PHYSICAL REVIEW B VOLUME 17, NUMBER 6 15 MARCH 1978

Electrical conduction and current noise mechanism in discontinuous metal films. I. Theoretical

M. Celasco,* A. Masoero, P. Mazzetti,† and A. Stepanescu
Istituto Elettrotecnico Nazionale Galileo Ferraris–Torino, Italy
and Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Ricerche,
U. R. 24–Corso Massimo d'Azeglio, 42–Torino, Italy
(Received 13 July 1977)

A new model of conduction mechanism in discontinuous metal films which gives the temperature dependence of conductivity over a very large temperature interval, from −250 to 450°C, and explains the origin of the conductivity fluctuations responsible for the current noise in these films is presented. It is shown that the conductivity change with temperature is due to a shift of the position of the Fermi level at the surface of the insulator. This changes the height of the potential barrier which must be overcome by tunneling of the conduction electrons. Such a shift, required at each temperature to ensure electron thermal equilibrium, is generated by a partial depletion of surface donor states located on the insulator surface between metal islands. It is further shown that the fluctuation of the surface charge created by the ionization of the surface states modulates the height of the tunneling barrier giving rise to a corresponding modulation of the electrical conductivity originating the current noise typical of discontinuous films. In paper II the results of the theory are compared with experiments.

I. INTRODUCTION

The mechanism of electrical conduction in discontinuous metal films has been the object of several papers.[1-20] The authors were faced with the problem of explaining such features as anomalously large resistivity, a negative thermal coefficient of resistivity, and the presence of a current noise, which is absent in bulk metals. Only in the case of an extremely high current density has a small current noise having $1/f$ spectrum been detected on very thin continuous metal films.[21] This noise has, however, an origin completely different from the one observed in discontinuous films, which form the object of the present paper.

While the problem of the anomalous conductivity behavior on discontinuous metal films has been explained in different ways by many authors,[1-14] only very few papers are dealing with current noise, even though this is a very important aspect of the electrical conduction mechanism in such films. Most of these last papers are experimental only,[15-18] showing that the power spectrum of the noise is $1/f$ type.

To our knowledge only the authors of two papers[19,20] propose a theoretical explanation of this noise, on the basis of the trapped assisted tunneling mechanism of conduction.[7]

However, while the shape of the power spectrum of the noise can be explained by such a theory, by assuming a suitable distribution of trapping times, the intensity of the noise from this model turns out to be many orders of magnitude lower than has been observed. Actually, none of the published theories on electrical conductivity explains the observed intensity of the noise (see Appendix in paper II).

In the present paper a new model of conduction mechanism is proposed which provides the correct order of magnitude for the noise. It also gives a new interpretation of the negative temperature coefficient of resistivity in these films, without assuming that the electrical conduction is a thermally activated process.

This theory assumes that the main conduction mechanism is a direct-tunneling process of electrons between metal islands within the insulator through a potential barrier which changes with the temperature. An experimental support of the assumption of direct tunneling derives from the fact that the electrical conductivity of the film drops rapidly from about infinity to a very low value during the film evaporation when a critical thickness of the film is reached, as in a percolation process.

The reason that the potential barrier is temperature sensitive will be fully described and quantitatively checked in the text. Very briefly, it can be anticipated that this dependence arises from the possibility that electrons bound either to surface states or to donor centers located within the insulator in the gap between metal islands will tunnel in metal states, giving rise to a double-layer charge at the interface between metal islands and substrate.

The corresponding potential drop shifts the position of the Fermi level with respect to the bottom of the conduction band of the insulator and changes the height of the tunneling barrier.

At the highest temperatures the contribution of thermally injected charges on the conduction band

of the insulator will be also taken into account in the computation of the Fermi level position, as well as the contribution of the thermionic current to the total current of the film.

In this model the noise arises from the statistical fluctuations of the number of ionized donors which give rise to a corresponding fluctuation in the conductivity of the film through a modulation mechanism of the potential barrier. This indirect effect of barrier modulation on conduction carriers is much more effective in producing conductivity fluctuation than with previous model and gives the correct order of magnitude for the current noise.

In the following, a detailed calculation of the current noise intensity and power spectrum versus temperature on the basis of the above model will be given. In paper II, which reports many experimental results on the conductivity and on noise of discontinuous gold films evaporated on different substrates, a comparison between theory and experiments is also given.

II. THEORETICAL MODEL

First, the electrical conductivity and the current noise are computed for two neighbor islands only; hence, the discussion can be extended to the entire metal film.

As pointed out in the Introduction the conduction process is assumed to be generated by four different types of mechanisms: (a) a direct tunneling of electrons between metal islands, which should be responsible for the main conductivity process at low temperature; (b) a thermionic current, which contributes to the conduction process at the highest temperatures; (c) a tunneling of electrons from surface states of the insulator to the metal islands, which is necessary to obtain thermal equilibrium of the electrons at the metal insulator interface; (d) a thermal injection of electrons from the metal to the conduction band of the insulator, which contributes to the balance of the displaced charge and becomes important at the highest temperatures.

Both the direct tunneling and the thermionic effect take place between metal islands within the insulator and are limited to a very thin layer of the insulator itself (20 Å) below the surface. The conductance between two metal islands can thus be easily calculated at each temperature value T once the potential barrier $\varphi_B(T)$ (see Fig. 1) is found.

The only important assumption of this theory concerns the existence of donor-like levels located at the surface of the insulator, of the type found in chalcogenide glasses.[22,23] In the present case these donor centers are probably due to broken bonds at the surface of the insulator, but they could also be related to impurity ions present on the substrate or diffused at the surface during substrate preparation. As will be seen in the following, the surface density of these states, which correctly accounts for the experimental results, is roughly equal to 10^{15} states/cm^2, corresponding to assuming that all the molecules of a monolayer at the surface of the insulator become donor centers.

In the following, for the sake of simplicity, it is assumed that these surface states form a single highly degenerate level φ_1 below the conduction band, as shown in Fig. 1.

The facts that in reality the surface levels may form a bundle or that other impurity levels having a much lower degeneracy are present will hardly affect the conclusions. Therefore, the behavior

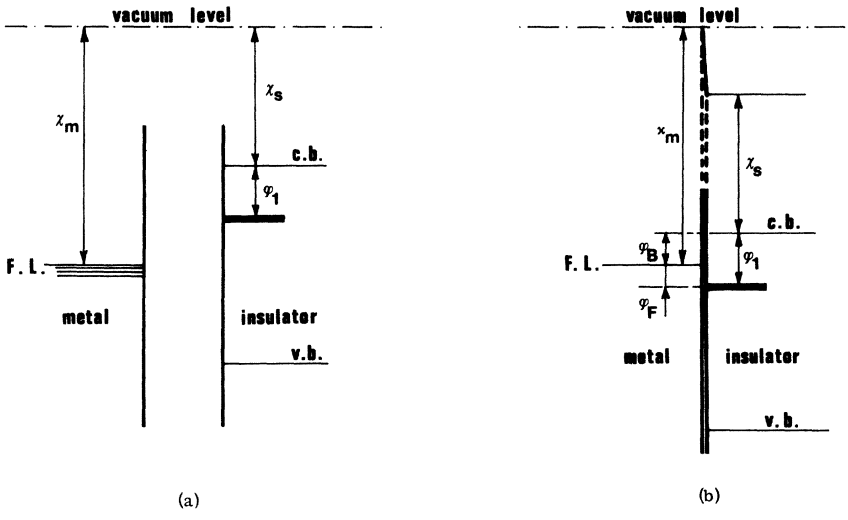

FIG. 1. Energy diagrams for the metal-island to substrate contact: (a) before contact is made; (b) after contact is made.

versus temperature of the barrier height $\varphi_B(T)$ is computed first. Then, through this function, the surface conductance and the conductivity noise spectrum of the film will be calculated.

A. Barrier height versus temperature

The barrier height, $\varphi_B(T)$, in thermal equilibrium is computed by using the model shown in Fig. 1 and by taking into account that the electrons can easily tunnel from the donor states located at the surface of the insulator to the metal islands and vice versa. In the calculation, the effect of the charge thermally injected from the metal into the insulator conduction band will also be taken into account.

The tunneling space-charge potential, which is much less dependent on temperature than the thermionic-charge potential, will be considered as a constant and incorporated in the χ_s. Let γ be the positive surface density charge due to the ionization of donor states which are below the Fermi level, of the amount φ_F, as shown in Fig. 1(b).

Further, let ρ be the thermionic volume charge density injected from the metal into the substrate conduction band. Under conditions of thermal equilibrium, the average potential barrier in the insulator between metal islands[24] can be written

$$\varphi_B(T) = \chi_m - \chi_s + E_1 + E_2, \tag{1}$$

where χ_m and χ_s are the electron work functions for the metal and the insulator, respectively, and E_1 and E_2 are the average potential-energy drops for one electron which moves from the metal islands into the gap due to surface charge density γ and to the volume charge density ρ, respectively. These last quantities are functions of temperature and of $\varphi_B(T)$, so that Eq. (1) should be interpreted as an implicit equation for $\varphi_B(T)$. The surface charge density γ can be easily calculated at each temperature once the surface states density δ for the insulator in known.

The ionization probability of a donor center P_i can be written, according to the Fermi-Dirac statistics;

$$P_i = 1 - \frac{1}{e^{-\varphi_F(T)/kT} + 1}, \tag{2}$$

where $\varphi_F(T)$ is the quantity shown in Fig. 1(b), k is the Boltzmann constant, and T is the absolute temperature. Thus the surface charge density γ, due to the donor states ionization, is simply given by

$$\gamma = e\delta P_i \simeq e\delta e^{-\varphi_F(T)/kT} = e\delta e^{-[\varphi_1 - \varphi_B(T)]/kT}, \tag{3}$$

e being the electron charge, and having taken into account that in the present case the value of the exponential is at any temperature $\ll 1$.

The thermionic injected charge ρ can also be calculated as a function of $\varphi_B(T)$ and T through the expression[25]:

$$\rho = 2e(m^*kT/2\pi\hbar^2)^{3/2} e^{-\varphi_B(T)/kT}, \tag{4}$$

where m^* is the effective electron mass at the bottom of the conduction band of the insulator, and $\hbar$ is the reduced Planck constant.

Since electron tunneling between islands takes place in a volume a few atomic layers below the surface, to calculate E_1 it can be assumed that the surface charge density γ is actually distributed in the gap between islands in a volume which extends below the surface for a depth roughly equal to the gap between metal islands (typically 20 Å). Simple electrostatic considerations yield

$$E_1 = -\frac{e\gamma d}{12\epsilon_0\epsilon_r^*} = -\frac{e^2\delta e^{-[\varphi_1 - \varphi_B(T)]/kT} d}{12\epsilon_0\epsilon_r^*} \tag{5}$$

and

$$E_2 = \frac{e\rho d^2}{12\epsilon_0\epsilon_r^*} = \frac{e^2 d^2}{6\epsilon_0\epsilon_r^*}\left(\frac{m^*kT}{2\pi\hbar^2}\right)^{3/2} e^{-\varphi_B(T)/kT}, \tag{6}$$

where d is the spacing between metal islands, ϵ_0 is the vacuum permittivity, and ϵ_r^* is the effective dielectric constant of the substrate.

Equation (1) gives

$$\varphi_B(T) = \chi_m - \chi_s + \frac{e^2 d}{12\epsilon_0\epsilon_r^*}\left[2d\left(\frac{m^*kT}{2\pi\hbar^2}\right)^{3/2} e^{-\varphi_B(T)/kT} - \delta e^{-[\varphi_1 - \varphi_B(T)]/kT}\right]. \tag{7}$$

In this equation ϵ_r^* is an effective dielectric constant which is proportional to the actual one for each type of substrate, the reduction coefficient β taking into account the fact that the active trapped charge is only a few atomic layers apart from the metal islands. Therefore, we set

$$\epsilon_r^* = \beta\epsilon_r, \tag{8}$$

where β has been estimated in the practical case to be approximately $\frac{1}{3}$.

B. Electrical conductance versus temperature

The electrical conductance G through the gap between two metal islands, assuming that the carriers move below the surface of the substrate in a layer having a depth approximately equal to the island separation, d(≈15–25 Å), can be written

$$G = G_1 + G_2, \tag{9}$$

where G_1 and G_2 are, respectively, the conductance due to the tunneling and the thermionic currents.

These terms can be easily calculated once $\varphi_B(T)$ has been found at each temperature T through Eq. (7). If l is the length of the gap between the two islands[26] to a good approximation it follows, from well-known equations,[27,28] that

$$G_1 = l(2m)^{1/2}(e/h)^2\varphi_B^{1/2}(T) \cdot \times \exp\{-[4\pi(2m)^{1/2}/h]d\varphi_B^{1/2}(T)\}. \quad (10)$$

On the other hand, the thermionic component G_2, assuming that in this case also the active length of the gap between metal islands is l and that the thermionic current flows in a layer of depth d, is given by the Richardson-Dushman equation,

$$G_2 = dl(4\pi m^* ek^2/h^3)T^2 e^{-\varphi_B(T)/kT}. \quad (11)$$

In both cases the image-charge effect on the potential barrier has been neglected. The thermionic component G_2 become comparable with G_1 at the highest temperature (above 500°K) only. A full discussion is given in paper II. However, it may be anticipated that an accurate comparison of this theory with the experimental results obtained at very high temperature (up to 725°K) shows that G_2 should be much smaller than that given by Eq. (11). The reason may be that this equation is true if the mean free path of an electron in the conduction band of the insulator is larger than the gap between metal islands, which probably is not the case near surface of the substrate.

C. Electrical noise

As anticipated in the Introduction, the current noise in the present model is generated by the thermodynamical fluctuations of $\varphi_B(T)$ due to the fluctuation of the number n of ionized donor states in the active area between metal islands. This number fluctuates in time around the mean value $\bar{n}(T)$ given at each temperature by

$$\bar{n}(T) = dl\delta e^{-[\varphi_1 - \varphi_B(T)]/kT}. \quad (12)$$

The effect of the fluctuation of the thermionically injected charge on the barrier $\varphi_B(T)$—which could have some influence at the highest temperatures, where it becomes comparable with the charge due to ionized donors—will be neglected in the noise calculation because the involved time constant (relaxation time) is much too short to give an appreciable contribution to the noise in the relevant spectral bandwidth. Actually, the spectral density of the noise generated by the fluctuation of such a space charge is very low because it extends up to very high frequencies, while the contribution due to the charge of ionized donor centers is mainly limited to a low-frequency band, owing to the fact that such a trapped charge communicates with the metal islands mainly through a tunneling process.

In order to develop a quantitative theory of the current noise, $\varphi_B(T)$ must be considered as a function of the actual value $n(T)$ of ionized donors. [In Eq. (7) $\varphi_B(T)$ is expressed in term of $\bar{n}(T)$.]

The power spectrum $\phi_G(\omega)$ of the conductance fluctuation between two metal islands can be expressed in terms of the fluctuation power spectrum $\phi_n(\omega)$ of the number of ionized donors n in the gap between the two islands.

Since the barrier $\varphi_B(T)$ depends on n and the fluctuation δn of ionized donors with respect to the average value is small, it follows that, to the first order in δn, the conductance $G(n)$ can be written

$$G(n) = G(\bar{n} + \delta n) = G(\bar{n}) + G'(\bar{n})\delta n, \quad (13)$$

where $G'(\bar{n})$ is the derivative of $G(n)$ with respect to n and the term $G'(\bar{n})\delta n$ represents the conductivity fluctuation ΔG.

The power spectrum of ΔG can thus become

$$\phi_G(\omega) = [G'(\bar{n})]^2\phi_n(\omega), \quad (14)$$

with

$$n(T) = \bar{n}(T) + \delta n = dl\delta e^{-[\varphi_1 - \varphi_B(T)]/kT} + \delta n. \quad (15)$$

Equation (7) yields

$$\varphi_B(T, n) = \varphi_B(T, \bar{n} + \delta n) = \chi_m - \chi_s + \frac{e^2 d}{12\epsilon_0\epsilon_r^*} \times \left[2d\left(\frac{m^* kT}{2\pi\hbar^2}\right)^{3/2} e^{-\varphi_B(T)/kT} - \frac{\bar{n}(T) + \delta n}{dl}\right]. \quad (16)$$

Thus, to the first order in δn,

$$G'(\bar{n}) = \frac{dG}{d\varphi_B}\frac{d\varphi_B}{dn} = \frac{dG}{d\varphi_B}\frac{e^2}{12\epsilon_0\epsilon_r^* l} = \frac{e^2\varphi_B^{-1/2}}{24\epsilon_0\epsilon_r^* l}\left(\frac{4\pi(2m)^{1/2}}{h}d - \varphi_B^{-1/2}\right)\langle G\rangle, \quad (17)$$

where $\langle G\rangle = G(\bar{n})$. Finally,

$$\phi_G(\omega) = \frac{e^4\varphi_B^{-1}}{576\epsilon_0^2\epsilon_r^{*2}l^2}\left(\frac{4\pi(2m)^{1/2}}{h}d - \varphi_B^{-1/2}\right)^2 \times G^2(\bar{n})\phi_n(\omega), \quad (18)$$

where the conductivity term due to thermionic current has been neglected for the reason pointed out above.

For comparison with the experimental results, it is more convenient to introduce the power spectrum

$$\psi(\omega) = \frac{\phi_G}{\langle G\rangle^2} = \frac{e^4\varphi_B^{-1}}{576\epsilon_0^2\epsilon_r^{*2}l^2}\left(\frac{4\pi(2m)^{1/2}}{h}d - \varphi_B^{-1/2}\right)^2\phi_n(\omega), \quad (19)$$

which refers to the relative conductance fluctuation $\Delta G/\langle G\rangle$.

Hence the power spectrum $\phi_n(\omega)$ shall be computed. Let us first assume that the ionization processes of donors centers are statistically independent events. Indicating with an asterisk the quantities

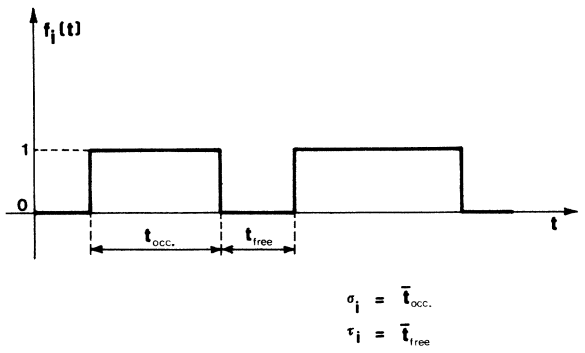

FIG. 2. Occupation function $f_i(t)$ vs time of the ith donor center: t_{occ}, donor center occupied; t_{free}, donor center ionized.

which refer to this approximation, it can be written

$$\phi_n^{(*)}(\omega) = \sum_i S_i(\omega) = \delta d l S_n(\omega) , \tag{20}$$

where $S_i(\omega)$ is the power spectrum of the occupation function $f_i(t)$ of the ith donor center, defined as that function of time, which is 1 if the donor center is occupied by an electron and zero if the center is ionized (see Fig. 2).

In the same equation, $S_n(\omega)$ is an average spectrum obtained by an averaging equation over the index i. It should be noted that $S_i(\omega)$ strongly depends on the location of the donor center in the gap as shown below. In order to calculate $S_i(\omega)$ let σ_i and τ_i be the variables representing the average occupation time and the average ionization time for ith donor center, respectively.

In terms of these variables the power spectrum is given by the Machlup expression[29]

$$S_i(\omega) = \frac{1}{\pi} \frac{\vartheta(T)}{1+\vartheta(T)} \frac{\tau_i'}{\tau_i'^2\omega^2+1} , \tag{21}$$

where

$$\vartheta(T) = \tau_i/\sigma_i \tag{22}$$

and

$$\tau_i' = \frac{1}{1+\vartheta(T)} \tau_i . \tag{23}$$

From Eq. (21) it is seen that the power spectrum $S_i(\omega)$ has the appearance of a Lorentzian noise with a time constant given by Eq. (23). To evaluate the spectrum $S_n(\omega)$ it is necessary to estimate the form of the distribution function of τ_i' and to average with respect to that function. The quantity $\vartheta(T)$ is actually a function of the temperature only and does not depend on the index i.

Using Eq. (2), which gives the ionization probability of a donor center, the occupation probability is easily obtained, namely,

$$\frac{\sigma_i}{\sigma_i+\tau_i} = \frac{1}{e^{-[\varphi_1-\varphi_B(T)]/kT}+1} . \tag{24}$$

From Eq. (21) it is seen that, at each temperature, the distribution function of τ_i' is proportional to the distribution function f of the ionization times τ_i. To evaluate the distribution function it is necessary to estimate the average ionization time of a donor center having a distance x_i from the nearest metal island. Taking into account that only those processes where an electron is exchanged between metal islands and donor centers must be considered, and that this requires a tunneling process through a barrier width x_i and height φ_1, we can write, to a good approximation,

$$\tau_i \propto e^{[2(2m\varphi_1)^{1/2}/\hbar]x_i} . \tag{25}$$

The effect of direct transitions from donor centers to the conduction band of the insulator, which can be active at the highest temperatures, will change the distribution function for the ionization times, a fact which has an experimental support, and will be discussed later.

From Eqs. (23) and (25), assuming a uniform distribution for the x_i variables, the distribution function $p(\tau_i')$ for the τ_i''s is easily found:

$$p(\tau_i') = \left[\ln\left(\frac{\tau_M'}{\tau_m'}\right)\right]^{-1} \frac{1}{\tau_i'} , \tag{26}$$

where τ_M' and τ_m' are the largest and the smallest values of τ', respectively.

The distribution given by Eq. (26) is equal to that used by McWhorter[30,31] to explain $1/f$ noise in semiconductors. In that case such a distribution was generated by tunneling of carriers to trapping states located on the surface oxide layer.

The power spectrum $\phi_n^{(*)}(\omega)$ defined in Eq. (20) can thus be found through an averaging operation over Eq. (21) by using the distribution given by Eq. (26), namely,

$$\phi_n^{(*)}(\omega) = \frac{1}{\omega} \frac{\vartheta(T)}{1+\vartheta(T)} \left[\ln\left(\frac{\tau_M'}{\tau_m'}\right)\right]^{-1} \times [\tan^{-1}(\omega\tau_M') - \tan^{-1}(\omega\tau_m')] . \tag{27}$$

The spectrum $\phi_n^{(*)}(\omega)$ is thus of the $1/f$ type[30] in a frequency range which covers the interval from $1/\tau_m'$ to $1/\tau_M'$.

It follows from Eq. (19) that

$$\psi^{(i)}(\omega) = \frac{e^4\varphi_B^{-1}(T)\delta d}{576\pi\epsilon_0^2\epsilon_r^{*2}l} \frac{\vartheta(T)}{1+\vartheta(T)} \times \left[\frac{4\pi(2m)^{1/2}}{h} d - \varphi_B^{1/2}(T)\right]^2 \left[\ln\left(\frac{\tau_M'}{\tau_m'}\right)\right]^{-1} \times \frac{1}{\omega} [\tan^{-1}(\omega\tau_M') - \tan^{-1}(\omega\tau_m')] . \tag{28}$$

Equation (28) represents the power spectrum of the

relative conductance fluctuation between two islands with the basic assumption that the ionization processes of different donor centers are independent events.

Actually it must be considered that a fluctuation of the number of ionized centers changes the height of the potential barrier $\varphi_B(T)$, which in turn has a feedback effect on the fluctuation itself, giving rise to a smoothing effect on the noise. This effect is similar to the one produced by space charge on the shot noise in a nonsaturated vacuum diode,[32] which gives rise to a noise intensity reduction by a factor Γ without practically affecting the spectral density distribution. An approximate expression of the power spectrum for the relative conductance fluctuation which takes into account the feedback effect described above can thus be written

$$\psi(\omega) = \Gamma \psi^{(i)}(\omega). \tag{29}$$

In the present case the smoothing factor Γ can be calculated by making use of the Nyquist theorem, which allows one to evaluate the thermal equilibrium fluctuation of the potential barrier $\varphi_B(T)$. Actually it must be considered that in the present model the ionization of donor centers is a thermal equilibrium process which is not influenced by the current flowing in the film.

Integrating Eq. (28) with respect to ω, and taking into account that τ_M is many orders of magnitude larger than τ_m, we get as a good approximation,

$$\frac{\langle \Delta G^2 \rangle}{\langle G \rangle^2} = \int_{-\infty}^{\infty} \psi(\omega) d\omega \simeq \Gamma \frac{e^4 \varphi_B^{-1}(T) \delta d}{576 \epsilon_0^2 \epsilon_r^{*2} l} \frac{\vartheta(T)}{1+\vartheta(T)} \times \left[\frac{4\pi (2m)^{1/2}}{h} d - \varphi_B^{1/2}(T) \right]^2 . \tag{30}$$

This equation should be compared with the one obtained by using Nyquist's theorem. In the Appendix the generalized form of Nyquist's theorem is developed, suitable for application to physical systems where energy fluctuations are associated with entropy fluctuations. Application to the present case yields

$$\int_{-\infty}^{\infty} \psi(\omega) d\omega = \frac{e^2 \varphi_B^{-1} kT}{48 \epsilon_0 \epsilon_r^* l} \left(\frac{4\pi}{h} (2m)^{1/2} d - \varphi_B^{1/2} \right)^2 . \tag{31}$$

From Eqs. (30) and (31), the following expression for the smoothing factor Γ is obtained:

$$\Gamma = \frac{12kT}{e^2} \frac{\epsilon_0 \epsilon_r^*}{\delta d} \frac{1+\vartheta(T)}{\vartheta(T)} . \tag{32}$$

The value of this factor can be easily calculated at any temperature whenever the values of δ, d, and $\varphi_F(T)$ are known. With the values given below for these quantities in the case of gold film on sapphire having a room-temperature surface resistivity of about 50 k$\Omega\square$ the value $\Gamma \approx 0.24$ is obtained. It should be noted that the temperature dependence of Γ is very small, because $\vartheta(T)$ (which is always smaller than 1) is approximately a linear function of temperature.

Equation (29), taking into account Eqs. (28) and (32) yields

$$\psi(\omega) = \frac{kTe^2 \varphi_B^{-1}(T)}{48\pi \epsilon_0 \epsilon_r^* l} \left[\frac{4\pi (2m)^{1/2} d}{h} - \varphi_B^{1/2}(T) \right]^2 \times \left[\ln \left(\frac{\tau_M'}{\tau_m'} \right) \right]^{-1} \frac{1}{\omega} \left[\tan^{-1}(\omega \tau_M') - \tan^{-1}(\omega \tau_m') \right] . \tag{33}$$

Equations (31) and (33) give, respectively, the total noise spectrum and the power spectrum for the relative conductance fluctuation between two metal islands separated by a gap of width d and length l. The extension of this formula to the whole film is made in the Sec. III.

III. EXTENSION OF THE THEORY TO THE ACTUAL FILM STRUCTURE

Theoretical results relative to the conductivity and noise worked out in the preceding sections refer to only two metal islands. In order to define quantities like the surface conductance G_s and the relative conductance spectrum $\psi_F(\omega)$ for the whole film, some more information is needed about the island structure of the film.

For what concerns G_s, the relation between this quantity and G is simply

$$G_s = G. \tag{34}$$

The structure of the film, however, is involved in the definition of the quantity l, which has been defined as the active gap length between contiguous metal islands along the current lines. What really matters here is the definition of a new quantity D, which has the meaning of the average linear dimension of the metal islands for the particular film considered.

As is seen from the electron microscope photographs shown in paper II, the actual structure of the islands of gold films annealed at high temperature (above 700°K) is very complicated.

In this case D should be chosen to characterize the average linear dimension along the current direction of the equipotential areas metallically connected as schematically represented in Fig. 3. According to this scheme, even the gaps having a width which is rather smaller than the average should be considered as metallic connections because, owing to the strong nonlinear dependence of the electrical conductance between metal islands on the gap width, the potential drop in this case be-

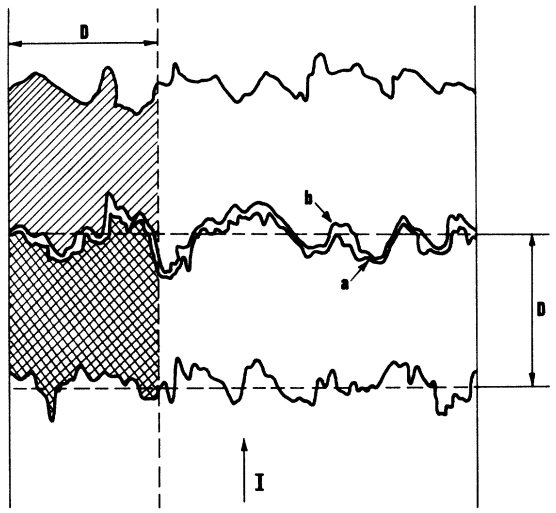

FIG. 3. Schematic representation of different equipotential lines (a and b) in a discontinuous gold film having a structure of the type shown in Fig. 1 of paper II. The parameter D can be interpreted as the average linear dimension of the metal islands.

comes negligible. The length l in this scheme represents the average total length of all the active gaps between two different contiguous equipotential lines in a strip of width D along the current direction (shadowed area in Fig. 3). A final point concerns the relation between relative conductance noise spectrum $\psi(\omega)$ relative to an island pair and the spectrum $\psi_F(\omega)$ which refers to the whole film.

By representing the film as a network of resistors, each having a conductance G, independently fluctuating with a relative conductance fluctuation spectrum $\psi(\omega)$, for a film L wide and L' long, the following relation can be obtained:

$$\psi_F(\omega) = (D^2L/L'^3)\psi(\omega). \tag{35}$$

The coefficient D^2L/L'^3, which appears in this equation, has a very simple interpretation in the case of a square film where $L = L'$. In this case it represents the inverse of the total number of the metal islands constituting the film.

Equations (29) and (31) will be used to compare experimental and theoretical results. Owing to the very rough approximation made in the definition of the quantities l and D, it is obvious that in comparing the theoretical results with the experimental ones only the correct order of magnitude can be hoped for as concerns the absolute values of the conductivity and noise intensity. However, the theory also gives the correct temperature dependence of these quantities as well as the correct shape of the noise power spectrum.

IV. DISCUSSION

The theoretical results are being discussed from a rather general point of view giving the order of magnitude of the parameters involved and the general trend of the basic quantities like the barrier height $\varphi_B(T)$, the film surface conductance $G_s(T)$, and the relative conductance noise power spectrum $\psi_F(\omega)$. With regard to $\varphi_B(T)$, in practical cases a good fit of the experimental results for gold on silica glass substrates is obtained by assuming in Eq. (7) $\chi_m - \chi_s = \varphi_1$, $\delta = 10^{15}$ states/cm^2, $\varphi_1 = 0.25$ eV, $d = 26$ Å. The position $\chi_m - \chi_s = \varphi_1$, which corresponds to assuming that the effective electron work function for the substrate (that is, the work which must be done to remove one electron from a donor state into vacuum) is very nearly equal to χ_m, has some independent experimental support[7,16,17] and in any case is not critical for $\varphi_B(T)$.

The density of surface states chosen above, which corresponds to assuming that almost all the molecules of a single monolayer on the free surface of the substrate become donor centers, is roughly equal to that found in semiconductors.[33] The value of φ_1 was actually picked up by taking the best fit for the experimental results, but it is not far from the energy value of donor centers found in chalcogenides glasses, which form a bundle located from 0.2 to 0.5 eV below the conduction band.[34] Finally, the average width of the tunneling barrier d is chosen in each case to yield the correct absolute value of the conductivity of the particular film, once the length l of the gap between neighbor islands has been estimated.

It should be noted that a change of d of a few angstroms changes the conductivity of an order of magnitude without appreciably changing the behavior of $\varphi_B(T)$, so that a wrong estimation of l will simply yield slight variation of d. The behavior of $\varphi_B(T)$ versus temperature for the values reported above is given in Fig. 4. In this figure, the case of sapphire is also reported. In this case the fit with the experimental results was found with a slightly different value of $\varphi_1(0.2$ eV) and of d (16 Å).

In each case, the effective dielectric constant ϵ_r^* has been taken to be proportional to the true value after applying a reduction coefficient $\beta = \frac{1}{3}$, to take into account that the active trapped charge is only a few atomic spacings apart from the metal islands.

As is seen from Fig. 4, $\varphi_B(T)$ decreases with temperature, because of an increase of the charge of ionized donor states. However, above a given temperature the thermionic injected charge, represented by the first term within the large squared brackets in Eq. (7), begins to become im-

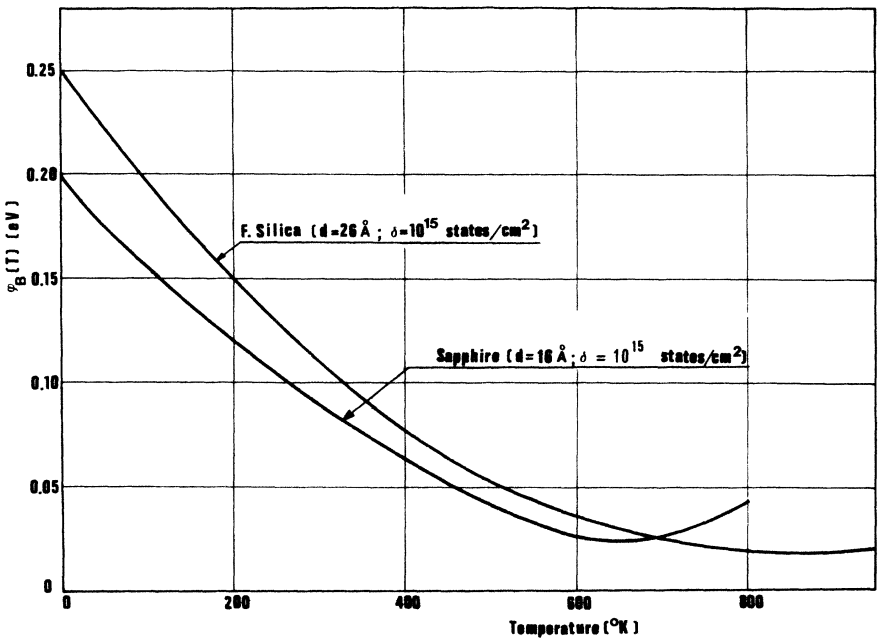

FIG. 4. Potential barrier height $\varphi_B(T)$ vs temperature from Eq. (7) with $\chi_m - \chi_s = \varphi_1$, $\epsilon_r^* = \frac{1}{3}\epsilon_r$. For sapphire substrate $\varphi_1 = 0.2$ eV, $d = 1.6\times10^{-9}$ m, $\delta = 10^{19}$ states/m^2. For fused-silica substrate, $\varphi_1 = 0.25$ eV, $d = 2.6\times10^{-9}$ m, $\delta = 10^{19}$ states/m^2. The values of the dielectric constants ϵ_r for sapphire and silica have been taken as a function of temperature by von Hippel's (Ref. 35) and MIT (Ref. 36) reports.

portant and the barrier behavior is inverted.

Such an inversion in the film resistivity at very high temperatures has been actually observed in experiments at temperatures above 500°C. Equation (10), taking into account the temperature dependence of $\varphi_B(T)$, gives a surface conductance versus temperature which fits well the experimental behavior in a very large temperature range (from about 20°K up to 700°K), as seen in paper II. The generally assumed exponential behavior, typical of an activated process, seems thus an approximation valid in a reduced-temperature range.

Another important point, which also has well established experimental support, is the dependence of the thermal coefficient of resistance on the conductivity of the film, i.e., on the gap width d between metal islands. Equation (10), together with Eq. (7), fits the experimental results very well, as shown in Fig. 12 in paper II.

Concerning the noise power spectrum given by Eq. (28), the most important point to be discussed is the lifetime-distribution function of the ionized donor centers. As already pointed out, such a $1/\tau$ distribution function has been obtained by neglecting the electron transition from donors to the conduction band of the insulator. This approximation is valid when the temperature is so low that the probability of an electron being excited from a donor center to the conduction band of the insulator is much smaller than the tunneling probability from the donor center to a metal island.

To a rough approximation, it can be assumed that Eq. (31) stands for $T < T^*$, where T^* is given by the relation

$$\exp(-\varphi_1/kT^*) \approx \exp\{-[2(2m\varphi_1)^{1/2}/\hbar]d/2\}, \quad (36)$$

that is,

$$T^* = \sqrt{\varphi_1}\,\hbar/k(2m)^{1/2}d. \quad (37)$$

With the values of φ_1 and d given above, Eq. (37) gives for sapphire $T^* = 464$ °K and for silica $T^* = 436$ °K.

For temperature above T^*, it is easily seen that the longest ionization times, which refer to donor centers farthest away from metal islands, are reduced and contribute only partially to the occupation function of the center. This is because, to the barrier height fluctuation, only the transitions from donor centers to metal islands are active. On the other hand, donor centers located very near the metal islands, which supply the shortest ionization times to the distribution function, are hardly affected by thermal excitation.

Thus, the expected result is that the distribution function decreases more rapidly than $1/\tau$ and the power spectrum $\psi_F(\omega)$ becomes more "white" on account of weakening in the low-frequencies range. This is what actually happens in experiments, as reported in paper II. No exception has been found to this effect, which is one of the best reproducible results observed in metal film at high temperature. For the same reason the integral spectrum, given by Eq. (28) for temperatures below T, drops when temperature is well above T^*. In the following paper it will be shown that not only the shape of the

power spectrum but also its absolute value agree well with the experimental results.

APPENDIX

First, Nyquist's theorem is derived in a rather generalized form, which applies to systems in which a fluctuation in the energy is associated with a fluctuation in the entropy, as in the specific case considered in the text.

Let us consider a physical system whose Hamiltonian $H(V)$ depends explicitly on a quantity $V(t)$ which is a function of time and can fluctuate for thermodynamical reasons around its equilibrium value V_0. It is assumed that averaging over any other possible variable deriving from other degrees of freedom of the system has already been performed on H, which is thus reduced to a function of V only. Similarly let us define the entropy of the system $S(V)$ as

$$S(V) = -k \ln P(V), \tag{A1}$$

where $P(V)$ is the number of states of the system corresponding to the energy $H(V)$ defined above. It follows from statistical thermodynamics considerations that the probability density $q(V)$ of a definite value V of the quantity $V(t)$ is given at a temperature T by

$$q(V) = CP(V)e^{-H(V)/kT} = Ce^{-F(V)/kT}, \tag{A2}$$

where C is a normalizing constant and $F(V) = H(V) - TS(V)$ is the free energy of the system, which is also a function of V and has a minimum for $V = V_0$, if during a fluctuation of V the temperature and the volume of the system remain constant.

Expanding $F(V)$ in power series of ΔV, where $\Delta V = V(t) - V_0$ represents a fluctuation of $V(t)$ with respect to its mean value V_0 and is a small quantity, the following relation is obtained:

$$F(V) = F(V_0) + \tfrac{1}{2}F''(V_0)\Delta V^2, \tag{A3}$$

where the primes mean derivative with respect to V and the terms in ΔV to a power higher than 2 have been dropped. The first derivative $F'(V_0)$ is zero, owing to the equilibrium condition at $V = V_0$. From Eqs. (A3) and (A2) it follows that

$$\langle V^2 \rangle = kT/F''(V_0), \tag{A4}$$

which represents the generalized Nyquist theorem for the fluctuation of $V(t)$.

To apply this expression to the specific case considered in the text about the fluctuation of the number of ionized donor centers in the insulator, consider a slide of dielectric material contained in the gap between metal islands as shown in Fig. 5. As in the text, it is assumed that the electric charge due to ionized donors is uniformly spread out with

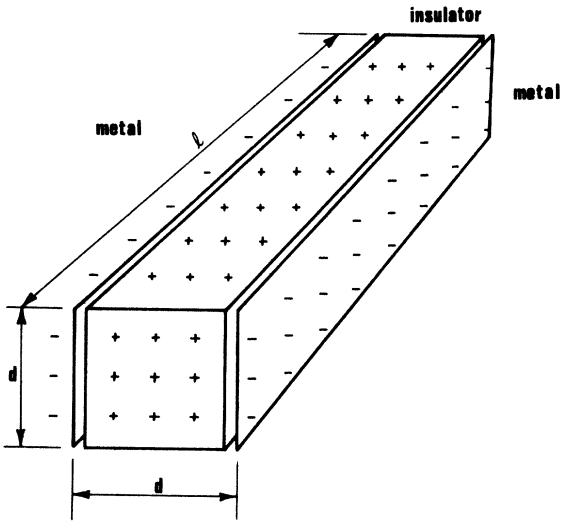

FIG. 5. Schematic representation of the dielectric between two metal islands showing the electric charge due to donor centers ionization.

the dielectric, which is only a few atomic layers thick, while an opposite charge is localized on the metal at the interface with the dielectric.

Using the same notation as in the text, the free energy part of the system depending on n, where n is the number of ionized donors in the slide, can be written

$$F(n) = H_1(n) + H_2(n) - TS_c(n), \tag{A5}$$

where $H_1(n)$ represents the electrostatic energy associated with n ionized donors, $H_2(n)$ is the energy change of n electrons which have left the donor centers in the insulator to populate electronic levels near the Fermi level in the metal, and $S_c(n)$ represents the configurational entropy of the system, related to the degree of ionization of the donor centers. The thermal part of the entropy does not appear in Eq. (A5) because transitions from donors to the insulator conduction band have been considered negligible and furthermore in the metal, for fermions, it is practically independent of n. From simple electrostatics considerations and from the energy plot of Fig. 5 we obtain

$$H_1(n) = \frac{1}{24}\,\frac{e^2n^2}{\epsilon_0\epsilon_r^* l}, \tag{A6}$$

$$H_2(n) = -n(\chi_m - \chi_s - \varphi_1), \tag{A7}$$

$$S_c(n) = -kN\left(\frac{n}{N}\ln\frac{n}{N} + \frac{N-n}{N}\ln\frac{N-n}{N}\right)$$

$$\simeq -kn\ln\frac{n}{N}, \tag{A8}$$

where, in Eq. (A8), N is the total number of donor

centers in the slide and account has been taken that $n \ll N$. Equation (A8) is a well-known expression for the configurational entropy of a system of $N-n$ electrons which can occupy N different states, all having the same energy.

To apply Eq. (A4) it should be considered that the fluctuation of n is around the value $\bar{n}$ given, as a good approximation, by

$$\bar{n} = Ne^{-\varphi_F(T)/kT}. \tag{A9}$$

Substituting Eqs. (A6), (A7), and (A8) in Eq. (A5) and taking the first derivative with respect to n, one gets

$$F'(\bar{n}) = \frac{1}{12}\,\frac{e^2}{\epsilon_0\epsilon_r^* l}\,\delta de^{-\varphi_F(T)/kT} - \chi_m + \chi_s + \varphi_1 - \varphi_F(T). \tag{A10}$$

Taking into account that $\varphi_F(T) = \varphi_1 - \varphi_B(T)$, the condition

$$F'(\bar{n}) = 0, \tag{A11}$$

thus exactly corresponds to Eq. (7) of the text, if the first term within large square brackets, which takes into account electron transitions to the conduction band of insulator, is dropped. This is a consequence of the fact that the thermal entropy has not been considered in Eq. (A5), which is thus correct if the temperature is not too high.

The second derivative of the free energy becomes

$$F''(n) = \frac{1e^2}{12\epsilon_0\epsilon_r^*} - \frac{kT}{\bar{n}} \simeq \frac{1}{12}\,\frac{e^2}{\epsilon_0\epsilon_r^* l}. \tag{A12}$$

The last step in Eq. (A12) has been made taking into account that from Eqs. (A11), and (A12), with the position $\varphi_1 - \chi_m + \chi_s = 0$ made in the text, it turns out

$$\frac{1}{12}\,\frac{e^2}{\epsilon_0\epsilon_r^* l} = \frac{\varphi_F(T)}{\bar{n}} \gg \frac{kT}{\bar{n}}. \tag{A13}$$

Finally, from Eq. (A4), we get

$$\langle \Delta n^2 \rangle = 12\epsilon_0\epsilon_r^* lkT/e^2. \tag{A14}$$

From Eq. (19) of the text, taking into account that

$$\langle \Delta n^2 \rangle = \int_{-\infty}^{\infty} \phi_n(\omega)\, d\omega = \frac{12\epsilon_0\epsilon_r^* lkT}{e^2}, \tag{A15}$$

Eq. (31) is finally obtained.

---

*Permanent address: Istituto di Fisica Generale, Università di Torino, Italy.
†Permanent address: Istituto di Fisica Sperimentale, Politecnico di Torino, Italy.

[1]S. S. Minn, J. Rech. C. N. R. 51, 131 (1960).
[2]R. C. Neugebauer and M. Webb, J. Appl. Phys. 33, 74 (1962).
[3]T. E. Hartman, J. Appl. Phys. 34, 943 (1963).
[4]D. S. Herman and T. E. Rhodin, J. Appl. Phys. 37, 1594 (1966).
[5]A. A. Milgram and C. Lu, J. Appl. Phys. 37, 4773 (1966).
[6]K. Van Steensel, Philips Res. Rep. 22, 246 (1967).
[7]R. M. Hill, Proc. R. Soc. A 309, 377 (1969).
[8]R. Kiernan and D. W. Stops, Nature 224, 907 (1969).
[9]R. Kiernan and D. W. Stops, J. Vac. Sci. Technol. 9, 433 (1971).
[10]A. Barr and R. D. Finney, Thin Solid Films 24, S11 (1974).
[11]L. L. Kazmerski and D. M. Racine, J. Appl. Phys. 46, 791 (1975).
[12]L. L. Kazmerski, D. M. Racine, and M. S. Ayyagary, J. Appl. Phys. 46, 2658 (1975).
[13]B. Abeles, Ping Sheng, D. M. Coutts, and Y. Arie, Adv. Phys. 24, 407 (1975).
[14]K. Uozumi, M. Nishiura and A. Kinbara, J. Appl. Phys. 48, 818 (1977).
[15]J. P. Borel, J. Phys. Radium 17, 224 (1956).
[16]A. Blanc-Lapierre and N. Nifontoff, J. Phys. Radium 17, 230 (1956).
[17]M. S. Osman, MIT Rept. No. ESL-R-149, 1962 (unpublished).
[18]L. S. Kremenchugsky, A. F. Mlnyev, and V. R. Samoilov, Ukr. Fiz. Zh. 86, 762 (1963).
[19]J. L. Williams and R. K. Burdett, J. Phys. C 2, 298 (1969).
[20]J. L. Williams and I. L. Stone, J. Phys. C 5, 2105 (1972).
[21]F. N. Hooge, Phys. Lett. 29 A, 139 (1969).
[22]N. M. Tallan and D. P. Detwiler, J. Appl. Phys. 34, 1650 (1963).
[23]R. C. Nelson, J. Appl. Phys. 34, 629 (1963).
[24]In this paper reference is made to the small volumes of the order of $10\times10\times100$ Å^3 dominating the electrical conduction between metal islands.
[25]J. M. Ziman, *Principles of the Theory of Solids* (Cambridge U.P., Cambridge, England, 1965), p. 122.
[26]A more precise definition of , taking into account the actual very complicated island structure is discussed in Sec. III.
[27]J. G. Simmons, J. Appl. Phys. 35, 2472 (1964).
[28]J. G. Simmons, J. Appl. Phys. 35, 2655 (1964).
[29]S. Machlup, J. Appl. Phys. 25, 341 (1954).
[30]A. L. McWhorter, MIT Lincoln Lab., Rept. No. 80, 1955 (unpublished).
[31]A. L. McWhorter, *Semiconductor Surface Physics* (University of Pennsylvania Press, Philadelphia, 1957).
[32]A. Van der Ziel, *Source, Characterization, Measurement* (Prentice-Hall, Englewood Cliffs, N. J., 1970), p. 101.
[33]A. Many, Y. Goldstein, and N. B. Grover, *Semiconductors Surfaces* (North-Holland, Amsterdam, 1971), p. 432.
[34]T. Botila, S. T. Sladaru, and N. Croitoru, *Proceedings of the Fifth International Conference on Amorphous,*

Liquid, Semiconductors, (Garmisch-Partenkirchen, (1973), Vol. 2, p. 799.
[35]R. von Hippel, *Les dielectriques et leurs applications* (Dunod, Paris, 1961), p. 373.
[36]MIT, Technical Rept. No. 119, "Tables of dielectrical materials," Vol. V, p. 70, 1957 (unpublished).