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Features of the nonlinear microwave response of ultrathin YBaCuO films

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Received April 18, 2024
Revised April 18, 2024
Accepted May 8, 2024

Using the method of nonlinear near-field microwave microscopy, the temperature dependences of the third harmonic power were studied for different orientations of the $a-b$ axes of ultrathin YBaCuO films with a thickness of 4−5 nm. The experimental results obtained indicate that the nonlinear response at low temperatures may be associated with the presence of Andreev bound states.

Keywords: high-temperature superconductors, near-field microwave microscopy, nonlinear Meissner effect, Andreev bound states, ultrathin films.

DOI: 10.61011/PSS.2024.06.58684.19HH

1. Introduction

The nonlinear Meissner effect (NLME) is one of the fundamental properties of a superconductor. This mechanism of nonlinearity is attributable to the destruction of Cooper pairs and a reduction of the concentration of superconducting electrons under the action of a current flowing through a superconductor. This results in the dependence of the surface impedance of the superconductor on the current or magnetic field or the generation of higher harmonics of the fundamental frequency [1,2].

NLME is known to be sensitive to the internal properties of a superconductor, such as, for example, the symmetry of the order parameter. In particular, high-temperature superconductors with d symmetry of the gap order parameter can have a strong NLME when the temperature T tends to zero due to low-lying excitations near the nodes of the superconducting gap [3]. Moreover, the state with d pairing also results in a dependence of the nonlinear response on the angle between the direction of current flow and the nodes of the superconducting gap [4]. At first, the anisotropy of the plane $a-b$ for the dependence of the London penetration depth on the magnetic field was predicted at $T = 0$. Later, this theory was generalized to the entire temperature range within the framework of the nonlinear microwave response of an anisotropic superconductor and a practical method for studying NLME was proposed [5–7]. It was also found that the anisotropy of NLME in cuprate superconductors becomes significant only at temperatures of $T/T_c < 0.6$. At the same time, at low temperatures, the nonlinearity coefficient in a clean superconductor increases according to the law of $1/T$ [7]. The saturation of the temperature dependence is observed at lower temperatures, which is associated with non-local effects attributable to impurities in superconductors [6].

Many experimental attempts have been made to observe NLME in cuprate superconductors. The first indirect confirmation of the existence of nodes of the superconducting gap of single crystals of $Bi_2Sr_2CaCu_2O_{8-x}$ (Bi-2212) was obtained in the study of Maeda et al. that demonstrated the linear behavior of $\lambda(H, T)$ depending on the constant magnetic field H [8]. Later, the first experimental evidence of the existence of NLME in high-temperature superconductors YBCO [9,10] was obtained using a sensitive method based on measurements of intermodulation microwave distortions. It was also found that the observation of NLME strongly depends on the sample and the sensitivity of the measurement methods.

An additional contribution to the NLME may be attributable to the Andreev bound states (ABS) [11], which occur, for example, as a result of the presence of a (110)-oriented surface in high-temperature superconductors. Such interfaces at twin boundaries can be formed spontaneously during the growth of the epitaxial film. Changing the sign of the order parameter results in a strong Andreev reflection of quasiparticles on the surface of the superconductor. A bound state is formed as a result of the interference of electronic and hole excitations, which results in a paramagnetic surface current in a superconductor.

The ABS was studied experimentally using transport [12] and magnetic measurements [13,14]. In particular, it was shown that the appearance of a peak in differential conductivity at zero displacement is attributable to the formation of ABS at the boundaries of high-temperature superconductors. Furthermore, it was found that the temperature dependence of the effective penetration depth demonstrates a local minimum. It is related to the fact that, on the one hand, the depth of penetration due to volumetric currents decreases monotonously with a decrease of the temperature of the HTSC film. On the other hand, the surface current associated with quasiparticles results in an increase of the effective penetration depth. This minimum can be observed at a relatively high temperature $T/T_c \cong 1/\sqrt{\kappa} \cong 0.1$ for cuprate superconductors (κ — Ginzburg–Landau parameter). The paramagnetic Meissner effect was also observed in YBCO thin films with oriented inner surfaces obtained by bombardment with heavy ions [14].

Additionally, the theory [15,16] predicted a strong temperature dependence of the nonlinearity coefficient $\sim 1/T^3$ at low temperatures for this NLME mechanism. It was also shown in Ref. [16] that the nonlinear response attributable to ABS can lead to a reversal of the sign of the nonlinear current flowing through the superconductor and a decrease of the temperature dependence of the third harmonic power.

However, few papers have been devoted to local studies of NLME in high-temperature superconductors. Sensitive near-field nonlinear microwave microscopes have recently been developed for this purpose [17–19]. These methods make it possible to study the nonlinear Meissner effect using different current orientations relative to the directions of the nodes of the superconducting gap.

The temperature dependences of the third harmonic power were studied in this paper using the method of nonlinear near-field microwave microscopy at different orientations of the axes of $a-b$ ultrathin films.

2. Method of study and samples

The nonlinear microwave properties of ultrathin YBaCuO films were studied in this paper using the method of nonlinear near-field microwave microscopy. This method is based on the registration of a nonlinear microwave response using an inductive type probe. The probe is a thin copper wire with a length of 2 mm and a diameter of 40 μm shorting the inner and outer conductors of the coaxial cable [17]. The frequency of the first harmonic in the experiment was 472 MHz. The microwave signal is amplitude modulated with 1 kHz signal. The level of incident power at the frequency of the first harmonic during measurements ranged from 2 to 100 mW. The maximum power of the microwave signal is limited by the power of the generator, power amplifier and losses in the microwave path, and the minimum microwave power is determined by the sensitivity of the receiver. A high-density alternating current flows in the copper wire since the wave impedance of the coaxial cable is significantly greater than the impedance of the shorting wire, when the microwave signal is reflected from the near-field probe. When a strong high-frequency field interacts with the sample under study, higher harmonics of the fundamental frequency arise in the spectrum of the reflected signal because of the nonlinear properties of the superconductor. The receiver measures the amplitude of the reflected microwave signal at the frequency of the third harmonic. This signal is isolated using bandpass filters with a bandwidth of 1.41 − 1.43 GHz. A low-noise intermediate frequency amplifier with a bandwidth of 0.1 − 2 GHz is used to amplify this signal. Then it is detected, amplified by the SR830 Lock-In Amplifier at a frequency of 1 kHz and registered using the computer. The local method of study of nonlinearity makes it possible to study the spatial distribution of the third harmonic power $P_{3\omega}(x, y)$ of YBCO films in the temperature range from 4.2 to 100 K.

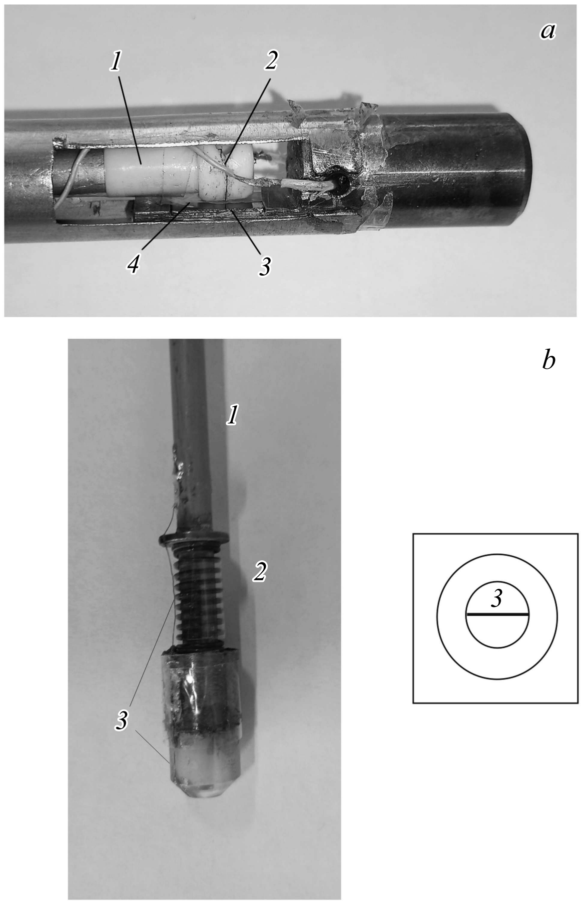

Figure 1. *a*) Image of the near-field microwave probe for scanning along the film: *1* — coaxial cable, *2* — wire of the near-field microwave probe, *3* — HTSC film, *4* — Teflon film; *b*) image of a near-field microwave probe for measuring the anisotropy of nonlinear microwave properties of superconductors: *1* — coaxial cable, *2* — spring, *3* — wire of a near-field microwave probe. The insert for the case (*b*) shows the bottom view.

Two modifications of the near-field microwave probe of the inductive type are used in the experiment. The first probe is designed for scanning in one direction along the sample (Figure 1, *a*). The wire of the near-field microwave probe is parallel to the edge of the film during scanning. The probe is moved mechanically by turning the screw. The scanning step is 0.5 mm. The initial distance from the probe wire to one of the four edges of the square film is recorded using an electronic vernier caliper. The displacement of the position of the near-field microwave probe after cooling is controlled by the angle of rotation of the screw. The microwave cable is placed inside a stainless steel tube to compensate for temperature drifts in the size of the insert. It should be noted that the magnitude of the third harmonic power signal strongly depends on the height of the probe wire above the film surface in the case of a nonlinear near-field microwave microscope. Therefore, its height is set using a $10\,\mu$m thick Teflon film, which is placed between the sample and the near-field microwave probe.

The second probe is used to measure the anisotropic nonlinear microwave properties of superconducting films (Figure 1, b). In this case, the near-field microwave probe is placed in the center of the film, and the coaxial cable is directed orthogonally to its surface. At the same time, the probe wire is symmetrical with respect to the center of the film (Figure 1, b). The microwave cable tube is centered with foam rings to reduce the displacement of the probe relative to the center of the film when it is rotated. A spring is used to press the probe wire evenly.

An epitaxial film was grown from a high-temperature YBCO superconductor with a thickness of 4−5 nm on a substrate $LaAlO_3$ with a size of $10 \times 10\,mm^2$ at a temperature of 665°C. Then it was coated with a thin layer of gold 20 nm thick to protect it from moisture. The gold film was formed at a temperature of about 100°C with a slight vacuum gap after sputtering of the YBCO film. It should be noted that both double-layer YBCO/Au structures and YBCO films were studied in the work.

The orientation of the axes $a-b$ and the thickness of the YBaCuO films were determined by X-ray diffraction analysis using Bruker D8 Discover diffractometer.

3. Experimental results

It was previously shown that the temperature dependence of the nonlinear microwave response of superconducting films of $YBa_2Cu_3O_7$ with a thickness of 100 nm has a maximum near the critical transition temperature [17]. No increase of the third harmonic power in $P_{3\omega}(T)$ with a decrease of temperature was observed in the low temperature region. Therefore, ultrathin YBCO films were studied to increase the sensitivity of the setup. This is attributable to the fact that a decrease of the thickness of the film results in an increase of the nonlinear microwave signal due to an increase of the density of the superconducting current flowing through it.

Figure 2 shows the temperature dependences of the third harmonic power $P_{3\omega}(T)$ of the double-layer YBaCuO/Au structure at different distances from the edge of the film with a thickness of 5 nm. Figure 2 shows that the temperature dependence of the nonlinear microwave response in this case demonstrates a maximum near the temperature of 80 K. In addition, $P_{3\omega}(T)$ in the low temperature range

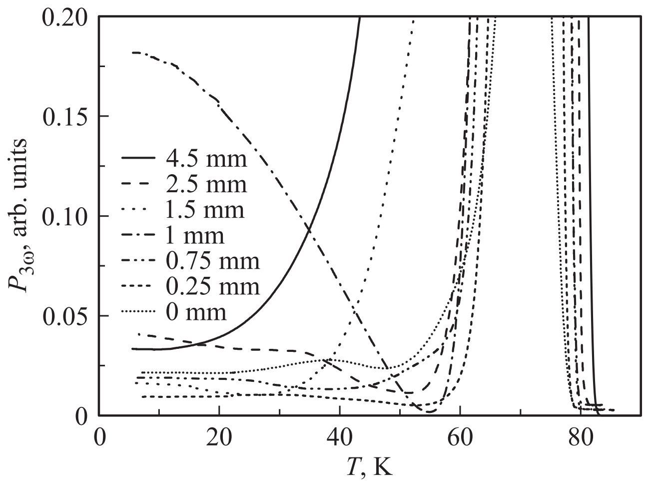

Figure 2. The temperature dependence of the third harmonic power $P_{3\omega}(T)$ of the double-layer YBaCuO/Au structure at different distances from the edge of the film. The angle between the direction of the axes $a-b$ and the edge of the sample is 45°. The thickness of the HTSC film is 5 nm. Microwave signal power is 7 dBm.

demonstrates an increase of the nonlinear microwave response with a decrease of the temperature. As is known, the increase of the nonlinear microwave response at low temperatures in high-temperature superconductors can be associated with both the nonlinear Meissner effect and the nonlinear response due to the presence of Andreev bound states. Zeroing of the third harmonic power and a strong increase of the nonlinear microwave response in the low temperature region is observed with a probe distance of 1 mm from the edge.

Figure 3 shows the temperature dependences of the third harmonic power $P_{3\omega}(T)$ of the double-layer YBaCuO/Au structure at different microwave signal powers at a distance of 1 mm from the edge of the superconducting structure. Figure 3 shows that the zeroization temperature $P_{3\omega}(T)$ depends on the microwave power and shifts towards lower temperatures when it increases.

Figure 4 shows the results of scanning a 4 nm thick YBaCuO film. The figure shows the temperature dependences of the third harmonic power $P_{3\omega}(T)$ at different distances from the edge of the film. Figure 4 demonstrates an increase of the nonlinear microwave signal at temperatures of the order of 20−25 K. In addition, a non-linear microwave response is also observed with a probe distance of 3 and 4 mm from the edge, as in the case of a double-layer structure of YBaCuO/Au.

It is known that zeroization of the power of the third harmonic can occur due to the nonlinear response associated with Andreev edge states. In this case, the phase of the nonlinear current of the Andreev states is shifted by 180° compared, for example, with the volumetric nonlinear Meissner effect [16]. At the same time, these states in films of a high-temperature superconductor can occur both at the

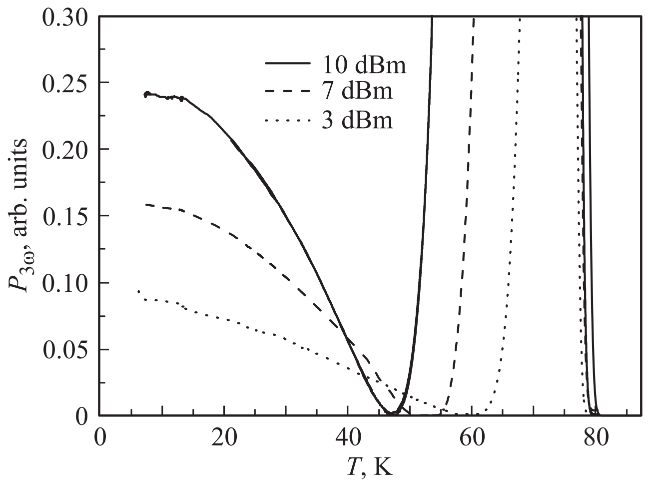

Figure 3. The temperature dependence of the third harmonic power $P_{3\omega}(T)$ of the double-layer YBaCuO/Au structure at different microwave signal levels. The angle between the direction of the axes $a-b$ and the edge of the sample is 45°. The thickness of the HTSC film is 5 nm. The distance of the probe wire to the edge of the film is 1 mm.

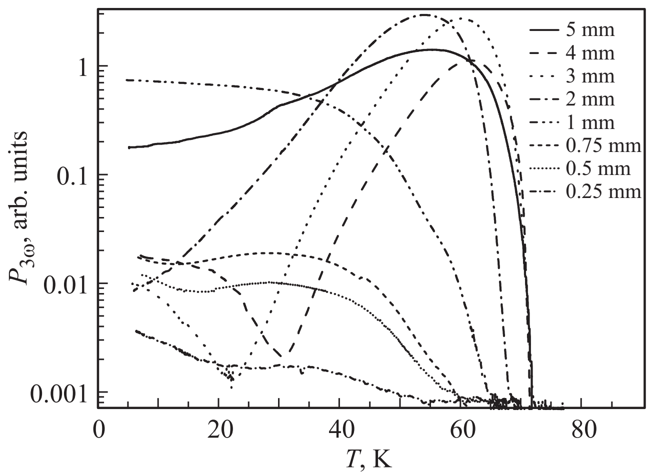

Figure 4. The temperature dependence of the third harmonic power $P_{3\omega}(T)$ of the YBaCuO film with a thickness of 4 nm at different distances from the edge of the film. The angle between the direction of the axes $a-b$ and the edge of the sample is 45°. Microwave signal power is 3 dBm.

boundary of the sample and in the center of the film. This is attributable to the fact that such interfaces at the twin boundaries can be formed spontaneously during the growth of the epitaxial film [20]. Figure 5 schematically shows the process of occurrence of a response associated with ABS at the twin boundaries.

The dependence of the third harmonic power on the orientation of the wire of the near-field microwave probe in the center of the film was studied for studying the anisotropy of nonlinearity (Figure 6). Figure 6 shows that the maximum value of the nonlinear microwave response is observed in the direction of the axes $a-b$. It should be noted that an increase of the nonlinear signal along the directions of the axes $a-b$ was also associated with the nonlinearity of the Andreev states in Ref. [19]. On the other hand, the anisotropy of the sample shape in the center of the film, in our opinion, does not affect the nonlinear microwave response, since, as can be seen from Figure 6, an increase of the nonlinear microwave response is observed at a distance of 1 mm from its edge (Figure 2). Therefore, the angular dependence of the nonlinear microwave response is not related to the influence of the film edges.

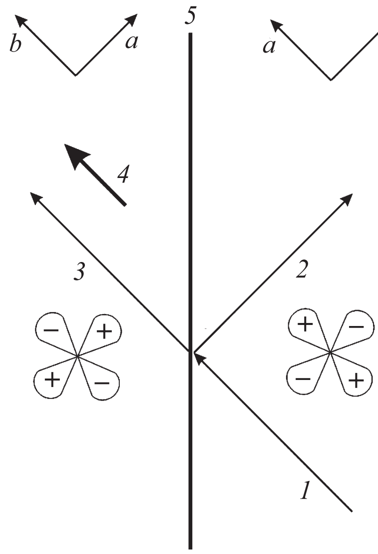

Figure 5. Schematic representation of the process of occurrence of the response associated with ABS: *1* — incident electron, *2* — reflected electron, *3* — passed electron, *4* — microwave current, *5* — the boundary of the twinning. The angle between the axis *a* and the twin boundary is 45°.

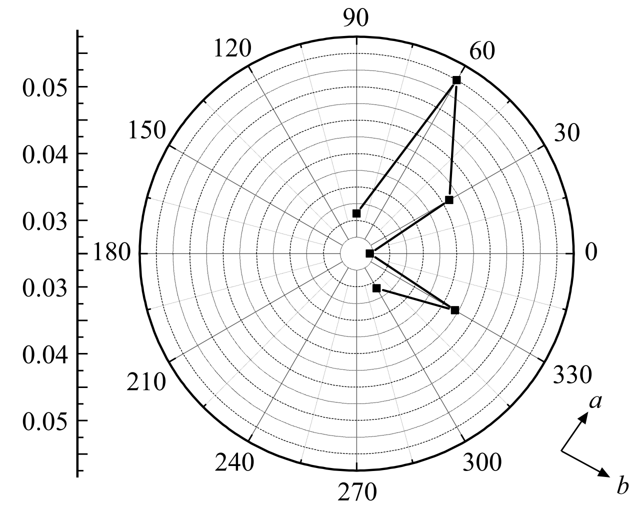

Figure 6. Dependence of the third harmonic power $P_{3\omega}(T)$ on the orientation of the probe wire relative to the axes $a-b$ at temperature of $T = 4.2$ K. The probe is located in the center of the film. Microwave signal power is 10 dBm.

In conclusion, it should be noted that the microwave properties are quite sensitive to various types of structural defects [21,22]. This is attributable to the short coherence length of YBCO films (in the order of 2 nm) compared to the sizes of these defects. Therefore, despite the fact that the characteristic dimensions of the twin are in the order of 50 nm, the microstructure of the films affects the nonlinear microwave response. At the same time, the inhomogeneous distribution of the power of the third harmonic at millimeter scales in the film plane is apparently attributable to the inhomogeneous distribution of the twinning boundaries in the YBCO film (Figures 2 and 4).

Therefore, firstly, the increase of the nonlinear response at low temperatures in ultrathin YBCO films, secondly, the presence of zeroization in $P_{3\omega}(T)$ at temperatures below the critical and, thirdly, the anisotropy of the power of the third harmonic indicate that the nonlinear microwave response is attributable to the nonlinearity of the Andreev states, arising on the interfaces of the twin boundary.

4. Conclusion

An increase of the power of the third harmonic in the low temperature region was detected using the method of nonlinear near-field microwave microscopy in ultrathin YBCO films. The temperature dependence of the nonlinear microwave response demonstrates zeroization at temperatures below T_c. The anisotropy of the nonlinear response of ultrathin YBCO films was studied by near-field microwave microscopy. The experimental results obtained indicate that the nonlinear microwave response in the low temperature region may be associated with the presence of Andreev boundary states occurring at the interfaces of the boundaries of the twins.

Acknowledgments

The author is grateful to A.S. Melnikov for useful discussions.

Funding

This study was carried out under the state assignment of Institute of Physics of Microstructures, Russian Academy of Sciences (SA topic:FFUF-2022-0006). The equipment provided by the Center for Collective Use „Physics and Technology of Micro- and Nanostructures“ was used for the study.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by A.Akhtyamov