# Unusual features in the nonlinear microwave surface impedance of Y-Ba-Cu-O thin films

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Striking features have been found in the nonlinear microwave (8.0 GHz) surface impedance $Z_s = R_s + j \cdot X_s$ of high-quality YBaCuO thin films with comparable low power characteristics [$R_{res} \sim 35-60\ \mu\Omega$ and $\lambda_L(15\ \text{K}) \sim 130-260$ nm]. The surface resistance $R_s$ is found to increase, decrease, or remain independent of the microwave field $H_{rf}$ (up to 60 mT) at different temperatures and for different samples. However, the surface reactance $X_s$ always follows the same functional form. Mechanisms which may be responsible for the observed variations in $R_s$ and $X_s$ are briefly discussed. [S0163-1829(98)03242-1]

Measurements of the nonlinear microwave surface impedance $Z_s$, of high-temperature superconductors (HTS's) is a powerful tool for studying nonequilibrium processes in these materials. Nonlinear impedance measurements allow one to investigate peculiarities of the rf-vortex nucleation, and to study the vortex dynamics at elevated microwave fields. Such measurements may also discriminate between d-wave and s-wave mechanisms of pairing symmetry in HTS's, and indicate the presence of magnetic impurities in the materials.[1,2]

In the present paper, we report observations of nonmonotonous behavior of $R_s$ and the penetration depth $\lambda$ (or, equivalently, the surface reactance $X_s = \omega\mu_0\lambda$), of high-quality epitaxial YBaCuO thin films, in microwave fields up to 60 kA/m ($\sim$700 Oe) using the coplanar resonator technique[3] at 8 GHz. For all samples, depending on temperature $T$, $R_s$ demonstrates completely different behavior, whereas $\lambda$ always preserves the same $H_{rf}$ dependence, irrespective of sample and $T$. Measurements are presented for very high-quality samples over a wide temperature range (12–75 K) which at first time reveal *nonmonotonous* and *uncorrelated behavior* in $R_s$ and $X_s$ as a function of $H_{rf}$. Such a behavior does not agree with any of the existing models for the nonlinear microwave impedance.[4–9] In the following we discuss several mechanisms relevant to these observations.

The films are deposited by $e$-beam coevaporation onto polished (001)-orientated MgO single crystal substrates $10 \times 10\ \text{mm}^2$. The films are 350 nm thick. The $c$-axis misalignment of the films are typically less than 1%, and the dc critical current density $J_c$ at 77 K is around $2 \times 10^6\ \text{A/cm}^2$. More detailed information on the growth technique can be found in Ref. 10. The values of $R_s$ and $\lambda$ at 15 K are 60, 35, 50 $\mu\Omega$ and 260, 210, 135 nm for samples TF1, TF2, and TF3, respectively.

Changes in $R_s$ and $X_s$ with $H_{rf}$, $\Delta R_s = R_s(H_{rf}) - R_s(0)$ and $\Delta X_s = X_s(H_{rf}) - X_s(0)$, are plotted in Figs. 1 and 2 for all three samples. For sample TF1 for all $T$ and in the whole field range $\Delta R_s \sim H_{rf}^2$, whereas for samples TF2 and TF3 the behavior of $\Delta R_s(H_{rf})$ changes dramatically with $T$. For sample TF2 $R_s$ changes from decreasing at 15 K to almost $H_{rf}$-independent behavior at 35 K, and to a rapidly increasing function of $H_{rf}$ between 40–75 K. At 15 K $\Delta R_s$ diminishes noticeably only at $H_{rf} > 10$ kA/m showing no features of saturation up to the highest available $H_{rf}$ of $\sim$40 kA/m. At higher $T$ (70 K) a transition to a characteristic sublinear field dependence ($\sim H_{rf}^n$, $n < 1$) occurs. A similar behavior is also observed for sample TF3 at 15 and 35 K over the whole field range, whereas at $T > 70$ K [see Fig. 1(c)] a minimum in $\Delta R_s(H_{rf})$ at low fields appears, after which the usual sublinear $H_{rf}$ dependence is recovered. As regards to $\Delta X_s(H_{rf})$, it is always a sublinear function of $H_{rf}$ at low fields with a characteristic kink and superlinear $H_{rf}$ dependence ($\sim H_{rf}^n$, $n > 1$) at higher fields (see Fig. 2). This dependence of $\Delta X_s$ on $H_{rf}$ persists for all samples and for almost all temperatures, and in general *no correlation is observed* between $\Delta X_s(H_{rf})$ and $\Delta R_s(H_{rf})$. The only exception is sample TF3 for which $\Delta X_s(H_{rf})$ qualitatively correlates with $\Delta R_s(H_{rf})$ at all $T$, and even the minimum at low fields is reproduced in both dependences at 75 K [see Figs. 1(c) and 2(c)]. In Figs. 1 and 2 some of $\Delta R_s$ and $\Delta X_s$ data are fitted to the function $\sim H_{rf}^n$ which is predicted by Halbritter's model of Josephson vortex motion in weak links (WL's)[8] ($0.5 < n < 2$) and the Ginzburg-Landau theory for the pair breaking mechanism ($n = 2$).

In Fig. 3, we plot the temperature dependence of the $r$ parameter ($r = \Delta R_s / \Delta X_s$) for all the samples at three microwave power levels. These data are often used to distinguish

 

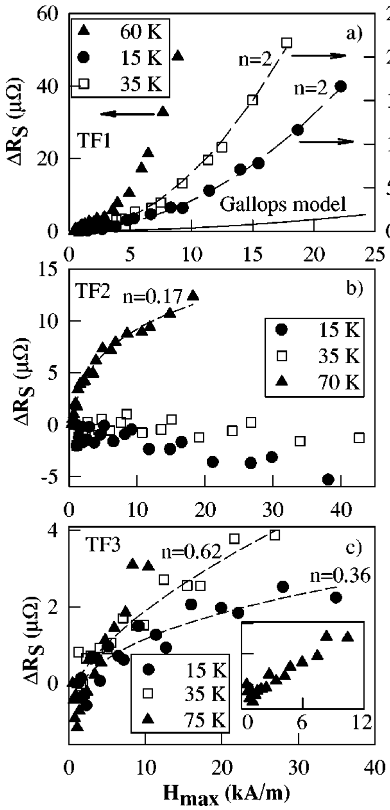


FIG. 1. Microwave field $H_{\mathrm{rf}}$ dependences of the change in the surface resistance $\Delta R_s$ for three samples TF1, TF2, and TF3 at different temperatures $T$. The dashed lines are fitting curves using a function $\sim H_{\mathrm{rf}}^n$, which is discussed in the text. $T$ and $n$ values are given in the figure. The solid line in (a) is a fit using the modified model of Gallop *et al.* Ref. 14. The parameters of the fit are as follows: normal resistance $R_n = 1.83\ \Omega$, zero field critical current density $J_{c0} = 10^{10}$ A/cm$^2$, grain size $\alpha = 6.4\times10^{-7}$ m, grain penetration depth $\lambda_{ab0} = 2.51\times10^{-7}$ m. The inset shows the data at 75 K on an expanded scale.

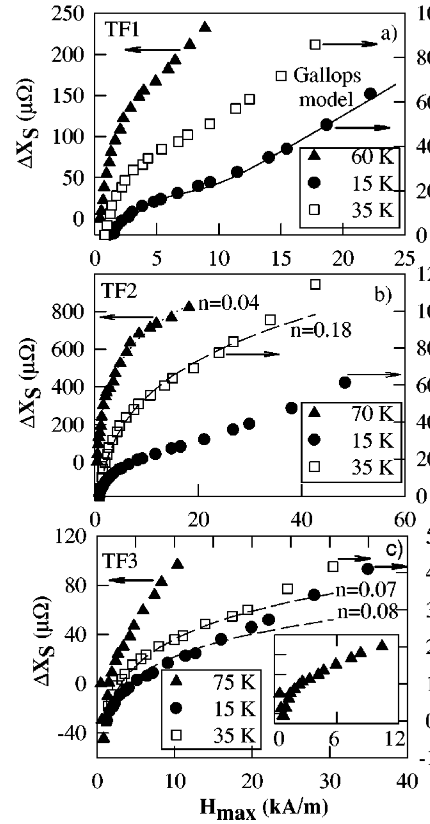


FIG. 2. The change in the surface reactance $\Delta X_s$ as a function of $H_{\mathrm{rf}}$ for three samples TF1, TF2, and TF3 at different temperatures. The fits are the same as those plotted in Fig. 1. The inset shows the data at 75 K on an expanded scale.

between various nonlinear mechanisms.[5,8] The general trend of $r(T)$ for all the samples is a decrease in the absolute value of $r$ with increasing $T$, gradually saturating at high $T$. For samples TF2 and TF3 the most pronounced change in $r$ occurs at low $T$, where it has a large negative value of about $-1$ and rapidly levels off with temperature approaching a value of 0.01–0.06 [see Figs. 3(b), 3(c)]. One can see that the initial negative value of $r$ is reduced with enhanced power. Unlike other samples, TF1 in the high field regime ($H_{\mathrm{rf}} \sim 7600$ A/m) displays an increase in the $r$ parameter at high temperatures ($T > 45$ K) and reaches a value of $\sim 0.15$. In addition, the initial low-$T$ $r$ value for sample TF1 depends nonmonotonously on the microwave field [see Fig. 3(a)].

For explanation of the observed nonmonotonous field dependences of $\Delta R_s$ and $\Delta X_s$ we involve three different mechanisms. Each mechanism is capable of describing only particular features in $H_{\mathrm{rf}}$ dependences of $\Delta R_s$ and $\Delta X_s$. First, the modified weakly coupled grain model,[11] proposed by Gallop *et al.*,[12] assumes that for high-quality HTS films a WL between two superconducting grains is shunted by another third grain, which serves as an additional path for both dc and rf currents. This model presents a highly simplified picture (not least, because it makes no distinction between

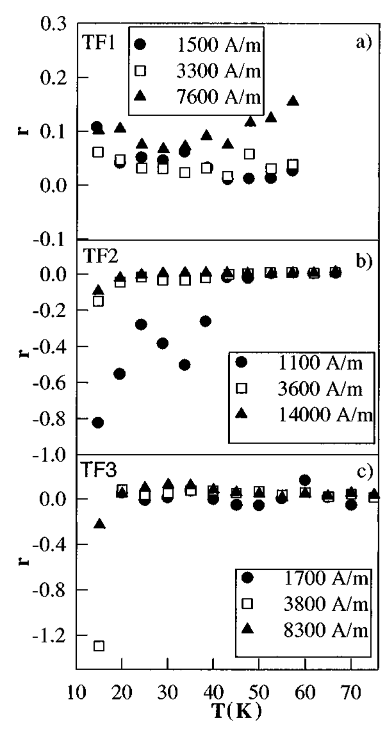


FIG. 3. Temperature dependences of the $r$ parameter ($\Delta R_s/\Delta X_s$) for samples TF1, TF2, and TF3 at different $H_{\mathrm{rf}}$ (specified in the figure).

the Meissner and the mixed states). Nevertheless it enables one to reproduce a reduction of $R_s$, such as we observe, given a certain set of the material parameters. However, it is unable to describe two important features observed by us; sublinear $\Delta X_s(H_{rf})$ dependence at low fields with a characteristic change in curvature at higher $H_{rf}$, and the decrease of $\Delta X_s$ with $H_{rf}$ [see Fig. 2(c)]. We have managed to overcome in part the first drawback of the model by introducing an effective local flux density $B_{eff}$, which interacts with the rf current. When both the junction and grains are in the Meissner or mixed state, the magnetic flux is quasihomogeneously distributed throughout the region to which it penetrates, and $B_{eff}=B$. However, in the mixed state of the WL only, the flux will be concentrated inside the junctions due to screening currents in the grains, and hence, in the WL $B_{eff}>B$. The ratio $B_{eff}/B$ should increase with $B$ and reach a maximum at $B_{eff}=\mu_0 H_{c1}$ ($H_{c1}$ is the lower critical field of the grains) after which it should decrease rapidly. Adopting a simple function for $B_{eff}/B(B)$ which possesses the properties specified above (we took the Gaussian function) we have managed to get an excellent fit to our $\Delta X_s(H_{rf})$ data [see Fig. 2(a)], but we failed to reproduce the $\Delta R_s$ field dependence [see Fig. 1(a)]. Moreover, such a model cannot reproduce the decrease in $\Delta X_s$ with $H_{rf}$ observed for TF3 sample at high temperatures [see Fig. 2(c)].

Another model applicable to our results is the model of Eliashberg[13] for superconductivity stimulated by high power microwave irradiation. As shown by Eliashberg,[13] in a superconductor with a homogeneous order parameter distribution, microwave radiation of a certain power can induce a new quasiparticle distribution function with an increased gap, which in turns leads to an enhancement in superconducting properties. A similar effect is predicted by the Aslamazov-Larkin (AL) theory[14] for inhomogeneous WL, due to the radiation-induced diffusion of quasiparticles out of the junction region which occurs for a certain level of microwave power. Since the AL theory, contrary to the Eliashberg model, predicts a suppression of the order parameter at low rf fields, we can exclude this mechanism immediately, since we observe a *reduction* of $R_s$ at the lowest fields [see Figs. 1(b), 1(c)]. With regards to the Eliashberg theory, it predicts a decrease of the stimulation effect with lowered temperature, and a suppression of superconductivity by a static magnetic field.[15] In fact, for sample TF2 we see that the decrease in $R_s$ with $H_{rf}$ is reduced with increasing temperature and completely disappears at high $T$, whereas for sample TF3 the decrease in $R_s$ is observed only at high $T$ [see Fig. 1(c)]. Moreover, additional experiments performed by us in low dc magnetic fields $H_{dc}\leqslant 12$ mT (to be published elsewhere), showed that while for sample TF2 $H_{dc}$ causes an even more pronounced decrease in $R_s$, for sample TF3 the dc field *always leads to an enhanced* $R_s$.[16] However, in accordance with Ref. 13, stimulation of superconductivity is expected only in highly uniform narrow and thin superconducting channels with a homogeneous order parameter and microwave field distribution, which is hardly the case for our wide and ''quasibulk'' samples.

Finally, the third mechanism which may account for our results is the recovery of superconductivity due to the field-induced spin alignment of magnetic impurities which are likely to be present in most HTS's (particularly in YBaCuO).[1] Magnetic impurities are a source of Cooper pair breaking due to the spin-flip scattering process. However, at low temperatures the decrease in thermal motion leads to the appearance of spin-spin correlation of the impurity atoms which becomes strong and may frustrate the spin-flip scattering. An external magnetic field also leads to ordering via alignment of the magnetic impurity spins and hence, can also lead to a reduction of pair breaking.

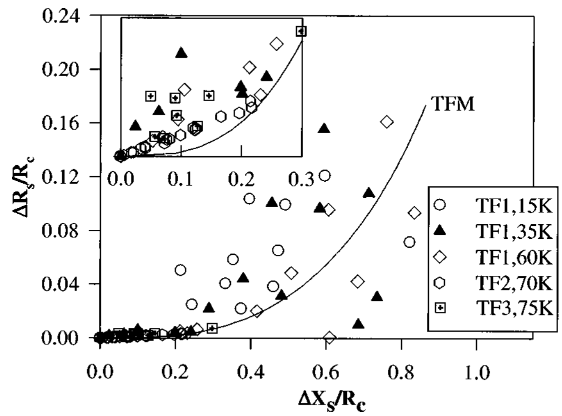


FIG. 4. Parametric plot of $\Delta R_s/R_c$ vs $\Delta X_s/R_c$ for various samples at different temperatures (specified in the figure). The solid line is a fit to the two-fluid model discussed in the text. The inset shows the low power data on an expanded scale.

Recently Hein *et al.*[2] observed a correlated reduction of $R_s$ and λ in both dc and rf fields of the same order of magnitude ($\leqslant 20$ mT). They performed an analysis of the function $\Delta R_s/R_c(\Delta X_s/R_c)$ (where $R_c=\sqrt{\omega\mu_0/2\sigma_n}$, and $\sigma_n$ is the normal electron conductivity) in terms of the two-fluid model (TFM) and found that their data collapsed onto a single TFM curve. In addition, the conductivity ratio $y=\sigma_1/\sigma_2$ (where $\sigma_1$ and $\sigma_2$ are quasiparticle and superfluid conductivities, respectively) was found to decrease with increased magnetic field, which was attributed to the field-induced reduction of pair breaking. The major difference between our results and those of Hein *et al.*[2] is that they did not observe a reduction of $R_s$ with $H_{rf}$ without an accompanying reduction of λ; but at the same time they observed a reduction of λ with $R_s$ being almost independent of $H_{rf}$. In contrast, we observed a reduction of $R_s$ for a monotonously increasing λ, and moreover, only in rare cases did we observe a decrease in $R_s$ correlated with a decrease in λ [see Figs. 1(c) and 2(c)]. In addition, a similar analysis based on the TFM was performed by us which showed rather poor scaling of our $\Delta R_s/R_c(\Delta X_s/R_c)$ data, as plotted in Fig. 4. One further distinctive feature of our data compared to those of Hein *et al.*[2] is the significant discrepancy (up to several times) in the $R_c$ values extracted from the $\Delta R_s$ and $\Delta X_s$ data (see Table I). In addition, our conductivity ratio $y=\sigma_1/\sigma_2$, extracted from the fitting to the TFM curve, was found to increase with $H_{rf}$, rather than decrease, as expected for the mechanism of the impurity spins alignment.[1] Nevertheless, our preliminary measurements in weak dc magnetic fields ($\leqslant 12$ mT) in field cooled regime at constant $H_{rf}$ showed a decrease of $R_s$ and λ with $H_{dc}$ for samples TF1 and TF2 (to be published elsewhere). This suggests that magnetic impurities may play a significant role in our samples and might also affect our nonlinear measurements.

TABLE I. Two fluid model fitting parameters of $\Delta R_s/R_c$ vs $\Delta X_s/R_c$ dependences for various samples at different temperatures. Here $y(H_{rf}=0)$ and $y(H_{rf}^{on})$ are the conductivity ratios ($y=\sigma_1/\sigma_2$) at low and high microwave powers, and $R_c(\Delta R_s)$ and $R_c(\Delta X_s)$ are the $R_c$ values extracted from $\Delta R_s(H_{rf})$ and $\Delta X_s(H_{rf})$ data, respectively.

| Sample | $y(H_{rf}=0)$ | $y(H_{rf}^{on})$ | $R_c(\Delta R_s)$, m$\Omega$ | $R_c(\Delta X_s)$, m$\Omega$ |
|---|---|---|---|---|
| TF1, 15 K | 0.183 | 0.173 | 0.103 | 0.046 |
| TF1, 35 K | 0.028 | 0.123 | 0.215 | 0.084 |
| TF1, 60 K | 0.050 | 0.111 | 0.700 | 0.352 |
| TF2, 70 K | 0.008 | 0.012 | 0.021 | 0.004 |
| TF3, 75 K | 0.010 | 0.022 | 0.0008 | 0.0003 |

We proceed with an analysis of our high-temperature data for increasing $\Delta R_s(H_{rf})$ and $\Delta X_s(H_{rf})$ in terms of the $r$ parameter.[5,8] It is essential to consider not only the $r$ value, but also the power dependence of the impedance.[17] A mechanism, such as the response of Josephson vortices, for which $\Delta R_s, \Delta X_s \sim H_{rf}^n$ $(0.5<n<2)$ and the $r$ value is about unity, can be excluded immediately, since for our experiments $r$ never exceeds 0.15. Moreover, the fit of our rf field dependences with a function $\sim H_{rf}^n$ [see Figs. 1(b), 1(c), 2(b), 2(c)] has revealed uncorrelated values of $n$ for $\Delta R_s$ and $\Delta X_s$ data (while from the theory[8] they should be the same), saying nothing about an apparent departure of the $H_{rf}^n$ fit from $\Delta X_s(H_{rf})$ data at high fields [Figs. 2(b), 2(c)]. The same conclusion is valid for the heating of weak links ($r_{HE}<1$, $\Delta R_s$, $\Delta X_s \sim H^2$) and the RSJ model[8] ($r_{RSJ}<1$, $\Delta R_s$ increasing and $\Delta X_s$ oscillating with $H_{rf}$). A value of the $r$ parameter consistent with our data could follow from either uniform heating or intrinsic Ginzburg-Landau nonlinearity[5] (for both mechanisms $r<10^{-2}$), but the rf dependence $\sim H_{rf}^2$ is generally not observed for our samples [except sample TF1, for which $\Delta R_s \sim H_{rf}^2$, but $\Delta X_s$ is not $\sim H_{rf}^2$, see Fig. 2(a)]. Moreover, the $r$ value should increase with $T$ for the above two mechanisms, while we see almost $T$-independent behavior at high temperatures [Figs. 3(b), 3(c)]. Thus, our high-temperature data which shows an increase in $\Delta R_s$ and $\Delta X_s$ with $H_{rf}$ are apparently not explained by any of the known theoretical models.

In conclusion, in our experiments we appear to observe a complicated interplay of several nonlinear mechanisms. At low temperatures, the observed reduction in $R_s$ may arise due to the effect of the magnetic impurity spins alignment by the rf field, while at higher $T$ stimulation of superconductivity by microwave irradiation and vortex mechanisms may also come into play. However, universal temperature- and sample-independent $H_{rf}$ dependence of the penetration depth $\lambda$ (or, equivalently, the surface reactance $X_s$) and similar values of $r \sim 0.01-0.06$ for all the samples over a broad temperature range ($35<T<75$ K) do not rule out the possibility that all the observed features may arise due to one and the same mechanism, the origin of which is not known at the moment. At the same time, absence of correlation between $\Delta R_s(H_{rf})$ and $\Delta X_s(H_{rf})$, for some of the samples particularly TF1, implies that the microstructure of the samples may interfere with the intrinsic behavior in the nonlinear response.


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