Quasiparticle relaxation in optically excited high-Q superconducting resonators

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The quasiparticle relaxation time in superconducting films has been measured as a function of temperature using the response of the complex conductivity to photon flux. For tantalum and aluminum, chosen for their difference in electron-phonon coupling strength, we find that at high temperatures the relaxation time increases with decreasing temperature, as expected for electron-phonon interaction. At low temperatures we find in both superconducting materials a saturation of the relaxation time, suggesting the presence of a second relaxation channel not due to electron-phonon interaction.

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The equilibrium state of a superconductor at finite temperatures consists of the Cooper pair condensate and thermally excited quasiparticles. The quasiparticle density \( n_{qp} \) decreases exponentially with decreasing temperature. These charge carriers control the high frequency (\( \omega \)) response of the superconductor through the complex conductivity \( \sigma_1 - i\sigma_2 \). At nonzero frequencies the real part \( \sigma_1 \) denotes the conductivity by quasiparticles and the imaginary part \( \sigma_2 \) is due to the superconducting condensate \( \sigma_{cp} \). When the superconductor is driven out of equilibrium it relaxes back to the equilibrium state by the redistribution of quasiparticles over energy and by recombination of quasiparticles to Cooper pairs. The recombination is a binary reaction, quasiparticles with opposite wavevector and spin combine, and the remaining energy is transferred to another excitation. The latter process is usually controlled by the material dependent electron-phonon interaction \( g \). With decreasing temperatures the recombination time increases exponentially reflecting the reduced availability of quasiparticles. Here, we report relaxation time measurements in superconducting films far below the critical temperature \( T_c \). We find strong deviations from exponentially rising behavior, which we attribute to the emergence of an additional relaxation channel in the superconducting films.

We have measured the time dependence of the complex conductivity of superconducting films after applying an optical photon pulse. In addition, the noise spectrum is measured in the presence of a continuous photon flux \( \delta \). The superconducting film is patterned as a planar microwave resonator. The resonator is formed by a meandering coplanar waveguide (CPW), with the central line 3 \( \mu \)m wide and the slits 2 \( \mu \)m wide, and is coupled to a feedline, see Fig. 1a. The complex conductivity results in a kinetic inductance \( L_k \propto 1/d\omega \sigma_2 \), for thin films with thickness \( d \), which is due to the inertia of the Cooper pair condensate. It sets together with the length of the central line the resonance frequency: \( \omega_0 = 2\pi/4\sqrt{(L_g + L_k)/C} \), with \( l \) the length of a quarterwave resonator, \( L_g \) the geometric inductance and \( C \) the capacitance, both per unit length. The variation in kinetic inductance due to photons is connected to the quasiparticle density \( n_{qp} \), by \( \delta L_k/L_k = \frac{1}{2}\delta n_{qp}/n_{cp} \), with \( n_{cp} \) the Cooper pair density.

FIG. 1: (a) A quarter wavelength resonator, capacitively coupled to a feedline, formed by the superconducting film (gray) interrupted by slits (black). (b) The resonator exhibits a dip in the magnitude and circle in the complex plane (inset) of the feedline transmission \( S_{21} \). (c) The feedline transmission is converted into a phase \( \theta \) and amplitude \( A \) using the equilibrium resonance circle as reference (right inset). The response to an optical pulse of length 0.5 \( \mu \)s (at \( t=0 \)) (open circles) exhibits an initial rise due to the response time (3.7 \( \mu \)s) of the resonator and subsequently follows an exponential decay (34 \( \mu \)s) (dashed), reflecting the restoration of equilibrium (Eq. 1). The response is measured with a signal generator, low noise amplifier (LNA) and quadrature mixer (upper inset).
Resonance frequencies used lie between 3-6 GHz. For a quarterwave resonator at 6 GHz, the length of the meandering superconducting CPW-line is 5 mm. The resonator is capacitively coupled by placing a part parallel to the feedline.

The resonators are made from superconducting materials with different electron-phonon interaction strengths, tantalum (strong interaction) and aluminium (weak interaction). The tantalum film, 150 nm thick, is sputtered on a high resistivity silicon substrate. A 6 nm thick niobium seed layer is used to promote the growth of the desired tantalum alpha phase. The critical temperature $T_c$ is 4.43 K, the low temperature resistivity $\rho$ is 8.4 $\mu$Ωcm and the residual resistance ratio (RRR) is 3.0. A 100 nm thick aluminium film is sputtered on silicon ($T_c=1.25$, $\rho=1.3$ $\mu$Ωcm, RRR=3.7). Alternatively, a film of 250 nm thick is sputtered on silicon ($T_c=1.22$, $\rho=1.0$ $\mu$Ωcm, RRR=6.9) and another one of 250 nm is sputtered on sapphire ($T_c=1.20$, $\rho=0.25$ $\mu$Ωcm, RRR=11). The samples are patterned using optical lithography, followed by wet etching for aluminium and reactive ion etching for tantalum. For both materials quality factors in the order of $10^6$ are reached. The sample is cooled in a cryostat with a conducting magnetic shield. Alternatively, the sample space is surrounded by a cryoperm and a superstat with an adiabatic demagnetization refrigerator. The observed energy gap is 8.4 $\mu$eV.

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The phase $\theta$ with respect to the resonance center frequency $\omega_0$ is given by $\theta = \arctan(\text{Im}(S_{21})/(\omega - \text{Re}(S_{21})))$ and is related to the change in resonance frequency $\omega$ by: $\omega - \omega_0 = -\frac{\omega_0}{\delta \omega} \delta \omega$, with $Q$ the resonator loaded quality factor.

A related change in $L_k$ is given by $\delta L_k/L_k = -\frac{\omega_0}{Q} \delta \omega$, with $\alpha$ the ratio of the kinetic to the total inductance. The phase $\theta$ is therefore a direct measure of the change in complex conductivity (given in the dirty limit by):

$$\theta = -2\alpha Q \frac{\delta \sigma_2}{\sigma_2} (f(E), \Delta),$$

with $f(E)$ the electronic distribution function characterizing the non-equilibrium and $\Delta$ the superconductor energy gap.

The amplitude $A$ depends predominantly on $\sigma_1$ and $\sigma_2$ to a smaller degree on $\sigma_2$. The amplitude is determined by the complex transmission $S_{21}$ by: $A = \sqrt{|\text{Re}(S_{21}) - x_c|^2 + \text{Im}(S_{21})^2/(1 - x_c)}$. On resonance $S_{21} = Q_e/(Q_e + Q_u)$ with $Q_u \propto \sigma_2/\sigma_1$ the unloaded resonator quality factor and $Q_e$ the coupling quality factor, leading to

$$A = 1 - 2 \frac{Q}{Q_u} \left[\frac{\delta \sigma_1}{\sigma_1} (f(E), \Delta) - \frac{\delta \sigma_2}{\sigma_2} (f(E), \Delta)\right].$$

By measuring $A$ and $\theta$ in the frequency- and time-domain we obtain direct information on the relaxation through the complex conductivity of the superconducting films.

A typical pulse response is shown in Fig. 1a. The initial rise of the phase $\theta$ is due to the response time of the resonator. The relaxation shows up as an exponential decay. The right inset of Fig. 1c shows the evolution of the response in the transformed polar plane. These data are interpreted as governed by one relaxation time. This is justified by performing measurements of the noise spectrum and applying the analysis by Wilson et al. Since the superconducting condensate and the quasiparticle excitations form a two-level system a Lorentzian spectrum is expected, with the relaxation dominated by resonator noise, rolling off at a frequency proportional to the coupling quality factor.

The measured noise power spectra of the amplitude and phase of a tantalum sample are shown in Fig. 2. In equilibrium the amplitude noise spectrum (dashed blue line) is flat over the full range, and the phase noise (solid blue line) follows $1/f^\alpha$ with $\alpha \approx 0.25$. The amplitude noise is due to the amplifier, remaining unchanged at frequencies far away from $\omega_0$ while the phase noise is dominated by resonator noise, rolling off at a frequency corresponding to the resonator response time ($0.5 \mu$s). Under a continuous photon flux we observe excess noise
The measured relaxation times for temperatures down to 50 mK are displayed in Fig. 3. The data shown are representative for the relaxation times found in all samples of different films. In the high temperature regime \((T/T_c \gtrsim 0.175)\) the relaxation times increase for decreasing bath temperature in a similar manner for both tantalum and aluminium samples until a new regime is entered around \(T/T_c \sim 0.15\). The tantalum samples clearly show a non-monotonic temperature dependence, exhibiting a maximum near \(T/T_c \sim 0.15\). Two aluminium films show a less pronounced non-monotonic temperature dependence. We do not see a non-monotonic temperature dependence in samples of aluminium with the lowest level of disorder (highest RRR). Below \(T/T_c \sim 0.1\) the relaxation times become temperature independent at a plateau value of 25-35 \(\mu s\) for Ta, 390 \(\mu s\) for 100 nm thick Al on Si, 600 \(\mu s\) for 250 nm thick Al on Si and 860 \(\mu s\) for 250 nm thick Al on sapphire.

The relaxation times for aluminium are measured in half wavelength resonators where the central line is isolated from the ground plane. For the directly connected quarter wavelength resonators a length dependence was found. For tantalum the values are found to be length independent in both cases. Consequently, the data shown are not influenced by quasiparticle outdiffusion. Also, the relaxation times remain unchanged when instead of an optical pulse a microwave pulse at frequency \(\omega_0\) is used. In this method only quasiparticle excitations near the gap energy are created by the pair-breaking current.

This observation leads us to believe that the observed decay is due to recombination of quasiparticles with energies near the gap.

The exponential temperature dependence for \(T/T_c \gtrsim 0.175\) is consistent with the theory of recombination by electron-phonon interaction [4]. The dotted lines in Fig. 2 follow the expression for the recombination time,

\[
\frac{1}{\tau_{rec}} = \frac{1}{\tau_0} \sqrt{\pi \left( \frac{2\Delta}{kT_c} \right)^{5/2} \sqrt{T/T_c} e^{-\frac{\Delta}{kT}}},
\]

with \(\tau_0\) a material-specific electron-phonon scattering time. We find for 150 nm Ta on Si \(\tau_0 = 42 \pm 2\) ns.
and for 250 nm Al on Si $\tau_0 = 687 \pm 6$ ns. The deviation from the exponential rise and the low temperature behavior is incompatible with the established theory for electron-phonon relaxation. We assume that an additional relaxation channel \cite{11} is dominant at low temperatures, where the electron-phonon mechanism becomes too slow.

In previous experiments using superconducting tunnel junctions a similar saturation in the quasiparticle loss has been reported. For photon detectors inverse loss rates in the order of tens of microseconds have been found for tantalum \cite{12, 13, 14, 15} and hundreds of microseconds for aluminium \cite{8}. Some of these experiments also indicated a non-monotonic temperature dependence \cite{10}. Most of these observations have been attributed to trapping states at surfaces or in dielectrics. The fact that our similar experimental results occur in simple superconducting films and two different materials suggests that processes in the superconducting film itself lead to the observed low temperature behavior.

The observed saturation in the relaxation times in our samples is reminiscent of experiments in normal metals on inelastic scattering in non-thermal distributions and on dephasing in weak localization studies. The apparent saturation of the dephasing time and the strong quasiparticle energy exchange at low temperatures have been shown to be caused by dilute concentrations of magnetic impurities \cite{17, 18, 19, 20}. It is known that in superconductors a large density of magnetic impurities decreases the critical temperature. For dilute magnetic impurities the local properties are most important. In experiments with magnetic adatoms impurity bound excitations arise \cite{21}, tails in the density of states within the gap might form and the formation of an intragap band with growing impurity concentration are predicted \cite{22, 23}. In ongoing experiments we observe a gradual decrease of the relaxation time with an increasing ion-implanted magnetic impurity concentration (0-100 ppm). However, disorder plays a role as well and further experiments are needed to clarify possible relaxation processes \cite{24}.

In conclusion, we find that the quasiparticle relaxation times, probed by means of the complex conductivity, saturate for both tantalum and aluminium, below a tenth of the critical temperature. We suggest that the saturation of the relaxation time is due to the presence of a relaxation channel, which is not caused by the conventional process dominated by electron-phonon interaction.

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