1/*f* noise in the half-metallic oxides CrO_2, Fe_3O_4, and $La_{2/3}Sr_{1/3}MnO_3$

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The excess low-frequency ($1/f$) electrical noise of three representative half-metallic oxides, CrO_2, Fe_3O_4, and $La_{2/3}Sr_{1/3}MnO_3$, has been studied as a function of the temperature (5–300 K) and a magnetic field (up to 12 T). In CrO_2 and $La_{2/3}Sr_{1/3}MnO_3$, the resistance fluctuations are investigated in the metallic regime, below T_c. The magnetite noise is measured across the Verwey transition, in the hopping regime. We find, in all these oxides, a high normalized noise level which roughly follows the temperature dependence of the resistivity. For CrO_2 and $La_{2/3}Sr_{1/3}MnO_3$, the form of the density of state of the responsible excitations is inferred. Oxygen displacement around grain boundaries, magnetic fluctuations, and reversed-spin electronic excitations in the band structure are discussed as possible explanations of the low-frequency fluctuations. For Fe_3O_4, the $1/f$ noise and its temperature dependence are attributed to fluctuations of the number of carriers in a critical network. [S0163-1829(99)04219-8]

INTRODUCTION

Half-metallic ferromagnets are a class of materials of great interest for their potential in spin electronics. The definition of a half metal is a metal where the spin polarization of electrons at the Fermi level is 100%: one spin subband is metallic, whereas the Fermi level falls into a gap of the other one. Experimental studies such as spin-resolved photoemission and band structure calculations support the half-metallic picture for some Heusler alloys (NiMnSb, PtMnSb),[1–5] CrO_2,[3,6–9] and ferromagnetic manganites such as $La_{2/3}Sr_{1/3}MnO_3$.[10,11] Magnetite, Fe_3O_4, is thought to be a half-metallic ferrimagnet.[4]

The bulk intrinsic magnetoresistance of half metals at low temperature, directly linked to their spin anisotropy, is rather low. However, in relation to devices such as spin-polarized tunnel junctions, half-metallic films are of great interest as injectors of fully spin-polarized electrons into the junction.[12–14] The large ratio $(n_\uparrow/n_\downarrow)$ of the spin-polarized density of states at the Fermi level should allow, in principle, the ultimate enhancement of tunneling magnetoresistance (TMR) and its applications.[15]

Recent transport measurements on magnetic oxides have revealed a high level of $1/f$ resistance fluctuations,[16–19] several orders of magnitude larger than the electrical noise typically observed in metallic films. If one thinks of including a half-metallic ferromagnetic oxide in a magnetic sensor, its intrinsic $1/f$ noise could limit the low-frequency sensitivity of the device for detection of resistance changes induced by a magnetic signal. At present, the strength of the electrical $1/f$ noise in metallic oxides and its microscopic origin are not well understood and no universal cause for the $1/f$ noise has been isolated.[20] Research is needed to identify the intrinsic processes which couple to the resistivity to produce the $1/f$ noise. Besides, there is the possibility that these noise measurements may provide insight into the electronic transport properties of oxide films.

The goal of the present study is to characterize the $1/f$ electrical noise in half-metallic oxides. Attempts are made to understand its microscopic origin and to link the noise behavior to the conductivity regime, the structure, and the half metallicity.

We have measured the resistance fluctuations as a function of the temperature (5–300 K) and a magnetic field (up to 12 T) in the three representative half-metallic oxides: CrO_2, Fe_3O_4, $La_{2/3}Sr_{1/3}MnO_3$. The manganite and the chromium dioxide resistance fluctuations are studied in the metallic regime, below T_c. The magnetite noise measurements are performed across the Verwey transition, in the hopping regime of conductivity.

We find that the normalized noise in all these oxides has a $1/f$ spectral density, whose magnitude roughly tracks the temperature dependence of the resistivity. The origin of the noise is analyzed in terms of a broad superposition of thermally activated processes which couple to the resistivity to give the $1/f$ noise spectrum. In CrO_2, the Dutta-Dimon-Horn model[21] appears consistent with the data and leads to a distribution of activation energies of an ensemble of fluctuators that increases above 0.5 eV. For Fe_3O_4, the $1/f$ noise and its temperature dependence are attributed to the fluctuations of the number of carriers in a critical network. In the manganite, the study of the temperature dependence of the noise and transport properties suggests that, below T_c, the resistance fluctuations are directly coupled to the thermally activated spin fluctuations. A drastic decrease of the noise level is observed when a continuous magnetic field (up to 12 T) is applied on the manganite.

SAMPLES AND EXPERIMENTAL TECHNIQUE

1 μm CrO_2 films were deposited on (001) and (110) TiO_2 substrates by high-pressure decomposition of CrO_3.[22] For the (001) orientation, the resulting material has a granular structure with needlelike grains having their long axis (the *c* axis) perpendicular to the surface. The average size of the grains in the plane is about 0.5 μm. The ferromagnetic Curie temperature is 390 K. Below a temperature $\Delta \approx 80$ K, the carriers are mainly scattered by impurities and grain boundaries[23] and the residual resistivity is around 75 $\mu\Omega$ cm. At higher

temperatures, there is a T^2 variation of the resistivity and at room temperature and above, the intrinsic behavior of the oxide is that of a bad metal.[9,23]

2 μm Fe_3O_4 films were prepared by molecular beam epitaxy on (100) MgO substrates. The temperature dependence of the resistivity is dominated by a first-order electronic phase transition at the Verwey temperature ($T_V \approx 123$ K), below which the electrical conductivity decreases by a factor of 100. Below and above T_V, conduction is by hopping of down-spin electrons among the Fe^{3+} cores on the octahedral sites of the cubic inverse spinel structure.

The mixed-valence manganites, (LaSr)MnO_3, are 1 μm thin films deposited on a heated (100) MgO substrate by pulsed laser deposition; the nominal composition is $La_{2/3}Sr_{1/3}MnO_3$. The residual resistivity at 4 K is about 100 $\mu\Omega$ cm and the Curie temperature is approximately 360 K.

For the electrical measurements, CrO_2 and $La_{2/3}Sr_{1/3}MnO_3$ are patterned by photolithography and wet chemical etching to form a Hall bar consisting of three Hall crosses with one common current leg. Each contact area is isolated from the voltage measurement region of the sample to eliminate the effects of contact noise. Electrical contacts are made either by silver paint, or by indium solder. In each case, the same noise level was found. The final sample volume involved in the noise measurements is 2.5×10^{-8} cm^3. Due to the high resistivity of the magnetite below T_V, the Fe_3O_4 film was simply patterned in a large bar of 3×10^{-6} cm^3.

The electrical noise is measured with a standard five-probe configuration using a dc and/or an ac current.[24] With a dc current, the fluctuations of the voltage about 0 are amplified by a SR560 low-noise preamplifier and its output is plugged into a HP 35660A commercial spectrum analyzer. Using an ac current, the carrier frequency is chosen between 400 Hz and 10 kHz depending on the environment. A PAR 116 preamplifier and a PAR 124A lock-in amplifier are used as a demodulator, with minimum time constant ($\tau<1$ ms) and flat mode configuration.

A large driving current is usually needed to increase the ratio of excess noise to Johnson noise. Nevertheless, we were able to measure the flicker noise in our samples with reasonable current densities between 10^2 and 10^4 A/cm^2, avoiding any substantial heating of the films. The same results were obtained with ac or dc current. The standard tests were made to check for errors due to contact noise: the measured power spectral density, normalized by the applied current, was found to be independent of the ratio R/r, where R is the ballast resistance and r is the specimen resistance. This proves that contact noise has been eliminated.[25] The background noise level corresponded to the Johnson noise of the sample and for each sample we checked the squared current dependence of the excess noise spectral density S_V. All these tests made us confident that the measured $1/f$ noise comes from the sample and is related to its resistance fluctuations. Let us emphasize that the noise data we present are the normalized noise spectral density S_V/V^2 where the noise level is normalized by the values of the applied current and the resistance of the sample.

RESULTS

CrO_2

Figure 1 shows the normalized noise spectral density

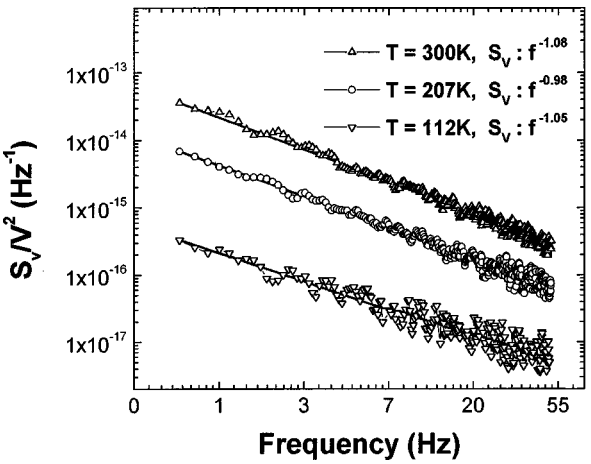

FIG. 1. Normalized power spectral density of the excess noise in CrO_2, showing the $1/f$ frequency dependence at three temperatures.

S_V/V^2 over the frequency band 0.1–50 Hz measured on the CrO_2 film at selected temperatures 112, 207, and 300 K. Hooge's empirical relation is generally used to normalize the data in order to compare the noise levels in different materials. Hooge's equation is[26]

$$S_V/V^2 = \gamma/N_c\Omega f^{\alpha}, \tag{1}$$

where V is the applied voltage across the sample, N_c the density of carriers, and Ω the noisy volume between the probes. The α exponent usually varies in a range between 0.8 and 1.4. The Hooge parameter γ represents the strength of the normalized $1/f$ noise. Taking the density of carriers to be 3.5×10^{22} cm^{-3}, corresponding to one carrier per formula unit,[27] yields a γ value of 2000 at 300 K. This is at least five or six orders of magnitude greater than normally found in metal [$\gamma_{\text{metals}} \approx 10^{-3}-10^{-2}$ (Refs. 28–30)], and is comparable to the γ values recently obtained in colossal magnetoresistance (CMR) magnetic oxides.[16–19]

The temperature dependence of the normalized noise at 20 Hz reveals an increase of the noise by a factor of 50 between 50 and 300 K (Fig. 2). There is a small maximum of

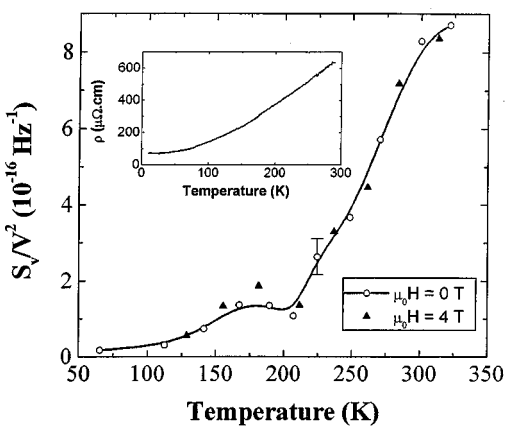

FIG. 2. Temperature dependence of the normalized noise in CrO_2 at 20 Hz (▲: with a 4 T magnetic field perpendicular to the film. ○: in zero field, after the magnetization of the sample). The solid line is to guide the eyes. In the inset, the resistivity of a 1 μm film of CrO_2 on (001) TiO_2.

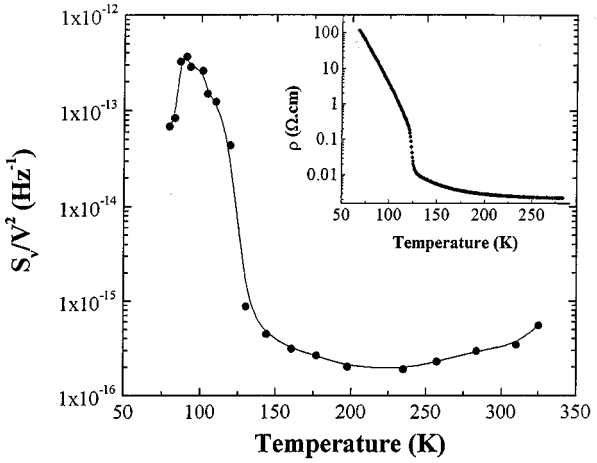

FIG. 3. Temperature dependence of the normalized noise in Fe_3O_4 at 20 Hz. The solid line is to guide the eyes. In the inset, the resistivity of a 2 μm film of Fe_3O_4 on (100) MgO.

the noise around 175 K, followed by a drastic increase above 210 K. The corresponding $\alpha(T)$ exponent of the $1/f^{\alpha}$ spectral density varies from 0.97 to 1.16. The noise level variation as a function of the temperature roughly follows the temperature dependence of the intrinsic CrO_2 resistivity (inset, Fig. 2) which varies by a factor of 30 from 40 K to room temperature. This occurs despite the fact that the trivial dependence of the noise on resistance is eliminated by using the normalized expression of the noise spectral density.

When we apply a 2 T magnetic field perpendicular to the film, along the *c* axis of the CrO_2 sample, the $1/f$ noise level is decreased by a factor of 2 from the value in the demagnetized state. As we decrease the field down to zero, the magnitude of the noise remains more or less constant, revealing a hysteretic effect due to the sample magnetization. In Fig. 2 we notice that, whatever the temperature, the noise has much the same level at 4 T and zero field after the sample has been magnetized. We conclude, therefore, that one part of the electrical noise comes from magnetic domain structures which disappear when a strong enough field is applied. Once this noise is suppressed by applying a field, the resistance fluctuations remain very high ($\gamma \approx 2000$) and are not linked to electron scattering by unstable orientations of magnetic domains.

Fe_3O_4

The $1/f$ noise in the magnetite film was measured below and above the Verwey transition (Fig. 3). At room temperature, using the number of carriers around $0.4\times10^{22}\,\mathrm{cm}^{-3}$ (see discussion below), the strength of the noise is estimated to be $\gamma \approx 80$, which is still a high level. Its temperature dependence reveals a huge decrease of its magnitude by a factor of 100 around T_V, following once again the temperature dependence of the resistivity (inset, Fig. 3). At room temperature, one notices a slight increase of S_V/V^2 even though the resistivity keeps decreasing. The $\alpha(T)$ exponent of the $1/f^{\alpha}$ spectral density varies between 0.88 and 1.32. No particular feature in the temperature dependence of $\alpha(T)$ is observed around T_V. Despite the ferrimagnetism of the magnetite, an applied field of 2 T has no significant influence on the amplitude of the noise.

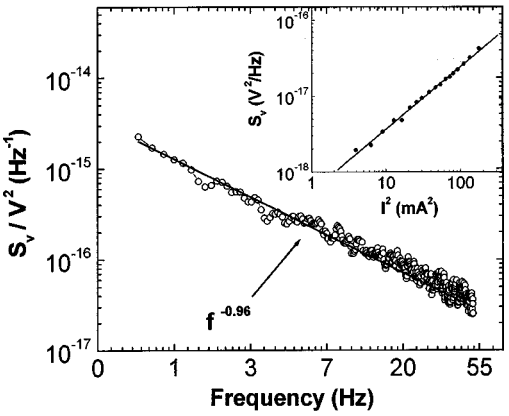

FIG. 4. Normalized power spectral density of the excess noise in $La_{2/3}Sr_{1/3}MnO_3$, showing the $1/f$ frequency dependence at 86 K. In the inset, bias current of the noise power spectral density S_V for $La_{2/3}Sr_{1/3}MnO_3$ at 86 K and 20 Hz.

$La_{2/3}Sr_{1/3}MnO_3$

Figure 4 shows the noise spectral density as a function of the frequency and its current dependence at 86 K. The proportionality of the noise power to the square of the current is again proof that the $1/f$ noise is originating from the resistance fluctuations in the sample, since the amplifier noise does not have any bias current dependence. The temperature dependence of the noise below T_c, and measured at 20 Hz, is presented in Fig. 5. One observes the drastic increase of the noise by a factor of 10^4 as the temperature varies from 10 to 300 K, tracking the temperature dependence of the resistivity (inset, Fig. 5), except that the noise variation is larger by a factor of 500. The γ parameter at room temperature is about 80, taking the number of carriers equal to $0.5\times10^{22}\,\mathrm{cm}^{-3}$. Let us note that the strength of the noise in the metallic phase is a little bit smaller than recently reported by others[16–19] but it remains three or four orders of magnitude greater than in metals. The $\alpha(T)$ exponent varies between 0.8 and 1.2.

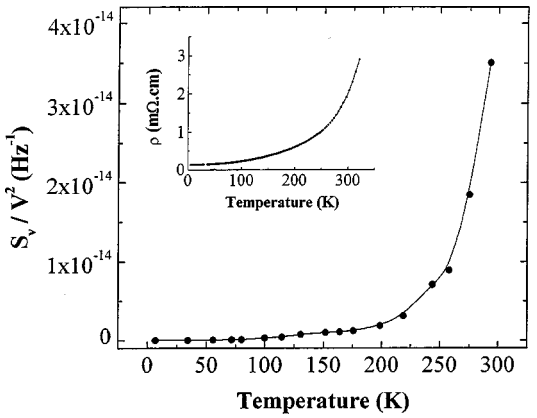

FIG. 5. Temperature dependence of the normalized noise in $La_{2/3}Sr_{1/3}MnO_3$ at 20 Hz. The solid line is to guide the eyes. In the inset, the resistivity of a 0.1 μm film of $La_{2/3}Sr_{1/3}MnO_3$ on (100) MgO.

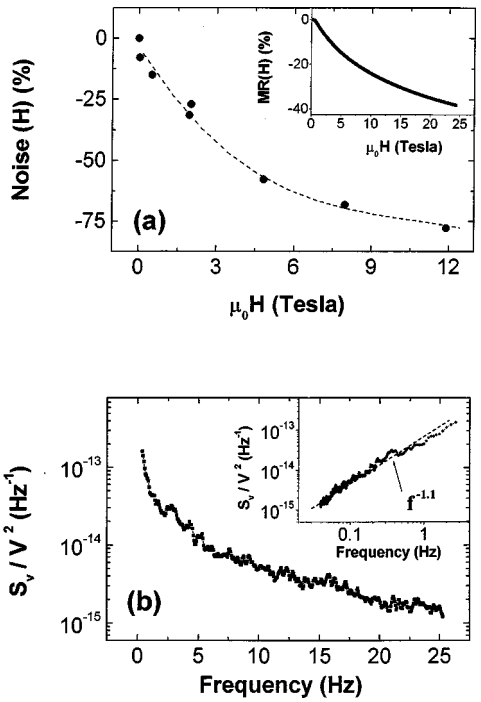

FIG. 6. (a) Magnetic field dependence of the normalized noise power spectral density: Noise $(H)=[S_V/V^2(H) - S_V/V^2(0)]/[S_V/V^2(0)]$, for $La_{2/3}Sr_{1/3}MnO_3$ at room temperature and at 20 Hz. In the inset, the high magnetic field magnetoresistance of the same sample at 300 K. (b) Frequency dependence of the normalized noise power spectral density at 12 T and 300 K for $La_{2/3}Sr_{1/3}MnO_3$.

The $1/f$ noise was also measured in a continuous magnetic field, up to 12 T at the National High Magnetic Field Laboratory in Tallahassee. The field dependence reveals a 75% decrease of the noise level as the field is increased up to 12 T [Figs. 6(a) and 6(b)]. No significant variation of the α exponent as a function of the field was observed.

THEORETICAL BACKGROUND AND DISCUSSION

It is now well known that pure $1/f$ resistance fluctuations are mathematically produced by superimposing a large number of random processes coupled to the resistivity and with an appropriate distribution of characteristic times $D(\tau)$.[20,31–33] This general feature was first applied by McWhorter to model the charge fluctuations at a semiconductor-oxide interface[32] and later by Dutta, Dimon, and Horn (DDH), who proposed that the approximate $1/f$ spectrum in metals was due to a broad distribution of activation energies for the random processes.[21] In metals, such processes are usually related to thermally activated motion of structural defects disturbing the effective cross section of the electron scattering centers. Currently, the notion of superposition of thermally activated processes coupled to the resistivity is widely invoked to explain the origin of the $1/f$ noise, and this in many different systems: metals,[21,30,33] semiconductors,[34–37] magnetic materials,[38,39] superconductors in the normal state,[40,41] etc.

In the normally activated fluctuation model of DDH, the total noise spectrum is simply deduced from an integration of the Lorentzian spectra over the distribution of the activation energies:

$$S(f)\propto \int \frac{\tau(E)}{1+\tau(E)^2 4\pi^2 f^2} D(E)\, dE, \tag{2}$$

where $\tau(E)$ is the characteristic time of one process, defined by an Arrhenius law. DDH made the assumptions, among others, that the net mean-square fractional resistance fluctuation (integrated over all frequency) is temperature independent and the same distribution of energies is involved to describe the nearly $1/f$ noise and its temperature dependence. Therefore, the $\alpha(T)$ exponent of the $1/f^{\alpha}$ spectral density and the temperature dependence of the noise are related by the following expression:[21]

$$\alpha(T)=1-\frac{1}{\ln(2\pi f\tau_0)}\left[\frac{\partial \ln S(f,T)}{\partial \ln T}-1\right], \tag{3}$$

where τ_0 is the ''attempt time'' which, in solids, is of the order of the inverse phonon frequency, typically 10^{-12}–10^{-14} s. If $D(E)$ varies slowly compared to kT, the distribution of energy may be deduced directly from the temperature dependence of the noise spectrum by[21]

$$D(E)\approx 2\pi f S(f,T)/kT. \tag{4}$$

It should also be noted that the $\alpha(T)$ values (which can be greater or less than 1) reflect the shape of the distribution of the activation energies [respectively, $\partial D(E)/\partial E>0$ and $\partial D(E)/\partial E<0$].

If the predicted $\alpha(T)$ values from Eq. (3) are in agreement with experimental results, it means that the assumptions of the model are consistent and the distribution of energies is reliably extracted from Eq. (4). But we should bear in mind that the energy range on which the distribution is estimated directly depends on the experimental conditions. The transition energies we probe are roughly linked to the frequency bandwidth and the temperature by

$$E\approx -kT\ln(2\pi f\tau_0). \tag{5}$$

This means, in our case, that the $1/f$ noise we measure between 0.1 and 100 Hz and 4 and 300 K arises from high transition energies between 0.02 and 0.9 eV, in agreement with slow thermally activated relaxation processes.

CrO_2

In Fig. 7(a) (dots), the noise exponent α is shown as a function of the temperature. In the same figure are plotted (lines) the predicted $\alpha(T)$ values according to the DDH model [Eq. (3)], using a value of $\tau_0\approx 10^{-14}$ s. In this plot, there are no adjustable parameters. Clearly, the overall shape of $\alpha(T)$ is very well reproduced by the DDH model. In view of this good agreement and Eq. (4), we can infer the value of $D(E)$ from $S_V(T)$ [Fig. 7(b)]. A large increase of the distribution is observed above 0.5 eV with an energy peak beyond 0.8 eV. These transition energies are related to some processes responsible for the $1/f^{\alpha}$ spectrum of the noise and its drastic increase above 210 K. The smaller peak centered around 0.4 eV corresponds to the process responsible for the noise anomaly near 175 K (Fig. 2). In the inset in Fig. 7(b) is presented the simulated distribution of transition energy de-

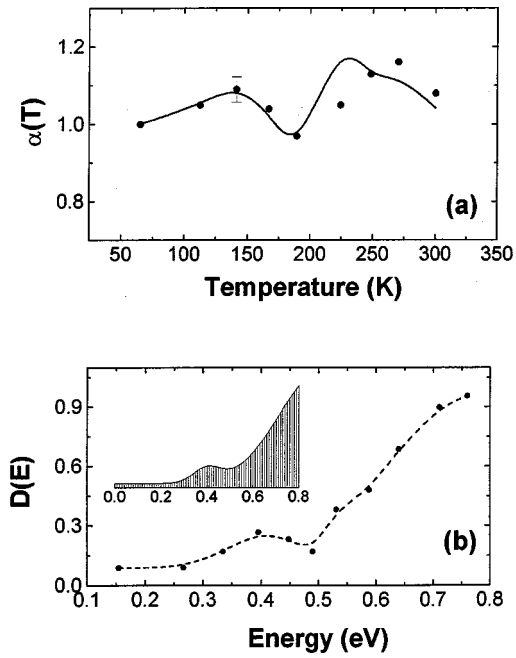

FIG. 7. (a) Frequency exponent $\alpha(T)$ versus temperature for CrO_2. The solid line is the prediction of Eq. (3) using the temperature dependence of the noise (Fig. 2). (b) Energy distribution calculated from Eq. (4) and the curve in Fig. 2. In the inset, numerical calculation of the energy distribution from Eq. (2) and the curve in Fig. 2, without the DDH mathematical approximation.

duced from a numerical calculation of Eq. (2). A satisfactory agreement is found between the simulation and the distribution deduced from the DDH expression. We conclude that the resistance noise is well described by the DDH model despite its assumptions and that random processes with a large activation energy, greater than 0.5 eV are involved.

In metals, it is believed that the distribution of energies is mostly related to fluctuations of lattice defects which make slow thermally activated transitions between states of nearly equal energy.[20,33,42] It is possible that the exceptionally high $1/f$ noise in CrO_2 is attributable to the behavior of oxygen defects and grain boundaries. The distribution of energies we found is consistent with the activation energy of 0.93 eV for oxygen diffusion.[43] Various experimental evidence shows that oxygen is widely involved in defect or vacancy motion.[40,44,45] Local oxygen motion and also oxygen deficiencies create thermally activated vacancies and defects. Either they may act like dynamic traps of free carriers and induce carrier fluctuations, or they may jump between atomic sites that are not equivalent in their contribution to the electrical resistivity. Such effects are usually more pronounced in the grain boundaries which are one of the most probable places in a polycrystalline sample to find mobile atoms.[46] The structure dependence of the noise in CrO_2 has been studied in a 700 nm (110) textured film. The growth on (110) TiO_2 (Ref. 23) induces a needlelike structure with grains of approximately 30 μm length and 0.5–2 μm width. Atomic force and optical microscopy revealed a very good alignment of the long grain axis along the in-plane [110] direction of the substrate. Two Hall bars have been patterned at 90° from one another on the surface of the sample[47] in an orientation such that transport measurements with a current either perpendicular or parallel to the long axis of the grains can be

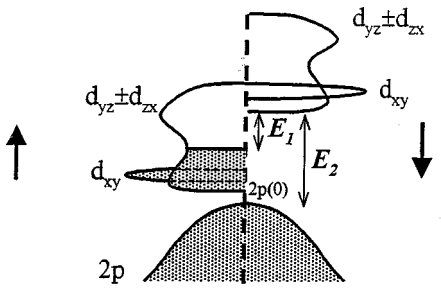

FIG. 8. Schematic electronic band structure in CrO_2 after (Ref. 9). The activation energy for creation of down-spin carriers in the t_{2g} band is E_1, for $d\rightarrow d$ excitation and E_2 for $p\rightarrow d$ excitation.

performed. We found a significant decrease by a factor of 4 of the $1/f$ noise at room temperature, when the current is applied along the path with the lowest density of grain boundaries. Another possible explanation of the huge $1/f$ noise is related to the half-metallic character of CrO_2; the fluctuation providing the noise may be of electronic rather than structural origin. The band structure of the CrO_2 is shown schematically in Fig. 8. The activation energy for creation of down-spin carriers in the t_{2g} band is E_1 for $d\rightarrow d$ excitation and E_2 for $p\rightarrow d$ excitation. Any reversed spin in the conduction band has a severe effect on the conductivity because it will be associated with a reversed Cr^{4+} site moment, which not only withdraws that ion from the conduction process, but also reduces the bandwidth of the local density of states of neighboring sites[23] producing a localized cluster which acts as a strong dynamic scattering center. No significant effect of an applied magnetic field is expected because the energy shift of the spin-polarized bands (0.67 K/T) is much less than their separation ($\geqslant$1000 K). In this model, we associate E_1 and E_2 with the peaks at 0.4 and $\geqslant$0.8 eV [Fig. 7(b)].

Further work is needed to study the influence of the grain boundary density on the $1/f$ noise level and determine the extent to which structural and electronic excitation contribute to the noise. Noise experiments at high temperature, above 400 K, are in progress to investigate the processes around 1 eV and therefore, to see the whole shape of the distribution.

Fe_3O_4

In the magnetite film, an obvious discrepancy is observed near T_V between the theoretical predictions of $\alpha(T)$ [Eq. (3)] and the experimental results (Fig. 9). This disagreement is not unexpected, because near the first-order transition, corresponding to an ordering of the Fe^{3+} and Fe^{2+} cores, we should expect a drastic change of the number of fluctuators responsible for the noise and/or their coupling to the resistivity. Such a feature is not allowed in the simplest form of the DDH model.

Previous transport studies in Fe_3O_4 films show that the mobility of carriers is roughly constant below 250 K and, incidentally, does not significantly affect the temperature dependence of the resistivity at low temperatures.[48] The drastic increase of the resistivity around T_V is mainly attributed to an increase of the activation energy related to the electronic hopping process. Therefore, the $R(T)$ behavior reflects a sig-

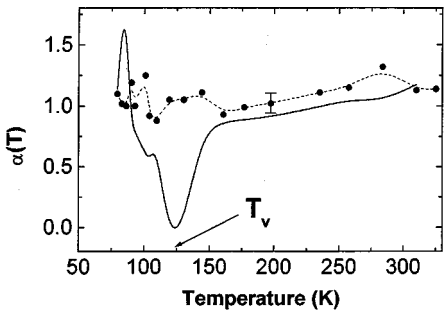

FIG. 9. Frequency exponent $\alpha(T)$ versus temperature for Fe_3O_4 (solid dots and dashed line to guide the eyes). The solid line is the $\alpha(T)$ prediction using Eq. (3) and the temperature dependence of the noise (Fig. 3).

nificant decrease in the number of carriers below T_V. In a model gap, the density of carriers is

$$N_c(T) = n_0 \exp\left(-\frac{\Delta}{kT}\right). \tag{6}$$

We assume n_0 to be equal to $1.35\times10^{22}\,\mathrm{cm}^{-3}$, the number of Fe_3O_4 molecules/cm^3. From the resistivity measurement as a function of the temperature, we deduce two average activation energies Δ, respectively, 70–90 and 20–40 meV below and above T_V. Note that the decrease of 50 meV in the gap around T_V is consistent with the two-order-of-magnitude conductivity jump. Furthermore, our Δ values are in agreement with previous work.[49]

Figure 10 presents the temperature dependence of the noise spectral density multiplied by the number of carriers $N_c(T)$. To produce this plot, we make the reasonable assumption that $R(T)$ gives a good qualitative picture of the temperature dependence of the number of carriers around T_V. Clearly, the product $[S_V(T)/V^2][N_c(T)]$ is almost temperature independent over a wide range of temperature, including T_V. From Eq. (6) and the Δ values, we infer the number of carriers equal to 8.8×10^{18} and $0.11\times10^{22}\,\mathrm{cm}^{-3}$, just below and above T_V. We deduce the corresponding Hooge parameter γ, respectively, 50 and 40, which is nearly constant, compared to the several orders of variation of the

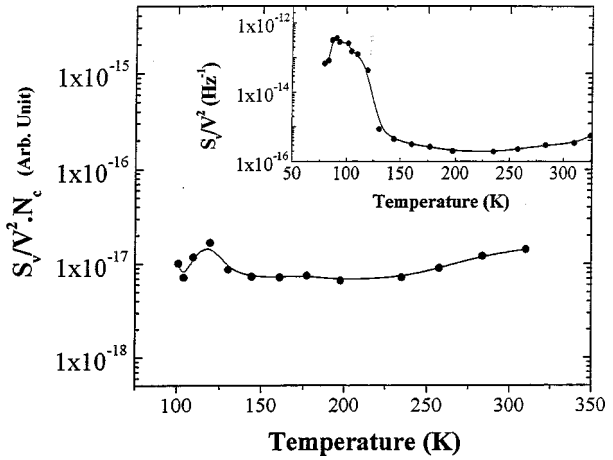

FIG. 10. Product S_V/V^2. N_c as a function of the temperature in Fe_3O_4. In the inset, the temperature dependence of the normalized noise in Fe_3O_4 at 20 Hz.

normalized noise near T_V (inset, Fig. 10). In other words, the noise spectral density scales inversely with the number of carriers in the noisy volume, in agreement with the empirical Hooge relation.

Consequently, we primarily attribute the temperature dependence of the noise in Fe_3O_4 to the variation of the density of carriers in the critical network of conduction. When the number of carriers involved in the hopping conduction decreases, the conductivity becomes more sensitive to the fluctuation of the number of charges in the network; it provokes an increase of the $1/f$ electrical noise level. Such an interpretation is fully consistent with the fact that both the noise magnitude and the resistivity have the same temperature dependence near T_V.

To deal with the microscopic origins of the $1/f$ noise in hopping conductivity, we can refer either to the McWhorter model[32] or to the Shklovskii model.[50] They both showed that a strong $1/f$ noise at low frequencies arises from slow electronic exchanges between a random spatial distribution of traps or donors and the critical electronic network responsible for the conduction. The same argument can apply to a distribution of the energy levels of impurities or excitations. In Fe_3O_4, we have no experimental evidence concerning the microscopic nature of the traps or donors which behave as two-level systems (empty or occupied) and induce fluctuations in mobility and carrier number in the critical network. However, it is necessary to point out that in the hopping regime, the *B*-site electron hopping time τ ranges from 7×10^{-9} s at 120 K to 1.1×10^{-9} s at 300 K.[51] Such relaxation times are obviously too short to contribute to the $1/f$ noise. Furthermore, the weak temperature dependence of the noise once it is scaled to the number of carriers proves that there is no significant temperature dependence of the number of active impurities or excitations. We infer therefore that the resistance fluctuations we measure are caused by slow exchange of electrons between the $Fe^{3+}-Fe^{2+}$ *B* sites and isolated traps or donors widely distributed in energy.

$La_{2/3}Sr_{1/3}MnO_3$

From the $S_V(T)/V^2$ curve obtained on the manganite film (Fig. 5), we deduce the predicted $\alpha(T)$ values from the DDH model and we compare them again to the $\alpha(T)$ exponent extracted from each temperature measurement [Fig. 11(a)]. The ''bumps'' and ''dip'' evident in $\alpha(T)$ are seen to follow the predicted values, but the most striking feature is a relatively constant offset between the levels of the experimental and the theoretical curves. Such a vertical shift is clear evidence that we cannot assume that there is no net temperature dependence of the mean-square fractional resistance fluctuations $\overline{(\delta R)^2}/R^2$. It means that a temperature-dependent function $g(T)$ must be introduced in the general noise integral, as follows:[35,36]

$$S_V(f,T) \propto \int g(T)\,\frac{\tau(E)}{1+\tau(E)^2 4\pi^2 f^2}\,D(E)\,dE. \tag{7}$$

In other words, this function quantifies the temperature dependence of the total noise level and may reflect the variation of the number and/or the strength of the fluctuators as a

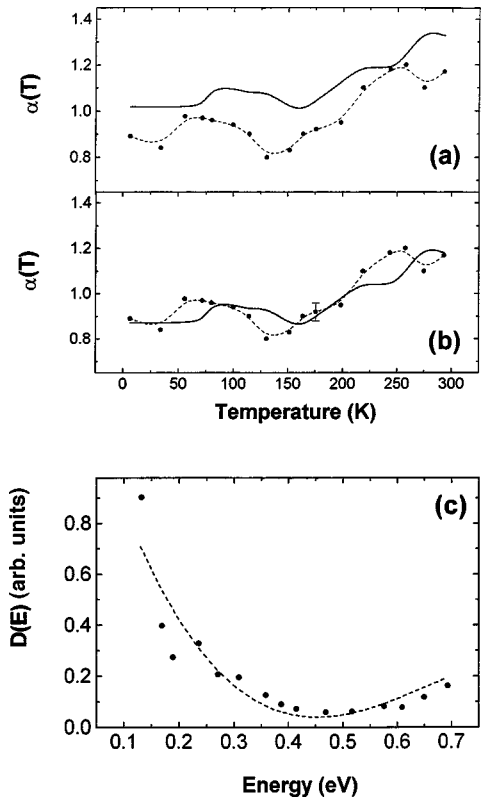

FIG. 11. (a) $\alpha(T)$ exponent as a function of the temperature for $La_{2/3}Sr_{1/3}MnO_3$ (dashed line plus solid dots). The solid curve is the predicted $\alpha(T)$ values from the data shown in Fig. 5 and Eq. (3). (b) The best fit between the experimental $\alpha(T)$ values and the predicted values from Eq. (8), with $g(T)=aT^{4.2}$. (c) Energy distribution calculated from the curve in Fig. 5 and Eq. (9). The dotted line is to guide the eyes.

function of the temperature. It is straightforward to generalize the DDH relation to include this temperature dependence, and one obtains

$$\alpha(T)=1-\frac{1}{\ln(2\pi f\tau_0)}\left[\frac{\partial \ln S_V(f,T)}{\partial \ln T}-\frac{\partial \ln g(T)}{\partial \ln T}-1\right]. \tag{8}$$

The distribution of the activated energies $D(E)$ is now expressed by

$$D(E)\approx\frac{2\pi f S_V(f,T)}{kT}\frac{1}{g(T)}. \tag{9}$$

According to Fig. 11(a), $\partial \ln g(T)/\partial \ln T$ is roughly temperature independent and a direct integration gives a simple variation law for $g(T)$, $g(T)\approx aT^b$. The exponent b is unambiguously deduced from the vertical shift. Figure 11(b) presents a satisfactory agreement between $\alpha(T)$ and the new predicted values according to Eq. (8) using $b\approx 4.2\pm 0.3$ as a fitting parameter. We conclude therefore that the number and/or the strength of the fluctuators responsible for the electrical noise drastically increase as a function of the temperature according to a $T^{4.2}$ law.

Following this analysis, we infer the shape of the distribution of energy $D(E)$ from $S_V(T)$, $g(T)$, and Eq. (9). It can be seen in Fig. 11(c) that $D(E)$ may possess a double-peaked

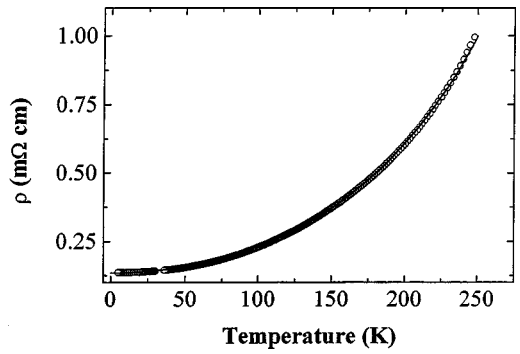

FIG. 12. Temperature dependence of the resistivity of $La_{2/3}Sr_{1/3}MnO_3$ (open circles). The solid line is the fit to $R_0+R_2T^2+R_{4.5}T^{4.5}$, up to 250 K.

structure with a minimum around 0.5 eV. Note that such a shape is perfectly consistent with the experimental $\alpha(T)$ values we measure: the $\alpha(T)$ values, lower than 1, between 10 and 190 K agree with the corresponding decrease of $D(E)$ between 0.1 and 0.5 eV. In the same way, the $\alpha(T)$ values greater than 1, above 190 K, are consistent with the increase of $D(E)$, above 0.5 eV.

In an attempt to understand the physical origin of the $g(T)$ function, we have analyzed our temperature resistivity data with a fit to $R_0+R_2T^2+R_{4.5}T^{4.5}$.[52] The $T^{4.5}$ term was predicted by Kubo and Okata for electron-magnon scattering in the double exchange theory.[53] The result of the fit is plotted in Fig. 12. Good agreement is observed and suggests that the mechanism responsible for the $T^{4.5}$ temperature dependence of the resistivity may be linked to the $g(T)$ function via the number and the strength of the fluctuators. In other words, the fluctuators could be associated with spin fluctuations whose coupling to the resistivity varies as $T^{4.5}$. Such magnetic spin fluctuations as the main origin of the noise in the ferromagnetic phase of $La_{2/3}Sr_{1/3}MnO_3$ also make sense with the drastic initial decrease of the electrical noise level as a magnetic field is applied [Fig. 6(a)]. Moreover, the link we established between the noise behavior and the resistivity data within the framework of the DDH model provides a direct interpretation of the similarity between the temperature dependence of the electrical noise and the resistivity, as the same scattering centers seem to be responsible for the resistivity and its fluctuations.

Again, we may speculate that the activation energy deduced from the DDH model [Fig. 11(c)] is associated with electronic excitations in the band structure. The first peak of the distribution, centered at low energies (below 0.1 eV), may be related to the transition energies involved in the spin fluctuation process, where electrons are excited from the Fermi level to the bottom of the $t_{2g}\downarrow$ band.[10] The second peak of $D(E)$, above 0.5 eV, might correspond to the activation energy for electronic excitation from the top of the filled $2p$ band to the Fermi level. These energies are the analogs of E_1 and E_2 in CrO_2 (Fig. 8). Alternatively, the peak above 0.5 eV may be related to oxygen diffusion.

CONCLUSION

We have examined the $1/f$ noise behavior in CrO_2, Fe_3O_4, and $La_{2/3}Sr_{1/3}MnO_3$. In all these oxides, the normal-

ized noise roughly follows the temperature dependence of the resistivity. For CrO_2, the high noise level measured below T_c is independent of an applied magnetic field once the sample is magnetized. Magnetic domain fluctuations cannot therefore be involved to explain these low-frequency resistance fluctuations. In view of the form of the density of states of the activation energies responsible for the electrical noise in CrO_2 and $La_{2/3}Sr_{1/3}MnO_3$, we have suggested two possible explanations. The first one involves a structural effect relating to thermally activated motion of oxygen defects around the grain boundaries with activation energy below 1 eV. The other possible explanation depends on the half-metallic character of the oxides. The resistance fluctuations may be due to electronic excitations with reversed spin in the band structure. The activation energies for creation of down-spin carriers in the t_{2g} band for $d \rightarrow d$ and $p \rightarrow d$ excitation are in agreement with the activation energies deduced from the DDH approach. Any reversed spin in the conduction band is thought to have a drastic influence on the conductivity. While these provide a fairly natural interpretation of our results, further investigation including studies of nonmagnetic metallic oxides is needed to test these possible explanations.

For Fe_3O_4, it appears that the number of carriers is thermally activated with a narrow gap, despite the half-metallic band structure. A two-order-of-magnitude increase of the normalized noise observed as the temperature is decreased through the Verwey transition can only be explained by a significant change in the number of carriers and/or their coupling strength to the resistivity. We find that the normalized noise around T_V is inversely proportional to the number of carriers involved in the hopping conduction. We therefore infer that the excess noise is related to slow electronic exchanges between the critical network of conduction made by the Fe^{2+} and Fe^{3+} B sites and traps or donors widely distributed in energy.

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