Finite quasiparticle lifetime in the surface layer of disordered superconductors

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We investigate complex conductivity of a highly disordered MoC superconducting film with \( k_F l \approx 1 \), where \( k_F \) is the Fermi wavenumber and \( l \) is being the mean free path, derived from experimental transmission characteristics of coplanar waveguide resonators in a wide temperature range below the superconducting transition temperature, \( T_c \). We find that the modified Mattis-Bardeen model with a finite quasiparticle lifetime, \( \tau \), offers a perfect description of the experimentally observed complex conductivity. We show that \( \tau \) is appreciably reduced by surface scattering effects. Characteristics of the surface scattering centers are independently found by the scanning tunneling spectroscopy and agree with those determined from the complex conductivity.

Disordered superconductors is a subject of an intense current attention. The interest is motivated not only by an appeal of dealing with the most fundamental issues of condensed matter physics involving interplay of quantum correlations, disorder, quantum and thermal fluctuations, and Coulomb interactions\(^1\)–\(^4\), but also by the high promise for applications. Existence of the states with giant capacitance and inductance in the critical vicinity of superconductor-insulator transition\(^2\)–\(^5\) breaks ground for novel microwave engineering exploring duality between phase slips at point-like centers\(^6\) and at phase slip lines\(^7\) and Cooper pair tunneling\(^8\)–\(^10\). Indeed, a possibility to build superconducting flux qubit employing quantum phase slips in a weak link realized by highly disordered superconducting wire was demonstrated by Astafiev et al.\(^5\). Yet, while there has been notable recent success in describing dc properties of disordered superconductors,\(^2\)–\(^9\),\(^11\) the understanding of their ac response remains insufficient and impedes advance in their microwave applications. In this Letter we discuss a model that enables practical calculations of complex conductivity of disordered superconductors and experimentally demonstrate its validity.

Having extended BCS theory\(^12\) Mattis and Bardeen derived complex frequency dependent conductivity\(^13\). They formally introduced an infinitesimal scattering parameter, \( s = 2\pi/\tau \), which was set to zero at the end of calculations. It has been shown\(^14\) that in disordered superconductors the finite \( s \) acquires physical meaning as a diffusion enhancement of the Coulomb contribution to one-particle scattering rate. Maintaining a finite value of \( s \) as a phenomenological inverse inelastic quasiparticle lifetime one can derive the modified formulas for the ratio of the superconducting complex conductivity \( \sigma_s/\sigma_n \) in the interval \( \omega < 2\Delta \) as

\[
\frac{\sigma_s}{\sigma_n} = 1 - \frac{i}{\hbar \omega} \int_{\Delta}^{\infty} \left[1 - 2f(E)\right] \left|g_+(E, \omega) - g_-(E, \omega)\right| dE
\]  

(1)

where \( f(E) \) is Fermi-Dirac distribution function and propagators \( g \) are defined as

\[
g_{\pm}(E, \omega) = \frac{E}{\sqrt{(E^2 - \Delta^2)}} \pm \frac{E \pm \hbar(\omega - is)}{\sqrt{(E^2 - \Delta^2)}} - \frac{\Delta}{\sqrt{(E^2 - \Delta^2)} - \sqrt{(E^2 - \Delta^2)}}
\]  

(2)

Here \( E = (\epsilon^2 + \Delta^2)^{1/2} \) is the excitation energy in superconductor with energy gap \( \Delta \). Inspecting Eq. 2 one sees that the first term is a product of the standard BCS quasiparticle density of states and the similar but smeared one, which can be viewed as resulting from
Coulomb and/or phonon interaction. A similar broadening was obtained by Nam\textsuperscript{15} for superconductors containing magnetic impurities.

To verify the applicability of the modified Mattis-Bardeen model we carried out the measurements of the complex conductivity of disordered 10 nm thin MoC films with the sheet resistance \( R_\square \approx 180 \, \Omega \).\textsuperscript{16} The films were fabricated by magnetron sputtering. We used a reactive sputtering process, where particles of molybdenum were sputtered from the Mo target in an argon-acetylene mixture. The MoC thin film was deposited on the sapphire \( c \)-cut substrate. The thickness of the film was controlled by tuning the sputtering time according to the respective sputtering rate calibrated to 10 nm/min. The details of the preparation are given in Ref.\textsuperscript{17}.

To reconstruct the complex conductivity \( \sigma = \sigma_1 - i \sigma_2 \) we designed the coplanar waveguide (CPW) resonators\textsuperscript{18} made of the 10 nm thin films (see Fig. 1). The chosen thickness of 10 nm is optimal for further patterning of superconducting nanostructures which are expected to exhibit quantum phase slips.\textsuperscript{5} The coplanar waveguide resonators were then patterned by the optical lithography and etched by the argon ion etching.

Transmission measurements of the CPW resonators yielded temperature dependencies of the resonant angular frequency \( \omega_0 \) and the quality factor \( Q \), both depending on the complex conductivity. The imaginary part of the impedance is mostly represented by the inductance of the CPW and, therefore, can be used to calculate the CPW resonant frequency. The capacitance of the CPW is explicitly defined by its geometry.\textsuperscript{18} The real part of the impedance is determined by the resistive losses in the CPW and therefore influences the internal quality factor.\textsuperscript{18} Taking into account the external quality factor \( Q_{ext} \) due to input/output coupling capacitances, one can recalculate the required internal quality factor, using the data on the loaded quality factor. To carry out the comparison of the obtained data with the theory, we calculated the complex impedance of the CPW with known geometry (see Fig. 1) using complex conductivity given by Eq. 1. To verify numerical calculations that were used for designing the CPW resonant frequency as well as its external quality factor, we fabricated the test resonator out of the “thick” (~200 nm) MoC film and took the corresponding measurements. The parameters were predicted to be \( \omega_0 = 2 \pi \times 2.5 \, \text{GHz} \) and \( Q_{ext} = 40000 \). The results of measurements completely agreed with the prediction, confirming the validity of our procedure.

We measured several MoC samples with different sheet resistances. The parameters of the samples are presented in Fig. 2. The most striking feature of our data is that while the Mattis-Bardeen model predicts that with an increase of the sheet resistance the quality factor would increase as well, the experiment reveals an opposite trend: decrease of the measured quality factors with the growth of the sheet resistance. Furthermore, the measured quality factors noticeably differ from those predicted by the Mattis-Bardeen model in the wide temperature range, see Fig. 3a. At the same time, the measured resonant frequency falls below the theoretical one (Fig. 3b) only slightly, but systematically. This deviation was studied in Ref.\textsuperscript{19} in the narrow temperature range.

![FIG. 1: Scheme of coplanar waveguide resonator with dimensions: W=50 \( \mu \text{m} \), S=30 \( \mu \text{m} \), C=10 \( \mu \text{m} \), t=10 nm, h=430 \( \mu \text{m} \).](image)

![FIG. 2: The thickness dependence of sheet resistances (squares) and internal quality factors (triangles) of MoC coplanar resonators. Internal quality factors calculated from the Mattis-Bardeen model for the corresponding \( R_\square \) and \( t \) (circles) exhibit the opposite trend. The solid lines are eye-guides.](image)
FIG. 3: The temperature dependence of the quality factor $Q$ (a) and the resonant frequency $f_0$ (b) of 10 nm thin CPW resonator. Circles are measured data and lines are data calculated from original Mattis-Bardeen relations ($s \to 0$) for parameters $T_c=5.8$ K, $R_\Box = 180$ $\Omega$, $\Delta_0 = 1.9kT_c$.

It is worth noticing that incorporation of mesoscopic fluctuations that leads to broadened superconducting density of states$^{20}$ does not improve significantly the agreement between the theory and the experiment.

One sees from Fig. 3 that the loss of a CPW resonator at low temperatures is much higher than it what is predicted by the Mattis-Bardeen model. One finds then that the theoretical and experimental results can be conveniently compared via using the ratio $\sigma_2/\sigma_1$, which can be expressed as a function of the resonant frequency and quality factor of the CPW resonator

$$\frac{\sigma_2}{\sigma_1} = Q \left( 1 - \left( \frac{\omega_0}{\omega_g} \right)^2 \right).$$

Here $\omega_0$ is a resonant frequency of the resonator in the normal state of the lossless metal. In Fig. 4 we compare the experimental data together with the $\sigma_2/\sigma_1$ temperature dependence calculated for different values of the parameter $s$.

The fits to the experimental results corresponding to different values of $s$ are presented in Fig. 4.

FIG. 4: Measured ratio $\sigma_2/\sigma_1$ (open circles) for 10 nm thin MoC CPW resonator compared with the ratio calculated from modified Mattis-Bardeen relations for various values of the parameter $s$. At low temperatures (below 1 K) the lines from top to bottom correspond to $\hbar s = \Delta_0/100, \Delta_0/10, \Delta_0/5, \Delta_0/2$.

At high temperatures $T > T_c/2$ the small values of the scattering parameter $s$ provide a fair agreement between the theoretical prediction and the experimental data, i.e in this temperature range the original Mattis-Bardeen model applies well. However, at very low temperatures the larger value of $s$ is required to fit the measured data, and it is not possible to find any intermediate value of $s$ to obtain a reasonable agreement with the experiment for the complete temperature range.

We propose that at low temperatures the main contribution to the conductivity comes from the surface scattering while the role of the bulk scattering diminishes. The similar idea has been put forward to explain the low temperature losses in the high quality CPW resonators made of conventional superconductors.$^{21}$ Following this guiding concept we express the total conductivity as a sum of the bulk and surface conductivities $\sigma = (1 - \kappa)\sigma_b + \kappa\sigma_s$. We take the surface scattering parameter $s_s$ as fitting parameter while the bulk one $s_b$ was taken to be zero.

The obtained agreement between the experimental results and the theoretical model is excellent, see Fig. 5, which fully justifies the concept of the finite quasiparticle lifetime in disordered superconductors.
Figure 5: The temperature dependence of the ratio $\sigma_2/\sigma_1$ of 10 nm thick CPW resonator. Circles are measured data fitted by modified Mattis-Bardeen relations (solid line) taking into account the surface layer of superconductor for parameters $\kappa = 0.3$, $\Delta_0 = 1.76kT_c$, $T_c = 6.4$ K, $s_s = 0.23\Delta_0$, $s_b = 0$.

To test further the model we employed the tunneling spectroscopy allowing for a direct determination of the superconducting DOS at the surface of the sample by using a subkelvin scanning tunneling microscope. The previous studies on disordered superconductors have shown that microwave measurements and tunneling spectroscopy provide consistent results on the superconducting density of states.\textsuperscript{22} Scanning tunneling spectroscopy measurements were done using a homemade low-temperature scanning tunneling microscope head. The Fig. 6 shows the normalized tunneling conductance spectra between the Au tip and the MoC sample measured at different temperatures ranging from 0.43 to 5.6 K. Each curve was normalized to the spectrum measured at 5.6 K with the sample in the normal state. Since the Au tip features a constant density of states, each of these differential conductance versus voltage spectra reflects the superconducting density of states (SDOS) of MoC, smeared by $2k_BT$ in energy at the respective temperature. Consequently, in the low temperature limit ($k_BT \ll \Delta$), the differential conductance measures the SDOS directly. As evidenced by the 0.43 K curve the measured SDOS differs from the BCS SDOS: it reveals a significant quasiparticle density of states at the Fermi level and broadened coherence peaks at the gap edges. This could be be accounted by the empiric Dynes formula:\textsuperscript{23,24}

$$N(E) = \text{Re} \left\{ \frac{E + i\Gamma_{in}}{\sqrt{(E + i\Gamma_{in})^2 - \Delta^2}} \right\}, \quad (4)$$

with the inelastic scattering parameter $\Gamma_{in} = hs$. The best agreement is achieved at $\Gamma_{in} = 0.12\Delta_0$, $\Delta_0 = 1.83kT_c$, $T_c = 5.85$ K. These values differ slightly from the results obtained from the fitting of the complex conductivity ($\Delta_0/kT_c = 1.76$ and $hs = 0.25\Delta_0$). This discrepancy can be explained by the fact that the tunneling spectroscopy is a local measurement, whereas microwave measurements give an ‘integral value’ of parameters averaged over a large area of the coplanar waveguide resonator with the length $\approx 2$ centimeters. Hence in disordered superconductors a reasonable spread of parameters is expected. Moreover, the metallic tip of the scanning tunneling microscope can screen Coulomb interactions which can, in turn, influence measured spectra.\textsuperscript{25} Our model can also be used for thin films with lateral inhomogeneity with ‘alternating’ regions of very low and very high disorder\textsuperscript{26} or for disordered superconducting thin films with strong spatial inhomogeneities of the superconducting gap in the density of states.\textsuperscript{3}

In conclusion, we demonstrated that the complex conductivity of the disordered superconductor is well described by the modified Mattis-Bardeen model in the wide temperature range between 300 mK and just below $T_c$. The validity of our approach is supported by the data from the tunnel spectroscopy.

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