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Unusual low-frequency noise in irradiated $SrTiO_3$

R. Guerrero,[1] A. Solignac,[1] M. Pannetier-Lecoeur,[1] Y. Apertet,[2] P. Lecoeur,[2] and C. Fermon[1]
[1]*DSM/IRAMIS/SPEC, CEA Saclay, 91191 Gif sur Yvette Cedex, France*
[2]*Institut d'Electronique Fondamentale, Université Paris-Sud–CNRS, 91405 Orsay, France*
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We report the transport and noise properties as function of temperature and electric field of Ar^+ irradiated $SrTiO_3$ exhibiting at its surface a high-mobility electron gas. This system exhibits metallic properties with low-charge densities allowing mobility measurements. The low-frequency $1/f$ noise presents a linear dependence with the mobility. This nonintuitive variation requires a rewriting of the commonly used Hooge model.

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I. INTRODUCTION

Oxide-based electronics is a new emergent field that shows a great variety of new applications, covering, for example, magnetism or high critical-temperature superconductivity applications. Among them $SrTiO_3$ (STO) is one of the most interesting systems. STO is a band insulator with a perovskite-crystallographic structure. It has been widely used as substrate for the deposition of many perovskite materials, for instance, manganites or cuprates. Recently, field-effect transistors have been developed based on this material.[1–4] Besides that, the possibility of controlling the growing processes of STO and its surface has enabled the development of a high-mobility electron gas in the interface of STO and $LaAlO_3$ (LAO).[5] Even if the origin of this phenomenon is unclear[6–8] the properties of the interface are very interesting and promising for new potential applications.

It has been published that following an irradiation procedure the STO surface becomes conductive,[9–11] exhibiting similar properties to STO/LAO interface or bulk-reduced STO.[12–15] The irradiation of STO produces oxygen vacancies without modifying the perovskite structure[11] and due to the low penetration depth of the ions into the material, it produces a conducting layer with a thickness of the order of nanometers.[9,11] For practical applications of these highly conductive films, the knowledge of their noise as function of frequency is essential.

We have measured a surprising strong increase in the noise with the decrease in temperature. In order to understand this behavior, we have measured resistance and Hall Effect in the same samples and then determined the carrier density and their mobility.

II. SAMPLE PREPARATION

Samples were prepared using STO substrates from Crystec GmbH with dimensions 5 mm×5 mm×0.5 mm. On the top of the substrate we define several lines of different widths: 10, 20, 50, and 100 μm using standard photolithography. After that, we irradiate the substrates in order to define lines of conductive STO. The irradiation was done using an Ar plasma accelerated by 0.7 kV, provoking a current density of 1.28 mA/cm^2 incident on the surface. A cap layer of Si_3N_4 was deposited *in situ* to avoid any reoxidation of the sample. Finally electrical contacts composed of Ta(10 nm)/Cu(150 nm)/Ta(10 nm) were sputtered in a classical hall bar geometry (inset of Fig. 1). All the samples were contacted to a sample holder through Al wire bonding in order to perform their characterization.

We could estimate the thickness of the conductive layer using the empirical equation used in[9,10]

$$L = 1.1E^{2/3}W/[\rho(Z_i^{1/4} + Z_t^{1/4})^2], \qquad (1)$$

where ρ is the density of the substrate, Z_i is the atomic number of the Ar, and E is the energy of the Ar ions used during the etching, and W and Z_t are the atomic weight, in a.m.u., and number of the material, respectively, because STO is a compound we used and weighted average. The estimated thickness is about 10 nm.

These films are very sensitive to their exposure of oxygen at room temperature. It is for this reason that we cover the irradiated surface with a 25-nm-thick layer of Si_3N_4 *in situ* under vacuum just after the irradiation. We have not observed any variation in resistance or noise of our samples after 3 months in air at room temperature and after several temperature cycling from 4 to 300 K.

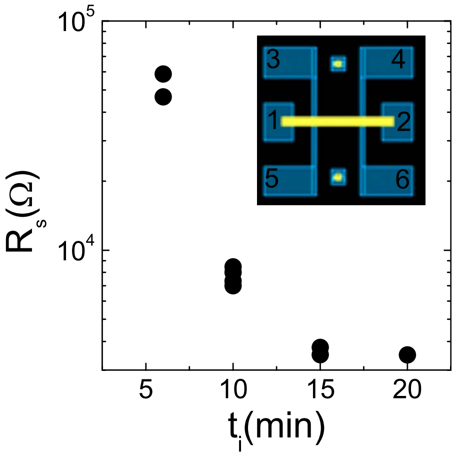

FIG. 1. (Color online) Sheet resistance at room temperature as a function of the irradiation time. The improvement of the R_s with the irradiation time might be related to an increment of the number of oxygen vacancies created. Inset: sketch showing the position of the contacts in the conducting STO line. Current is applied between contacts 1–2.

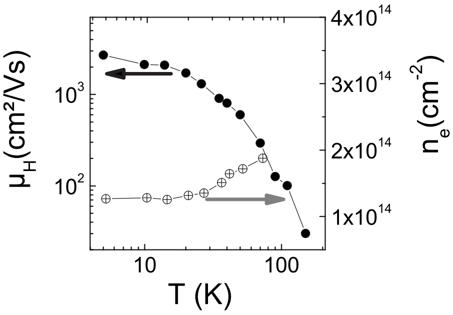

FIG. 2. Dependence of the mobility as a function of the temperature in a 50-μm-large sample irradiated during 10 min. The mobility increase monotonically up to about 10 K and then saturates. In the tested samples there is not a clear dependence on the Ar etching time and the low-temperature mobility. In the right axis is shown the dependence of the number of carriers per surface unit on the temperature for the same sample.

III. RESULTS

Resistance measurements were done using four probes method. We use a Keithley 6221 current source to bias our samples, the voltage was detected using a dc amplifier and an acquisition card. Hall-Effect measurement were done in a magnetic field up to 1 T using the same current source and a differential amplifier. In order to do the noise measurement we use an ac coupled low noise amplifier and a battery fed current source.

The irradiation process reduces the sheet resistance (R_s) at room temperature till a saturation value (Fig. 1). A similar R_s behavior was reported in Refs. 9 and 11. Although in all the cases the R_s behavior is qualitatively the same, there is no quantitative agreement. This difference may be related to the irradiation process and to the temperature of the substrate which is not well known.

All the studied samples exhibit metalliclike temperature dependence. The residual resistance in the samples is reached at $T < 10$ K. Whereas there is a clear correlation between the R_s at room temperature and the irradiation time (t_i) (see Fig. 1), the residual-resistance ratio, and thus the low-temperature resistance is not correlated with t_i. We think that the low-temperature resistance is very sensitive to a small amount of impurities or dislocations and they may vary from sample to sample.

From resistance and Hall-Effect measurements, we have deduced the surface density of carriers and their mobility (μ_H), see Fig. 2. The trend of μ_H in all the studied samples is similar to the observed in STO/LAO interface,[5] irradiated STO,[9–11] and reduced STO.[12–15] First an increase in μ_H is observed when temperature decreases, then the mobility saturates at a critical temperature. References 13 and 14 argued that this behavior in reduced STO is due to a crossover from a mobility controlled by impurities at low temperatures to a mobility controlled by optical phonons at high temperatures.

The number of carriers decreases with T, this behavior has been reported in reduced STO.[14] Considering a thickness of about 10 nm the order of magnitude of the carrier density is 10^{20} cm^{-3}, this value is about one order of magnitude less than the results obtained in irradiated STO (Ref. 9) and three orders of magnitude bigger than in bulk-reduced STO.[14]

The noise in the studied samples contains a white-noise part given by $\sqrt{4kTR}$ and a low-frequency $1/f$ noise. In all the measured samples the exponent of the power spectral density (PSD) was 1 or very close to this value. Even at low temperature the exponent remains close to the unity instead of presenting Lorentzian peaks, which are characteristics of random telegraph noise, for example, due to generation-recombination noise.[16]

The dependence of the PSD on the bias voltage (V) is proportional to V^2 in all the studied range of temperature. This kind of dependence is expected in the case of resistance fluctuations.[17] Well-known Hooge formula gives the spectral power of the low-frequency noise

$$S_V(f) = \frac{\alpha V^2}{Nf}, \tag{2}$$

where α is a phenomenological parameter which ranges from 4×10^{-6} up to 5×10^{-2} in metallic systems.[18] N is the total number of carriers involved the system.

In Fig. 3(a) we show the parameter α / n_e obtained in our experiments. Two main features can be observed in this figure, first a surprising increase in the noise when the temperature is decreased and second, a sharp peak at about 28 K.

Figure 3(b) gives α as function of the hall mobility and a linear variation is observed which has never been reported. The sharp peak at 28 K could be related to a structural-phase transition already detected between 35 and 55 K using different experimental techniques.[19,20] In this range of temperatures the STO is not exactly cubic, instead the crystallographic structure is distorted, leading to the formation of domains in the crystal. In our opinion at about 35 K the structure should be stable but probably the two different phases still coexists down to 28 K, thus leading to an increase of the scattering which yields the observed peak in noise. However this explanation is not very satisfactory and the question remains open. Another structural-phase transition happens at 105 K,[21] however in the presented results the noise does not exhibit any peak at this temperature.

In order to test the relation between mobility and noise, we have performed experiments where the temperature is maintained constant and an electric field is applied on the sample. We have applied a backgate voltage up to 250 V in several of the studied samples. The effect of the electric field in the irradiated STO is to modify slightly the number of electrons while the mobility is changed by a factor about 5. We have found the same linear dependence of the α parameter (Fig. 4). This measurement excludes a coincidence in the variation of the mobility and the α parameter.

In order to fit our data, we have to use the following modified Hooge formula:

$$S_V(f) = \frac{\alpha_m \mu V^2}{Nf}, \tag{3}$$

where $\alpha_m \approx 1.4$ V s/cm^2 and has the dimension of the inverse of a mobility in this particular sample. This value is not universal, although the relationship is the same in all the

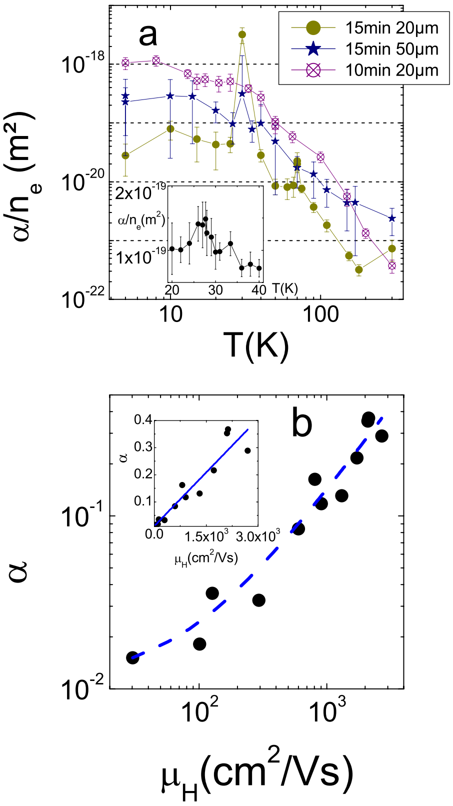

FIG. 3. (Color online) (a) The ratio between the Hooge parameter and the surface density of carriers is plotted as function of the sample temperature. Two features could be remarked: first the peak located at $T \approx 28$ K and increasing of the plotted parameter when T is diminished. The inset shows a zoom to the peak observed at about 30 K in a 50-μm-width sample irradiated 15 min. (b) α parameter versus the mobility of a single sample changing the temperature. The dashed line is a linear fit in linear scale, we use instead a logarithmic scale in order to make the plot more visible. The inset shows the same graph in linear scale.

studied samples, the constant α_m changes. This formula allows determining the effective noise of our system for the different irradiation times and as function of temperature and electric field.

IV. DISCUSSION

The origin of $1/f$ noise is still extensively discussed. A rather complete review of models and results is given in Ref. 18. In the first Hooge model, the noise is independent of the mobility. The simple argument behind is that the frequencies involved in the resistance fluctuations are much lower than the traveling time of electrons in the sample and hence there speed does not impact on the noise spectrum.

In an extension of Hooge model, fluctuations of mobility have been taken into account to explain results in semiconductors.[22] Then a dependence of μ^2 has been proposed, compatible with some results in semiconductors.

In metals, a large number of experiments have shown that the $1/f$ noise is related to the density and the motion of defects or impurities. A model has been proposed which gives a noise spectral density[18]

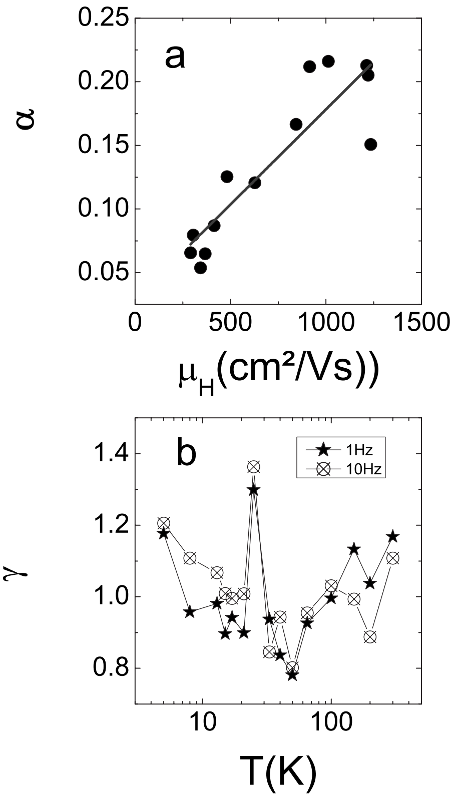

FIG. 4. (a) Dependence of α parameter on the mobility when a backgate voltage from -250 up to 250 V is applied at 5 K. The linear dependence obtained rule further support our hypothesis that there is an intrinsic relationship between the $1/f$ noise and the mobility in the presented samples. (b) We plot γ [Eq. (5)] at different frequencies as function of the temperature. In all the range of temperatures γ depends slightly on frequency. The increase in γ for $T < 50$ K rules out the DDH model (see text).

$$\frac{\alpha}{N} = \frac{f S_V(f)}{V^2} = \frac{k_B T l^2}{\mathrm{Vol}} F(E_\omega, T), \tag{4}$$

where $F(E_\omega, T)$ is the distribution function of the product of concentration of defects with a given activation energy (E_ω) and their scattering cross section, Vol is the volume of the sample and l is the mean-free path. The function $F(E_\omega, T)$ has a small variation with temperature and frequency in metallic systems[18] hence the main variation of α is given by Tl^2 variation. We have tried to fit our data with a $T\mu^2$ variation as μ increases with temperature but the fit is poor and this formula is not compatible with the variation of α when we vary the electric field at constant T.

The Dutta-Dimon-Horn (DDH) model[23] explains the noise dependence on the temperature by the variation in the above defined $F(E_\omega, T)$ function on temperature and frequency. This model relates the temperature dependence of the spectral density with the spectral exponent (γ) defined as

$$\gamma = -\frac{\partial \ln[S_V(f)]}{\partial \ln[f]}. \tag{5}$$

We have plotted the γ as function of temperature in the Fig. 4(b) at two different frequencies 1 and 10 Hz. There is a slight dependence of γ on the frequency in all the range of temperatures. In the DDH model, due to the thermally activated character of the $1/f$ noise, γ is related with the derivative of the noise dependence on temperature, which in our case is always positive and the temperature exponent is about 3 at temperatures less than about 40 K. Although the γ deduced from the temperature dependence does not fit with the obtained data, we could not rule out that the observed noise dependence on temperature is due to the function $F(E_\omega,T)$ due to the experimental incertitude.

At lower temperatures the DDH model is not valid anymore because the thermal energy would be smaller than the typical activation energies, thus no movement would occur. In this regime the tunneling of defects plays an important role and the noise does not depend on temperature. We could only guess that the saturation of the noise at low temperature comes from this phenomenon. However, it is difficult to say whether the tunneling plays a role in our process or not because we do not have access to the characteristic time of the defect motion and its dependence on temperature.

Around the peak at 28 K usually γ ranges from 1.5 down to 0.5. In our opinion, this is related to the appearance of a weak random telegraph noise at frequencies that do not lie in our detection bandwidth. In this case the spectral exponent would correspond to the tails of the Lorentzian peaks, that lead to a decreased or increased exponent dependent on the position of the peak in frequency. This kind of noise is due to the fluctuation of the resistance in the specimen between two stable states. We think that this kind of noise would support our hypothesis that the increase in the noise at 28 K is due to a structural-phase transition and the coexistence of two different phases. However, the origin of this peak and the strong deviation of the spectral exponent are unknown and further work should be performed in order to clarify this point.

A quasilinear variation in the α parameter as function of temperature has been measured in Bi thin films.[24] This variation has been explained by a coherence length (L_φ) larger than the mean-free path, inducing a change in formula (4) due to universal conductance fluctuations. In such a case the $1/f$ noise should be reduced by a factor of 2 when a magnetic field about Φ_0/L_φ^2 is applied. In our system, we have found no variation of the low-frequency noise with a perpendicular to the sample surface applied magnetic field up to 0.7 T at 10 K. Besides that, this kind of effect is accompanied by negative magnetoresistance at low field due to the suppression of the weak localization term in the resistivity. In the studied samples, instead only small positive magnetoresistance of about 3% for 0.7 T has been observed at low temperatures.

V. CONCLUSIONS

We have produced stable high-conductive electron gas at the surface of $SrTiO_3$ by Ar+ irradiation under vacuum and protection by an insulating layer. Low-frequency noise has been measured as function of different external parameters. This noise exhibits a nonusual variation with the electron mobility and can be well described by a modified Hooge formula (3) for different irradiation times, size, temperature, or applied gate voltage. Thus optimization in terms of signal to noise of electronic devices or just conductive links made with these films will be much easier.

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