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Electrical characterization and extraction of activation energies of the defect states in the LaAlO₃/SrTiO₃ heterostructure

Yoann Lechaux^{1,a)}, Yu Chen², Albert Minj³, Florencio Sánchez², Gervasi Herranz², Laurence Méchin¹ and Bruno Guillet¹

Abstract

In this work, we study the electronic properties of defects in the LaAlO₃/SrTiO₃ heterostructure, which is known to host a high mobility two-dimensional electron gas (2DEG) at the interface. This 2DEG also shows photoconductance, which could be related to defects that act as deep centers trapping and releasing carriers by interaction with light. This phenomenon has raised an interest for the identification of deep energy levels in the LaAlO₃/SrTiO₃ heterostructure. We have studied the defect state properties using electrical characterization such as capacitance-voltage (C-V), current-voltage (I-V) measurements and Deep-Level Transient Fourier Spectroscopy (DLTFS). From C-V and I-V analysis, a hysteresis was observed indicating an effect of mobile charges in the LaAlO₃. Using DLTFS we identify three defect states located at around 0.17 eV below conduction band and at 0.23 eV and 0.26 eV above the valence band. These defect states were attributed to defects in SrTiO₃ such as strontium vacancies or titanium vacancies. We identify a fourth defect state having an energy of about 0.69 eV below the conduction band that could be related to oxygen vacancies in LaAlO₃ or in SrTiO₃. In addition, the observation of an effect of the electric field with DLTFS indicated that oxygen vacancies might be involved in Fowler-Nordheim or trap-assisted tunneling through the LaAlO₃ the layer.

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Oxide-based peroskite heterostructures show a wide range of physical properties¹ such as ferroelectricity, ^{2,3} superconductivity ⁴⁻⁶ or ferromagnetism, ^{7,8} which make them appealing for the development of multifunctional materials for a wide range of applications⁹⁻¹². One particularly interesting example is that of LaAlO₃/SrTiO₃ heterostructures which host two-dimensional electron gase (2DEG)⁶ with strong Rashba spin-orbit coupling^{13,14} and large spin-to-charge conversion, ¹⁵ as well as two-dimensional superconductivity, with transition temperature around 300 mK^{16,17}. Other applications include their use as channel materials in transistors^{12,18} or exploit the tunability of the 2DEG by electrostatic gating, ^{16,17,19–21} or by light as in persistent photoconductance^{22–25}. In this regard, we have demonstrated recently that the carrier population of the LaAlO₃/SrTiO₃ interface can be increased or depleted using light pulses with different wavelengths,²² a property which could be potentially useful to mimic spike-timing-dependent plasticity using the 2DEG as artificial synapses stimulated by optical pulses with duration down to microsecond. We proposed that this unusual photoconductance could be explained by the presence of defects in the LaAlO₃/SrTiO₃ heterostructure, particularly to the so-called DX-centers, ²⁶ which would act as trapping centers that capture and release carriers by interacting with light.²² Furthermore, a longstanding question is the charge density inferred from Hall measurements, which is about one order of magnitude smaller than the value expected from the polar discontinuity model.^{27–30} Related to this issue, it has been proposed that polar discontinuity triggers the spontaneous formation of some defects³¹ that could be trapping centers in the LaAlO₃/SrTiO₃ heterostructure related to point defects³² which would induce electron accumulation at the interface³³ with a depth of few nanometers.³⁴ Moreover, the characterization of defect states is a central piece of research for applications of semiconductors in electronic devices.³⁵

These observations raise the interest of identifying trapping centers in the LaAlO₃/SrTiO₃ heterostructure. Several works have reported defect state properties in LaAlO₃³⁶⁻³⁸ or in SrTiO₃.³⁹⁻⁴¹ Regarding bulk LaAlO₃, some studies from K. Xiong *et al.* have been performed to estimate the energies of the defect states using the density functional theory (DFT).^{36,37} Their calculations showed that oxygen interstitial states such as Io⁻ lie at about 0.5 eV above the valence band while Io⁻² lie in the valence band. Moreover, they calculated the energy of the oxygen vacancy states, such as Vo⁰ to be about 0.8 eV below conduction band while Vo⁺ and Vo⁺² states lie at about 0.6 eV and 0.55 eV, respectively, below the conduction band. In the context of LaAlO₃/SrTiO₃ interfaces, L. Yu *et al.*³² reported on different defect states, including Ti_{Al} antisites in the LaAlO₃ layer, Al_{Ti} antisites at the interface and oxygen vacancies at the surface of LaAlO₃ that may cause the 2DEG.³¹ Moreover, oxygen vacancies in the LaAlO₃/SrTiO₃ heterostructure were studied using density functional theory^{42,43} and were experimentally related to the enhancement of electrical conductivity.⁴⁴

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Regarding SrTiO₃, A. K. Ghosh et al. evidenced four defect states from 0.26 eV to 0.43 eV above valence band³⁹ by thermally stimulated current method. Using the same technique, B. Wang et al. found five energies from 0.15 eV to 0.86 eV.41 Osawa et al. reported an activation energy of about 0.06 eV above valence band for strontium vacancies.⁴⁵ Y. Seo et al. made DLTS measurement to characterize Cr-SrTiO₃ on silicium substrate but all the identified traps were related to silicium.⁴⁶ F. A. Selim et al. used the positron annihilation lifetime spectroscopy to find the defect lifetimes in SrTiO₃⁴⁷ (however, this technique cannot give the defect state energies). On the other hand, T. Tanaka et al. calculated defect states energies from 0.11 eV to 0.20 eV above valence band for strontium vacancies, from 0.26 eV to 0.84 eV above valence band for titanium vacancies and from 0.08 eV to 0.21 eV below conduction band for oxygen vacancies. 48 A. Janoti et al. calculated the defect states in the n-doped SrTiO₃ and they found energies of about 0.51 eV below conduction for oxygen vacancies (V₀+), from 0.20 eV to 0.30 eV above valence band for strontium vacancies and from 0.58 eV to 0.81 eV above valence band for titanium vacancies depending on charge state.⁴⁹ Besides, D. Manger et al. reported oxygen vacancy energies from 0.8 eV to 1.2 eV below conduction band in the SrTiO₃ films grown by atomic layer deposition.⁵⁰ B Liu et al. calculated the energies for interstitial defect states in SrTiO₃ and showed a large variety of defect states.⁵¹

As pointed out above, defect formation and detection has been studied in LaAlO₃/SrTiO₃ heterostructures. A few experimental works have reported on defect state properties in the LaAlO₃/SrTiO₃ heterostructure, yet some fundamental aspects, such as their activation energy, have not been identified. For example, A. Kalabukhov *et al.* observed the impact of oxygen vacancies on the electrical properties using cathode and photoluminescence techniques.⁵² N. Palina *et al.* showed oxygen orbital states using X-ray spectroscopies,⁴⁴ A. Gloter *et al.* observed the distribution of point defects at the interface using scanning transmission electron microscopy⁵³ and G. Yuan *et al.* reported on the behavior of oxygen vacancies at the LaAlO₃/SrTiO₃ heterostrucutre using positron annihilation spectroscopy.⁵⁴ In our work we extracted defect properties that cannot be accessed using these methods, such as the activation energies and capture cross-sections of the defect states using Deep-Level Transient Fourier Spectroscopy (DLTFS).^{55,56}

All the measurements were performed on a isolated capacitor device based on the LaAlO₃/SrTiO₃ heterostructure, which was fabricated through a sequential deposition process combined with photolithography. First of all, pulsed laser deposition (PLD) was exploited to deposit an amorphous ~60 nm thick LaAlO₃ layer at room temperature on substrate SrTiO₃ with annular geometry defined by photolithography and lift-off. Afterward, an epitaxial LaAlO₃ layer with thickness of 12 unit cell (around 4.6 nm) was grown on the annularly patterned SrTiO₃ using PLD at 650°C and oxygen partial

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pressure about 10⁻⁴ mbar. At the end of the deposition, we cooled down the sample in the oxygen rich atmosphere to minimize the formation of oxygen vacancies. Subsequently, the photolithography was carried out again to define top electrodes position on the center of the annulars, followed by the gold top contact deposition using evaporation and lift-off. Thus, a capacitor structure is formed (Au/LaAlO₃/SrTiO₃) where the 2DEG at the LaAlO₃/SrTiO₃ interface, used as bottom electrode, was contacted by ultrasonic aluminium wire-bonding. FIG. 1 shows the heterostructure and the measurement sketch (FIG. 1(a)) and a top view of the circular devices (FIG. 1(b)) among which the 800 µm diameter size capacitor was measured. Details on the setup description and on the method for the Deep-Level Transient Fourier Spectroscopy analysis can be seen in supplementary material.

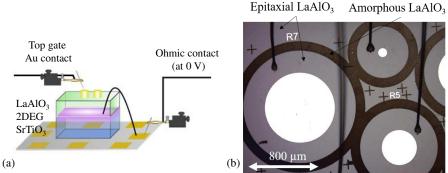


FIG. 1. (a) Schematics of the LaAlO₃/ SrTiO₃ heterostructure showing a 2DEG at the interface (in purple color) and (b) a top view of the fabricated circular devices.

The capacitance-voltage (C-V) and the current-voltage (I-V) characteristics were measured at room temperature with an AC signal at 1 MHz. Two voltage sweeps in opposite directions, from positive to negative values (1) and from negative to positive values (2), were performed one after the other to observe the impact of the defect states on the static characteristics, such as hysteresis effects.

Condidering an n-type 2DEG at the LaAlO₃/SrTiO₃ interface, the heterostructure would behave as a p-metal-oxide-semiconductor (MOS) capacitor. At high frequency, the capacitance generally increases with the positive voltage from the depletion regime to the accumulation regime. FIG. 2(a) shows the capacitances from parallel (FIG. 2(c)) and series (FIG. 2(d)) RC models as a function of the voltage at room temperature. A large difference between both capacitances was observed. Contrary to the parallel capacitance C_P , the series capacitance C_S is increasing with the voltage from about 12 pF at -5 V to about 68 pF at 1 V. This trend indicates that the series RC model might be more relevant,

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However, in these models, the capacitance is drastically reduced at high frequency (> 10 kHz) as mentioned in previous works.^{57–59} Conversely, S.K. Kim *et al.*⁵⁷ proposed two different models for the depletion and accumulation regimes. In accumulation, the model is a series combination of a resistance and a parallel *RC* circuit (FIG. 2(e)). The series resistance *Rs* comes from the 2DEG in the SrTiO₃ and the parallel *RC* circuit corresponds to the LaAlO₃ layer. In depletion, the model becomes more complex and it is a series combination of the 2DEG resistance and two parallel *RC* circuits corresponding to the SrTiO₃ and to the LaAlO₃ layers.

However, a few observations can be made from the *C-V* characteristics in FIG. 2(a). Even though

although no saturation of the series capacitance was observed for the range of analyzed voltages.

However, a few observations can be made from the C-V characteristics in FIG. 2(a). Even though the dielectric constant for LaAlO₃ cannot be extracted reliably, we can clearly observe the two regimes. More specifically, accumulation of the n-2DEG is observed for voltages above -1 V, while its depletion, associated with the reduction of C_S , is seen for voltages below -1 V, up to a saturation for complete depletion. Besides, a hysteresis is observed for voltages between 0.5 V to -3.5 V with a maximum difference of about 0.45 V between the two sweep at $C_S = 40$ pF. This voltage shift may hint towards charge transfer through defects under the influence of electric field, and such charged defects could be possibly linked to oxygen vacancies in LaAlO₃. S.K. Kim *et al.* and C.W. Bark *et al.* related this phenomenon to an electric-field induced redistribution of the oxygen vacancies in the LaAlO₃ up to the interface which leads to a switchable hysteretic response 61,62 as observed on the I-V characteristics.

FIG. 2(b) shows the *I-V* characteristics at room temperature. We observed different behaviors between the two voltage sweep directions, mainly for negative gate voltages. For sweep (1) and for voltages between 1.5 V and 0 V, the current is decreasing while the voltage decreases. Then, in the bias range between 0 V and -5 V, the current initially increases with the applied negative bias up to -2.5 V and decreases until -5 V forming a hump in the *I-V* characteristics as reported by S.K. Kim *et al.*.⁶² This observed hump indicates a sudden increase of the resistance for biases below -2.5 V. Then for sweep (2) from -5 V to 1.5 V, no hump is observed and the current level starts at the same level as that for the previous sweep. Similar features in *I-V* curve were previously observed and were attributed to oxygen vacancy drift⁶⁰ and might be related to the hysteresis observed in the *C-V* curve. This hump on the *I-V* curve must be associated to a drop in current due to full depletion of the *n*-2DEG which induced an increase of the resistance beneath of the electrode.

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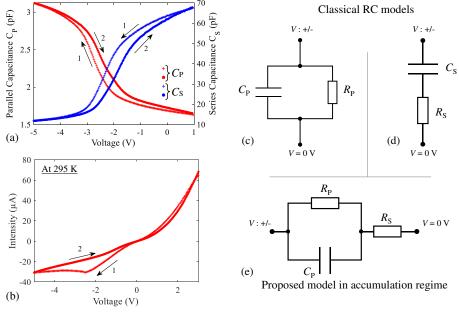
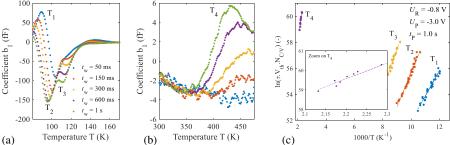


FIG. 2. (a) Capacitance-voltage and (b) current-voltage characteristics for two different voltage sweep directions, at room temperature. Different RC models: (c) parallel circuit, (d) series circuit and (e) the combination of a parallel and series circuits.

In order to evaluate the defect state properties, DLTFS measurements were performed for various time windows t_w . Fig. 3 shows the DLTFS spectra for $t_w = 49$ ms, 146 ms, 293 ms, 586 ms, 977 ms while $U_R = -0.8$ V, $U_P = -3.0$ V and $t_P = 1.0$ s. We observed two main different regions in the spectra, namely, one for temperatures below 150 K and another for temperatures above 300 K. For temperatures below 150 K, we found three significant peaks, one positive (T₁) and two negative peaks (T₂ and T₃), see Fig. 3(a). For temperatures above 300 K, a significant positive peak (T₄) was observed, see Fig. 3(b). We noticed an increase of T₂, T₃ and T₄ amplitudes with increasing t_w . In contrast, the T₁ amplitude decreases when t_w increases, which indicates a much lower relaxation time for T₁ in comparison to T₂, T₃ and T₄. Sesides, the temperatures of the DLTFS peaks shift toward lower values with increasing t_w , what is in agreement with a decrease of the emission rate. From the Arrhenius diagram (Fig. 3(c)), we first extracted the activation energy t_w . For the analysis we considered that the LaAlO₃ and SrTiO₃ are n-type majority materials. The different extracted energies are about 0.17 ± 0.03 eV below conduction band for T₁, 0.23 ± 0.03 eV above valence band for T₂, 0.26 ± 0.06

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eV above valence band for T_3 and 0.69 ± 0.03 eV below conduction band for T_4 . T_1 , T_2 and T_3 show shallow energy levels comparable to the ones observed in $SrTiO_3^{39,41,45,48,49}$ while T_4 shows a deeper

Fig. 3. DLTFS spectra of the LaAlO₃/SrTiO₃ heterostructure for various time windows t_w and for $U_R = -0.8$ V, $U_P = -3.0$ V and $t_P = 1.0$ s, at (a) low and (b) high temperature regions. (c) The Arrehnius diagram extracted for all the defect states.

To determine the origin of the observed defect states in the LaAlO₃/SrTiO₃ heterostructure, DLTFS measurements were carried out for various reverse and pulse voltages while keeping other parameters constant. These two variations allow, for example, to distinguish interface and bulk traps.^{64,65} In the following, we focus on the study of the T₄ trap center at high temperature—which, as we argue below, might be related to oxygen vacancies in the LaAlO₃-layer. A similar analysis for the T₁, T₂ and T₃ traps is more complicated, due to the overlap of the corresponding peaks.

Fig. 4(a) shows the DLTFS spectra for reverse voltages U_R from -0.4 V to -1.0 V, keeping other parameters fixed at $U_P = -3.0$ V, $t_P = 1$ s and $t_W = 977$ ms. The data indicate an increase of the T₄ peak amplitude and a shift to lower temperatures when the reverse bias is changed from -0.4 V to -1.0 V. These both observations indicate that the trap states associated with the T₄ peak cannot be related to band-like defect states at the interface between LaAlO₃ and SrTiO₃, otherwise a decrease of the peak amplitude should be observed. On the other hand, an increase of the reverse bias from -0.4 V to -1.0 V should enhance the upward band bending close to the interface at the SrTiO₃ side (Fig. 4(c)). This would be accompanied by a narrowing of the 2DEG and a lowering of the trapped electron density at the interface associated to the bulk oxygen vacancies in the SrTiO₃, which should manifest as a

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decrease of the DLTFS amplitude. However, this contradicts the increase of the peak amplitude with U_R observed in the DLTFS spectra in Fig. 4(a). We therefore conclude that the T₄ peak might not be related to bulk oxygen vacancies in SrTiO₃.

To explain the experimental data, we can invoke other phenomena based on trap assisted tunneling, Folwer-Nordheim tunneling or Poole-Frenkel effect. Indeed, an increase of the reverse bias from -0.4 V to -1.0 V should induce an increase of the electric field in the LaAlO₃ layer. In turn, this would be associated with a lowering of the effective barrier thickness between the oxygen vacancy levels located within the LaAlO₃ layer or in the bulk SrTiO₃ and the Au gate contact. For high negative pulse voltages, the levels of oxygen vacancies in both SrTiO₃ and LaAlO₃ are below the Au Fermi level. Thus, electrons can be transfered from the gate contact to the LaAlO₃ oxygen vacancies (Fig. 4(c)) by trap assisted tunneling, as similarly reported by K. Song *et al.* in their recent experimental work⁶⁶. Moreover, these electrons can also be transferred up to SrTiO₃ oxygen vacancies by Fowler-Nordheim tunneling. As the effective barrier thickness diminishes when the voltages goes toward -1.0 V, a higher trapped electron density will result in an increase of the DLTFS peak amplitude. Therefore, the variation of the T₄ peak with *U*_R would be in agreement with the assumption of either trap assisted tunneling or with Fowler-Nordheim tunneling through LaAlO₃.

In order to validate this assumption, DLTFS measurements were carried out for various pulse voltages U_P between -1 V to -3.5 V at U_R = -0.95 V, t_P = 1 s and t_W = 977 ms (Fig. 4(b)) to maximise the DLTFS signal. No shift is observed when changing U_P = -1 V to U_P = -3.5 V indicating that the emission rate is quasi constant, so the T₄ peak might be related to bulk traps, thus supporting the view that the T₄ peak should be related to defects spread over the SrTiO₃ or LaAlO₃ layers. Note that in the case of band-like states, an increase of the pulse voltage would induce a shift of the DLTFS peak accompanied by an increase of its amplitude. As no shift is observed, it means that the traps are not just located at the interface but distributed across the SrTiO₃ or the LaAlO₃ layers. Besides, we found an increase of the DLTFS peak amplitude with the increase of pulse voltage from -1.0 V to -3.5 V which is related to the increase of trapped charge density. This observation is consistent with the adscription of tunnel effects through LaAlO₃ although it is difficult to distinguish whether the T₄ peak is related to oxygen vacancies in the LaAlO₃ layer or in the SrTiO₃ substrate.

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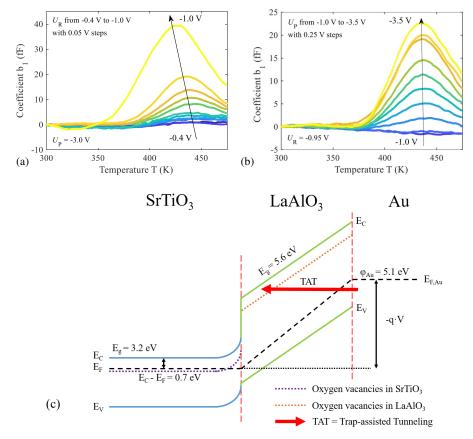


Fig. 4. (a) DLTFS spectra of the LaAlO₃/SrTiO₃ heterostructure for various reverse voltages U_R from -0.4 V to -1 V, keeping fixed $U_P = -3.0$ V, $t_P = 1.0$ s and $t_w = 977$ ms. (b) DLTFS spectra for various pulse voltages U_P from -1 V to -3.5 V at $U_R = -0.95$ V, $t_P = 1.0$ s and at $t_w = 977$ ms. (c) Schematic of the band diagram of the Au/LaAlO₃/SrTiO₃ heterostructure for a gate voltage of -3.0 V.

After analysing the T_4 peak, we now turn the discussion of the other peaks observed in the DLTFS spectra. Comparing the extracted activation energies in the previous literature^{48,49,51} with our observations, the T_1 peak energy close to the conduction band could be associated to interstitial titanium defect I_{T_1} in its neutral state. I_2 and I_3 peaks might be related to defect states in the I_3 such as strontium vacancies I_3 or antisites I_3 . Also, the extracted energy levels for I_3 to I_3 close to the valence band, lower than I_3 eV, could not be related to titanium vacancies I_3 as it shows deeper energy-level.

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In summary, we studied the defect state properties of the in a LaAlO₃/SrTiO₃ heterostructure. We detected four defect states and their energies and capture cross sections were extracted. Three defect states were attributed to SrTiO₃ showing shallow energy levels at 0.17 eV below the conduction band and from 0.23 eV to 0.26 eV above the SrTiO₃ valence band. A fourth defect was attributed to oxygen vacancies in the SrTiO₃ or LaAlO₃ layers with an energy of about 0.69 eV below the conduction band. More importantly, we demonstrated the DLTFS can be a reliable method, showing similar results in comparison to other experimental techniques. With this in view, we propose that DLTFS could provide useful insights into some problems that are still under debate. For instance, it is kown that carrier densities at the LaAlO₃/SrTiO₃ interface are significantly lower than the ones expected from the polar catastrophe model⁴, while larger values are observed at some RTiO₃/SrTiO₃ (R = rare earth) interfaces⁶⁷. In this regard, DLTFS could provide a means to explore whether the reduction of trapping centres could be correlated with the transport properties of those interfaces.

Supplementary material

See supplementary material for the setup description and the method for the Deep-Level Transient Fourier Spectroscopy analysis.

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Data availability

The processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

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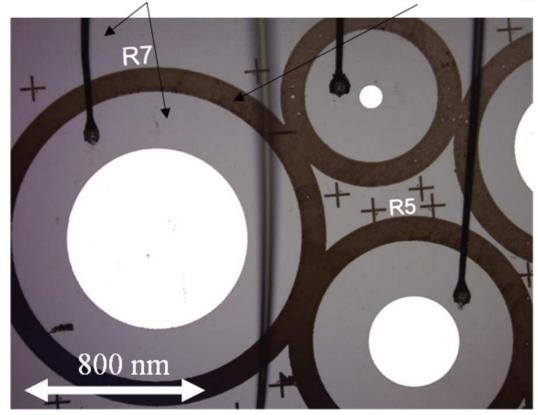
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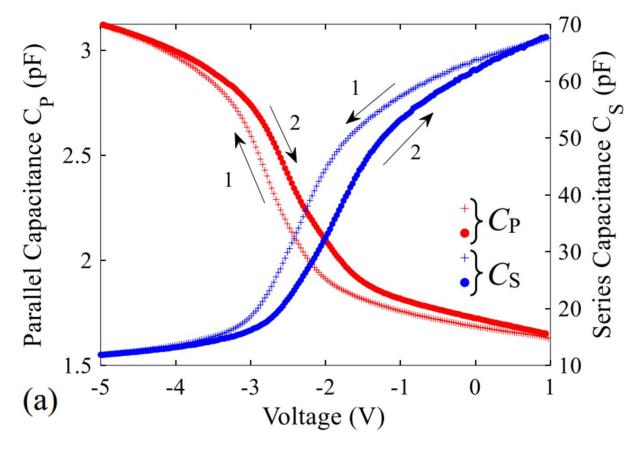
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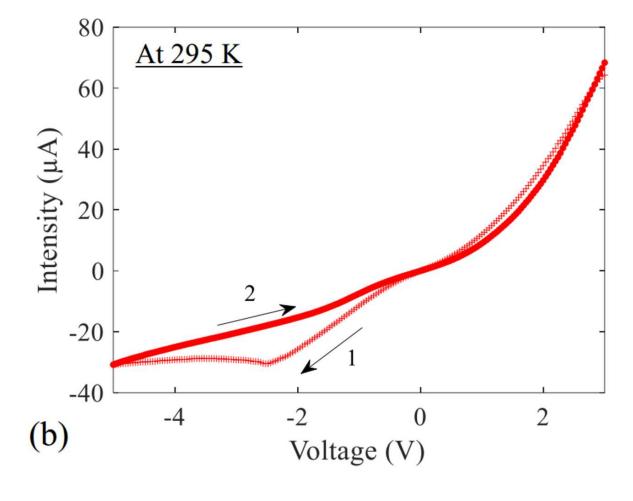
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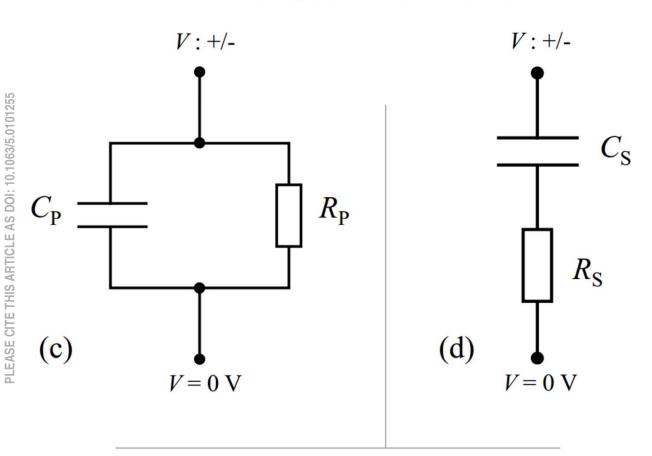
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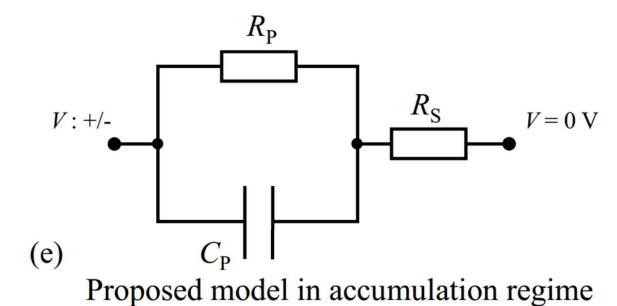


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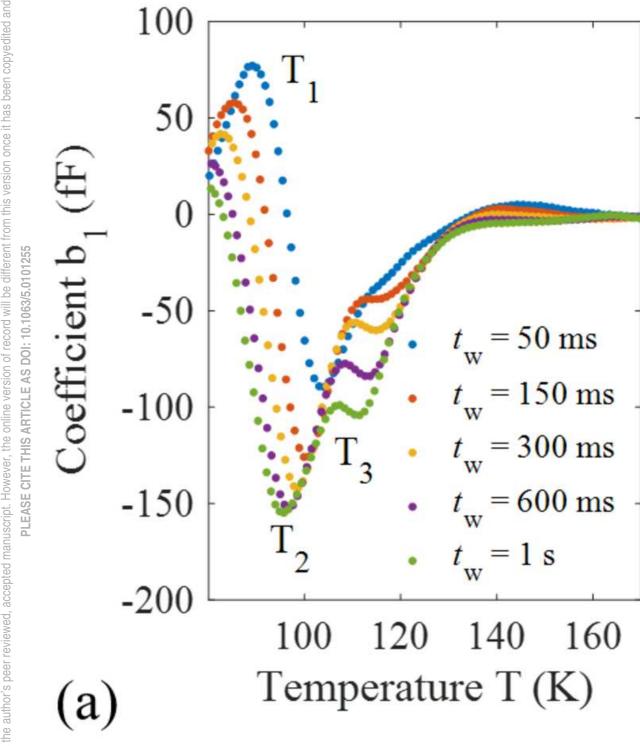


Classical RC models

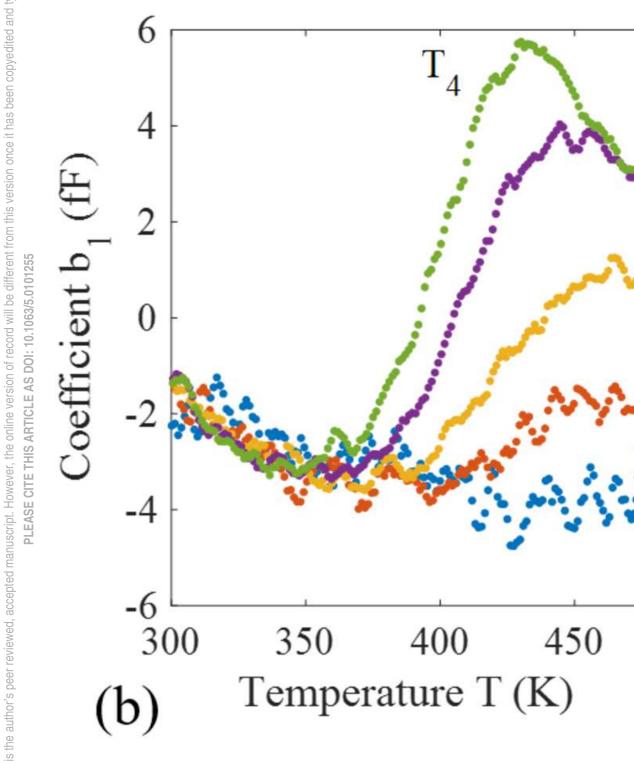




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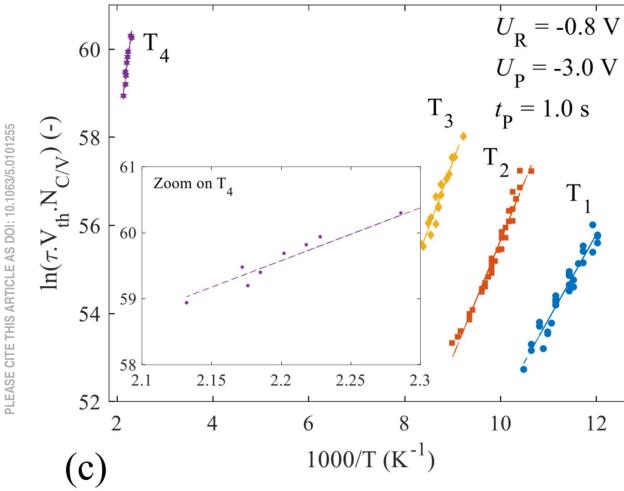
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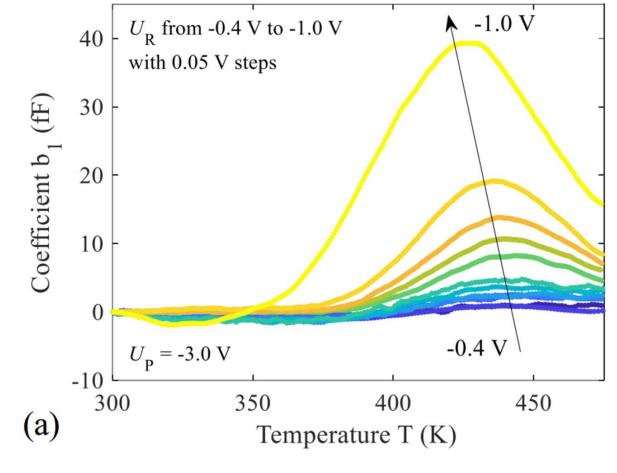


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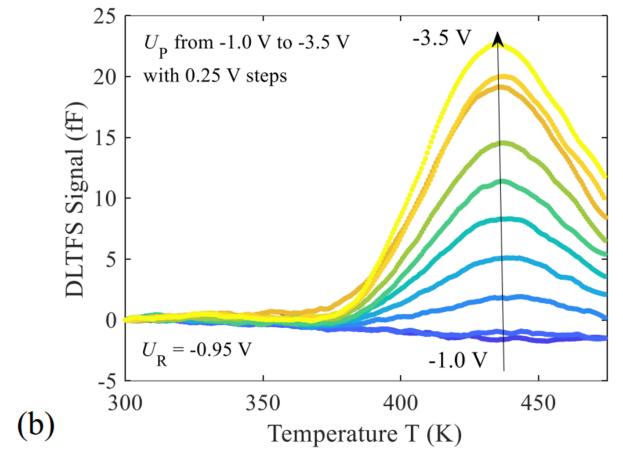
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