Non-linear Microwave Surface Impedance of Epitaxial HTS Thin Films in Low DC Magnetic Fields

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We have carried out non-linear microwave (8 GHz) surface impedance measurements of three YBaCuO thin films in dc magnetic fields $H_{dc}$ (parallel to c axis) up to 12 mT using a coplanar resonator technique. In zero dc field the three films, deposited by the same method, show a spread of low-power residual surface resistance, $R_{res}$ and penetration depth, $\lambda$, ($T=15$ K) within a factor of 1.9. However, they exhibit dramatically different microwave field, $H_{rf}$, dependences of the surface resistance, $R_s$, but universal $X_s(H_{rf})$ dependence. Application of a dc field was found to affect not only absolute values of $R_s$ and $X_s$, but the functional dependences $R_s(H_{rf})$ and $X_s(H_{rf})$ as well. For some of the samples the dc field was found to reduce $R_s$ below its zero-field low-power value.

I. INTRODUCTION

Understanding the mechanisms of the non-linearity of high-$T_c$ superconductors (HTS) at microwave frequencies is very important from the point of view of application of the materials in both passive and active microwave devices. Recently, unusual features such as decrease of the surface resistance $R_s$ and reactance $X_s$ of HTS thin films with microwave field $H_{rf}$ have been reported\textsuperscript{[1,2]}. Similar observations were made in weak ($\leq 20$ mT) static fields $H_{dc}$\textsuperscript{[3]} which have shown that a small dc magnetic field can cause a decrease of $R_s$ and $X_s$ in both the linear and nonlinear regimes.

In the present paper we report measurements of the microwave field dependences of $R_s$ and $X_s$ of high-quality epitaxial YBaCuO thin films in zero and finite ($\leq 12$ mT) applied dc magnetic fields. All the samples have rather different functional form of $R_s(H_{rf})$, but $X_s(H_{rf})$ is universal and nearly temperature-independent. At the same time, $H_{dc}$ applied parallel to the c-axis of the films has a qualitatively similar effect on both $R_s(H_{rf})$ and $X_s(H_{rf})$, giving evidence of non-monotonic behavior of $R_s$ and $X_s$ as a function of $H_{dc}$ both in the linear and nonlinear regimes. An even more striking feature is that for some of the samples the dc field can decrease $R_s$ below its low-power zero-field value, thereby offering a possible way of reducing the microwave losses of HTS thin films.

II. EXPERIMENTAL RESULTS

The films are deposited by e-beam co-evaporation onto polished (001)-oriented MgO single crystal $10 \times 10$ mm$^2$ substrates. The films are 350 nm thick. The c-axis misalignment of the films is typically less than 1%, and the dc critical current density $J_c$ at 77 K is around $2 \cdot 10^6$ A/cm$^2$. The films were patterned into linear coplanar transmission line resonators with resonance frequency of $\sim 8$ GHz using the technique described in\textsuperscript{[4]}. The nonlinear measurements were performed using a vector network analyzer with a microwave amplifier providing CW output power up to 0.3 W. The low-power values of $R_s$ and $A$ 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 Fig. 1 for all three samples. It is seen that the $H_{rf}$-dependence of $\Delta R_s$ is rather different for different samples, whereas $\Delta X_s(H_{rf})$ is universal. For sample TF1, $\Delta R_s \sim H_{rf}^3$ from the lowest $H_{rf}$. For sample TF2, a decrease in $R_s$ is observed with increased $H_{rf}$, and the absolute value of $R_s$ falls below the corresponding low-power value. Finally, for sample TF3, $\Delta R_s$ is rather independent of $H_{rf}$ up to sufficiently high fields ($\sim 60$ kA/m), after which a skewing of the resonance curve is observed. The surface reactance, $X_s$, for all three samples is a sublinear function of $H_{rf} (\sim H_{rf}^n, n < 1$) at low powers, then has a kink, followed by a superlinear functional dependence ($\sim H_{rf}^n, n > 1$).

The effect of dc magnetic fields ($\leq 12$ mT) on the microwave power dependence of $R_s$ and $X_s$ for all the samples is illustrated in Fig. 2 and Fig. 3. The common feature for all three samples is that the dependences of $R_s(H_{rf})$ and $X_s(H_{rf})$ upon $H_{dc}$ are non-monotonic. For samples TF1 and TF2 (for particular $H_{rf}$-range and $H_{dc}$-values), the static field leads to a decrease in $R_s$ compared to the low-power zero-field value. This means that both dc and rf fields can cause a reduction of the microwave losses in YBaCuO (see Fig. 2a and Fig. 3b). A possible mechanism of such a behavior is discussed later.

One can see that for sample TF1, a dc field of a certain strength (10 mT) can cause a decrease in $R_s$, whereas $X_s$ is always enhanced by a dc field. Similarly, for sample TF3, the behavior of $R_s(H_{rf})$ and $X_s(H_{rf})$ in $H_{dc}$ is also uncorrelated. However, for TF1 we observe a reduction of $R_s$ without an accompanying decrease in $X_s$, whereas for TF3 the effect is opposite; for particular values of $H_{dc}$ (5, 10 mT) the in-field ($H_{dc} \neq 0$) value of $X_s(H_{rf})$ is lower than the corresponding value for $H_{dc} = 0$ (Fig. 2b), while the in-field value of $R_s(H_{rf})$ is always higher than corresponding zero-field value (Fig. 3b). Here, the most pronounced decrease in $X_s$ for TF3 is observed at $H_{dc} = 5$ mT. Finally, for sample TF2 there is a well pronounced correlated behavior of $R_s(H_{rf})$ and $X_s(H_{rf})$ in a dc field; $H_{dc}$ of any value from 5 to 12 mT decreases both $R_s$ and $X_s$ (see Fig. 2b and Fig. 3b).
curves almost collapse over the entire range of $H_{3-7 \text{ kA/m}}$, where the $r$-dependence of $X$ is given by $X_s(H_{rf}) = X_s(0)$ as a function of peak microwave magnetic field $H_{rf}$ for three samples (specified in the figure) at $T = 15 \text{ K}, H_{dc} = 0$. The inset shows data for sample TF1 on an expanded scale.

### III. DISCUSSION AND CONCLUSIONS

A powerful approach in distinguishing between various nonlinear mechanisms is a parametric representation of the data in terms of the $r$ parameter, where $r = \Delta R_s/\Delta X_s$. In Fig. 3 we plotted the $H_{rf}$-dependence of the $r$ parameter for all three samples in different dc magnetic fields from 0 to 12 mT. One can see that for sample TF1, all the in-field $r(H_{rf})$ curves almost collapse over the entire range of $H_{rf}$, whereas the zero-field $r(H_{rf})$ data are clearly different from the in-field ones. This is especially noticeable at low $H_{rf}$ (between 3-7 kA/m), where the $r$ values differ by up to a factor of 10 between the zero-field and in-field $r(H_{rf})$ dependences. At the same time, at the lowest $H_{rf}$ (2-3 kA/m), the zero-field $r$-values match very well with the in-field ones (see Fig. 3). Therefore, the low-power nonlinearity for sample TF1 appears to have the same origin for zero-field and in-field regimes, whereas the high-power range mechanisms are likely to be different.

For sample TF2 at $H_{dc} = 5 \text{ mT}$ and 10 mT, $r(H_{rf})$ is rather noisy, which clearly correlates with the noisy dependence of $X_s(H_{rf})$ for this sample at the relevant dc fields (see inset in Fig. 3a). The $r$ parameter oscillates between $-4$ and 6 with an average values close to 0.3–0.4 and 0.2–0.3 for 5 and 10 mT, respectively. For zero field and 12 mT, $r(H_{rf})$ are quite consistent, starting to increase from large negative values $\sim -1$ at low powers, and saturating to the level of $-0.2$ to $-0.1$ at higher $H_{rf}$.

Finally, for sample TF3 at zero dc field, $r$ increases with $H_{rf}$ at low powers, whereas the 10 and 12 mT $r$-values decrease, but all three curves level off for $H_{rf} \geq 10 \text{ kA/m}$ to a value of $\sim 0.1$. However, $r(H_{rf})$ at 12 mT appears to tend to small negative values at high $H_{rf}$, consistent with the decrease in $R_s(H_{rf})$ at 12 mT in the relevant $H_{rf}$ range (Fig. 3c). Standing apart from other dependences is $r(H_{rf})$ at 5 mT, which shows very high values $\sim 2$ at low $H_{rf}$, saturating at a level of $\sim 0.4$ at higher $H_{rf}$. Note that this value is about a factor of 4 larger than the saturation level for other curves. This seems to imply that $H_{dc} = 5 \text{ mT}$ causes a switching of the mechanism of the nonlinearity in the film, as compared to the
mechanism at other fields, including zero-field results.

Recently Ma et al.\textsuperscript{9} have found that YBaCuO thin films deposited by the same method exhibit correlation of $R_s(\text{H}_{\text{rf}})$ with the values of low-power residual $\lambda_{\text{res}}$ and the normal-fluid conductivity $\sigma_n$. At the same time, they failed to note any correlation between the power dependence and $R_{\text{res}}$. A similar conclusion can be drawn from our results (see Fig. 1a). One can see that $R_s$ is almost independent of $\text{H}_{\text{rf}}$ for sample TF3, which has the lowest $\lambda(15 \text{ K}) = 135 \text{ nm}$, whereas sample TF1 with the largest $\lambda(15 \text{ K}) = 260 \text{ nm}$ exhibits the strongest $\text{H}_{\text{rf}}$-dependence. On the other hand, there is no strict correlation between $R_s(\text{H}_{\text{rf}})$ and low-power $R_s$ (see Sec. I and Fig. 1a), which is also consistent with the results of Ma et al. However, the strongest power dependence, $R_s \sim \text{H}_{\text{rf}}^2$, is observed for sample TF1 with both the highest $R_{\text{res}}$ and $\lambda_{\text{res}}$, in agreement with recent results on YBaCuO thin films with different low-power characteristics.\textsuperscript{10}

There are two further distinctive features for samples TF1, when compared with the two other samples. The functional form of $R_s(\text{H}_{\text{rf}})$ is noticeably changed by a dc field ($R_s \sim \text{H}_{\text{rf}}^n$, where $n = 2, 1.12, 0.8$ and $1.24$ for $0, 5, 10$ and $12 \text{ mT}$, respectively), whereas $X_s(\text{H}_{\text{rf}})$ is not affected by $H_{\text{dc}}$. In addition, for TF1, a dc magnetic field changes not only the power dependence of $R_s$, but the absolute value of the low-power $X_s$ (see Fig. 2a), while for TF2 and TF3 no such effect is observed (Fig. 2b, c). The effect of a dc field on $R_s(\text{H}_{\text{rf}})$ is also seen for sample TF3 at $H_{\text{dc}} = 5 \text{ mT}$ which, as will be argued later, may switch the mechanism of nonlinearity for this sample.

Recently Habib et al.\textsuperscript{11} have found that for a stripline resonator with a weak link in the middle, $R_s(\text{H}_{\text{rf}})$ is strongly affected by the junction, whereas $X_s(\text{H}_{\text{rf}})$ was found to be insensitive to the presence of the weak link. Based on this finding, we can suggest that the difference between $R_s(\text{H}_{\text{rf}})$
for our samples may originate from different microstructure (type, dimension and number of defects) of the samples, whereas the similar form of $X_s(H_{dc})$ appears to reflect the intrinsic behavior of each film, mostly exhibited by grains. This assumption is further supported by the strong effect of a small dc field on $R_s(H_{dc})$ for samples TF1 (Fig. 2a) and TF3 (at 5 mT, Fig. 2c), whereas the functional form of $X_s(H_{dc})$ is unchanged by $H_{dc}$.

**A. Analysis of Possible Mechanisms**

As we have shown earlier, such uncorrelated behavior of $R_s(H_{dc})$ and $X_s(H_{dc})$, as we observed for our samples (Fig. 1), cannot be explained by any of the known theoretical models, including Josephson vortices (where $r_{JF} < 1$, $\Delta R_s, \Delta X_s \sim H_{dc}^n$, $0.5 < n < 2$), heating of weak links ($r_{HE} < 1$, $\Delta R_s, \Delta X_s \sim H^2$) and the RSJ model ($r_{RSJ} < 1$, $\Delta R_s$ increasing in a stepwise manner and $\Delta X_s$ oscillating with $H_{dc}$), intrinsic pair breaking or uniform heating (for both mechanisms $r < 10^{-2}$, and $\Delta R_s, \Delta X_s \sim H^2$). We can also rule out the mechanism of the superconductivity stimulation by microwave radiation recently claimed by Hein et al. In this mechanism, the dc magnetic field decreases the order parameter, increasing both $R_s$ and $X_s$, which we do not observe for any of our samples. Moreover, we see that even in the low-power regime $H_{dc}$ can cause reduction of both $\Delta R_s$ and $\Delta X_s$, which is not explained by the above model at all.

The most plausible mechanism responsible for the decrease in $R_s$ and $X_s$ with both $H_{dc}$ and $H_{dc}$ fields seems to be field-induced alignment of the spins of magnetic impurities, which are likely to be present in most HTS (particularly in YBaCuO). This mechanism was recently claimed by Hein et al. to explain their results on non-monotonic behavior of $R_s$ and $X_s$ in $H_{dc}$ and $H_{dc}$ for YBaCuO thin films. However, because our non-linear results for $\Delta R_s \sim X_s$, which we observe for any of our samples. Moreover, we see that even in the low-power regime $H_{dc}$ can cause reduction of both $\Delta R_s$ and $\Delta X_s$, which is not explained by the above model at all.

In conclusion, we have presented here the results on non-monotonic microwave power dependence of $R_s$ and $X_s$ in both zero and weak ($\leq 12$ mT) dc magnetic field for very high-quality epitaxial YBaCuO thin films. Since this unusual behavior has come into being only owing to a significant progress in the thin film fabrication for the past few years, we conclude that the features observed by us seem to originate from the intrinsic properties of superconductors. However, different functional form of $R_s(H_{dc})$ for different samples and universal $X_s(H_{dc})$ behavior seem to imply that the microstructure still plays a significant role in the macroscopic properties of the samples. In addition, the observed decreases in $R_s$ and $X_s$ below their zero-field low-power values means that there is still room for improvement of the microwave properties of the thin films. This can be realized upon adequate understanding of the mechanisms responsible for the unusual behavior observed, and can lead to improved characteristics of HTS-based microwave devices.

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