Electric Noise Spectroscopy

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Electric noise spectroscopy is a non-destructive and a very sensitive method for studying the dynamic behaviors of the charge carriers and the kinetic processes in several condensed matter systems, with no limitation on operating temperatures. This technique has been extensively used to investigate several perovskite compounds, manganese oxides ($La_{1-x}Sr_xMnO_3$, $La_{0.7}Ba_{0.3}MnO_3$, and $Pr_{0.7}Ca_{0.3}MnO_3$), and a double perovskite (Sr_2FeMoO_6), whose properties have recently attracted great attention.

Keywords: noise spectroscopy ; magnetoresistance ; thin films ; quantum interference effects ; charge density waves

1. Introduction

Recently, perovskite materials have attracted great attention due to their electrical, transport, and magnetic properties [1][2][3][4]. In particular, the so-called magnetoresistance (MR) effect and the interplay between spin [5], orbital [6], charge, and structural degrees of freedom [7], have been investigated in polycrystals, single crystals, and thin films. All these phenomena have been the subject of a great deal of research, in view of possible applications in spin electronics and magnetism. In this respect, magnetoresistive materials are already used today in a number of commercially available devices, such as magnetic sensors [8][9], magnetic recording heads [10], and magnetic memories [11][12]. The magnetoresistance effect, when observed in metals, is normally very small and offers scarce possibilities for technological applications. However, the fast advancement of technology and new materials research in recent years may make them more feasible. Instead, larger magnetoresistive effects have been found in ferromagnetic metals and their alloys. The phenomenon is called anisotropic magnetoresistance (AMR) because the change in resistance, when a field is applied parallel to the current direction, is different from when the field is perpendicular to the current direction [13][14]. Moreover, typical components in modern read heads operate thanks to the so-called giant magnetoresistive (GMR) effect, where the magnetoresistance values are more than one order of magnitude larger than those seen in AMR materials. GMR compounds are made with thin layers of magnetic material separated by non-magnetic ones and, depending on their thickness, the magnetic layers couple either ferromagnetically or antiferromagnetically [15][16][17]. Finally, the colossal magnetoresistance (CMR) effect is also observed in pervoskite structure manganites. The term colossal has been chosen because of the very large change in resistance, essentially from an insulating to a conducting state, occurring on application of a magnetic field. Since large fields of the order of a few tesla are required to cause this resistance variation, CMR materials are still not currently considered for practical application as magnetic sensors and, in particular, as the reading element in recording heads. However, a number of other applications are being explored, including bolometers [18][19][20], where a change in temperature causes a change in conductivity driven by a metal-insulator transition, and in spin-tunneling devices, that exploit their half-metallicity [21][22]. Despite the large amount of studies about the magnetic and transport properties of these materials, electric noise characterizations, that can provide new insights on individual perovskite systems, have not been systematically reported [23][24][25][26][27][28].

The spontaneous charge random fluctuations in electron devices are usually called noise, and both terms, fluctuations and noise, are used interchangeably. The physics of fluctuations is of great conceptual importance and is a part of physical kinetics which studies the variations of physical quantities, occurring spontaneously or induced by external fields [29][30]. The investigation of fluctuation phenomena, which may be called “fluctuation spectroscopy”, is a very informative method for the study of kinetic processes in matter. It is often also a method that is much more sensitive than the measurement of average quantities, as demonstrated in superconducting thin films [31][32][33][34] and devices [35][36][37][38], in low-dimensional conductors [39][40][41][42], in carbon nanotubes [43][44][45][46][47] and magnetic composites [48][49], and in conventional [50][51] and innovative solar cells [52][53][54][55]. The role of the noise spectrum analyzer is similar to the role of a microscope: It enables us to visualize the microscopic motion and transitions of particles. For all these reasons, electric noise measurements can give interesting information on the conduction mechanisms and the dynamic behaviors of the charge carriers in the many physical systems.

Triggered by these motivations, noise spectroscopy experiments have been performed on several perovskite compounds (in the form of thin and ultrathin films) and the results of the analysis are presented in this work. In particular, in Sr_2FeMoO_6 (SFMO) thin films, a fluctuation-induced tunneling model satisfactorily explains the measured temperature dependence of the electrical conductance and the current-voltage - curves behavior. This model can be also extended to describe the resistance fluctuations, confirming the dominant role of intergranular tunneling processes in the conduction phenomena of SFMO polycrystalline samples [56]. Current-resistance (CR) effects in $La_{1-x}Sr_xMnO_3$ (LSMO) ultrathin films are also reported. The resistance vs. temperature curves show a negative CR effect in the whole investigated temperature range, while the - characteristics evidence a non-Ohmic regime. The noise measurements are explained in terms of a two-level tunneling systems (TLTS) model, involving different physical scenarios. Among them, the one developed in the case of manganite bi-crystal junctions seems to capture many of the obtained experimental results and applies naturally to the LSMO samples grown on $SrTiO_3$ (STO) substrates in the presence of miscut induced terraces [57]. Different conduction mechanisms are identified in $La_{0.7}Ba_{0.3}MnO_3$ (LBMO) thin films deposited on STO and MgO substrates, respectively. While a standard noise behavior is observed with STO substrates, an anomalous behavior is found in the MgO case. Such anomalous temperature dependence of the measured noise, in the ferromagnetic metallic region, for LBMO-MgO samples is interpreted by considering an enhanced spin ordering with increasing bias currents. This experimental evidence is explained in terms of a spin torque like model assuming that the metallicity of the system is improved by the application of increasing current [58]. Finally, it is also possible to use the electric noise spectroscopy to identify, among different transport mechanisms, the dominating one. This is the case of charge density waves (CDW) conduction in $Pr_{0.7}Ca_{0.3}MnO_3$ (PCMO) epitaxial thin films, and of weak-localization (WL) effects in ultrathin manganite samples. In both cases, the occurrence of unusual noise contributions has been observed, together with an overall increase of the noise level [59][60].

2. Perovskite Compounds: General Concepts and Applicative Indications

Perovskite magnetic materials have been studied for almost 50 years. These systems offer a degree of chemical flexibility which allows the relation between the oxides structure, and electronic and magnetic properties that can be controlled in various ways, such as: Doping [61][62][63], magnetic field [64][65][66], electric field [67][68], temperature [69][70][71], pressure, and photoexcitation [72][73][74][75]. Research on these compounds has revealed the relevant phenomenon of magnetoresistance [76][77][78][79], and has led to the formulation of important physical concepts such as double exchange and the Jahn–Teller polaron [80].

In particular, the double exchange (DE) mechanism was proposed by Zener as a way for the charge to move by the generation of a spin polarized state [81]. This DE process has been historically explained in two different ways. Originally Zener, starting from the insulating antiferromagnetic $LaMnO_3$ system, where electrons are localized on the atomic orbitals, showed how it should gradually become more ferromagnetic upon hole doping (introduction of Mn^{4+}). He considered the problem of the exchange between Mn^{3+} and Mn^{4+} ions via an oxygen ion and introduced the concept of simultaneous transfer of an electron from the Mn^{3+} to the oxygen and from the oxygen to the neighboring Mn^{4+}. Such a transfer was called double exchange. In the case of magnetic atoms, the configurations Mn^{3+}–O^{2-}–Mn^{4+} and Mn^{4+}–O^{2-}–Mn^{3+} are degenerate if the spins of the two *d* shells are parallel, and the lowest energy of the system at low temperature corresponds to parallel alignment of the spins of the two adjacent cations. If the manganese spins are not parallel or if the Mn–O–Mn bond is bent, the electron transfer becomes more difficult and the mobility decreases. It follows that there is a direct connection between conductivity and ferromagnetism. The second way to visualize DE processes was presented in detail by Anderson and Hasegawa [82]. It involves a second-order process in which the two states described above go from one to the other using an intermediate state Mn^{3+}–O^{2-}–Mn^{3+}. In this context, the effective hopping probability (HP) for the electron to move from one Mn-site to the next is proportional to the square of the HP involving the *p*-oxygen and *d*-manganese orbitals. In addition, if the localized spins are considered classical and with an angle between nearest-neighbor ones, the effective HP becomes proportional to [82]. If the HP is the largest, while if , corresponding to an antiferromagnetic background, then the HP vanishes. The quantum version of this process has been described by Kubo and Ohata [83].

The prevailing ideas to explain the magnetotransport behavior of perovskites changed in the mid-1990s from the simple double exchange scenario to a more elaborated picture, where a large Jahn–Teller (JT) effect, which occurs in the Mn^{3+} ions, produces a strong electron–phonon coupling that persists even at densities where a ferromagnetic ground state is observed. In fact, in the undoped limit, and even at finite but small doping, it is well-known that a robust static structural distortion is present in the manganites. In this context, it is natural to imagine the existence of small lattice polarons in the paramagnetic phase above the Curie temperature , and it was believed that these polarons lead to the insulating behavior of this regime. Actually, the term polaron is somewhat ambiguous. In the context of manganites, it is usually associated with a local distortion of the lattice around the charge, sometimes together with a magnetic cloud or region with ferromagnetic correlations (magneto polaron or lattice-magneto polaron). A comprehensive description of theories on the transport and magnetic properties of the mixed-valence oxides is well reported in [2].

3. Electric Noise Spectroscopy: General Concepts and Measurement Techniques

Noise is a stochastic process described by a random function of the independent variable time . The deviation of from its mean value is the fluctuation . How evolves in time on average can be analyzed through the correlation function, which is one of the most important characteristics of any random process and is a nonrandom property of the kinetics of the random fluctuations. When is a sum of many () independent and identically distributed random quantities, it has the normal (Gaussian) distribution. For this class of random processes, the correlation function can be written in terms of a two-dimensional probability density representing the correlation between the values of the random process at two different times, and . This is commonly known as the autocorrelation function , which, in the case of stationary systems, depends only on the difference . It is clear, therefore, that in the time domain the basic properties of random data can be extracted from such autocorrelation functions. In the frequency domain, instead, similar information on random processes can be obtained by the spectral density function , derived, according to the Wiener–Khintchine theorem, as the Fourier transform of the autocorrelation function: *(w)* [84][85].

The main information, given by a power spectral density function computed from physical data measurements, can be found in its amplitude frequency dependence. This allows establishing correlations and relationships with the basic characteristics of the system involved. The common types of low-frequency noise are:

1. Johnson or thermal noise (Figure 1a), generated by the thermally induced motion of the charge carriers (usually the electrons) inside a conductor at equilibrium;
2. shot noise (Figure 1b) modeled by a Poisson process and originating from the discrete nature of electric charge;
3. pink or 1/noise (Figure 1c), characterized by a frequency spectrum which is inversely proportional to the frequency of the signal; and
4. random telegraph noise (Figure 1d), consisting of sudden step-like transitions between two or more discrete voltage or current levels.

In uniform conductors, the dominant noise component at low frequencies is the flicker 1/-type, usually modeled by the Hooge empirical relation. This qualitative rule establishes a direct proportionality between the noise amplitude and the square of the average voltage or current , and is essentially connected to random resistance fluctuations [86]. However, although useful to compare the noise level in different materials of different sizes, the Hooge formula does not have a general physical base and, especially in complex systems, cannot be applied, since resistance fluctuations are not the unique sources of the noise mechanisms in action.

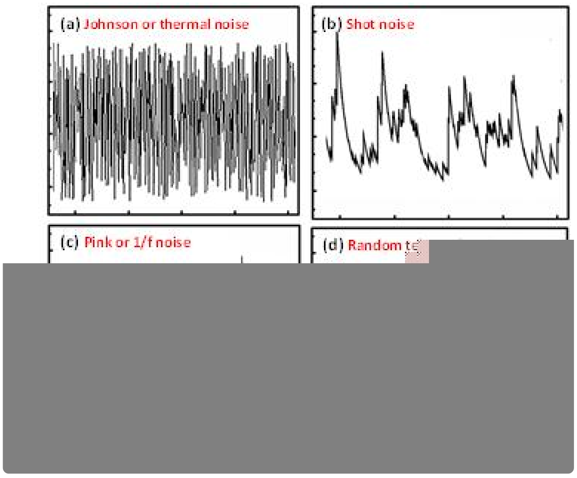

Figure 1. Examples of different types of voltage fluctuations due to: Johnson noise (**a**), shot noise (**b**), 1/ noise (**c**), and random telegraph noise (**d**). The voltage and time units are arbitrary.

As far as noise spectral measurement, one of the most important parameters to be controlled is the system temperature. For the experimental investigations reported in the following, the temperature has been varied with a closed-cycle cold finger refrigerator, operating in a range between 300 and 8 K, and stabilized with a proportional–integral-derivative (PID) algorithm. The output ac voltage has been amplified by a low-noise preamplifier, Signal Recovery model 5113, and the spectral analysis has been realized by a dynamic signal analyzer, Hewlett-Packard model HP35670A. Moreover, standard four-probe measurements have been used to investigate the transport and noise properties of the analyzed samples. Using four probes eliminates measurement errors due to the probe resistance, the spreading resistance under each probe, and the contact resistance between each metal probe and the specimen material. However, in noise studies, the four-probe technique alone does not eliminate completely external spurious and unwanted contributions. In this case, indeed, the method has several limitations due to the fact that each component of the bias and measurement circuit generates its own noise, producing additional current fluctuations in the sample. In principle, the use of an ideal current source in the four-probe configuration can reduce the effect of current contact resistance fluctuations [87]. Unfortunately, the electronic solutions based on feedback circuit often act as ideal sources only at dc. Therefore, in most cases, a second option is considered, that is, the use of a battery in series with a low-noise resistor of a value much higher than the sample resistance. This method fails when the sample resistance is large. Starting from these considerations, it is clear that the development of an experimental procedure, useful to separate and to subtract noise components due to contact resistance fluctuations and to all active instrumentation of the experimental setup ("background noise"), is a fundamental requirement to perform high-quality voltage-spectral density measurements. This can be realized by resorting to a specific analytical correction based on a sequence of two—and four—probe measurements, followed by a mathematical manipulation of the data [88][89].

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