Broadband microwave study of $SrRuO_3$ and $CaRuO_3$ thin films

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Broadband microwave study of $SrRuO_3$ and $CaRuO_3$ thin films

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Abstract. We study the microwave properties of $SrRuO_3$ and $CaRuO_3$ thin films in the frequency range 45 MHz to 40 GHz, and at temperatures between 80 K and 300 K, using a broadband Corbino technique. The films are grown on $SrTiO_3$ and MgO substrates, and we discuss how dielectric substrate resonances complicate the study of the film properties, in particular in the case of $SrTiO_3$ with its large dielectric constant. For $SrRuO_3$ on MgO substrate we find signatures of the ferromagnetic transition throughout our spectral range.

1. Introduction

Quantum phase transitions (QPTs) and their role for non-Fermi-liquid (NFL) behavior in electronic systems are a highly investigated topic, both theoretically and experimentally [1, 2]. While antiferromagnetic QPTs are found in a number of different materials (e.g. heavy fermion compounds [3]), one of the established examples of a ferromagnetic QPT [4] is the doped system $Sr_{1-x}Ca_xRuO_3$ [5, 6, 7] at the critical concentration $x \approx 0.8$. In optical measurements the frequency acts as tuning parameter which can be adjusted to probe the relevant energy scales of the system. For the pure compounds of our material system, $SrRuO_3$ and $CaRuO_3$, NFL behavior of the optical conductivity has been reported for infrared and THz frequencies [8, 9, 10], but it is unclear to which extent this is related to the ferromagnetic QPT. To match the characteristic low energy scales of QPTs and strongly correlated electron systems [11], in the present study we address the optical properties of $SrRuO_3$ and $CaRuO_3$ at very low energies, in the microwave frequency range.

2. Experiment

We have grown epitaxial thin films of $SrRuO_3$ and $CaRuO_3$ on $SrTiO_3$ substrates and polycrystalline $SrRuO_3$ films on MgO substrates using metalorganic aerosol deposition [7, 12, 13]. These films are in the range of 50 nm thick, i.e. much thinner than their microwave skin depth. We have measured the microwave properties of these samples in the frequency range 45 MHz to 40 GHz and at temperatures between 80 K and 300 K with a broadband Corbino technique as described in [14], but with a different cryostat that was optimized for intermediate and higher temperatures (similar to the one in [15]). Here the thin film sample under investigation terminates a coaxial cable: a vector network analyzer (NWA) sends the microwave signal through the cable onto the sample, where it is reflected and therefore probes its electromagnetic

properties. The reflected signal is detected by the NWA. To allow for a precise data analysis, we perform a thorough three-standard calibration procedure [14]. From the calibrated reflection coefficient we directly calculate the impedance and conductivity of the sample.

3. Results

In Fig. 1 we present reflection coefficient S_{11}, impedance Z, and conductivity $\sigma = \sigma_1 + i\sigma_2$ spectra for $SrRuO_3$ films on $SrTiO_3$ and MgO substrates, measured at 300 K. In this frequency range and at this temperature we expect a nearly constant σ_1 and a vanishing σ_2, consistent with metallic Drude behavior [16]. This is what we find for the film on MgO, see Fig. 1(f): $\sigma_1 \approx 0.23 \cdot 10^4\ \Omega^{-1}\ \mathrm{cm}^{-1}$ is almost constant and σ_2 is smaller. On top of these flat curves, there are two weak resonant features around 7 GHz and 13 GHz; these are not intrinsic to the $SrRuO_3$ film, but they are due to dielectric resonances in the substrate [17]. As these resonances are limited to small frequency regions, we can still analyze the microwave properties of the $SrRuO_3$ film in the broad frequency range outside the substrate resonances.

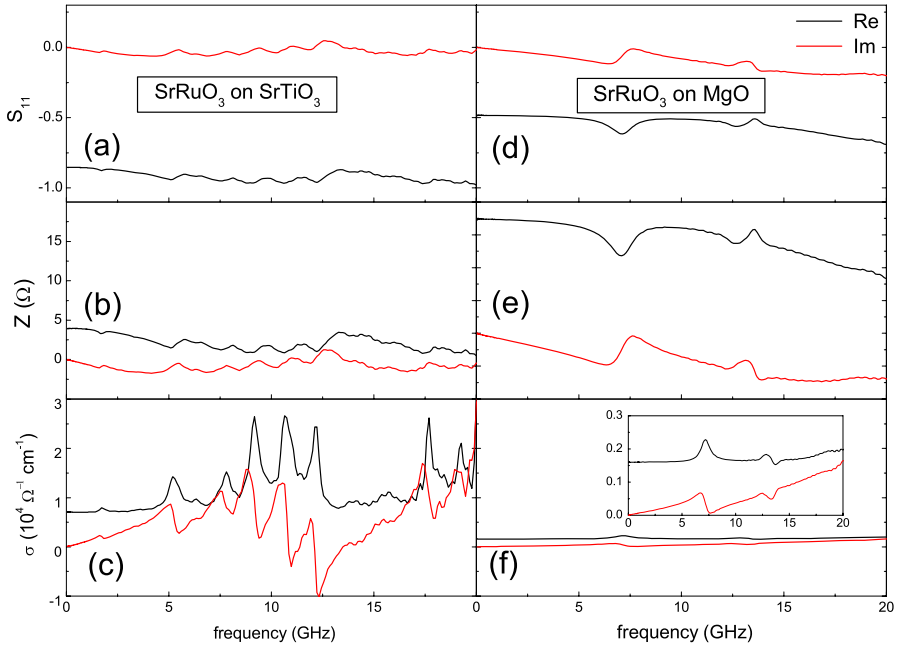

Figure 1. Real (black) and imaginary (red) parts of reflection coefficient S_{11}, impedance Z, and conductivity at 300 K for $SrRuO_3$ on $SrTiO_3$ and $SrRuO_3$ on MgO.

The $SrRuO_3$ on $SrTiO_3$ spectra in Fig. 1(a)-(c) show a rather different behavior: strong resonances cover the entire frequency range. These numerous resonances are a result of the very high dielectric constant of the $SrTiO_3$ substrate [18, 19], which shifts substrate resonances from higher frequencies into the measured microwave frequency range [17]. Here the influence of the substrate overwhelms the contributions of the $SrRuO_3$ film, and we cannot deduce the intrinsic microwave properties of the film from the measured overall response.

With decreasing temperature, the influence of the $SrTiO_3$ substrate gets even stronger, due to its increasing dielectric constant [18, 19]. This is shown in Fig. 2, where we plot $\mathrm{Re}(Z)$ of the measured impedance for $SrRuO_3$ and $CaRuO_3$ on $SrTiO_3$ substrates as a function of frequency and temperature. Upon cooling, substrate resonances move to lower frequencies, thus increasing the total number of resonances in our frequency range. With this strong influence of the substrate at GHz frequencies, we find signatures of the intrinsic response of the metallic films only at frequencies below 1 GHz, with a temperature dependence consistent with dc measurements [7].

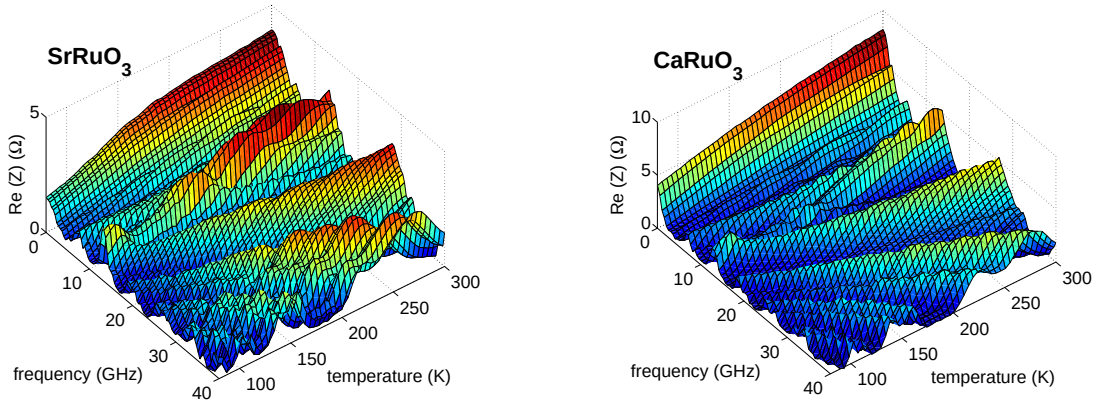

Figure 2. Real part of the impedance for $SrRuO_3$ and $CaRuO_3$ on $SrTiO_3$.

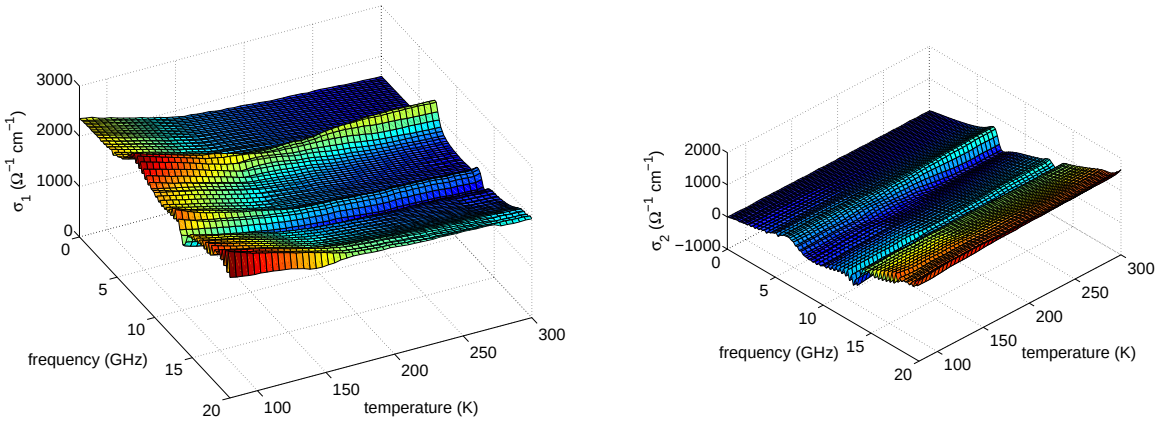

Figure 3. Real and imaginary parts of the conductivity of $SrRuO_3$ on MgO.

The situation is quite different for the $SrRuO_3$ film on MgO, as evident from the frequency and temperature dependence of σ_1 in Fig. 3. The surface of this plot is smooth up to 20 GHz, and the two substrate resonances in this range remain at the same frequency for all temperatures. For this sample, we measure intrinsic properties of the $SrRuO_3$ film. In Fig. 4 we show the temperature dependence of the real part ρ_1 of the resistivity ($\rho = 1/\sigma$) for different frequencies. The kink around 150 K indicates the ferromagnetic transition in the thin film and is known from the dc resistivity of $SrRuO_3$ [6, 7]. With increasing frequency, the kink slightly shifts toward lower temperatures. This is reminiscent of the behavior of a ferromagnetic antiresonance (FMAR) [20] in the sense that at higher probing frequencies the observed onset of ferromagnetism moves to lower temperatures. But contrary to FMAR, here we are not sensitive to the magnetic permeability of the sample and only sense the conductivity or resistivity, respectively. This kink at the ferromagnetic transition can also be identified for the $SrRuO_3$ film on $SrTiO_3$, see Fig. 2 at low frequencies, but the strong temperature dependence of the substrate excludes a detailed analysis. For $CaRuO_3$ no such kink is observed, consistent with dc data and the absence of ferromagnetic ordering [6, 7].

While we can easily determine the microwave properties of the $SrRuO_3$ film on MgO, this choice of substrate is disadvantageous compared to $SrTiO_3$ from a film growth point of view:

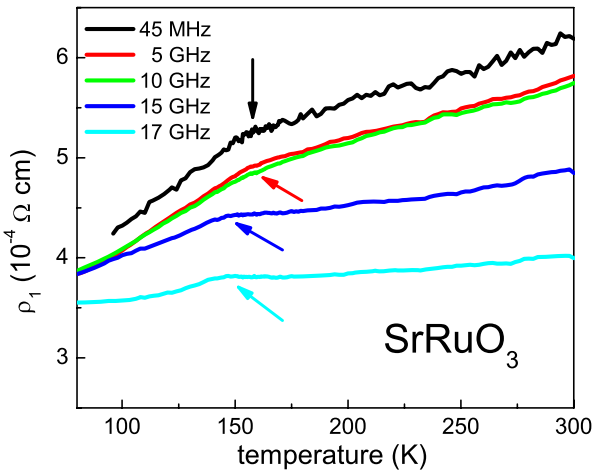

Figure 4. Temperature dependence of the real part of the resistivity of $SrRuO_3$ (on MgO substrate) at different frequencies. Arrows indicate the kink which heralds the ferromagnetic transition.

the lattice mismatch to the $Sr_{1-x}Ca_xRuO_3$ system is larger, and as a result the film quality is reduced [13]. This is evident from our data for the case of $SrRuO_3$, where for MgO substrate compared to $SrTiO_3$ the room temperature conductivity is much lower (Fig. 1); also the resistivity ratio upon cooling is smaller. For $CaRuO_3$ the lattice mismatch to MgO is even too big for successful film growth.

4. Conclusions and Outlook

We have measured the frequency-dependent impedance of $SrRuO_3$ and $CaRuO_3$ thin films on $SrTiO_3$ substrate and a $SrRuO_3$ thin film on MgO substrate at microwave frequencies. For $SrRuO_3$ on MgO, we clearly observe the signature of the ferromagnetic transition throughout our frequency range. While thin films on MgO substrate are well suited for optical measurements at low frequencies, thin films on $SrTiO_3$ show strong substrate contributions, which hamper a proper analysis. Since the high lattice mismatch of MgO and $Sr_{1-x}Ca_xRuO_3$ limits the film quality, an alternative substrate with low and temperature-independent dielectric constant and lower lattice mismatch should be tried for future studies e.g. $NdGaO_3$ [21].

Acknowledgements

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