# Impedance of $SrTiO_3$ thin films upon bias voltage: Inductive loops as a trace of ion motion

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## INTRODUCTION

Over the last few decades a vast amount of experimental work was performed on perovskite-type materials, with the focus on clarifying defect chemical questions and understanding electrochemical behaviour. In this context $SrTiO_3$ acts as a model material for other - more complex - large-band gap oxides such as $BaTiO_3$ or $Pb(Zr,Ti)O_3$. Fundamental experiments performed on single crystalline, poly- and even bi-crystalline materials revealed the importance of oxygen vacancies, electrons and holes as relevant charge carriers [1,2]. Current research is focused on the investigation of nanocrystalline materials and thin films. This field of research is eminently attractive due to the current demand of smaller devices. Nanocrystalline $SrTiO_3$ in particular is known to exhibit transport properties that differ from those of the bulk material [3]. Especially oxygen vacancies are discussed as important charge carriers which are involved in polarization phenomena such as resistance degradation and Wagner-Hebb polarization primarily investigated in bulk material. These properties make $SrTiO_3$ and its use in several new applications extremely attractive, e.g. memory devices based on resistive switching and sensoring applications [4].

In terms of investigating charge transport properties of electroceramics, electrical impedance spectroscopy and DC measurements are known as two powerful measurement tools. In this study, both methods were applied to slightly Fe-doped $SrTiO_3$ thin layers in order to investigate mass and charge transport in such films. Special emphasis is put on transient effects caused by a bias load at elevated temperatures (325-700°C). A major outcome of these measurements is the fact that bias induced ion motion in thin films often leads to inductive loops in impedance spectra.

## EXPERIMENTAL

Slightly Fe-doped $SrTiO_3$ thin films were prepared by pulsed laser deposition. (PLD) Nb-doped $SrTiO_3$ single crystals served as substrate material (electronic conducting but negligible ionic conducting = ionically blocking). As top (working) electrode $(La,Sr)CoO_{3-\delta}$ (LSC) was deposited by PLD. A sketch of the investigated sample set-up is shown in Fig. 1.

Circular microelectrodes were structured by photolithography and subsequent chemical etching with diluted HCl.

Impedance measurements and DC measurements were performed in the set-up shown in Fig. 2a.

Deposition parameters for the Fe:STO thin films (varying film thickness): T= 650°C, $p_{O_2}$= 0.15 mbar, pulse rate= 5 Hz

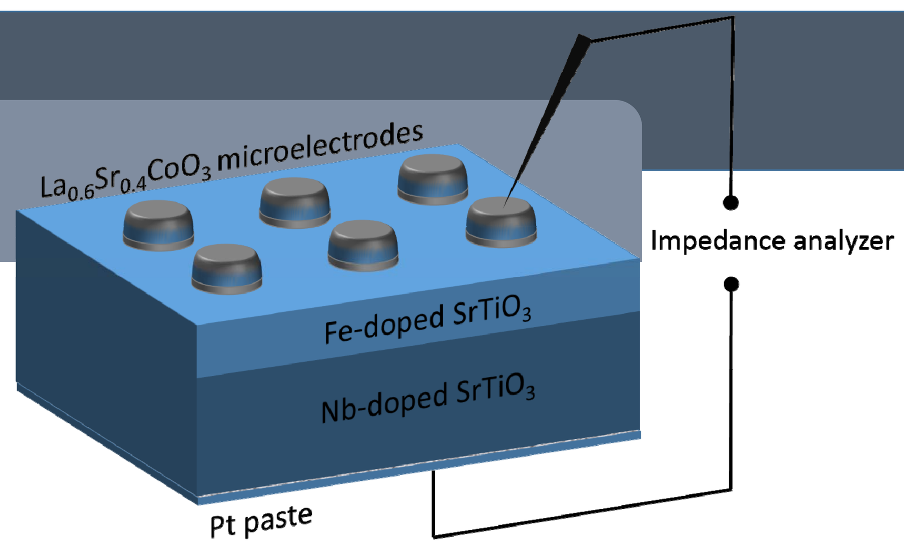


**Fig. 1.** Sketch of the sample set-up

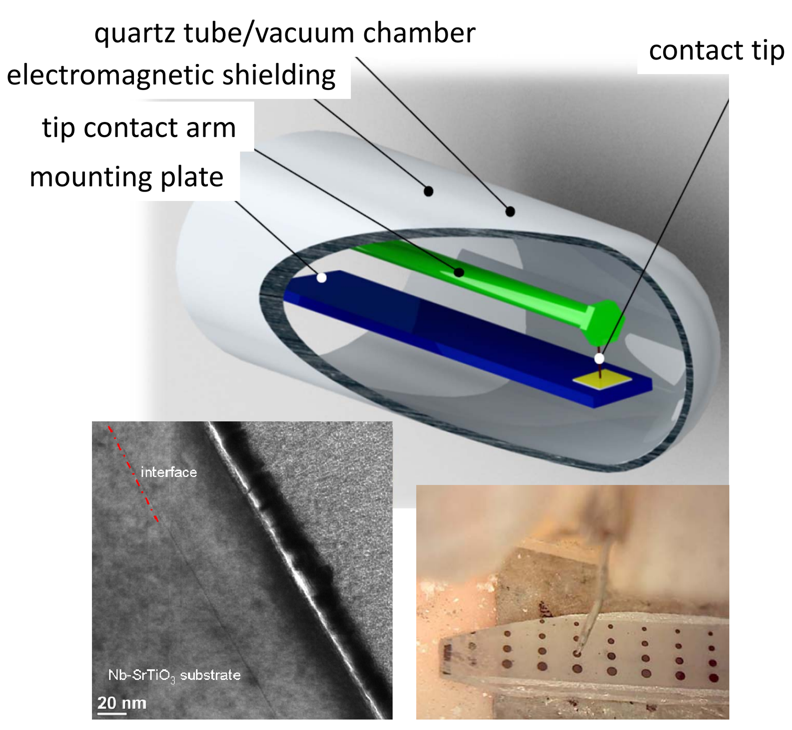


**Fig. 2a.** Measurement set-up, **b.** TEM image of a thin layer on Nb:STO, **c.** contacted sample

## U-I CHARACTERISTICS

Basically we can distinguish between two different measurement modes:

- Slow U-I curves (one cycle takes several hours)
- Fast U-I curves (measurement cycle in the range of seconds)

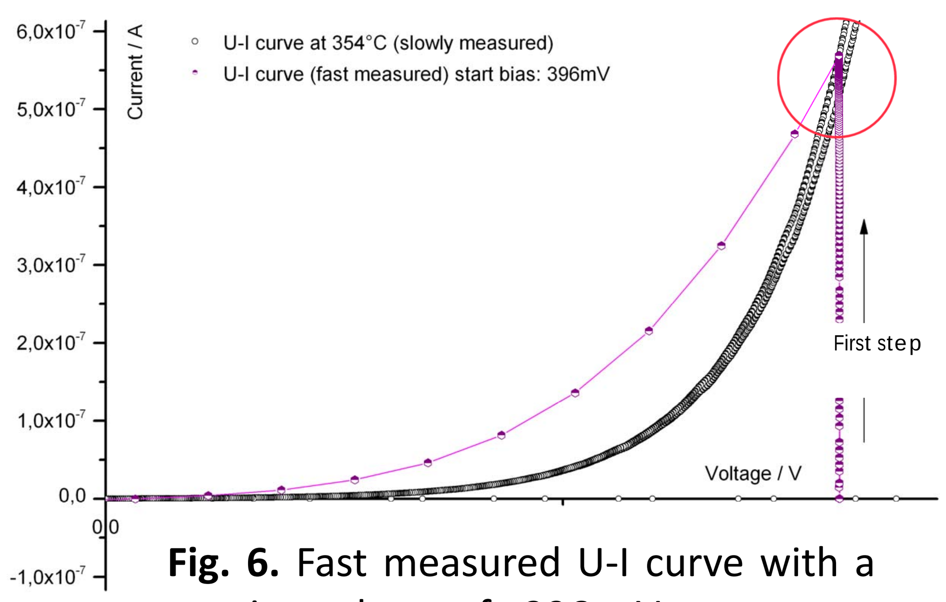


**Fig. 6.** Fast measured U-I curve with a starting voltage of +396 mV

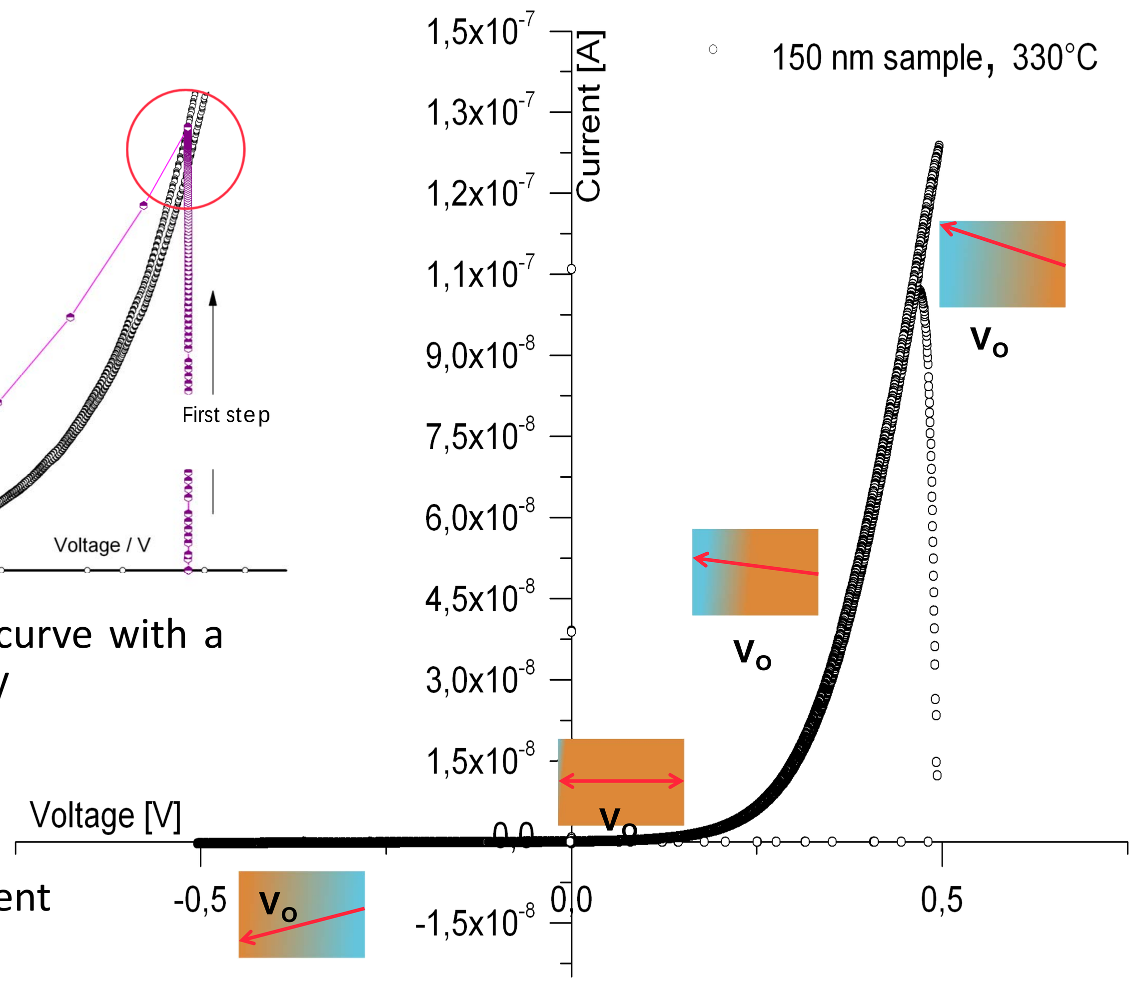


**Fig. 5.** Slowly measured U-I curve, building up an oxygen vacancy gradient

The non-linear behavior observed for the slowly conducted curves can be attributed to a continuous change in the oxygen vacancy distribution. (see Fig. 5. stoichiometry polarization)

For the fast measured curve a particular bias voltage is applied for a certain time (reaching steady state condition in the first step – see Fig.6.). After this initial step the fast measurement is performed, resulting in a curve that differs from the slowly measured curve. This behavior was found for the anodic as well as for the cathodic bias regime.

## IMPEDANCE MEASUREMENTS

The relation between resistance and film thickness (see Fig. 3.) revealed that the „bulk" resistance is measured. The applied bias voltage results in a resistance change of the high frequency arc. In addition a second feature (low frequencies) can be observed under bias. (see Fig. 4.)

An inductive loop is found for the anodic bias, and the cathodic bias regime is characterized by a second semicircle.

Parameters for the electrochemical characterization: T= 325-700°C, frequency range= $10^6$ Hz - $10^{-2}$ Hz, bias voltage= -/+500 mV

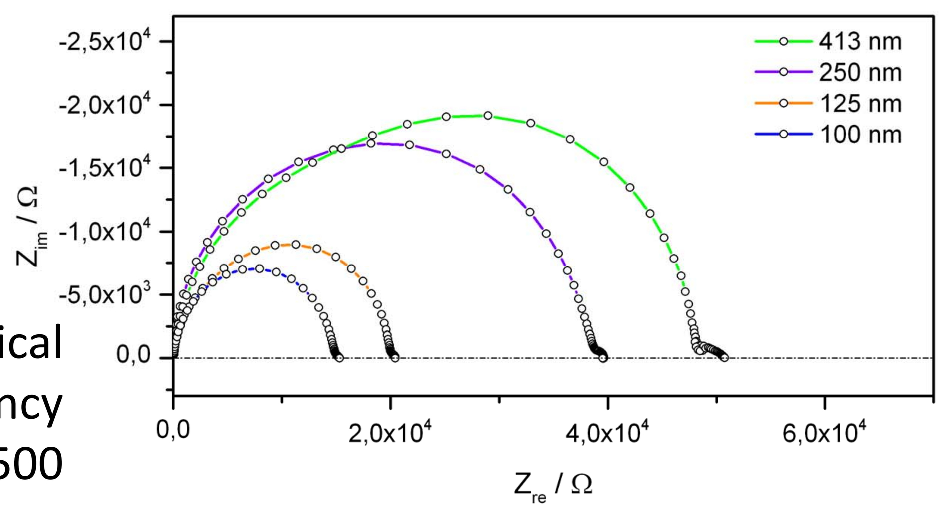


**Fig. 3.** Thickness dependent EIS measurements

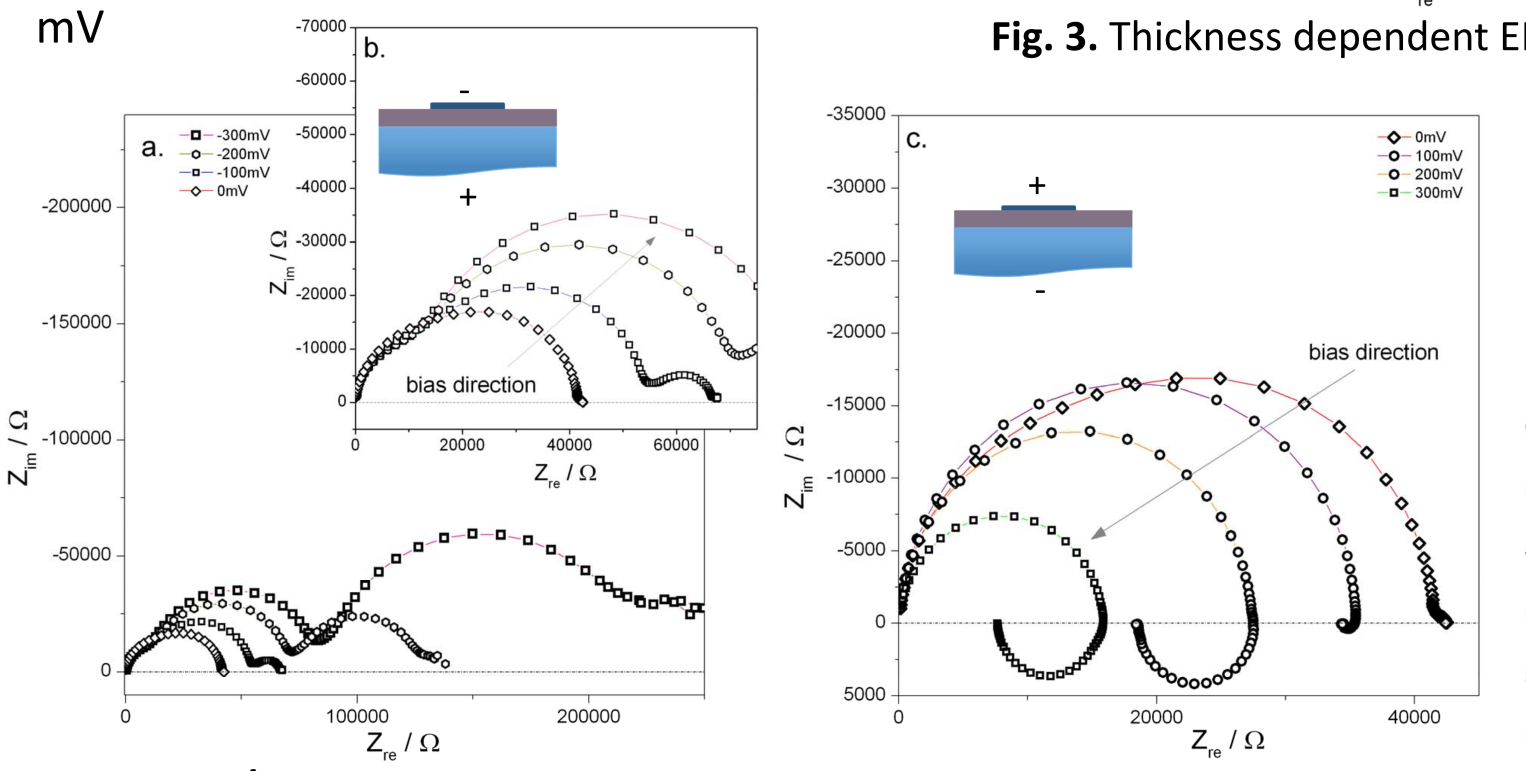


**Fig. 4a/b.** Impedance spectra recorded under cathodic bias, **c.** and under anodic bias

## RELATION – EIS AND U-I CURVES

Comparing the slopes of fast and slowly measured U-I curve in the working point of +400 mV and -400 mV (see Fig.7. and Fig.8.):

**Anodic case** – the slope of the fast measured curve is smaller, corresponding in a higher differential resistance. As a consequence in impedance spectra the larger resistance is obtained for intermediate frequencies.

**Cathodic case** – the inverse situation is observed, steeper slope of the fast measured curve correlates with a smaller differential resistance. Therefore, in impedance data the smaller resistance can be found for intermediate frequencies.

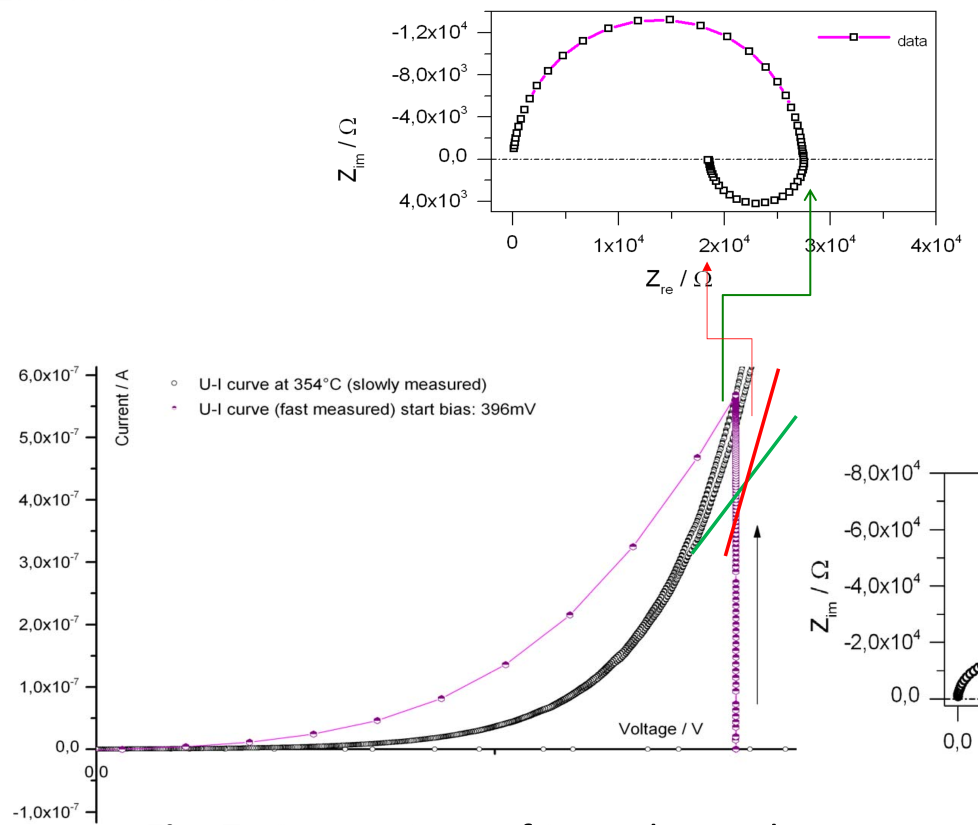

**Fig. 7.** Comparison of Impedance data with U-I curves (slow and fast) – anodic regime

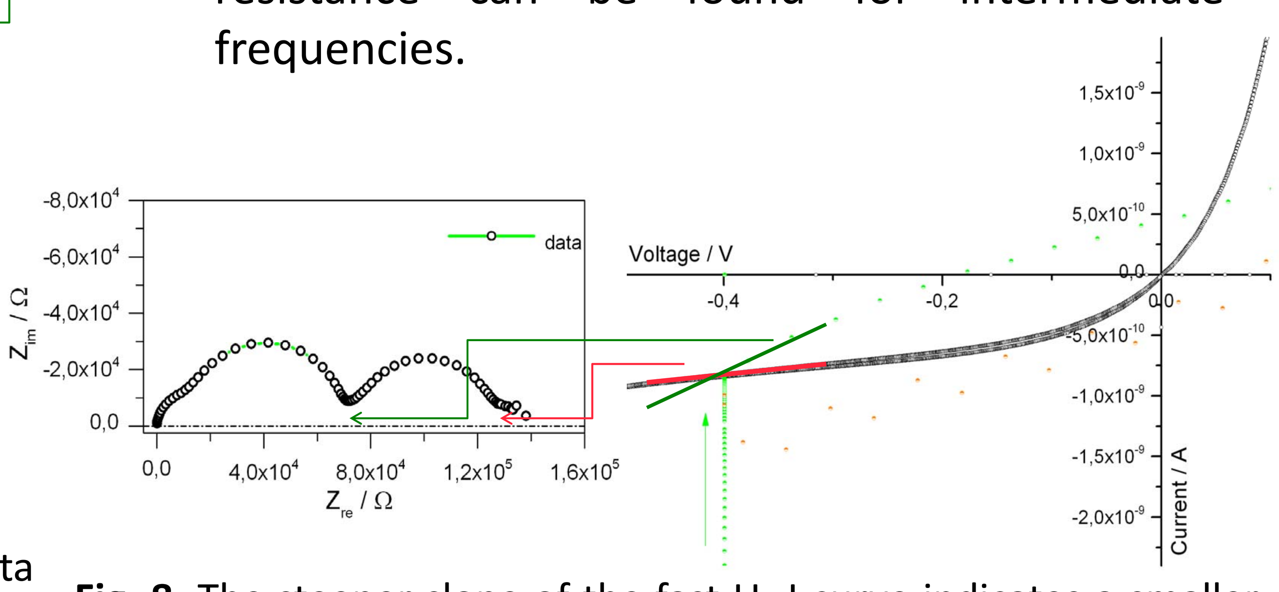

**Fig. 8.** The steeper slope of the fast U- I curve indicates a smaller differential resistance resulting in a second semicircle – cathodic regime

## CONCLUSION

- **Fe:STO thin films show reproducible temperature and bias dependent impedance results**
- **The phenomena are not electrode effects which could be confirmed by thickness dependent measurements**
- **Inductive loops and second semicircle are a trace for ion motion in the investigated thin Fe:STO films:**

**At low frequencies the vacancy distribution changes in phase, at intermediate frequencies ions cannot follow the applied ac voltage**

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