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Energy relaxation time in NbN and YBCO thin films under optical irradiation

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Abstract. For a systematic study of energy relaxation processes in thin NbN and YBCO films on sapphire substrates, a frequency domain technique has been set up and employed. The magnetron sputtered NbN films of 3 nm to 22 nm thickness and pulsed-laser deposited YBCO films with thicknesses between 20 nm and 45 nm were excited by amplitude-modulated optical radiation (λ = 850 nm). The response spectra were analyzed on basis of the two-temperature model of the energy dynamics in the interacting electron and phonon subsystems at quasi-equilibrium conditions. An increase of the energy relaxation time with increasing film thickness has been obtained for both NbN and YBCO thin film samples.

1. Introduction
Highly sensitive and fast superconducting Hot-Electron Bolometer (HEB) detectors are currently of high concern in research aiming to develop detectors for spectroscopy, security and astronomy applications in a broad spectral range. Detectors made from low-temperature superconducting materials such as niobium nitride (NbN) meet the requirements of high sensitivity up to the single photon regime. High-Τc materials like YBa₂Cu₃O₇₋δ (YBCO) are suitable for the development of very fast detectors since their electron-phonon interaction time is about one magnitude smaller (τₑp ≈ 1-2 ps for YBCO [1]) compared to low-Τc materials (τₑp ≈ 10 ps for NbN at 10.5 K [2]). Another advantage of high-temperature superconductors is the device operation at liquid nitrogen temperatures allowing the use of powerful and compact cryocoolers.

The photoresponse of superconductors already attracted considerable attention and was investigated by several groups either by using optical pulses [1], [3], [4], [5] or amplitude modulated THz radiation [1], [6], [7]. In both cases the experiments were carried out at high laser pulse energy or high local oscillator power, respectively, resulting in significant variation of the electron and phonon temperatures in the thin films with respect to the bath temperature [8]. Also, for sub-THz radiation a contribution from vortex motions to the response was observed for both materials [9], [10], [11], which have a strong influence on the intermediate frequency bandwidth of HEB mixers made from NbN and YBCO films. In order to investigate the speed-limiting processes of detectors, it is essential to study the energy relaxation processes under quasi-equilibrium conditions, i.e. for small excitation power, when a change of the effective temperatures of the electron and the phonon subsystems is much smaller than the operation temperature. Therefore, we performed our measurements using optical radiation to avoid any influence of moving vortices. The frequency domain technique supports
the detailed determination of the energy relaxation time at quasi-equilibrium conditions, since high enough resolution can be reached even at low excitation power.

In this report, we present in detail the frequency domain measurement setup as a tool to study energy relaxation processes by measuring and analyzing the response spectra to optical radiation. We demonstrate the application of this technique to NbN and YBCO thin films as two representatives for similar application areas, which are very different in their material composition, microstructure and electron and phonon properties.

2. Fabrication and characterization of NbN and YBCO samples

The YBCO and NbN samples have the same geometry consisting of several parallel stripes of 2 µm in width and 2 µm space in between. The length and amount of stripes are adjusted to realize a normal state resistance \( R_N \) above the superconducting transition of about 100 Ω. The stripes are embedded into gold contact pads of a coplanar design, which also serves to reflect the photons incident on the contact areas, so that the absorbed radiation is confined to the central part of the sample.

The NbN thin films were deposited by DC reactive magnetron sputtering of a Nb target in an Ar/N\(_2\) atmosphere. As substrate, one-side polished R-plane sapphire was used and heated to 750°C during the deposition of 3 nm to 22 nm thick films (for more details on NbN thin film fabrication and properties see [12]). To pattern the central part of the NbN samples, a standard photolithography process was used with subsequent reactive ion etching. After a second lithography step, the sample surface was cleaned \textit{in-situ} with an ion beam before sputtering the gold contacts on top of the NbN films.

The YBCO thin films were fabricated using on-axis pulsed-laser deposition (PLD) technique. A 50 nm thick CeO\(_2\) buffer layer was deposited on sapphire substrate at a temperature of 820°C. The YBCO layers with different thicknesses between 10 nm and 100 nm were \textit{in-situ} deposited on top of the CeO\(_2\) layer. Finally, a 60 nm Au protection layer was grown \textit{in-situ} using the same PLD technique. The Au contact pads of the YBCO films were patterned by standard photolithography and argon ion milling. At the final step of the sample fabrication the gold passivation layer was removed from the center part of the sample by wet etching with an I\(_2\)-KI solution.

The fabricated samples were characterized in a DC experimental setup using a quasi four-probe measurement configuration. The temperature dependence of the resistance \( R(T) \) of the thin NbN films showed a steep superconducting transition of about 1 K in width, taken from 10% to 90% of \( R_N \), resulting in a temperature derivative of the resistance \( dR/dT \) of up to 1000 Ω/K. The width of the superconducting transition of YBCO was broader and thus the \( dR/dT \) values were not larger than 40 Ω/K.

The critical temperature \( T_C \) is defined as the highest temperature at which the resistance reaches zero value. The \( T_C \) values of the NbN films with thicknesses \( d \geq 10 \) nm increased gradually from about 15.5 K up to 16.2 K for the thickest films, which is only 1 K lower than \( T_C \) of the bulk NbN samples. For the films thinner than 10 nm we observed a strong reduction of \( T_C \) down to 12.5 K for the thinnest 3 nm film. The YBCO films with \( d \geq 30 \) nm showed almost a constant value of \( T_C \) about 83 K. The critical temperature of thinner films decreased significantly down to 25 K for the 13 nm thick film. The decrease of \( T_C \) with decreasing film thickness is discussed in detail in [13] for NbN and in [9] for YBCO thin films.
3. Experimental setup

The contact pads of the samples are designed in a grounded coplanar waveguide layout, where the gap between the readout line and the ground is engineered to match to 50 Ω impedance. The samples are bonded with indium to a bias-tee circuit which is implemented with Surface Mount Device components. The schematic diagram of the experimental setup for measurements of the response of thin film superconducting samples to amplitude modulated optical radiation is shown in figure 1. The amplitude modulated optical power is generated by a commercial Toptica laser system [14], consisting of two temperature-controlled cw-laser diodes with 850 nm wavelength, coupled into the same single-mode optical fiber. The system has a maximum output power of 30 mW, which can be tuned down arbitrarily. The radiation frequencies $f_1$ and $f_2$ of the two laser diodes are slightly detuned, resulting in an amplitude modulation of the output power at $\Delta f = f_2 - f_1$ with a modulation depth of the power of about 24%. During measurements, one of the diodes operates at constant conditions, i.e. bias current and operation temperature. The temperature of the second laser is changed resulting in a shift of its radiation frequency. A change of the laser diode temperature which corresponds to a shift of the modulation frequency from 10 MHz up to 10 GHz results in a variation of the output power less than 5%. The total optical power measured by an optical power-meter (Thorlabs PM30) and the modulation depth measured by a fast GaAs photodiode (Thorlabs SM05PD2) [15] stay the same within the accuracy of the used instruments throughout the considered range of modulation frequency. The amplitude modulated laser power is passed into the experimental insert into a liquid helium transport dewar by an optical multimode fiber (105 µm core diameter) that ends at $\approx 1$ mm above the sample position. Due to the numerical aperture ($NA = 0.22$) of the fiber, the beam spot at the sample position has a diameter of $\approx 1$ mm, thus ensuring a homogeneous irradiation of a typical sample area of about 50 x 50 µm$^2$. The modulated radiation power was measured at room temperature with a photodiode placed at the sample position. At typical measurement conditions, the density of the modulated power amounts to $\Delta P = 210$ pW/µm$^2$.

The temperature of the sample is controlled by adjustment of the contact gas pressure in the insert and the power applied to a resistive heater placed in the vicinity of the sample. The DC bias current from a low-noise current source is applied through the DC path of the bias-tee. The response of the sample to modulated radiation was amplified by a two-stage low noise amplifier (50 Ω impedance, 18 dB gain, 60 K noise temperature at 4.2 K) with low power consumption (8 mW) that was designed for cryogenic applications [16]. Then, the pre-amplified signal is led out of the cryostat by a rigid stainless steel high-frequency cable and a special vacuum feedthrough. The response of the sample is measured by a N9020A MXA signal analyzer from Agilent Technologies. To calibrate the RF readout path, the sample was substituted by a frequency generator to measure the $S_{21}$ parameter of the whole readout line. The rigid cable shows a continuous decay of transmission as the frequency increases, up to $\approx 8$ dB damping at 10 GHz. The amplifier was calibrated at cryogenic temperatures using a network analyzer and is set to a working point with optimal flatness ($< 0.5$ dB) in the considered frequency range. The read-out electronics limit the system bandwidth to a frequency of 10 GHz.
4. Results and discussion

When a photon is absorbed in a superconducting thin film, its energy $h\nu$ will be quickly redistributed within the electron subsystem by inelastic electron-electron scattering with a time constant $\tau_{ee}$. Since this time in disordered superconductors is much shorter than the electron-phonon and phonon-electron interaction times ($\tau_{ep}$ and $\tau_{pe}$), the electron and phonon subsystems are essentially decoupled. In this quasi-equilibrium condition, each subsystem can be assigned a separate temperature $T_e$ and $T_p$, where $T_e > T_p$. Then the absorbed energy is transferred from the electrons to the phonons due to inelastic electron-phonon scattering on the time scale $\tau_{ep}$. Finally, the thin film sample returns to equilibrium with the bath temperature (substrate at temperature $T_S$) by diffusion and escape of the non-equilibrium phonons from the film to the substrate with the characteristic time $\tau_{es}$. It has been shown that this process of energy relaxation in thin film samples can be well described by the two-temperature (2T) model [4], [17]. If Joule heating produced by an applied bias current can be neglected and only small deviations of the electron and phonon subsystems from the equilibrium are considered, the differential equations of the model can be linearized

\[ C_e \frac{dT_e}{dt} = \frac{\alpha P_{in}(t)}{V} - \frac{C_e}{\tau_{ep}} (T_e - T_p) \]
\[ C_p \frac{dT_p}{dt} = \frac{C_p}{\tau_{pe}} (T_e - T_p) - \frac{C_p}{\tau_{es}} (T_p - T_S) \]

where $C_e$ and $C_{ph}$ are the electron and phonon specific heats, respectively, $\alpha$ is the radiation absorption coefficient, $V$ is the volume of the device and $P_{in}(t)$ is the incident optical power.

The change of the electron temperature incurred by the absorbed radiation power can be measured by the change of voltage due to a change of the temperature dependent resistance of the sample:

\[ \Delta U = I_{bias} \frac{dR}{dT} \Delta T_e \]  

For a periodic excitation $P_{in}(t) = P_0 \cos \omega t$, the time evolution of the electron temperature change $\Delta T_e$ can be calculated from the 2T model [17]. The resulting equation can be simplified to a single roll-off function:

\[ \Delta U(\Delta f) = \frac{\Delta U(0)}{\sqrt{1 + (\Delta f / f_c)^2}} \]

where $f_c$ is the characteristic frequency determined by the rate of energy relaxation processes and $\Delta U(0)$ is the voltage response of the sample at $f \ll f_c$. 

Figure 2. Dependence of voltage response $\Delta U$ of 22 nm thick NbN thin film sample to 850 nm radiation on bias current $I_{bias}$ at a modulation frequency of 150 MHz. The dependence of $\Delta U$ on $I_{bias}$ for YBCO films looks similar.
The dependence of the voltage response on bias current was measured at a fixed modulation frequency as shown in figure 2. For small currents, the voltage response increases linearly with an increase of the bias current as expected from equation (2). At higher currents the signal reaches the maximum and then decreases due to overheating of the sample and change of the operation point. The operation bias current for further measurements of the sample response to modulated optical power was chosen at the linear part of the $\Delta U(I_{\text{bias}})$ dependence. The typical values of the optimal operation current $I_{\text{bias}}$ were around several hundred micro-amperes for NbN and a few tens of milli-amperes for YBCO films, depending on the sample cross-section. The final operation point of the samples was adjusted by temperature to about 0.3 $R_C$. The change of the electron temperature $\Delta T_e$ caused by the modulated part of the radiation power can be calculated with equation (2), using the $dR/dT$ values given in section 2. Taking into account the gain of the high frequency readout, $\Delta T_e$ values were around 125 $\mu$K for the NbN films and 5 $\mu$K for the YBCO films. This further shows that the deviation from the equilibrium temperature is very small, especially when compared to pulse response measurements where the electrons are typically heated by several Kelvins [18].

The measurements of the response spectra for NbN and YBCO samples (figure 3) were made at constant bias current by sweeping of the modulation frequency $\Delta f$. A clear plateau for the lower frequencies was observed for all samples, followed by a decay of $\Delta U$ at higher frequencies. The parasitic modulation with a periodicity of 500 MHz on top of the signal is caused by the remaining impedance mismatch of the samples to the readout circuit. The photoresponse amplitude of all YBCO samples was below the $\Delta U$ values of the NbN films. There are several reasons for that. The first one is the much smaller temperature derivative of the resistance of YBCO films (see section 2). Another is that the absorption coefficient $\alpha$ in the near-infrared range is higher in NbN films compared to YBCO [1], [19]. In addition, due to the larger film thicknesses the volume of the YBCO samples presented in this report is much larger compared to the samples made from ultra-thin NbN films.

The response spectra were measured at various bias current values ($I_{\text{max}}/I_{\text{min}} = 5$) from the linear part of $\Delta U(I_{\text{bias}})$ dependence (see figure 2) and different level of the excited modulated power ($\Delta P_{\text{max}}/\Delta P_{\text{min}} = 4$). In both cases the change of the $I_{\text{bias}}$ or $\Delta P$ values results in a correspondent change of the amplitude of the response $\Delta U$. At the same time the frequency dependence of $\Delta U$ (position of plateau and roll-off of the sample response) remain unaffected for both NbN and YBCO thin film samples. Thus we can argue that changes of $T_p$ and $T_e$ by applied $I_{\text{bias}}$ and/or $\Delta P$ were small in comparison to the operation temperature $T = T_C$, and did not influence on the temperature dependent characteristic scattering times (see equation 1) in thin superconducting films.

The experimentally measured modulation frequency spectra of the voltage response are fitted by equation (3), where $\Delta U(0)$ and $f_c$ are the fitting parameters. The solid lines in figure 3 show these fits for the response spectra measured on NbN and YBCO samples with different thicknesses. From these fits we extracted the characteristic roll-off frequencies $f_c$, indicated by arrows in figure 3. As it is seen the $f_c$ values increase with decrease of thickness for both NbN and YBCO films. We can directly calculate the energy relaxation time $\tau_e = (2\pi f_c)^{-1}$ from the frequencies $f_c$ obtained by the fits. For NbN, we obtain values between 115 ps and 284 ps for the 4.2 nm and 22 nm thick films, respectively. In case of the YBCO thin film samples, the $\tau_e$ values range between 512 ps and 2 ns for the 23.5 nm and 44 nm films, respectively.

Since in YBCO the heat capacity of the phonons $C_{\text{ph}}$ is much larger than that of the electrons $C_e$ by a factor of 38 [5], the bolometric and electron processes are indeed strongly decoupled. The phonon diffusion can be neglected for these films since the phonon mean free path $l_{\text{ph}}$ is much larger than the film thickness. In this case, we can assume that the relaxation processes are only due to the heat transport performed by phonons crossing the thermal boundary from the film into the substrate, so that $\tau_e = \tau_{\text{ep}}$. However, for NbN where the ratio $C_{\text{ph}}/C_e$ is much smaller = 6.5 [2], the observed energy relaxation time $\tau_e$ is a combination of both escape time and electron-phonon interaction and can be expressed as $\tau_e = \tau_{\text{ep}} + (1 + C_e/C_{\text{ph}})\tau_{\text{es}}$ [20]. A separation and detail analysis of the electron-phonon
and the phonon escape characteristic times in these films will require further improvement of our
technique and especially an extension of the modulation frequency range which is currently limited by
the readout electronics.

5. Conclusion
An experimental frequency domain measurement setup for high resolution studies of energy relaxation
processes in superconducting thin films at quasi-equilibrium conditions was realized and described in
detail. This technique has been applied to study optical response of thin NbN and YBCO films with
different thicknesses deposited on sapphire substrates. Although these two materials are very different
in their physical properties and operated at different temperature ranges, we succeeded to measure the
samples in the same experimental setup. We have found that due to the difference in steepness of the
superconducting transition \( \frac{dR}{dT} \) and the different operating conditions, the response of the YBCO
samples to optical radiation is much lower than that of NbN even for comparable film thicknesses.
Response spectra in the modulation frequency range between 10 MHz and 10 GHz were measured and
the characteristic energy relaxation time \( \tau_\varepsilon \) has been extracted for NbN and YBCO thin films. For both
materials we observed a clear decrease of \( \tau_\varepsilon \) with decreasing film thickness: from 2.0 to 0.5 ns for 44
to 24 nm thick YBCO films and 284 to 115 ps for 22 to 3 nm thick NbN films, respectively.

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