

Temperature dependence of noise processes in metals

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Here we consider the analysis of the temperature and frequency dependences of the power spectral density $S_V(f, T)$ of excess low-frequency noise. Two limiting cases that arise naturally from a superposition of thermally activated relaxation processes are investigated. The Lorentzian spectra associated with various relaxation times $\tau = \tau_0 e^{E/kT}$ are supposed to arise from either (i) a distribution $D(E)$ of activation energies E (invoked in the Dutta-Horn model), or (ii) a distribution $H(\tau_0)$ of prefactors τ_0 (as required for activated diffusion processes). Much can be learned from the analysis of noise spectra with distinctive spectral features or known temperature-dependent variance $\langle \delta V^2(T) \rangle$, as the analysis of noise from H^+ diffusion in Nb films has illustrated. In the absence of such distinguishing features or independent data, particularly with $1/f$ noise, we find that there is generally insufficient information to distinguish even these limiting models, much less to determine their parameters. We have analyzed the resistance fluctuations of small-metallic-film conductors. We find that existing $1/f$ noise data from noble metals can be fit by either model with a wide range of assumptions about the variance $\langle \delta V^2(T) \rangle$. In contrast new $1/f$ noise data from Cr films are inconsistent with either model unless strong *ad hoc* temperature-dependent variance is postulated.

I. INTRODUCTION

Low-frequency conductance fluctuations in a variety of systems are thought to be associated with thermally activated processes. The temperature dependence of spectral features in the low-frequency excess noise of silicon on sapphire,¹ Bi films,² Si inversion layers,³ thick-film resistors,⁴ and metal-insulator-metal tunnel junctions⁵ provide direct evidence for such a claim. Measured symmetry properties of the local conductivity fluctuations of Bi films suggest a microscopic model connecting their $1/f$ noise to mobility fluctuations associated with "Snoek-like" internal friction mechanisms.^{2,6} Less direct evidence supports the claim that the featureless $1/f$ noise spectra $S_V(f, T)$ of metal films arises from a superposition of thermally activated Lorentzian spectra.^{7,8} In this case the only evidence available is qualitative agreement of the observed relationship between $(T/S)\partial S/\partial T$ and $(f/S)\partial S/\partial f$ with that predicted by such a model.

We have recently reported the temperature and frequency dependence of the excess noise of Nb films;⁹ there is considerable evidence that the noise arises from conductance fluctuations associated with scattering from hydrogen impurities. The fluctuation dynamics are that of one-dimensional diffusion with temperature dependence due to a diffusion coefficient $\mathcal{D}(T) = \mathcal{D}_0 e^{-E/kT}$. As such, the excess noise spectrum $S_V(f, T)$ may be written as a superposition of thermally activated Lorentzian spectra, analogous to the Dutta-Horn^{7,8} representation for $1/f$ noise, but with the various relaxation times $\tau = \tau_0 e^{E/kT}$ arising from a distribution of prefactors $\tau_0 = (\mathcal{D}_0 k^2)^{-1}$ determined by the spatial Fourier transform of the specimen geometry, $\Omega(k)$.

In this paper we consider the analysis of noise spectra arising from a superposition of thermally activated processes. This general problem has been a central issue for many years in the analysis of anelastic relaxation.¹⁰ The mathematics of our analysis is not new, but does aim to resolve some active questions of interpretation of electrical noise experiments. In particular, we examine the relationship between the temperature and frequency dependences for the two limiting cases where the various relaxation times $\tau = \tau_0 e^{E/kT}$ arise from (i) a distribution $D(E)$ of activation energies E , and (ii) a distribution $H(\tau_0)$ of prefactors τ_0 . The first case is similar to the $1/f$ noise model of Dutta, Dimon, and Horn,^{7,8} a generalization of an idea first proposed by van der Zeil¹¹ and Du Pre.¹² Predictions for the two models are compared with the $1/f$ noise of Ag films,¹³ hydrogen diffusion noise in Nb,⁹ and new Cr $1/f$ noise data. Analysis of the noise from H^+ in Nb is particularly interesting since the physical mechanism is understood. In this case we find that pronounced spectral features and knowledge of the variance $\langle \delta V^2(T) \rangle$ allow discrimination between the two models. Such discrimination is not possible with featureless $1/f$ noise. Furthermore, without independent knowledge of $\langle \delta V^2(T) \rangle$ the large number of free parameters makes it possible to fit most any "(1/f)-like" $S_V(f, T)$ with either model.

In the next section we introduce the necessary notation, define what is meant by a superposition of thermally activated processes, and with appropriate limiting assumptions, derive relationships between the scaling of the noise with frequency and with temperature. Noise from diffusion processes is discussed in Sec. III. In Sec. IV we compare the scaling relationships with the observed excess noise of various metal films. Finally, in the last section the results are discussed and summarized.

II. SUPERPOSITION OF THERMALLY ACTIVATED PROCESSES

Random fluctuations $\delta x(t)$ in a quantity x that relaxes to equilibrium with a single relaxation time τ have an autocorrelation

$$G_x(t) = \langle \delta x(0)\delta x(t) \rangle = \langle \delta x^2 \rangle e^{-|t|/\tau},$$

where $\langle \delta x^2 \rangle$ is the variance. The corresponding power spectrum $S_x(\omega)$, obtained using the Wiener-Kintchine theorem, has the Debye-Lorentzian form

$$S_x(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} G_x(t) = \langle \delta x^2 \rangle 2\tau / (1 + \omega^2\tau^2). \quad (1)$$

The linear superposition $X(t) = \sum_j x_j(t)$ of independent fluctuators x_j has a power spectrum

$$S_X(\omega) = \sum_j \langle \delta x_j^2 \rangle 2\tau_j / (1 + \omega^2\tau_j^2), \quad (2)$$

where $\langle \delta x_j^2 \rangle$ is the variance and τ_j is the relaxation time of the j th fluctuator.

Temperature dependence enters $S_X(\omega, T)$ through both the $\langle \delta x_j^2(T) \rangle$ and the $\tau_j(T)$. Recent discussion has focused on the temperature dependence due to the thermally activated relaxation processes $\tau_j(T) = \tau_{0j} e^{E_j/kT}$. To display that part of the problem, we assume that

$$\langle \delta x_j^2(T) \rangle = \langle \delta X^2(T) \rangle \langle \delta s_j^2 \rangle$$

so that the temperature dependence of the noise magnitude may be factored out of the summation.¹⁴ The normalized spectrum

$$S(\omega, T) = S_X(\omega, T) / \langle \delta X^2(T) \rangle = \sum_j \langle \delta s_j^2 \rangle 2\tau_j(T) / [1 + \omega^2\tau_j^2(T)] \quad (3)$$

depends on T only through the $\tau_j(T)$. Defining a temperature-dependent distribution of relaxation times

$$F(\tau, T) = \sum_j \langle \delta s_j^2 \rangle \delta(\tau - \tau_j(T)),$$

we have

$$S(\omega, T) = \int_0^{\infty} d\tau F(\tau, T) 2\tau / (1 + \omega^2\tau^2). \quad (4)$$

To present the issues, we need consider only two limiting cases for $F(\tau, T)$ where $\tau(T)$ is attributed to (i) a distribution of activation energies^{7,8} or (ii) a distribution of prefactors. For case (i), a distribution of energies

$$D(E) = \sum_j \langle \delta s_j^2 \rangle \delta(E - E_j)$$

yields the distribution of relaxation times

$$F(\tau, T) = kTD(E)/\tau, \quad (5)$$

where the T , E , τ , and τ_0 satisfy

$$\tau = \tau_0 e^{E/kT}. \quad (6)$$

For case (ii) we define a distribution of prefactors

$$H(\tau_0) = \sum_j \langle \delta s_j^2 \rangle \delta(\tau_0 - \tau_{0j})$$

which yields the distribution of relaxation times

$$F(\tau, T) = \tau_0 H(\tau_0) / \tau, \quad (7)$$

where again Eq. (6) applies.

Before proceeding it is appropriate to comment on Eq. (4). By definition, $F(\tau) \geq 0$ and $\int_0^{\infty} d\tau F(\tau) = 1$ at all T . Clearly, $S(-\omega) = S(\omega)$, $S(\omega) \geq 0$, and $-2 \leq (\omega/S) dS(\omega)/d\omega \leq 0$ (for $\omega \geq 0$). Any analytic function $S(\omega)$ satisfying these conditions is uniquely represented by a distribution $F(\tau)$ obtained by inverting Eq. (4), where

$$F(\tau) = -1/(\pi\tau^2) \text{Im}[S(i/\tau)]$$

(Ref. 15). In general, a numerical inversion of Eq. (4) is a nontrivial practical problem which lies outside the scope of this paper. However, for the relevant case of $1/f$ noise, $S(f) \propto f^{-\alpha}$ ($\alpha \approx 1$); therefore, $\tau F(\tau) \approx \text{const}$ and Eq. (4) simplifies approximately to

$$fS(f, T) \approx \tau F(\tau, T), \quad (8)$$

where $\tau = (2\pi f)^{-1}$ and $S(f, T) = 2[S(\omega, T)]_{\omega=2\pi f}$ is the one-sided spectrum $f \geq 0$. The approximation introduces no more than 50% error (2 dB) whenever $fS(f)$ does not vary by more than 50% over a decade in frequency. For sharper structure a more sophisticated inversion technique must be employed.¹⁶ No matter how $F(\tau)$ is obtained, it is ultimately tested by comparing the spectrum it generates via the exact Eq. (4) with the measured $S(f)$ it is supposed to represent.

Given an activation-energy distribution $D(E)$ and a single prefactor τ_0 for case (i), or alternatively $H(\tau_0)$ and a single activation energy E for case (ii), the distribution of relaxation times $F(\tau, T)$ may be calculated for any temperature T using Eqs. (6) and (5) [or Eq. (7)]. The integral in Eq. (4) is readily performed numerically to obtain $S(\omega, T)$, which, when combined with a given variance $\langle \delta X^2(T) \rangle$, gives $S_X(\omega, T)$.

We are interested in the inverse problem, to test whether observed noise spectra $S_X(\omega, T_j)$ at temperatures T_j arise from a single limiting distribution $D(E)$ or $H(\tau_0)$, and if so, to determine that distribution. The temperature dependence of the noise magnitude $\langle \delta X^2(T_j) \rangle$ is supposed to be independently determined. Equation (4) is inverted for each $S(\omega, T_j)$ to obtain corresponding $F(\tau, T_j)$. Next, Eqs. (5)–(7) are used to postulate a $D(E)$ or an $H(\tau_0)$ for each T_j , using any assumed prefactor τ_0 (or activation energy E). Of course, a finite frequency range $f_1 \leq f \leq f_2$ defines a finite energy range

$$-kT_j \ln(2\pi f_2 \tau_0) \leq E \leq -kT_j \ln(2\pi f_1 \tau_0),$$

or a finite range of prefactors

$$(2\pi f_2)^{-1} e^{-E/kT_j} \leq \tau_0 \leq (2\pi f_1)^{-1} e^{-E/kT_j}.$$

The observed frequency and temperature derivatives $\partial S/\partial \omega$ and $\partial S/\partial T$ may be compared to check for consistency. Using the approximate inversion equation (8) to obtain $F(\tau)$ from $S(\omega)$, it has been shown that, for a distribution of activation energies,

$$\alpha(\omega, T) \approx 1 - [(T/S)\partial S/\partial T - 1]/\ln(\omega\tau_0), \quad (9)$$

where $\alpha(\omega, T) = -(\omega/S)\partial S/\partial \omega$.⁷ Without approximation, a distribution of prefactors implies a relationship

$$\alpha(\omega, T) = 1 + (kT/E)(T/S)\partial S/\partial T. \quad (10)$$

TABLE I. Comparison of the assumptions and predictions of the Dutta-Horn and thermally activated scatterer diffusion models for noise spectra.

	Dutta-Horn	Diffusion
$S(\omega, T) = S_\rho(\omega, T) / \langle \delta\rho^2(T) \rangle$	ρ^2 / N_a	ρ_0^2 / N_0
$S(\omega, T) = \int_0^\infty d\tau F(\tau, T) (2\tau) / (1 + \omega^2\tau^2)$	$kTD(E)$	$\tau_0 H(\tau_0)$
$fS(f, T) \simeq \tau F(\tau, T)$	τ_0	E
$\tau = \omega^{-1} = (2\pi f)^{-1} = \tau_0 e^{E/kT}$		
Parameter		
$-(\omega/S)\partial S/\partial\omega$	$\simeq 1 - [(T/S)\partial S/\partial T - 1] / \ln(\omega\tau_0)$	$= 1 + (kT/E)(T/S)\partial S/\partial T$

The assumptions and predictions of the two models are summarized in Table I.

III. DIFFUSION NOISE

A. Definition of the problem

Number fluctuations associated with diffusion have been calculated for a variety of geometries and boundary conditions.¹⁷ Case (ii) applies with the prefactor distribution $H(\tau_0)$ determined by the diffusion geometry for diffusion with a single activation energy E_0 . To proceed, a measured property of some subvolume (Ω_0) of the system, in this case the resistivity (ρ), is assumed to be linearly coupled to the number (N) of diffusion "particles" contained in that subvolume. It is convenient to describe Ω_0 with a spatial weighting function $\Omega(\mathbf{x}) = 1$ for $\mathbf{x} \in \Omega_0$ and $\Omega(\mathbf{x}) = 0$ otherwise, so that

$$\delta\rho(t) = \xi \delta N(t) = \xi \int_{-\infty}^{\infty} d^3\mathbf{x} \Omega(\mathbf{x}) \delta n(\mathbf{x}, t), \quad (11)$$

where $\xi = \partial\rho/\partial N$ and $n(\mathbf{x}, t)$ is the local number density of diffusing particles. The two-time correlation function

$$G_\rho(t_2, t_1) = \langle \delta\rho(t_1) \delta\rho(t_2) \rangle$$

is completely determined by the space-time correlation function of the density fluctuations

$$c_n(\mathbf{x}_1, t_1; \mathbf{x}_2, t_2) = \langle \delta n(\mathbf{x}_1, t_1) \delta n(\mathbf{x}_2, t_2) \rangle.$$

The fluctuation-dissipation theorem gives the latter as the Green's function for the equation governing the relaxation of $n(\mathbf{x}, t)$ to equilibrium, namely, the diffusion equation (subject to appropriate boundary conditions)

$$\left[\mathcal{D} \nabla_1^2 - \frac{\partial}{\partial t_1} \right] c_n(\mathbf{x}_1, t_1; \mathbf{x}_2, t_2) = -\Delta n \delta(\mathbf{x}_1 - \mathbf{x}_2) \delta(t_1 - t_2), \quad (12)$$

where \mathcal{D} is the diffusion coefficient. The magnitude Δn is chosen to give the thermodynamic variance. Temporal- and spatial-translation invariance imply that c_n depends only on the differences $t \equiv t_2 - t_1$ and $\mathbf{x} \equiv \mathbf{x}_2 - \mathbf{x}_1$. The power spectral density

$$S_\rho(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} G_\rho(t)$$

is then

$$S_\rho(\omega) = \xi^2 \int_{-\infty}^{\infty} d\mathbf{k} (2\pi)^{-d} |\Omega(\mathbf{k})|^2 c_n(\mathbf{k}, \omega), \quad (13)$$

where

$$\Omega(\mathbf{k}) = \int_{-\infty}^{\infty} d\mathbf{x} e^{-i\mathbf{k}\cdot\mathbf{x}} \Omega(\mathbf{x}),$$

$$\mathbf{x} = \mathbf{x}_2 - \mathbf{x}_1,$$

and

$$c_n(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \int_{-\infty}^{\infty} d\mathbf{x} e^{-i\mathbf{k}\cdot\mathbf{x}} c_n(\mathbf{x}, t).$$

Systems that have rotation and translation invariance are particularly simple to deal with since

$$c_n(\mathbf{x}, t) = \langle \delta N^2 \rangle / \Omega_0 (4\pi \mathcal{D} t)^{-d/2} e^{-x^2/4\mathcal{D}t},$$

where $d=1,2,3$ is the dimensionality of the system and $\langle \delta N^2 \rangle$ is the variance of the number fluctuations in the subvolume Ω_0 . In this case the kernel $c_n(\mathbf{k}, \omega)$ is simply a Debye-Lorentzian spectrum

$$c_n(\mathbf{k}, \omega) = (\langle \delta N^2 \rangle / \Omega_0) (2\tau_k) / (1 + \omega^2 \tau_k^2), \quad (14)$$

where $\tau_k \equiv (\mathcal{D} k^2)^{-1}$ is the relaxation time for density fluctuations of wave vector \mathbf{k} . Substituting Eq. (14) into Eq. (13), we find

$$S_\rho(\omega) = (\langle \delta\rho^2 \rangle / \Omega_0) \times \int_{-\infty}^{\infty} d\mathbf{k} (2\pi)^{-d} |\Omega(\mathbf{k})|^2 (2\tau_k) / (1 + \omega^2 \tau_k^2), \quad (15)$$

where $\langle \delta\rho^2 \rangle \equiv \xi^2 \langle \delta N^2 \rangle$ is the variance. Equation (15) describes $S_\rho(\omega)$ as a superposition of Lorentzian spectra with the weighting of the various relaxation times τ_k determined by the Fourier transform $\Omega(\mathbf{k})$ of the specimen geometry.

B. Temperature dependence

Temperature dependence enters $S_\rho(\omega, T)$ on the right-hand side of Eq. (15) through the coupling coefficient $\xi(T)$ and the number variance $\langle \delta N^2(T) \rangle$ in $\langle \delta\rho^2(T) \rangle$, and through the diffusion coefficient $\mathcal{D}(T)$ in the relaxation times $\tau_k = [\mathcal{D}(T) k^2]^{-1}$. A change of integration variables in Eq. (15) shows that

$$S(\omega, T) = S_\rho(\omega, T) / \langle \delta\rho^2(T) \rangle$$

is represented by a temperature-dependent distribution $F(\tau, T)$ of relaxation times $\tau = (\mathcal{D}k^2)^{-1}$ satisfying all of the assumptions that led to Eq. (4). Further, for a diffusion coefficient $\mathcal{D}(T) = \mathcal{D}_0 e^{-E/kT}$, $F(\tau, T)$ may be expressed in terms of a distribution $H(\tau_0)$ of prefactors $\tau_0 = (\mathcal{D}_0 k^2)^{-1}$ through Eqs. (6) and (7).

Explicitly inserting temperature dependence into Eq. (15) and multiplying both sides by $\omega / \langle \delta\rho^2(T) \rangle$, we see that

$$\omega S_\rho(\omega, T) / \langle \delta\rho^2(T) \rangle = P(\Theta), \quad (16)$$

where P is a function of the single variable $\Theta(\omega, T) \propto \omega / \mathcal{D}(T)$. Equation (16) shows that the frequency and temperature dependences of $\omega S(\omega, T)$ are not independent, but instead enter through the combination $\omega / \mathcal{D}(T)$.⁹ This scaling holds for diffusion generally, independent of boundary conditions, dimensionality, and geometry because \mathcal{D} and t enter the diffusion equation only as the product $\mathcal{D}t$.

C. Noise magnitude

The temperature dependence of $\langle \delta\rho^2(T) \rangle$ depends on the particular scattering mechanism. For instance, enthalpy fluctuations couple to the resistivity through lattice scattering to produce a variance

$$\langle \delta\rho^2(T) \rangle \simeq \gamma^2(T) k T^2 / C_v(T),$$

where $\gamma(T) \equiv \partial\rho_L / \partial T$ and C_v is the heat capacity of the specimen volume Ω_0 . These fluctuations are hardly observable in ordinary metals at room temperature.¹⁸ Fluctuations in the equilibrium number of vacancies in the conductor are expected to produce

$$\langle \delta\rho^2(T) \rangle \simeq (\beta^2 / N_a) e^{-E_f/kT},$$

where $\beta = \partial\rho / \partial c_0$ is the resistivity increment per atomic fraction of vacancies, N_a is the number of atoms in the conductor, and E_f is the enthalpy for vacancy formation. E_f is too large for equilibrium vacancy noise to be observed at room temperature.¹⁹ A third system that does give measurable resistivity fluctuations at room temperature is hydrogen impurities in Nb films.⁹ Here the surface oxide prevents hydrogen from leaving the film, so that fluctuations in the resistivity of a film segment have a variance $\langle \delta\rho^2 \rangle \simeq \beta^2 c_0 / N_a$, independent of temperature.

For the above examples we have assumed Matthiessen's rule to calculate the variance $\langle \delta\rho^2 \rangle$ for fluctuations in a particular scattering mechanism, independent of the other scattering mechanisms that might contribute to the resistivity $\rho(T)$. For the last two examples, vacancies and impurities, $\langle \delta\rho^2 \rangle$ may be written as

$$\langle \delta\rho^2(T) \rangle = \rho_0^2(T) / N_0(T),$$

where $N_0(T) = c_0(T) N_a$ is the number of "noisy" scatterers in the conductor and $\rho_0(T)$ is their mean contribution to the resistivity. If $\rho(T) \simeq \rho_0(T)$, that is, if the noisy scattering mechanism also dominates the resistivity, and if c_0 is independent of T , then $\langle \delta\rho^2(T) \rangle \propto \rho^2(T)$. Dutta, Di-
mon, and Horn have made the implicit assumption that

$\langle \delta\rho^2(T) \rangle \propto \rho^2(T)$ in analyzing the temperature and frequency dependence of the excess noise of noble-metal films in terms of a distribution of energies $D(E)$ and a single prefactor τ_0 .⁷ The resistivities of their films, in the temperature range considered, were dominated by lattice scattering. These examples suggest that the assumption is not justified unless the noise mechanism is also associated with lattice scattering.²⁰

D. More complicated diffusion spectra

The characteristic frequencies ω_j of the diffusion problem are generally determined by the specimen dimensions; an open rectangular subvolume $\Omega_0 = L_1 L_2 L_3$ has $\omega_j \simeq \mathcal{D} / L_j^2$, $j=1,2,3$. It is easy to imagine physical situations where boundary conditions introduce additional length scales. For instance, dislocations and grain boundaries, in addition to the specimen surface, may act as sources and/or sinks for vacancies, interstitials, or impurities. Such defects, combined with inhomogeneities in metal films, yield additional physical lengths ranging from tens of angstroms to the largest grain size. We therefore suggest that $1/f$ noise in metals might be described by Eq. (13), where $\Omega(\mathbf{k})$ represents the distribution of \mathbf{k} vectors necessary to describe all of the important physical lengths of the diffusion problem.

IV. EXPERIMENTS

Below we analyze data from Nb, Ag, and Cr films to determine whether their excess noise spectra may be represented by a superposition of activated Lorentzians arising from either a $D(E)$ or an $H(\tau_0)$. The Ag data are taken from the literature.¹³ The Nb data⁹ and the Cr data²¹ were measured in our laboratory.

Niobium and chromium specimens were rectangular four-probe resistors (with fifth center-tap probe) of length L and width w subtractively patterned (following photolithography) from metal films deposited onto single-crystal sapphire substrates. Niobium films were deposited onto room-temperature substrates at a rate of 12 Å/s in a magnetron sputter system (base pressure 3×10^{-8} Torr), had a thickness $h = 120 \pm 20$ nm, resistivity $\rho(300 \text{ K}) = 28 \pm 5 \mu\Omega \text{ cm}$, and a residual resistivity ratio of $\rho(300 \text{ K}) / \rho(10 \text{ K}) = 2.0 \pm 0.1$.⁹ The chromium was e -beam-evaporated (base pressure $\simeq 1 \times 10^{-7}$ Torr) onto substrates held at 530°C at a rate of 40 Å/s, had $h = 120 \pm 10$ nm, $\rho(300 \text{ K}) \simeq 25 \pm 2 \mu\Omega \text{ cm}$, and $\rho(300 \text{ K}) / \rho(10 \text{ K}) = 2.0 \pm 0.1$ (significantly better than Cr films deposited at room temperature). Excess voltage noise spectra $S_v(f)$, measured using an ac technique, were reproducible to about 20% and accurately scaled with the mean-square current i_{rms}^2 ; excess noise spectra are conveniently represented by resistance-fluctuation spectra $S_r(f)$ and resistivity-fluctuation spectra $S_\rho(f)$ defined by

$$S_r(f) / r^2 \equiv S_\rho(f) / \rho^2 \equiv S_v(f) / (i_{\text{rms}} r)^2,$$

where r and ρ are the specimen resistance and resistivity, respectively. Details of the specimen preparation and noise measurement are given elsewhere.²¹ For measurement temperatures in the range $-20 \leq T \leq 90^\circ \text{C}$ the speci-

mens were mounted in an evacuated copper cylinder and placed in an ethylene glycol bath stable to ± 100 mK. Lower temperatures were achieved using a scanning Dewar capable of $4 \leq T \leq 300$ K with better than 5-mK stability.²² The absence of contact noise was verified throughout the entire temperature range by changing the ballast resistance.²³

We have measured the $S_p(f, 300 \text{ K})$ of 40 Nb specimens for frequencies $300 \mu\text{Hz} \leq f < 300 \text{ Hz}$.⁹ The resistivity $\rho(T_j)$ and excess noise of two specimens (N7-2 and N6-1) were measured at five temperatures T_j ($j=1,2,\dots,5$), $-20 \leq T_j \leq 90^\circ\text{C}$. The relative resistivity-fluctuation spectra of the smallest of these (N7-2) are graphed in Fig. 1; T_j (in K), $\rho(T_j)$ (in $\mu\Omega \text{ cm}$), and the number of atoms, N_a , in the conductor are given in the figure. Each of the spectra were integrated numerically to obtain $\langle \delta\rho^2(T_j) \rangle$ and $F(\tau, T_j)$ calculated using the approximate inversion of Eq. (8).²⁴ To test whether the $F(\tau, T_j)$ may be expressed in terms of a distribution of prefactors $H(\tau_0)$ through Eqs. (5) and (7), the value of the activation energy E must be specified. Here we used E as a fitting parameter, calculating and comparing the $H(\tau_0)$ for a trial value of E from the five $F(\tau, T_j)$. For most values of E the five calculated $H(\tau_0)$ were distinctly different. Had this been the case for all values of E , we would have concluded that the normalized spectra $S(f, T_j)$ could not be expressed in terms of a distribution of prefactors $H(\tau_0)$. As E approached the value $E_p = 230$ meV, however, the $H(\tau_0)$ began to agree; the resulting five distributions $H(\tau_0)$ are plotted in Fig. 2(a). The five spectra fit the same distribution of prefactors $H(\tau_0)$ as expected since the $f/f_c(T)$ diffusion scaling of Eq. (15) has already been demonstrated for this data.⁹ A weak temperature dependence is observed for the variance;

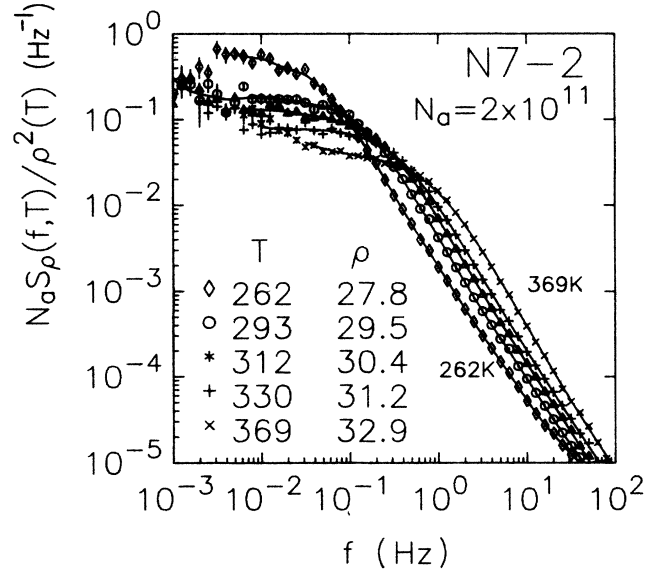


FIG. 1. Relative resistivity-fluctuation spectra of Nb specimen N7-2 at five temperatures (T) between -20 and 90°C . The lines are guides to the eye. Tabulated T and resistivities (ρ) have the units of K and $\mu\Omega \text{ cm}$, respectively. The specimen dimensions are $h=120 \pm 30$ nm, $w=0.7 \pm 0.3$ μm , and $L=40 \pm 1$ μm .

values of T_j (in K), $N_a \langle \delta\rho^2(T_j) \rangle$ [in $(\mu\Omega \text{ cm})^2$], and N_a are tabulated in the figure.

These same data have also been compared with predictions for noise arising from a distribution of energies $D(E)$. Equations (5) and (6) were used to calculate $D(E)$'s from the five $F(\tau, T_j)$ for a trial value of τ_0 . The

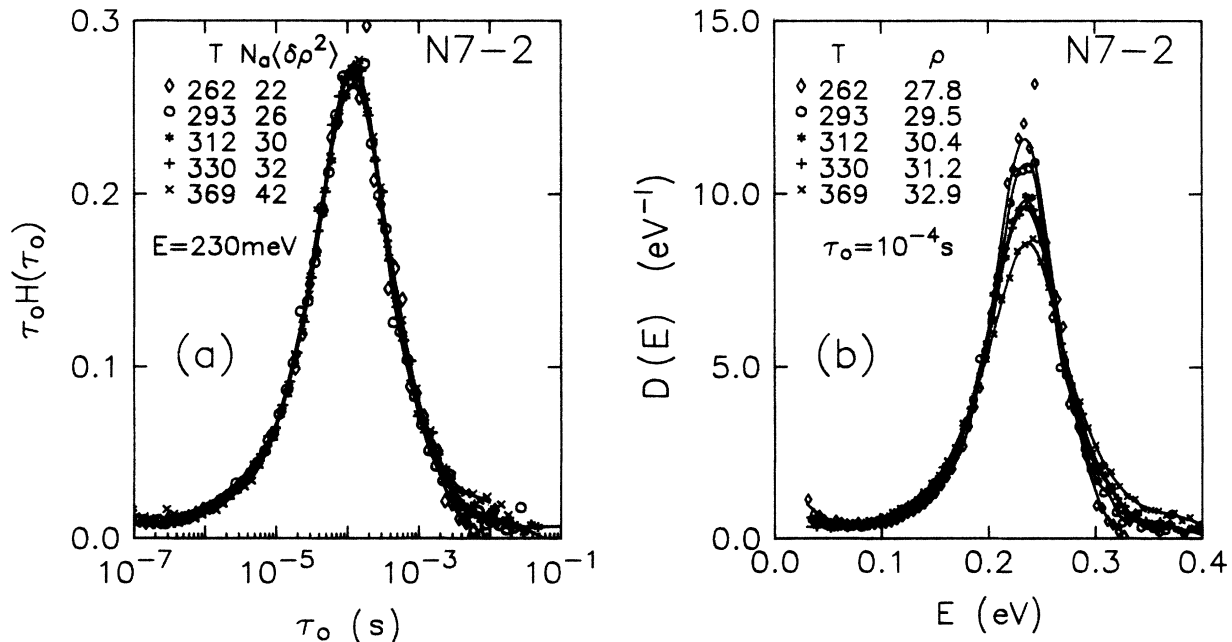


FIG. 2. (a) Graph of the five prefactor distributions implied by the noise spectra of Fig. 1 with $E=230$ meV (see text). (b) Graphs of the five energy distributions implied by the same data with $\tau_0=10^{-4}$ s (see text). Tabulated temperatures (T) and variances ($N_a \langle \delta\rho^2 \rangle$) have the units of K and $(\mu\Omega \text{ cm})^2$, respectively. The lines are guides to the eye.

best agreement was obtained for $\tau_0 \approx 10^{-4}$ s; the resulting five $D(E)$ are plotted in Fig. 2(b). The previous agreement with a diffusion model guarantees that the five $D(E)$ will peak at the same energy, $E_p = 230$ meV. Comparison of Figs. 2(a) and 2(b) leads us to conclude that the data fit a distribution $H(\tau_0)$ slightly better than a distribution $D(E)$, but that the difference is not compelling. Of course, noise from H^+ diffusion in Nb is expected to be described by an $H(\tau_0)$.

Eberhard and Horn have measured the excess noise of noble-metal films for temperatures $80 \leq T \leq 600$ K and frequencies $200 \text{ mHz} \leq f \leq 200 \text{ Hz}$.¹³ At all temperatures they report $(1/f)$ -like spectra

$$N_a S_\rho(f, T) = \rho_*^2(T) / f^{\alpha(T)}$$

with $\rho_*^2(T)$ depending strongly, and $\alpha(T)$ depending weakly, on temperature.²⁵ The temperature dependences of ρ_*^2 and α for one Ag film are shown in Fig. 3. Figure 3(a) is a semilogarithmic plot of the relative noise level ρ_*^2/ρ^2 versus T ; the solid line is a guide to the eye. The data in Fig. 3(b) are the measured frequency exponents $\alpha(T)$. Dutta, Dimon, and Horn have analyzed these data in terms of a superposition of thermally activated processes arising from a single prefactor τ_0 ($\approx 10^{-14}$ s) and a distribution of activation energies $D(E)$.⁷ By assuming $\langle \delta\rho^2(T) \rangle \propto \rho^2(T)$ and ignoring data below 175 K and above 530 K, they find satisfactory agreement between measured $\alpha(T)$ and those calculated from the temperature dependence of $\rho_*^2(T)/\rho^2$ using Eq. (9).

Here the Ag data are reanalyzed to determine (i) if the data may also be described by a distribution of prefactors

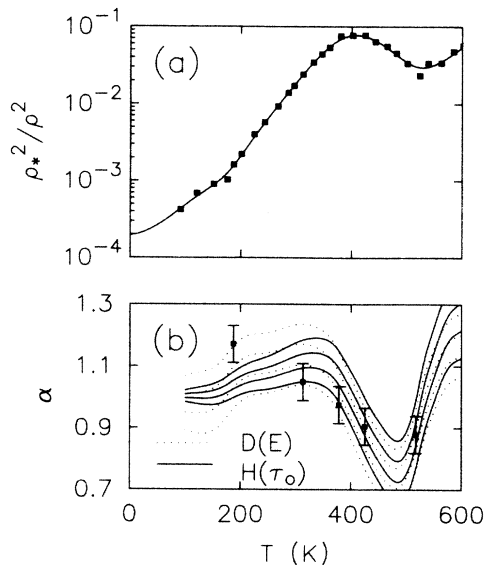


FIG. 3. Temperature dependence of the low-frequency excess noise of a Ag film (data taken from Ref. 10). (a) Semilogarithmic plot of relative noise level $\rho_*^2/\rho^2 \approx [f N_a S_\rho(f, T) / v^2(T)]_{f=20 \text{ Hz}}$ versus temperature T ; the line is a guide to the eye. (b) Comparison of measured frequency exponents α (symbols) with those implied by ρ_*^2/ρ^2 , assuming a $D(E)$ (dotted lines) or an $H(\tau_0)$ (solid lines). Each of the four pairs of lines corresponds to a different assumption for $\langle \delta\rho^2(T) \rangle$ (see text).

$H(\tau_0)$, and (ii) what justification there is for the assumption that $\langle \delta\rho^2(T) \rangle / \rho^2(T)$ is temperature independent. We find that the data can be fitted equally by either an $H(\tau_0)$ or a $D(E)$.²⁶ Further, both models work with a broad range of assumptions for the fitting parameters. These findings are illustrated in Fig. 3(b), where the measured α are compared with those calculated from the temperature dependence of $\rho_*^2(T)/\rho^2(T)$ for one set of assumptions about $\langle \delta\rho^2(T) \rangle$. The solid lines are calculated assuming a distribution $H(\tau_0)$, while the dotted lines are calculated for a distribution $D(E)$. The four pairs of lines in Fig. 3(b) were generated using Eqs. (9) and (10) along with the arbitrary assumption $\langle \delta\rho^2(T) \rangle / \rho^2(T) \propto T^{-\delta}$, for $\delta = -2, 0, 2$, and 4 ; τ_0 and E were arbitrarily set to 10^{-14} s and 1.2 eV, respectively. Here it is impossible to rule out one distribution in favor of the other by any optimization of these parameters. Furthermore, the absence of spectral features, the large uncertainties in the measured frequency exponents $\alpha(T)$, and ignorance of the variance $\langle \delta\rho^2(T) \rangle$ invalidate such analysis.

We have measured the room-temperature excess noise of four Cr specimens (C3-1, -2, -3, and -4) for frequencies $1 \text{ mHz} \leq f \leq 100 \text{ Hz}$. It is assumed that the specimens all have the same thickness and resistivity since they were fabricated from the same film. The excess noise and resistivity of two of these (C3-1 and -2) have been measured for temperatures $10 \leq T \leq 360$ K. At all temperatures resistivity-fluctuation spectra $S_\rho(f, T)$ were inversely pro-

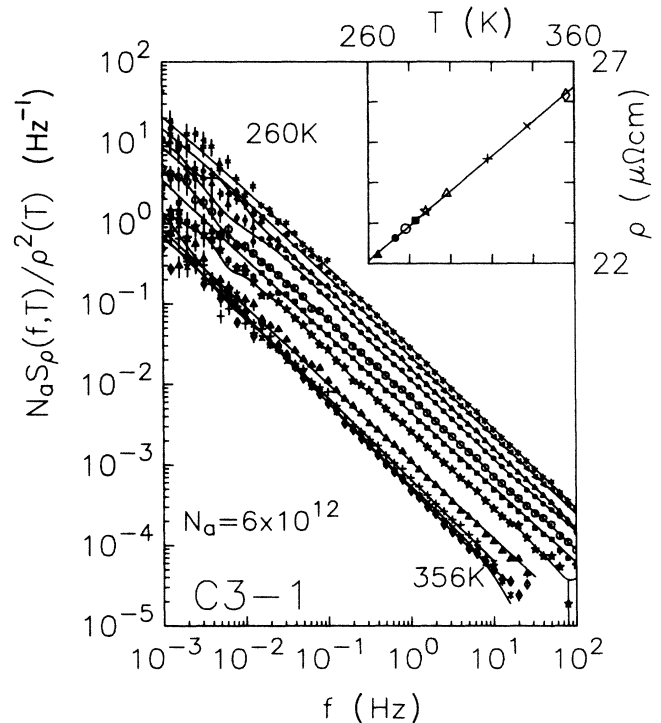


FIG. 4. Relative resistivity-fluctuation spectra of Cr specimen C3-1 at ten temperatures (T) between -13 and 83°C . The temperature dependence of the resistivity (ρ) is plotted in the figure inset using the same symbols. The lines are guides to the eye. The specimen dimensions are $h = 120 \pm 20$ nm, $w = 4.2 \pm 0.3$ μm , and $L = 160 \pm 2$ μm .

portional to the specimen area $A \equiv Lw$. This size dependence was conveniently removed by multiplying S_ρ by the number of atoms in the specimen (N_a). The relative resistivity-fluctuation spectra of specimen C3-1 ($L=160 \mu\text{m}$, $w=4.1 \pm 0.3 \mu\text{m}$) are graphed in Fig. 4 for temperatures $-20 \leq T_j \leq 100^\circ\text{C}$. Each temperature is represented by a different symbol (with error bars); the lines are guides to the eye. Notice the rapid increase in noise level with decreasing temperature while $\alpha = -\partial \ln S / \partial \ln \omega$ hardly changes from 1.0. The temperature dependence of the resistivity ρ is shown in the figure inset using the same symbols.

The full temperature dependence of the excess noise of both Cr specimens is conveniently represented by $\rho_*^2(T)$ and $\alpha(T)$, where

$$N_a S_\rho(f, T) = \rho_*^2(T) / f^{\alpha(T)}.$$

The temperature dependences of the resistivity ρ , noise level $\rho_*^2(T)$, and frequency exponent $\alpha(T)$ are plotted in Fig. 5. The triangle data were measured for specimen C3-2 ($L=320 \mu\text{m}$, $w=8.0 \pm 0.3 \mu\text{m}$) in the scanning Dewar seven months after the other data (circles and stars) had been measured from specimen C3-1. The solid lines in Figs. 5(a) and 5(b) are guides to the eye. The noise

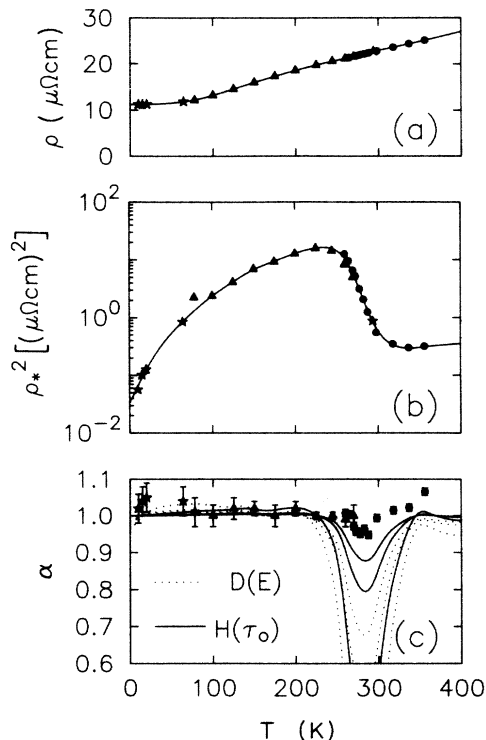


FIG. 5. Temperature dependence of the (a) resistivity ρ , (b) noise level ρ_*^2 , and (c) frequency exponent α for Cr film C3. Data are reported for two specimens, C3-1 (circles and stars) and C3-2 (triangles). The solid lines in (a) and (b) are guides to the eye. The lines in (c) are calculated from $\rho_*^2(T)/\rho^2(T)$ assuming $\langle \delta\rho^2(T) \rangle \propto \rho^2(T)$. The dotted lines correspond to a $D(E)$ for trial values $\tau_0=10^{-12}$, 10^{-24} , and 10^{-36} s. The solid lines correspond to an $H(\tau_0)$ for trial values $E=1, 3$, and 5 eV.

level increases with decreasing temperature, reaching a peak level $\rho_*^2(240 \text{ K})$ approximately 30 times its room-temperature value $\rho_*^2(300 \text{ K})$. Below 240 K the noise level monotonically decreases with decreasing temperature.

To determine whether the data are consistent with either a $D(E)$ or an $H(\tau_0)$, the variance $\langle \delta\rho^2(T) \rangle$ must be shown. Unlike noise from hydrogen diffusion, the variance may not be determined by numerically integrating the measured noise spectra. The data cannot be fitted by either model with the Dutta-Horn assumption that $\langle \delta\rho^2(T) \rangle \propto \rho^2(T)$. This is illustrated in Fig. 5(c), where measured α (symbols) are compared with those calculated from $\rho_*^2(T)/\rho^2(T)$ assuming a $D(E)$ (dotted lines) or an $H(\tau_0)$ (solid lines) using Eqs. (9) and (10), respectively. The three dotted lines correspond to trial values $\tau_0=10^{-12}$, 10^{-24} , and 10^{-36} s; the solid lines correspond to trial values $E=1, 3$, and 5 eV. Unreasonably small τ_0 or large E are required to make either model consistent with the data.

Fleetwood *et al.* suggest writing $\langle \delta\rho^2(T) \rangle \propto T^6$ with δ chosen to give the best fit.^{27,28} This method will not improve the agreement between the data and the curves of Fig. 5 since they do not differ by a simple translation.²⁷ The only way to make either model work (with acceptable values for the parameters τ_0 or E) is to let $\langle \delta\rho^2(T) \rangle$ take on most of the temperature dependence of $\rho_*^2(T)$. In the absence of physical knowledge of a complicated $\langle \delta\rho^2(T) \rangle$, this approach leaves $D(E)$ or $\tau_0 H(\tau_0)$ undetermined.

V. DISCUSSION

Our analysis of the excess noise of Nb films containing H^+ demonstrates that one can experimentally distinguish between a distribution of prefactors $H(\tau_0)$ and a distribution of activation energies $D(E)$. The distinction between the two models, in this case, is quite small since most of the noise power is associated with a single activated relaxation time [$\tau_c(T) \propto L^2/\mathcal{D}(T)$] consistent with either model. The analysis was aided by (i) knowledge of the variance $\langle \delta\rho^2(T) \rangle$, and (ii) a pronounced knee in the noise spectrum. Useful analysis may proceed even if only one of these is available. Consider a log-log plot of

$$fS_j \equiv fS_\rho(f, T_j) / \langle \delta\rho^2(T_j) \rangle$$

versus $f/f_c(T_j)$, where $\langle \delta\rho^2(T_j) \rangle$ and $f_c(T_j)$ are not known. Arbitrary adjustments of $f_c(T_j)$ and $\langle \delta\rho^2(T_j) \rangle$ produce, respectively, horizontal and vertical translations of such a graph. If either the horizontal or vertical translations are predetermined, say by aligning some spectral feature or by scaling with a known $\langle \delta\rho^2(T) \rangle$, the remaining translation is not arbitrary. The distinction between horizontal and vertical translations, however, is lost for featureless $1/f$ noise for which $\langle \delta\rho^2(T) \rangle$ is not known. Such is the case for Ag films where the excess noise could be fitted by a wide range of *ad hoc* assumptions about $\langle \delta\rho^2(T) \rangle$.

The temperature dependence of the resistivity of chromium films might provide a valuable clue as to the temperature dependence of $\langle \delta\rho^2(T) \rangle$. As was pointed out in Sec. III C, there is reason to look for scaling of $\langle \delta\rho^2(T) \rangle$ with some component $\rho_0(T)$ of the resistivity

$\rho(T)$. It is customary to write the resistivity $\rho(T) = \rho_d + \rho_L(T)$, where the (nominally) temperature independent piece ρ_d is due to defect, boundary, and impurity scattering, and $\rho_L(T)$ is the intrinsic resistivity due to lattice scattering. Inspection of Fig. 5(a) suggests that the chromium resistivity might be written as $\rho(T) = \rho_d + \rho_L(T) + \rho_0(T)$, where $\rho_0(T)$ is a third component with temperature dependence similar to the noise level, $\rho_*^2(T)$. The difference $\rho(T) - \rho_L(T) - \rho_d$ is not known with sufficient accuracy to allow extraction of $\rho_0(T)$. The temperature range is suggestive of the antiferromagnetic transition in Cr,²⁸ in which case the noise might be associated with carrier number or mobility fluctuations.

VI. SUMMARY AND CONCLUSIONS

We have considered resistivity fluctuations associated with scattering of conduction electrons. For scattering from diffusing defects we have found a scaling law: the quantity $\omega S_\rho(\omega, T) / \langle \delta\rho^2(T) \rangle$ is a function of ω and T only through the diffusion-normalized frequency $f/\mathcal{D}(T)$. If Matthiessen's rule is followed and the scatterers are independent, then $\langle \delta\rho^2 \rangle \propto \rho_0^2/N_0$; this expression is independent of the scatterer-relaxation mechanism. For the particular case of thermally activated diffusion, we find that the $S_\rho(f, T)$ can be expressed as a superposition of thermally activated processes with a set of apparent frequencies fully determined by the diffusion pro-

cess; relationships have been derived between the frequency and temperature dependence of $S_\rho(f, T) / \langle \delta\rho^2(T) \rangle$.

Attempts to compare excess noise spectra from three metal film examples with both the Dutta-Horn model [with its distribution of activation energies $D(E)$] and with thermally activated diffusion [with its distribution of prefactors $H(\tau_0)$] have led us to several disturbing conclusions: (i) Meaningful comparison of noise data with theoretical models requires either independent information about $\langle \delta\rho^2(T) \rangle$ of the observation of spectral features; (ii) both limiting models are equally consistent with the $1/f$ noise observed from noble metals, but neither is convincingly established; (iii) our Cr $1/f$ noise data cannot be represented by such models without the assumption of a strong, *ad hoc* temperature dependence for $\langle \delta\rho^2 \rangle$.²⁹

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¹⁴There may be interesting physical situations where the $\langle \delta x^2(T) \rangle$ do not share the same temperature dependence; the

remaining analysis does not apply to them. It is not obvious how such cases might be treated in general. We do not believe that the implications of this assumption have been fully appreciated in the past.

¹⁵This formula is mathematically equivalent to expressions derived previously; see Raymond M. Fuoss and John G. Kirkwood, *J. Am. Chem. Soc.* **63**, 385 (1941); B. Gross, *J. Appl. Phys.* **18**, 212 (1947).

¹⁶See Chap. 4 of *Anelastic Relaxation in Crystalline Solids*, Ref. 10, and references therein; also see F. C. Roesler and W. A. Twyman, *Proc. Phys. Soc. London, Sect. B* **68**, 97 (1955); J. R. Cost, *J. Appl. Phys.* **54**, 2157 (1983).

¹⁷For instance, see K. M. van Vliet and J. R. Fassett, in *Fluctuation Phenomena in Solids*, edited by R. E. Burgess (Academic, New York, 1965), pp. 267–364; Douglas Magde, Elliot Elson, and W. W. Webb, *Phys. Rev. Lett.* **29**, 705 (1972); Michael B. Weissman, *ibid.* **35**, 689 (1975); Richard F. Voss and John Clarke, *Phys. Rev. B* **13**, 556 (1976); J. H. Scofield, D. H. Darling, and W. W. Webb, *ibid.* **24**, 7450 (1981); also see Ref. 9.

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²⁰Resistivity fluctuations due to fluctuations in the carrier density (i.e., number rather than mobility fluctuations) might very well scale with the resistivity, since all components of the resistivity are affected by a change in the carrier density. The relative $1/f$ noise level in metals is generally thought to be too large of an effect to be due to trapping; see, for instance, F. N. H. Robinson, *Phys. Lett.* **97A**, 162 (1983).

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²⁴The approximation $\tau F(\tau) \simeq fS(f)$ is not very good for the diffusion noise spectra of Fig. 1. Therefore, the analysis has been repeated, inverting Eq. (4) with an iterative technique [see F. C. Roesler and W. A. Twyman, *Proc. Phys. Soc. London, Sect. B* **68**, 97 (1955)] which does yield accurate $F(\tau)$. The accurate distributions $\tau F(\tau)$ are more sharply peaked and differ by as much as a factor of 2 from the approximate distributions. Both the approximate and accurate inversions, however, yield the same relationship *between* the $F(\tau, T_j)$ (for the five T_j). The approximate analysis has been presented for simplicity.

²⁵Here, $\rho_*^2/\rho^2 = [fN_a S_v(f, T)/v^2]_{f=20 \text{ Hz}}$, assuming the excess noise is due to resistance fluctuations. This ignores the slight v dependence of $S_v(f)/v^2$ observed by the authors in Ref. 13.

²⁶Weissman has made the same observation; see M. B. Weissman, in *Proceedings of the Sixth International Conference on Noise in Physical Systems*, Gaithersburg, Maryland, 1981, edited by P. H. E. Meijer, R. D. Mountain, and S. J. Soulen, Jr., NBS Spec. Publ. No. 614 (U.S. GPO, Washington, D.C., 1981), p. 133.

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