Chapter 4

Experimental Setup

4.1 Introduction

For the purpose of doing mm-wave spectroscopy on high T\textsubscript{c} superconductors, during my graduate research a multi purpose setup has been developed. With this setup experiments such as reflection and transmission can be performed in the frequency range of 4 to 6 cm\textsuperscript{-1}. In this chapter the setup will be explained in detail. Besides the above mentioned experiments a slightly modified version of the same setup has been used to perform Photo Induced Activation of Mm-Wave Absorption (PIAMA) experiments. These experiments were done at the FOM-Institute for Plasma Physics in Nieuwegein, where we were given the opportunity to make use of the Free Electron Laser (FELIX). Details of these experiments will be given along with a short explanation of the principles of a free electron laser.

4.2 mm-Wave Transmission Experiments

Doing experiments in the micro- and mm-wave range one has to face a "build-in" duality. On the one hand, coming from the lower frequencies where ordinary electronics can be used, the wavelength of the radiation is becoming smaller. This makes that waveguides, which are usually employed in the range from 1 to 100 GHz (N.B.: 300 Ghz = 10 cm\textsuperscript{-1} = 1 mm) become extremely small and hard to handle. On the other hand, coming from the high frequency side, the wavelength is becoming longer. This results in difficulties such as diffraction, since the wavelength becomes comparable to the size of the optical components such as mirrors and lenses. One solution to this problem is to use so-called quasi-optics, which means that one uses large mirrors and large lenses.

Another problem in this frequency range is to achieve a reasonable bandwidth. In many experiments resonant cavities are used, thereby exchanging the bandwidth for a higher sensitivity. However, in order to obtain amplitude and phase information in a direct transmission measurement, one needs to use a broadband source. Conventional sources such as Gunn oscillators and impatt diodes have an intrinsic bandwidth of only a few percent. Furthermore, at lower frequencies, where the wavelength becomes very long, the
use of waveguides is limiting the operational bandwidth. Therefore we have chosen to operate at a frequency where we can use quasi-optical methods to manipulate the beam, starting with a source that has a relatively broadband output spectrum. The complete experimental setup is shown in fig. 4.1. The part in the dashed box is not used for the ordinary transmission measurements and will be explained in detail in the following section.

As a source we use a Backward Wave Oscillator (BWO) with an output power several tens of milliwatts [1]. A schematic diagram of a BWO is given in fig 4.2. Its useful range of radiation is from 110 to 180 GHz. The frequency of the radiation is continuously tunable by changing the high voltage from 0.5 to 1.5 kV. Changing the high voltage alters the velocity of the electron beam (3) traveling along a fine grid (6). The electron beam is focused using a magnetic field coming from two permanent magnets (5) packetized with the actual tube, making the total source fairly compact and robust. The interaction of the electrons with the grid results in the production of Bremsstrahlung (7) that is traveling in the opposite direction. The radiation is coupled out via a waveguide (8). The power supply for the BWO is home-build, and has a high voltage stability of about $10^{-5}$, which is necessary in order to obtain monochromatic radiation. The range of frequencies enables us to measure complete Fabry-Perot resonant spectra of our samples, thereby yielding complete phase information via the amplitude and position of the peaks.

The BWO-output first traverses a modulator, thereby creating the ac-response necessary for the detector (usually 100 - 200 Hz for the Si-bolometer). Then an attenuator is used to avoid a nonlinear response of the detector. Next the radiation is coupled out of the waveguide using a Gaussian horn, yielding a fairly well directed beam and is treated quasi-
optically thereafter. The beam is focused into the cryostat through a quartz window, onto the (superconducting) thin film using an off-axis parabolic mirror, placed slightly away from the focal point. This creates a 3:1 image of the waveguide output (P) in the center of the cryostat. The use of mirrors facilitates the alignment and is therefore favored over the available polyethylene lenses. A good alternative would be the use of lenses made of TPX, since these are transparent for visible light and have a low refractive index in the mm-wave range.

The cryostat is a special Variox type with an enlarged sample space, made by Oxford Instruments, and can be regulated in temperature from 1.6 to 300 K. The temperature is measured using a RhFe sensor situated on the cold plate, and a Pt1000 resistor placed very close to the sample, ensuring a stable temperature. Apart from the quartz in- and output windows there are 6 additional optical accesses, two with polyethylene and two with KRS-5 windows, while the other two are blanked. All optical accesses consist of three different windows, one at room temperature, one at 77 K and one at 4 K. This reduces
the intensity of the radiation, but allows the use of exchange gas. During most of the experiments presented later in this thesis, the windows at 77 K have been removed, in order to increase the transmitted power. The sample is loaded from the top and can be taken out while the cryostat remains cold. Using a sample-change compartment on top of the cryostat samples can be changed within 5 minutes, without breaking the vacuum within the cold chamber.

After the cryostat the transmitted radiation is subsequently picked up using a second off-axis parabolic mirror. This creates a second image $P'$ (1:1) which can be used for room temperature measurements. A third parabolic mirror is used to produce a parallel beam, that can propagate over a reasonable distance before being focused onto either one of two detectors, a highly sensitive but slow Si-bolometer operating at 1.6 K, or a fast, but less sensitive waveguide diode detector. Both detectors are used in combination with a lock-in amplifier. The modulation frequencies are 100-200 Hz and about 6 kHz for the Si-bolometer and the diode detector respectively. The complete setup is being controlled by computer making use of either Viewdac or Labview as the operating system.

A recent addition is the implementation of a fully automated sample positioning system using a stepper motor. The construction, shown in fig 4.3, uses a tube for the downward motion and a wire for the upward motion, thereby minimizing the heat load on the sample-holder. Before measuring, the strain on the sampleholder is released. During measurements three sequential scans as a function of frequency are taken at a fixed temperature, namely sample (film + substrate), bare substrate and a reference aperture. All three are part of one sampleholder, which can be reproducibly moved into three different positions. The reference aperture is used to yield absolute transmission coefficients for both sample and substrate, which are then used in the rest of the analysis. The frequency dependence of other components in the setup (attenuator, BWO) is thereby taken into account. It is important to realize that we are dealing with interference effects, and therefore we divide both the sample and the bare substrate single beam spectra by a reference hole. This is different than the common practice for FIR-transmission measurements where the sample spectrum is divided by the bare substrate, in order to obtain the film-response. In that case one has to realize that it is essential to have exactly the same thickness for the substrate used in both measurements. Even though a low resolution will solve the problem of the interference effects, a strong absorption will cause a problem. Since we model the substrate properties separately and enter the experimentally obtained dielectric function into the film+substrate analysis we are not restricted to using the same thickness.

Another common problem in doing mm-wave spectroscopy, both in a quasioptical setup and with the use of waveguides, is the existence of standing waves. Since the wavelength is comparable to the distances in the setup, standing waves will appear, making it more difficult to compare spectra taken on different samples directly since the standing wave pattern will be modified. A solution to this problem in waveguides is the use of isolators, damping out the amplitude of the reflected beam. In quasioptics this is more complicated, and therefore we utilize a small ($\sim 10$ V) ac-voltage superposed on the high voltage. This slightly modifies the outgoing frequency, thereby also changing the standing wave pattern, thus effectively averaging over several different standing wave patterns.
Figure 4.3: Schematic diagram of the automated sample positioning system, minimizing the heat load on the sampleholder.

Apart from the above described frequency dependent measurements, we can also fix the frequency and measure the transmission as a function of temperature. In this case the absolute value of the transmission is harder to achieve since a separate reference run has to be made. This means that one is more sensitive to long term instabilities of for instance the source or the slight motion of the sampleholder as a function of pump rate. Therefore the information obtained in the frequency dependent measurements is used to set the absolute scale.

4.3 Photo Induced Activation of Mm-Wave Absorption (PIAMA)

The same setup as described above, has been used in another series of experiments performed at the FOM-institute for Plasma Physics in Nieuwegein, where we made use of the free electron laser (FELIX). FELIX was used as the pump source in a pump-probe like
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Experiment, Photo Induced Activation of Mm-Wave Absorption (PIAMA). Before proceeding with the physical details of this technique we will first explain the operation principle of a free electron laser [2]. In section 4.3.2 the principle of measurement will be explained, demonstrating the influence of the FIR-radiation on the response at mm-wave frequencies. Finally in section 4.3.3 some of the experimental specifics of this measurement will be discussed. Results using PIAMA on both a conventional (NbN) and a high $T_c$ superconductor (DyBa$_2$Cu$_3$O$_{7-\delta}$) will be portrayed in chapter 7.

4.3.1 Operation Principle of FELIX

The heart of a free electron laser is depicted in fig 4.4. The electron beam is created by an rf-gun, and is propagating in bunches. These electrons are then accelerated to achieve a relativistic electron beam, entering the actual laser section, where they are decelerated by the undulator. This is a series of magnets with alternating poles, creating a spatially periodic magnetic field. The electrons perform a wiggling motion causing them to radiate, having a wavelength ($\lambda_0$) equal to [3]:

$$\lambda_0 = \frac{\lambda_u}{2\gamma^2(1 + K^2)}$$  \hspace{1cm} (4.1)

Here $\lambda_u$ is the period of the undulator field and K is a dimensionless parameter that depends on the magnetic field amplitude. Furthermore $\gamma$ is determined by the energy of the incoming electron, given by $\gamma mc^2$. The optical radiation is confined within the cavity resonator, and is amplified on successive trips by the newly injected electrons. The remaining power of the electron beam after traversing the undulator is dumped.

Figure 4.4: Schematic setup of the free electron laser (FELIX). The accelerated electron beam is injected in the undulator where the infrared radiation is produced by a wiggling motion of the electrons in a periodic magnetic field.
One of the major advantages of a free electron laser is evident from eq. (4.1). This is the fact that a free electron laser is continuously tunable over a broad frequency range, by changing either the energy of the incoming electrons, or the undulator field strength, changing K. By using two accelerators FELIX can span the frequency range from 100 to 2000 cm\(^{-1}\). The first accelerator produces an electron beam having a maximum energy of 25 MeV, while the second one ranges up to 45 MeV. The latter is used to produce the higher frequencies. The generic time structure of the FELIX output is shown in fig 4.5. The output consists of 5 to 10 \(\mu\)s long macropulses separated by 200 ms. Within the so-

![Diagram of FELIX output](image)

**Figure 4.5:** Time structure of FELIX, showing the specifications of both the micropulse and the micropulse.

called *macropulse* there are a large number of *micropulses* with a duration of 1 to several picoseconds and a repetition rate of nominally 1 GHz, i.e. the frequency of the rf-gun. All the parameters mentioned above can be altered up to a certain extent, making the free electron laser suitable for temporally resolved experiments on multiple timescales.

### 4.3.2 Principle of Measurement

The principle of measurement largely resembles the idea of a pump-probe experiment. However, in most pump-probe experiments, two pulses from the same source are delayed with respect to each other, where the second pulse probes the effects induced by the first. In our case the setup consists of two separate parts utilizing two *different* frequency sources. The main part of the setup, depicted in fig. 4.1 is used to measure the transmission of mm-wave radiation through a superconducting thin film. The backward wave oscillator, described in the previous section, produces a *continuous* beam of mm-waves, since we are only interested in *changes* in the mm-wave intensity. To induce changes in the mm-wave
intensity an additional source, the free electron laser (FELIX, indicated in the dashed box in fig. 4.1) is utilized. A pulsed FIR-beam coming from FELIX, temporarily changes the state of the superconducting thin film. This changes the dielectric function and therefore results in a temporary change in the transmissivity at frequencies much lower than the pump frequency of FELIX.

We assume that the superconducting state can be described using a two fluid picture [4], where both the superconducting and the normal fluid coexist. The total conductivity of the superconductor can then be described by

\[ \sigma = \sigma_n(f_n) + \sigma_s(1 - f_n) \]  

(4.2)

where \( f_n \) is the normal fluid fraction and hence \( 1 - f_n \) is the superfluid fraction, \( f_s \). Putting this into the expression for the transmission through a superconducting film on a substrate (eq. (3.6), page 48) yields:

\[ T = \frac{t_s^2}{(1 - f_n)^2 + (2\pi \nu_{mm} \tau f_n + t_s)^2} \]

(4.3)

where we have assumed the thin film limit \( (2\pi \nu_{mm} n d / c \gg 1) \), \( \nu_{mm} \) is the mm-wave frequency and \( \tau \) is the quasiparticle scattering rate. The first term in the denominator \( \sim (1 - f_n)^{-2} \) represents the dispersive part of the conductivity, whereas the second term \( \sim f_n^{-2} \) is related to the dissipative part. Furthermore:

\[ t_s = \frac{4\pi \nu_{mm} \lambda^2(0)}{dc} \]

(4.4)

where \( \lambda(0) \) is the zero temperature penetration depth and \( d \) is the film thickness.

Most importantly, the FIR radiation will perturb temporarily the delicate balance between the superfluid and the normal fluid density, by creation of quasiparticles. This can be done both optically, provided the energy of the incident photons is large enough, or thermally by simply heating the sample, for instance due to phonon absorption. We see that changing \( f_n \) will also alter the dynamic impedance at low frequencies, and therefore the transmission at mm-wave frequencies will be affected. Since we are probing the system response at such low frequencies, in principle we are only indirectly sensitive to changes related to phonons, while the modified electronic properties are directly observed.

From eq. (4.3) we learn furthermore that also the sign of the photo-induced signal can provide important information about the quasiparticle dynamics. In fact, if \( \omega \tau \gg 1 \) the first term in the denominator will be dominant, and we see that \( T \sim (1 - f_n)^{-2} \), yielding an increase of transmission when the quasiparticle density increases. However, if \( \omega \tau \) is of the order or larger than 1, the second, dissipative term will become more prominent in the induced changes, causing the transmission to be reduced when \( f_n \) increases, since \( T \sim (f_n)^{-2} \). The interplay between the quasiparticle dynamics, in particular the scattering time \( \tau \), and the significance of the inductive and dissipative parts of the electromagnetic response function of the superconductor provides an interesting playground. This playground will be explored in more detail in connection with the observed responses in chapter 7.
4.3.3 Practicalities

As can be seen in fig. 4.1 the infrared beam coming from FELIX is entering the cryostat under an angle of 45°, through a KRS-5 window. In fig 4.6 we show the transmission through the window for the relevant frequencies. The transmission is featureless and fairly frequency independent for frequencies down to 250 cm\(^{-1}\). At this frequency the transmission goes to zero due to a strong phonon Reststrahlenband.

The beam is incident on the thin film having an unfocused diameter of about 10 mm. The diameter of the inner KRS-5 window is 10 mm, meaning that the beam covers the whole sample (10x10mm) making the excitation as homogeneous as possible. The infrared beam induces a change in the dynamic impedance of the sample and therefore changes the transmission at mm-wave frequencies. The temporal evolution of the Photo Induced Activation of Mm-Wave Absorption is monitored using a fast waveguide diode detector, and an (even faster) oscilloscope. The total detection system has a integrating time constant of approximately 1 \(\mu\)s, limiting our time resolution to the macropulse. Moreover, the time constant of the differentiating circuit connected to the diode, was measured independently to be 45 \(\mu\)s. All the data presented in chapter 7 have been corrected for this by convoluting the transients with an exponential response function.

![Graph showing transmission through the KRS-5 window](image_url)

Figure 4.6: Transmission through the KRS-5 window, used during the PIAMA experiments.
The optimization of the position of the beam is initially done by measuring the direct transmission through a reference hole replacing the sample. Thereafter the induced change in mm-wave transmission is used to improve the position further. The power and the stability of FELIX are checked by using a power meter directly in front of the first window, while at some frequencies the transmitted power directly behind the cryostat is measured. This yields the attenuation of the beam due to the windows and can then be used to obtain the absolute power incident on the sample. In case of the studies as a function of power we used two options to attenuate the beam, yielding very distinct information. First it is possible to shorten the pulse length, thereby lowering the average power, but leaving the pulse height of the individual micropulses unaltered. Second we used attenuating materials such as dielectric films to reduce the instantaneous power.

Using eq. (4.3) we can make a rough estimate of what the magnitude of the induced signals will be. For a superconductor at the lowest possible temperature, assuming that all electrons are paired eq. (4.3) reduces to

\[ T = \frac{t_s^2}{1 + t_s^2} \]  

(4.5)

Assuming \( \lambda(0) = 1500 \, \text{Å}, d = 1000 \, \text{Å} \) and \( \nu_{\text{mm}} = 5 \, \text{cm}^{-1} \) the transmission is approximately \( 10^{-5} \). Assuming furthermore that the induced changes due to the presence of the FIR pulses of FELIX are of the order of a few percent, we know that the changes to be detected will be approximately \( 10^{-7} \) times the starting output power. This means that the dynamic range of the setup needs to be at least one order of magnitude better than that. Furthermore this implies that this technique is limited to thin films, since for the thicker films the unperturbed transmitted signal will be insufficient.

References


