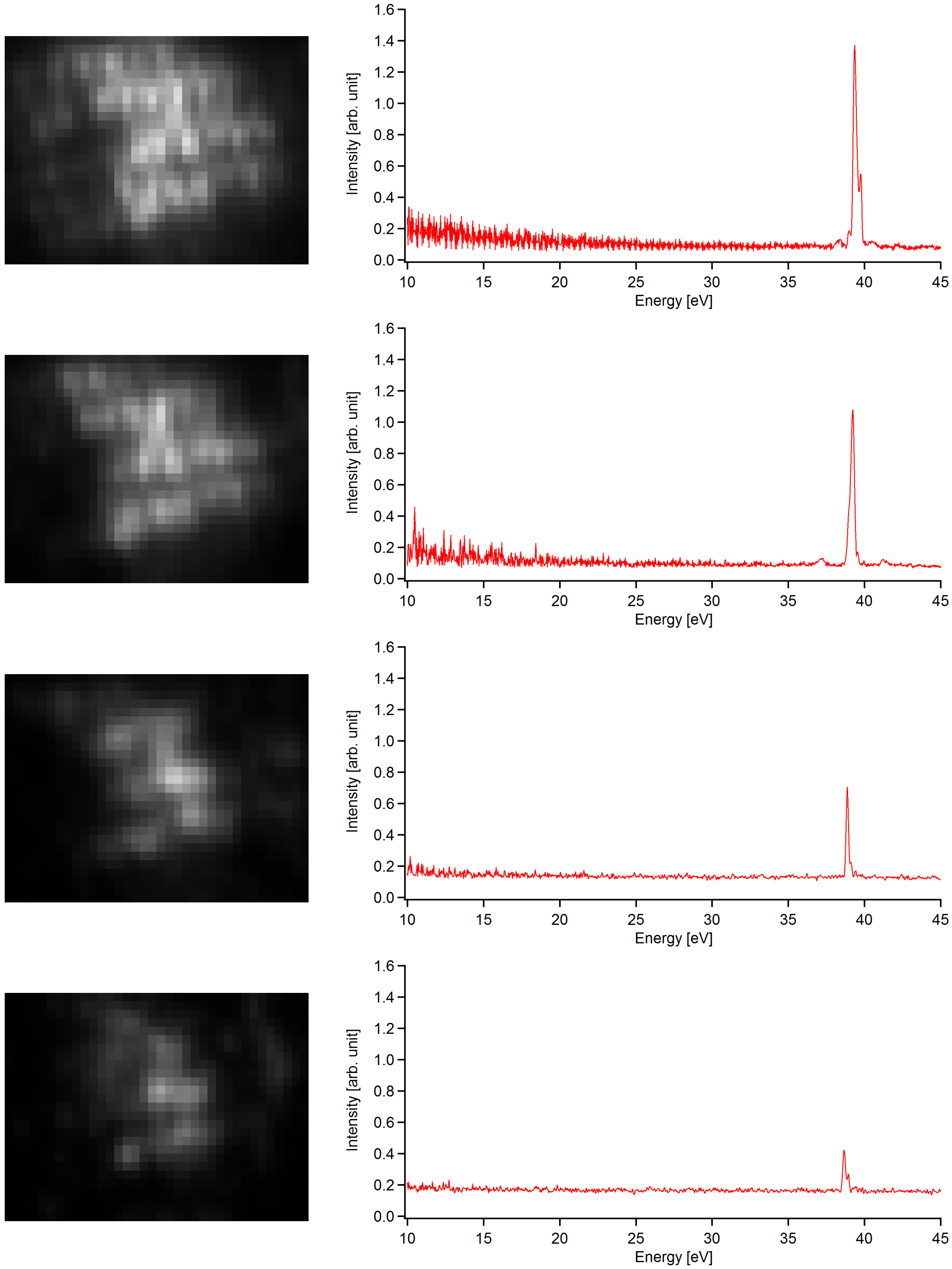

Figure 9: Reflection measurements with decreasing beam intensity. On the left hand side a single image of the interference pattern, on the right hand side its Fourier transformation. Note that not only the beam intensity was decreased but also the voltage of the amplifying MCP was increased.

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