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$1/f$ noise from surface generation and annihilation: Application to metal films

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A model in which a conserved quantity enters and leaves a diffusing medium at a bounding surface is discussed. Fluctuations in the rate at which the quantity enters or leaves lead to density fluctuations in the medium. These density fluctuations diffuse and eventually leave the medium with a time dependence correlated with the initial fluctuation. The Fourier transform of this time dependence gives $1/f$ noise. This model is applied to voltage noise in thin metal films. The diffusing quantities which are used and which affect the conductivity are phonon energy and vacancies. Order-of-magnitude agreement with experiment is obtained.

I. INTRODUCTION

At present there does not appear to be a satisfactory explanation of $1/f$ noise in metals. The trapping mechanism of McWhorter[1] does not seem applicable to such systems. Because diffusion effects can be long lived, many have attempted to explain flicker noise on the basis of diffusion. Unfortunately, it is found typically that diffusion transport noise levels off at low frequencies and goes as $f^{-3/2}$ or f^{-2} at high frequencies.[2]

A method of producing $1/f$ noise by generation and annihilation at a surface will be presented here. While diffusion will enter into the model, the usual transport noise will not be treated. It will be assumed that the entering and subsequent disappearance of a quantity, such as energy, at the surface of a material is the only effect that contributes to the noise. As long as this quantity is in the material, its effect on properties, such as electrical conductivity, is considered constant. Thus diffusion noise as such is not considered in this paper. However, diffusion does modify the time between the creation and annihilation at the surface and thus it affects the noise spectrum. Also the average production of the quantity at the surface, that would contribute to diffusion noise, will be shown not to contribute to the noise considered here. Only the random fluctuations from the average will contribute. Van Vliet, van der Ziel, and Schmidt[3] have recently considered a model similar to the one treated here, but they looked at the diffusion noise primarily.

Recently there have been measurements on thin metal films[4–6] that show a strong temperature dependence of the noise. Also it was shown that the substrate can have a strong influence on the noise at low temperatures. The general theory developed in Sec. II will be applied to the flow of energy in Sec. III to explain low-temperature noise, and to the flow of vacancies in Sec. IV to explain the high-temperature noise in the films.

II. MODEL FOR $1/f$ NOISE

The model to be used here is that of a diffusing medium assumed to have a bounding plane that is the source and a sink for the diffusing quantity. This quantity is assumed to be conserved in the bulk material. Thus it might be charged particles, energy, vacancies, etc. In the following discussion this quantity will simply be called "particles."

Let U_i and U_o be the number of particles per unit area per unit time going into and out of the medium, respectively, at the bounding plane. If noise measurements start at time $t = 0$, the change Δn in the number of particles per unit area in the medium up to time t is

$$\Delta n = \int_0^t (U_i - U_o)dt' . \tag{1}$$

The Fourier transform of Δn for measurements up to $t = T$ is

$$\Delta n_F = (2\pi)^{-1} \int_0^T e^{-i\omega t} dt \int_0^t (U_i - U_o)dt' . \tag{2}$$

For steady-state conditions since the particles are conserved, U_i and U_o are equal on the average so that Δn_F will depend only on the fluctuations of these quantities from the average. Suppose that in the time interval t_0 to $t_0 + \Delta t$ there is a fluctuation from the average in the number of ingoing particles, δn. Δt is assumed to be much less than $1/\omega$ with ω a frequency for which the noise is measured. However Δt is large enough so that $\delta n >> 1$. If coordinate axes are chosen such that the x axis is perpendicular to the bounding plane with this plane at $x = 0$, the fluctuation δn gives an excess of particles at $x = 0$ for $t = t_0$. A solution of the one-dimensional diffusion equation for the density σ of particles that corresponds to an initial distribution close to $x = 0$ with a perfect sink at $x = 0$ is

$$\sigma_1(x,\tau) = \alpha(x/\tau^{3/2}) \exp(-x^2/4D\tau) . \quad (3)$$

Differentiation with respect to x gives a particle current density at the plane of $D\alpha/\tau^{3/2}$. Here τ is $t - t_0$, D is the diffusion coefficient, and α is a constant. If there are moderate reflections of particles at the bounding surface, one may account for them by adding a solution of the diffusion equation that is nonzero at the origin and is proportional to $\tau^{-3/2}$ there. Such a solution is

$$\sigma_2(x,\tau) = \beta \frac{\partial \sigma_1(x,\tau)}{\partial x} , \quad (4)$$

with β a constant. Then

$$\sigma = \sigma_1 + \sigma_2 . \quad (5)$$

The term σ_2 simply redistributes the particles near $x = 0$ and does not give a net number of particles. It also gives no particle current at the bounding plane. Therefore, only the term σ_1 proportional to δn will be used in treating noise due to particles entering and leaving the medium. On the average the number of particles still in the medium at elapsed time $\tau >> \Delta t$ due to this fluctuation is

$$\delta n_\tau = \int_0^\infty \sigma(x,\tau) dx = 2D\alpha\tau^{-1/2} , \quad (6)$$

and the number disappearing at the plane sink per unit area per unit time is

$$\delta U_o = -\frac{d(\delta n_\tau)}{d\tau} = D\alpha\tau^{-3/2} \quad (7)$$

in agreement with the current density found above at the origin from Eq. (3). Equation (7) is the lifetime distribution for a pulse of particles at $\tau = 0$. With these equations one apparently can say immediately that the noise power is proportional to $f^{-3/2}$ by simply applying the results of the noise discussion by Halford.[7] However, this $f^{-3/2}$ noise is associated with the average U_o. In any case van Vliet *et al.*[3] have shown that the $f^{-3/2}$ dependence does not apply to surface noise but only to volume diffusion noise.

The constant α is difficult to evaluate exactly since it depends on the details of the first few scatterings leading to the diffusion. This is because most of the particles escape from the medium in the first few scatterings. The diffusion equation solution, Eq. (3), does not hold until a large number of scatterings or jumps leading to the diffusion has taken place. Therefore, to find α, one would need a detailed knowledge of the microscopic structure near the bounding plane and its effects on the first few scatterings. Ordinarily this is not known, so only order-of-magnitude calculations will be considered here. One may evaluate the order of magntiude of α by comparing with a one-dimensional random walk in which the time between steps is a constant τ_0 and each step is of length $l = (2D\tau_0)^{1/2}$. For δn particles at $x = l$ when $\tau = 0$ and for annihilation of all particles reaching $x = 0$, the distribution for $\tau >> \tau_0$ is that of the σ_1 term, Eq. (3), with

$$D\alpha = \delta n (\tau_0/2\pi)^{1/2} . \quad (8)$$

For order-of-magnitude calculations, $D\alpha$ for the actual diffusion will be chosen to be that of Eq. (8) with τ_0 the average collision or jump time leading to the diffusion. Again for order-of-magnitude calculations the expressions in Eqs. (6) and (7) will be assumed to be valid for $\tau > \pi\tau_0/2$ with no diffusion effects for smaller times.

Substitution of the δU_o of Eq. (7) for the U_o of Eq. (2) leads to a contribution to Δn_F from this fluctuation of

$$\delta\Delta n_F = \frac{\delta n}{\pi} \int_{\pi\tau_0/2}^{T-t_0} e^{-i\omega\tau} d\tau \left[\frac{\tau_0}{2\pi\tau}\right]^{1/2} e^{-i\omega t_0} . \quad (9)$$

Since in most noise measurements for the frequency range of $\omega/2\pi$ and for most t_0 the inequalities $\omega\tau_0 << 1$ and $\omega(T - t_0) >> 1$ will hold, the limits on the integral may be taken from zero to infinity. Then

$$\delta\Delta n_F = \delta n(1 - i)\tau_0^{1/2}/2\pi\omega^{1/2} . \quad (10)$$

The spectral density resulting from this fluctuation is then

$$S_{\delta n} = 8\pi^2 |\delta\Delta n_F|^2/\Delta t$$
$$= 4\delta n^2 \tau_0/\omega\Delta t \ . \quad (11)$$

Since the fluctuations at the various times t_0 are independent, the spectral density due to all such fluctuations is

$$S_i = \left[\sum_{t_0=0}^{T} S_{\delta n}\Delta t\right]/T = 4\langle\delta n^2\rangle\frac{\tau_0}{\omega\Delta t} \ . \quad (12)$$

Here $\langle\delta n^2\rangle$ may be considered to be either a time or ensemble average of the square of the fluctuation in the time Δt. This $1/f$ power spectrum results from particle creation pulses and subsequent correlated annihilation at a surface. The diffusion mechanism is used solely to give the time distribution of the annihilation. While the particles are in the medium no diffusion noise is taken into account.

There are statistical fluctuations also in U_o. These should lead to a resultant noise power spectrum identical to that of Eq. (12). This is because a fluctuation giving a decrease (or increase) in U_o will give the same effects in the medium as an equal increase (or decrease) in U_i. Since the noise power depends on the square of the fluctuation, the two kinds of fluctuation will give equal effects on this power. Hence, the total power spectrum per unit area is just twice that of Eq. (12), or

$$S = 8\langle\delta n^2\rangle\frac{\tau_0}{\omega\Delta t} \ . \quad (13)$$

III. NOISE FROM ENERGY FLUCTUATIONS

In the experiments of Dutta, Eberhard, and Horn[5] noise in metal films appears to arise from at least two different sources, depending on the temperature and substrate. At low temperatures with a fused quartz substrate the noise in copper is appreciably greater than with a sapphire substrate. It was suggested by those authors that this additional noise is associated with the equilibrium temperature fluctuations of Voss and Clarke.[8] This noise will be discussed using the results of Sec. II for phonon diffusion in the substrate.

Fluctuations of energy in a metal film cause changes in the conductivity, which lead to noise in the potential across the ends of the current-carrying film. The energy fluctuations can be produced by phonons going between the film and substrate. For the fused quartz substrate the film can act as a source and sink for phonons since the thermal conductivity of copper is large compared to that of quartz. Because of the large conductivity of sapphire, the film probably does not act as an energy sink for that substrate. While the noise in Eq. (13) was derived for the diffusing medium, in this case it also applies to the film since the energy fluctuations leading to the noise are equal and opposite in the substrate and film. If U_i corresponds to phonon energy per unit area per unit time, the voltage noise power divided by the square of the potential should be

$$\frac{S_v}{V^2} = AS\left[\frac{d\rho}{dT}\right]^2/(\rho C_v\Omega)^2 \ , \quad (14)$$

where ρ is the resistivity of the film, T is the absolute temperature, C_v is the heat capacity per unit volume, Ω is the film volume, and A is the area of contact between the film and substrate. The δn in Eq. (13) is now δE, the energy fluctuation. The equilibrium mean-square fluctuation of the energy in a volume $\Delta\Omega$ in the metal film is

$$\langle\delta E^2\rangle = k_B T^2 C_v \Delta\Omega \ . \quad (15)$$

For phonon energy transported toward the surface at an angle θ relative to the surface normal, $\Delta\Omega$ per unit area is $s_m\Delta t\cos\theta$, where s_m is an average sound speed in the film. Then summation over the angles gives

$$S = (\tfrac{1}{4}\langle\eta^2\rangle_{\rm av} s_m k_B T^2 C_v)(8\tau_0/\omega) \quad (16)$$

in which $\langle\eta^2\rangle_{\rm av}$ is the average of the square of the transmission coefficient of the phonons at the film-substrate interface. In the high-temperature limit where $\rho \propto T$ and $C_v = 3Nk_B/\Omega$ (N is the number of film atoms), the noise approaches a constant independent of temperature. For film thickness w

$$NS_v/V^2 \rightarrow 2\tau_0\langle\eta^2\rangle_{\rm av} s_m/3w\omega \ . \quad (17)$$

In copper $w = 800$ Å was assumed. The temperature dependence of C_v was found from the Debye theory with a Debye temperature of 343 K. Also the temperature dependence of the conductivity ρ was found from the Bloch-Gruneisen law[9] based on the Debye theory. The average speed of sound s_m was simply taken as the average over the longitudinal and two transverse modes in copper at room temperature, 3×10^5 cm/s. The transmission coefficient η at the interface was found by applying the classical boundary conditions to a plane interface and using the tabulated sound speeds of the various polarizations in the two media. The square of the

transmission coefficient was averaged over the angles of incidence (along with a factor of $\cos\theta$) and averaged over the polarizations. The result was $\langle \eta^2 \rangle_{av} \cong \frac{1}{4}$. The time τ_0 was found by using the order-of-magnitude kinetic formula for lattice thermal conductivity,

$$\kappa = Cs^2\tau_0/3 , \tag{18}$$

where κ for fused quartz was taken as 0.04 cal/s cm °C, the specific heat C of the quartz was chosen as 0.4 cal/cm^3 °C and the average speed of sound s in the quartz was found to be 4.5×10^5 cm/s. This gives $\tau_0 \cong 1 \times 10^{-12}$ s. No attempt was made to find a temperature dependence of τ_0. While the simple averaging methods and the method of finding $\langle \eta^2 \rangle_{av}$ described are not quite correct, they should give satisfactory order-of-magnitude values. The results of the calculations of NS_v/V^2 versus temperature are shown in the curve of Fig. 1 for a frequency of 20 Hz. The experimental points shown in this figure are those of Dutta, Eberhard, and Horn[5] for copper on quartz with the values for copper on sapphire subtracted to obtain the approximate energy fluctuation part. The sapphire substrate curve used was that of these authors multiplied by 0.8 so that the sapphire and quartz curves agree near 370 K. They assumed that the curves should agree near their peaks, but in Sec. IV it will be seen that the substrate might have appreciable effect near the peak. Dutta *et al.* state that the uncertainty in the absolute experimental values is around a factor of 3. The order-of-magnitude methods used in the theory also have at least this much uncertainty. Therefore, it appears that the experiment and theory are in satisfactory agreement.

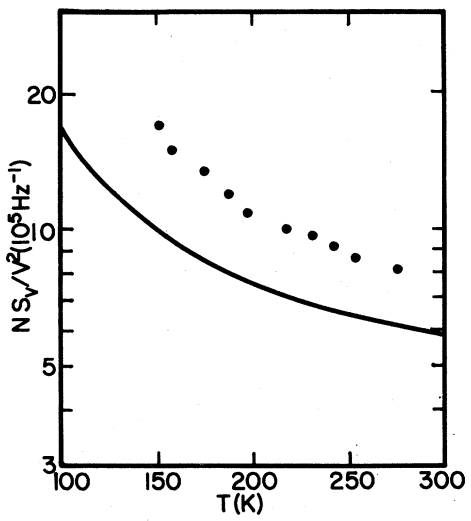

FIG. 1. The solid curve is a plot of energy fluctuation noise at 20 Hz calculated from Eq. (18). The experimental points are those of Dutta, Eberhard, and Horn.

IV. NOISE FROM VACANCY DIFFUSION

The part of the noise in metal films that varies rapidly with temperature does not appear to be explainable in terms of energy fluctuations. Eberhard and Horn[4] suggested that it might be due to vacancies. Therefore, the results of Sec. II will be applied to diffusion of vacancies to attempt an explanation of that part of the noise.

If it is assumed that vacancies are created primarily at the free surface of the metal film at random times, $\langle \delta n^2 \rangle / \Delta t$ of Eq. (13) is just $\langle U_i \rangle$. The density of vacancies σ_0 in the film should be of the order of

$$\sigma_0 = \tau_0 \langle U_i \rangle / b , \tag{19}$$

where b is the atomic nearest-neighbor distance in the film. Then Eq. (13) becomes

$$S = 8\sigma_0 b / \omega . \tag{20}$$

However, this expression should be modified to take account of the finite film thickness. To see qualitatively how this affects the noise, the unrealistic model that all vacancies reaching the film-substrate boundary are reflected will be assumed. Then an image method may be used to solve the diffusion problem. For thickness w of the film, in place of σ_1 of Eq. (3),

$$\sigma_1(x,\tau) = \frac{\alpha}{\tau^{3/2}} \sum_{p=-\infty}^{\infty} (-1)^p (x - 2pw) \times \exp\left[-\frac{(x-2pw)^2}{4D\tau} \right] . \tag{21}$$

Then in place of Eq. (9), one has

$$\delta\Delta n_F = (\tau_0/2\omega)^{1/2} e^{-i\omega t_0} (\delta n / \pi) I , \tag{22}$$

where

$$I = \int_{\pi\tau_0/2}^{T-t_0} \left[\frac{\omega}{\pi\tau} \right]^{1/2} e^{-i\omega\tau} d\tau \times \left[1 + 2\sum_{p=1}^{\infty} (-1)^p \exp\left[-\frac{p^2 w^2}{D\tau} \right] \right] . \tag{23}$$

This gives

$$S = 8\sigma_0 b \,|I|^2/\omega \,. \tag{24}$$

The quantity $|I|^2$ changes from one to zero as $\omega w^2/D$ goes from large values to zero. Of course, with the additional frequency dependence in I the spectrum will no longer be $1/f$ at higher temperatures. The voltage noise power spectrum is then given by

$$S_v/V^2 = AS\left[\frac{d\rho}{d\sigma_0}\right]^2 \Big/ (\Omega\rho)^2$$

$$= 8\,|I|^2 b\gamma \left[\frac{d\rho}{d\gamma}\right]^2 \Big/ (\omega w N \rho^2) \,. \tag{25}$$

Here γ is the atomic fraction of vacancies. Calculations were made for a silver film of 800 Å thickness. Mehrer and Seeger[10] give various diffusion parameters for silver. For monovancies the diffusion coefficient is

$$D_{1v} = a^2 \nu_{1v}^M \exp(-E_{1v}^M/k_B T) \,, \tag{26}$$

with $\nu_{1v}^M = 4 \times 10^{13}$/s and $E_{1v}^M = 0.86$ eV. The length a is the cubic cell edge length. For divacancies

$$D_{2v} = a^2 \nu_{2v}^M \exp(-E_{2v}^M/k_B T) \,, \tag{27}$$

with $\nu_{2v}^M = 4 \times 10^{13}$/s and $E_{2v}^M = 0.58$ eV. The atomic densities of mono- and divacancies γ_{1v} and γ_{2v} are related by

$$\gamma_{2v} = 6\gamma_{1v}^2 \exp[(\Delta S_{2v}/k_B) + E_{2v}^B/k_B T] \,, \tag{28}$$

with $S_{2v} = 2.6 k_B$ and $E_{2v}^B = 0.24$ eV. The factor 6 results from the six possible orientations of a divacancy in a face-centered-cubic lattice. Another parameter given by Mehrer and Seeger is $d\rho/d\gamma_{1v} = 1.6 \times 10^{-4}\ \Omega$ cm. These values are all for bulk material. Often in metal films the diffusion coefficients are greater because of "short circuits" resulting from greater densities of imperfections.[11] However, if the films are annealed on the order of an hour at temperatures comparable to the higher temperatures used in the noise experiments, the films have close to the bulk properties.[12] Therefore it will be assumed that the values listed above apply to these films. However, one set of bulk parameters cannot be applied to thin films. These parameters are the entropy and formation energy of about one electron volt for forming monovacancies. In the bulk material the vacancies are formed primarily at imperfections such as dislocations and grain boundaries. When the vacancies are formed primarily at the surface, the formation energy is presumably somewhat less, possibly on the order of E_{2v}^B, the energy to separate divacancies into monovacancies, Eq. (28). In addition to the constants already given the following were used: $a = 4.09$ Å, $b = 2.89$ Å, and $\rho = (5 \times 10^{-9} T)\ \Omega$ cm.

If the monovacancy constants are used and $\tau_0 \cong a^2/D_{1v}$, then for a 20 Hz frequency and for temperatures below 400 K it is found that $\omega\tau_0$ is of the order one or greater and the results of Sec. II do not apply. Also if the γ in Eq. (25) is assumed to correspond to that of monovacancies and this γ_{1v} is found by putting the experimental 400 K value of NS_v/V^2 into that equation, Eq. (28) then gives $\gamma_{2v} > \gamma_{1v}$. Both of these results indicate that γ in Eq. (25) should be associated with divacancies. For order-of-magnitude calculations the quantity $d\rho/d\gamma_{2v}$ will be chosen the same as $d\rho/d\gamma_{1v}$ $= 1.6 \times 10^{-4}\ \Omega$ cm.

One result may be found immediately from the divacancy constants. This is the position of the peak of the noise curve. One would expect that this would occur at a temperature for which the vacancies can diffuse the distance corresponding to the film thickness in the time $1/\omega$. In agreement with this idea it is found that for any reasonable choice of the divacancy formation energy the peak occurs when the parameter in Eq. (23) w^2/D_{2v} is approximately equal to $2/\omega$. Then for silver, Eq. (27) gives a temperature for the peak of 405 K, almost exactly the experimental value.[6] For copper Mehrer and Seeger[13] give $\nu_{2v}^M = 3 \times 10^{13}$/s and $E_{2v}^M = 0.66$ eV. The peak temperature for copper is then calculated to be 476 K as compared with the experimental value[6] of about 490 K. The uncertainty in the values of E_{2v}^M lead to an uncertainty of around 30° in these temperatures. If a model had been chosen in which all the vacancies were annihilated at the film-substrate boundary, these peak temperatures would have been calculated to be about 25° lower. Thus these temperatures do not depend strongly on the detailed model used for that boundary. Now to obtain a theoretical curve to compare with the experiment, let us assume that

$$\gamma_{2v} = \exp[(\Delta S/k_B) - E_{2v}^F/k_B T] \,, \tag{29}$$

with $\exp(\Delta S/k_B) = 1.6$ and $E_{2v}^F = 0.23$ eV to fit the experimental value at the peak and the experimental slope between 300 and 350 K where $|I|^2 = 1$. The value for γ_{2v} from the experimental noise value given by Eq. (25) at the 405-K peak that was used in this fit is 2×10^{-3}. The divacancy formation energy found here is very close to E_{2v}^B of

Eq. (28). The resulting curve of NS_v/V^2 is shown versus temperature at 20 Hz in Fig. 2. The experimental points are those of Dutta, Eberhard, and Horn[5] for a silver film on a sapphire substrate. The curve was plotted only down to 250 K because at that temperature $\omega\tau_0$ is comparable to one and the theory is no longer applicable. Presumably some other noise mechanism is important for lower temperatures, possibly diffusion of larger aggregates of vacancies. The primary lack of agreement is in the width of the peak. If a more realistic treatment of the film-substrate boundary had been made, the peak would have been broader. Vacancies trapped or diffusing in the substrate would cause the vacancies to arrive at the free silver surface at later times for the higher temperatures than in the total reflection model and thus the noise would drop less rapidly with increasing temperature. However, little is known about the vacancy diffusion properties of the substrate, so a combined treatment of diffusion in the film and substrate was not attempted.

The experiments show a temperature-dependent deviation from strict $1/f$ dependence of the noise. As pointed out above, this is to be expected at the peak of the curve. However, if there are other diffusion mechanisms that are important at lower temperatures, there could be finite thickness effects for them that would give some deviation from $1/f$ behavior for other temperatures. Of course, any other effects not considered in the diffusion model such as trapping or bulk annihilation of vacancies could also give such deviations.

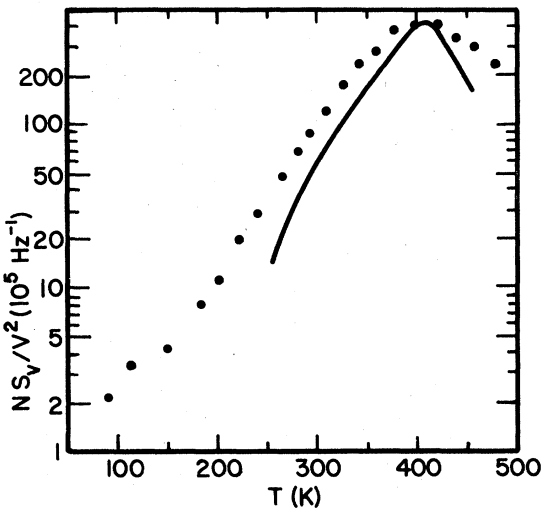

FIG. 2. The solid curve is a plot of noise at 20 Hz resulting from creation and annihilation of divacancies at the silver film surface as calculated from Eq. (29). The experimental points are those of Dutta, Eberhard, and Horn.

V. SUMMARY

Noise arising from a quantity entering and leaving a medium was investigated. The noise was generated by statistical fluctuations followed by correlated fluctuations as a result of diffusion in the medium. It was found that the noise had a $1/f$ spectrum. In the application to noise due to energy flow through the copper-film—quartz-substrate interface the order of magnitude of all parameters could be found. There was good agreement between theory and experiment, both in magnitude and in temperature dependence. For noise resulting from vacancy formation at the metal-film surface not all parameters were known. However, the peak temperature is largely independent of the unknown quantities and there was good agreement between theory and experiment for that temperature. The entropy and energy parameters necessary to obtain order-of-magnitude agreement with experiment did not seem unreasonable. Therefore, it appears that this diffusion model is successful in explaining much of the noise in thin metal films.

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