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Anomalous response of superconducting titanium nitride resonators to terahertz radiation

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We present an experimental study of kinetic inductance detectors (KIDs) fabricated of atomic layer deposited TiN films and characterized at radiation frequencies of 350 GHz. The responsivity to radiation is measured and found to increase with the increase in radiation powers, opposite to what is expected from theory and observed for hybrid niobium titanium nitride/aluminium (NbTiN/Al) and all-aluminium (all-Al) KIDs. The noise is found to be independent of the level of the radiation power. The noise equivalent power improves with higher radiation powers, also opposite to what is observed and well understood for hybrid NbTiN/Al and all-Al KIDs. We suggest that an inhomogeneous state of these disordered superconductors should be used to explain these observations.

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Superconducting resonators have been proposed as kinetic inductance detectors (KIDs) for sensitive multipixel radiation detection.1 Antenna-coupled hybrid niobium titanium nitride/aluminium (NbTiN/Al) KIDs2,3 and all-aluminium (all-Al)4 have shown generation-recombination noise and photon noise limited performance. KIDs can also be constructed as lumped element kinetic inductance detectors (LEKIDs)5 in which the KID is arranged as a photon absorbing area matched to free space. Aluminium LEKIDs have also shown generation-recombination and photon noise limited performance.6 However, the low normal-state resistivity of Al makes the design of the absorber for very high frequency radiation complex. Therefore, superconductors with a high normal state resistivity have recently become of particular interest.9

A figure of merit M to optimize the responsivity of KIDs is defined as $M = \frac{2\pi}{\tau} Q N(0) V F_{res}$ with $\tau$ the kinetic inductance fraction, $\tau$ the quasiparticle recombination time, $Q$, the internal quality factor, $F_{res}$ the resonance frequency, $N(0)$ is the single spin electron density of states at the Fermi level, and $V$ the volume of the KID. For example, Al KIDs have a long quasiparticle recombination time (a few milliseconds) and high internal quality factors (above one million), but their kinetic inductance fraction is low and their volume is large.

Superconducting materials with a high resistivity in the normal-state are promising because of their high quality factor and a long enough relaxation time. The high normal resistance implies a large sheet inductance, resulting in a large kinetic inductance fraction, which lowers the KID volume. The high surface impedance also eases matching to free space and optimises the photon absorption. Given the high quality NbTiN resonators pioneered by Barends et al.7,8 titanium nitride (TiN) has been proposed, because it has the previously mentioned properties in addition to a tuneable critical temperature, which facilitates a relatively long quasiparticle lifetime.9 Currently, several groups are studying the implementation of TiN KID devices and instruments.10–14

However, a material like TiN has also drawn the attention of the condensed matter physics community, interested in the disorder-induced superconductor-to-insulator transition.15 It has been shown that the superconducting state becomes gradually increasingly inhomogeneous for increasing disorder, a feature absent in the traditional description of the absorption of electromagnetic radiation by a superconductor. Consequently, a careful empirical study of the direct absorption of radiation by a strongly disordered superconductor such as TiN is urgently needed.

In this letter, we concentrate our experiments on atomic layer deposited (ALD) TiN exposed to 350 GHz radiation. A number of unanticipated trends were found in these experiments: (i) the responsivity to the 350 GHz radiation increases for increasing radiation power, opposite to what we observe in hybrid NbTiN/Al and all-Al KIDs, and what is expected for a conventional, uniform, superconducting state; (ii) the optical noise equivalent power (NEPopt) for 350 GHz radiation is much larger than the photon noise limited NEP at low radiation powers, whereas it becomes comparable at higher radiation powers; and (iii) the electrical NEP is much lower than the optical NEP, in contrast to what we find for the hybrid NbTiN/Al KIDs.

Several ALD TiN films have been deposited on high resistivity (>10 kΩcm) Si (100) substrates covered with a thin surface layer of native silicon oxide (for details see Coumou et al.17). The microwave properties of these TiN films have been analysed in detail by Driessen et al.18 and by Coumou et al.19 In the latter case, it was compared to a study of the local values of the superconducting density of states. The microwave response deviates from standard Mattis–Bardeen

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A modified Usadel equation was used, including a disorder-dependent pair breaking parameter, which smears the BCS density of states analogous to what is seen for magnetic impurities and a pair-breaking parameter. Physically, it refers to a ground state in which the Cooper-pairs have a finite momentum. The pair-breaking parameter modifies the quasiparticle DoS, smoothing the coherence peaks, which enter into the generalized Mattis-Bardeen expressions.

The local DoS of these films is independently determined using scanning tunnelling spectroscopy (STS). A clear correspondence between the STS measurements and the model is found for the least-disordered films. In contrast, the measured tunnel curves are no longer uniquely characterizing a film for the most-disordered superconducting films, but are found to vary laterally. For the present experiments, we use a 45 nm thick film because it has a relatively low disorder and, therefore, is expected to allow the application of a uniform model with a broadened DoS model (although it did not have the best properties as a detector). We expect this film to have similar properties as the film presented in Ref. 19. However, it is to be expected that effects of the tendency to non-uniformity of the superconductor with increasing disorder will depend on the parameters and the techniques used in the experiments.

The 45 nm ALD TiN film has a superconducting transition temperature $T_c$ of 3.2 K, a sheet inductance $L_s$ of 16.5 pH and a sheet resistance $R_s$ of 41.5 Ω. Two KIDs, capacitively coupled to a feedline of ~50 Ω, were fabricated in a single layer process. The KIDs were designed to absorb 350 GHz radiation and consist of three inductors and an interdigitated capacitor, all in parallel, forming a tank circuit with a resonance frequency $f_{res}$ of ~3.4 GHz. The width of the lines of the inductors is 24 μm, and the separation between adjacent lines is 60 μm, with a total area of the inductors of 1200 × 986 μm. The quality factor Q of the KIDs is $7 \times 10^4$, set by a coupling Q of $7 \times 10^4$, with an internal Q of $1.8 \times 10^6$.

The experiments are performed in a pulse tube precooled adiabatic demagnetisation refrigerator (ADR) with a temperature of 100 mK, well below the superconducting transition temperature of the film. The sample is mounted in a sample holder inside a light-tight box, in a box-in-a-box configuration. The 350 GHz radiation illuminating the KIDs is coming from a cryogenic black body placed inside the cooler at the 3 K stage, which temperature can be varied from 3 up to 35 K. Two stacks of metal mesh IR filters placed at the black body source and at the light-tight box determine the band pass of the radiation coupled to the KIDs, which is 50 GHz centered around 350 GHz. The same experimental setup is described in more detail by Janssen et al. and by de Visser et al. A schematic picture of the experimental setup and device is shown in Fig. 1.

The radiation power absorbed by the KIDs $P_{opt}$ is calculated by integrating the black body spectral radiance over the solid angle illuminating the KIDs, taking into account the filter transmission. Following the conventional approach, we use the number of excess quasiparticles $N_{qp}$ in a KID as the leading quantity. It is parametrized as $N_{qp} = \eta_{qp}P_{opt}/\Delta$, with $\eta_{qp}$ the so-called quasiparticle creation efficiency, i.e., the number of excess quasiparticles per incoming photon, and $\Delta$ the superconducting energy gap. The absorption of the superconductor is assumed to be given by the Mattis-Bardeen theory and the electron-phonon controlled recombination time as given in Kaplan et al. For a photon noise limited KID, the quasiparticle lifetime at a given radiation power is expected to be inversely proportional to the square root of the absorbed power $\tau \propto P_{opt}^{-1/2}$. Therefore, the 350 GHz radiation responsivity $\delta x/\delta P_{opt}$ (where $x$ is either amplitude response or phase response) is expected to be proportional to the square root of the absorbed power $\delta x/\delta P_{opt} \propto P_{opt}^{-1/2}$. These dependences were recently verified both for hybrid NbTiN/Al as well as and for the all-Al KIDs. However, given the known large deviations reported in highly disordered superconductors from Mattis-Bardeen theory, it is not clear how large the uncertainty of this assumption is. A spurious contribution in the phase response (shown in the inset in Fig. 2) is found. At temperatures below 250 mK, the response in the phase has an initial strong negative response followed by a gradually rising positive response, which is tentatively attributed to the temperature dependence of the

![Fig. 1. Schematic picture of the experimental setup.](Image)

![Fig. 2. Amplitude responsivity measured with a VNA as a function of radiation power at different bath temperatures.](Image)
dielectric material. Therefore, we only present and discuss the amplitude data. In the conventional response, the responsivity of a KID decreases with the increase of radiation power due to the reduction in life time of quasiparticles at higher quasiparticle densities. This has been clearly shown for both hybrid NbTiN/Al\(^3\) and all-Al\(^4\) KIDs. In contrast, the TiN KIDs reported in this paper become more responsive at higher radiation powers, as shown in Fig. 2.

In Fig. 3, we show the optical NEP for 350 GHz radiation as a function of radiation power from the black-body source. The optical NEP is defined as the power spectral density divided by the responsivity measured at a modulation frequency of 100 Hz (blue) compared to the electrical NEP calculated using the disordered-dependent pair breaking parameter model (black dot). Solid symbols were measured with a VNA, whereas open symbols were measured with an IQ mixer. The green line is the expected photon noise limited NEP.

\[\text{NEP}(\omega) = S_A^{1/2}(\omega) \left( \frac{\hbar \nu}{\Delta} \right)^{-1} \frac{(1 + \omega^2 \tau_{10}^2)}{\left(1 + \omega^2 \tau_{20}^2\right)}, \tag{1}\]

with \(\omega\) the resonator-frequency and \(\tau_{10}\) the resonator ring time. The noise and quasiparticle lifetime can be measured directly, but not the electrical responsivity \(\delta A/\delta N_{qp}\). Instead, the temperature dependence of the resonator \(\delta A/\delta T\) has been measured (Fig. 4) with \(T\) the temperature of the KID. The electrical response has been compared to Mattis-Bardeen theory.\(^{21}\) The electrical response was converted into responsivity calculating the number of thermally excited quasiparticles present at the given temperature. The number of quasiparticles was calculated using the relation shown in Eq. (2) with a standard BCS DoS

\[n_{qp} = 4N(0) \int_0^{\infty} N(E)f(E)dE \approx 2N(0)\sqrt{2\pi k_B T \Delta(0)e^{-\Delta(0)/k_B T}}, \tag{2}\]

where \(N(E)\) is the normalised quasiparticle density of states, \(f(E)\) the quasiparticle energy distribution, \(k_B\) the Boltzmann constant, and \(\Delta(0)\) the superconducting energy gap at zero temperature. At this point, we emphasize that for our material, the correct conceptual framework and theoretical model is unclear. It is not possible to describe the temperature dependence of the resonance-frequency with standard formulas, taking the measured \(T\) and a fixed relationship between \(k_B T\) and \(\Delta(0)\) of 1.76 valid for weak-coupling superconductors. This leads to the blue dotted curve in Fig. 4, and clearly signals a deviation. One can also take the energy-gap as a free adjustable parameter. This leads to an excellent fit and consequently to a DoS represented by the black dashed curve in the inset. In the more realistic model including the disorder\(^{18}\) which leads to the red solid line, we obtain an excellent fit as well, but with a broadened DoS, as also observed with local tunnelling spectroscopy, and shown in the inset.

![FIG. 3. Optical NEP for 350 GHz radiation in the amplitude signal defined as the power spectral density divided by the responsivity measured at a modulation frequency of 100 Hz (blue) compared to the electrical NEP calculated using the disordered-dependent pair breaking parameter model (black dot). Solid symbols were measured with a VNA, whereas open symbols were measured with an IQ mixer. The green line is the expected photon noise limited NEP.](image-url)
However, as we will argue below, such an approach is potentially misleading because it suggests that one is dealing with a uniform superconducting state.

We determine the quasiparticle lifetime by measuring the change of the amplitude signal, while equilibrium is being restored, after applying a short high power microwave pulse. The pulse decay time which we interpret as the quasiparticle lifetime is found to be \( \approx 40 \mu s \) at 250 mK (same temperature at which the optical NEP was measured). We find that it does not follow the exponential pattern expected from Kaplan et al., \(^{22}\) as reported by Coumou et al.\(^{17}\) The noise is determined with the black body source present at 3.5 K. We find that the electrical NEP equals to \( 7 \times 10^{-17} \) W/Hz\(^{1/2} \), which is in agreement with the electrical NEP reported by Leduc et al., \(^{9}\) taking into account the different superconducting transition temperatures and volumes. The difference between the electrical NEP and the measured optical NEP at the lowest radiation power is a factor of 100. Although room temperature transmission measurements with a vector network analyser and cryogenic FTS measurements show that TiN is a good absorber with absorption efficiencies of 75% and 90% respectively, we cannot understand its behaviour using standard BCS theory.\(^{16}\) No better understanding is achieved when a more realistic model including the disorder\(^{18}\) is used for the analysis.

We have calculated the electrical NEP using our current understanding of strongly disordered superconductors. However, in our analysis, we continue to assume an homogeneous superconducting state, whereas it has been clearly demonstrated that at a high enough degree of disorder, the superconducting energy gap \( \Delta \) varies spatially.\(^{15,19}\) In such an inhomogeneous system, excited quasiparticles might get trapped in low-gap puddles. In addition, the film may start to act as a random array of superconducting islands coupled by weak-link type Josephson junctions,\(^{27}\) which will have its own typical optical response.

Figure 4(b) shows a cartoon of such a spatially varying energy gap \( \Delta(\vec{r}) \). In this figure, \( \mu_p \) denotes the chemical potential, and the grey shade indicates the thermal distribution of quasiparticles. We estimate the root mean square variation of the gap \( \Delta_1 = \sqrt{\langle (\Delta(\vec{r}) - \langle \Delta(\vec{r}) \rangle)^2 \rangle} \) from the broadening parameter used to describe the electrodynamic response of the resonator: \( \Delta_1 \approx 44 \mu eV \). At a temperature of 100 mK, most quasiparticles will be trapped in the low-gap areas, since the thermal energy \( k_B T = 9 \mu eV \ll \Delta_1 \). At higher temperatures, the thermal energy and the variations of the energy gap become comparable and any (photon-induced) excess quasiparticle will be free to move. A similar effect will occur with increasing radiation power: At first, excess quasiparticles will be trapped in the low-gap regions, but at increasing power, the excess quasiparticles will be free to move, and will not be sensitive to the underlying gap inhomogeneity anymore. The superconductor will increasingly behave as an electronically homogeneous material. Note that in this qualitative model, there is still a coherent superconducting condensate present, which will determine the phase response of the KID, whereas the dissipative (amplitude) response will involve a combination of mobile and trapped quasiparticles. This model might therefore also explain the difference in phase and amplitude relaxation time that was observed by Gao et al. in TiN KIDs.\(^{12}\)

In summary, we have studied ALD TiN KIDs at a radiation frequency of 350 GHz using the same setup as used for hybrid NbTiN/Al\(^3\) and all-Al KIDs.\(^4\) A radiation power dependent responsivity was found, in contrast to the well understood hybrid NbTiN/Al and all-Al KIDs. Since the noise is independent of radiation power, the optical NEP is also a strong function of the radiation power. At high radiation powers, comparable to the incident radiation in ground-based telescopes, the optical NEP approaches the requirements for ground-based instruments (recently achieved by increasing the received power per unit volume of the KIDs\(^{24}\)). Despite the fact that the TiN is a good absorber at high radiation powers, the difference between the electrical NEP and the measured optical NEP at the lowest radiation power is a factor of 100.

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