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Quantitative "local-interference" model for $1/f$ noise in metal films

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Electron scattering calculations by Martin are used to predict the magnitude of resistivity fluctuations in metal films arising from the fluctuating interference of electrons in the local environment of a moving defect. A "local-interference" model based on these calculations accounts for the $1/f$-noise magnitude observed in irradiated Cu films and in room-temperature metal films. For relatively ordered metal films at room temperature this model predicts larger noise magnitudes than an alternative model based on universal conductance fluctuations, while the latter model predicts larger noise in metals that are sufficiently disordered and/or at lower temperatures.

The observation of flicker ($1/f$) noise in a wide variety of metal films has attracted considerable experimental and theoretical interest.[1] It is generally agreed that the noise is due to intrinsic resistance fluctuations,[2] and recent experimental evidence indicates that these fluctuations arise from the motion of defects.[3-10] The kinetics of the noise generation process are successfully described by the Dutta-Dimon-Horn thermal activation model,[11] but the microscopic mechanism and magnitude of the resistivity change due to the motion of a defect remain very much open questions.

Kogan and Nagaev[12] and Black, Restle, and Weissman[3] have discussed changes in the scattering cross section of asymmetric defects as they move, but did not carry out detailed calculations of the magnitude of these changes. We shall call this type of model a "local-interference" (LI) model for reasons which will become apparent. More recently, Feng, Lee, and Stone[13] proposed an alternative model based on "universal conductance fluctuations" (UCF's) in order to relate resistivity fluctuations to defect motion.

In this Rapid Communication we first use the defect-scattering calculations of Martin[14,15] to determine the magnitude of the resistivity fluctuations produced in the LI model. We then apply this model to measurements of the $1/f$ noise in thin films in which defects have been induced by electron irradiation.[7,8] Finally, we compare the predictions of the LI and UCF models to determine the conditions under which each is the predominant mechanism for $1/f$ noise.

The most complete LI model to date is that of Kogan and Nagaev,[12] who in particular considered *rotations* of point defects with symmetry lower than the crystal. To estimate the change, $\delta\sigma$, in the average cross section σ of a defect when it rotates, the authors considered electron scattering from anisotropic local defect potentials and set up a Boltzmann equation to determine the effect on the conductivity. However, they did not actually solve the equation, but rather used dimensional arguments to obtain the order-of-magnitude estimate $\delta\sigma \approx a_0^2 \approx \sigma$, where a_0 is a lattice constant. One clearly requires a quantitative calculation of $\delta\sigma$ to justify this estimate and to make a realistic comparison with both experiment and the UCF model.

In fact, Martin[14,15] performed such calculations some ten years prior to the work of Kogan and Nagaev. Martin considered scattering of conduction electrons from structural defects consisting of a number of vacant lattice sites and interstitial atoms, each of which acts as an isotropic scattering center. He showed that *interference* of electrons scattered by the centers causes the resistivity tensor $\rho_{\alpha\beta}$ to be anisotropic, and to change with varying separation of the centers. This interference can be included in the calculations in the form of a structure factor.[20] The total interference term for an arbitrary configuration can be expressed as a sum of dipole interference terms, one for each pair of scattering centers. Furthermore, for a given pair the dependence of $\rho_{\alpha\beta}$ on the dipole orientation and on the separation R of the centers is large only for $k_F R < 7$, where k_F is the Fermi wave vector. For $k_F R \gg 7$, the resistivity tensor becomes isotropic and independent of R. We accordingly designate a $1/f$-noise model based on this effect a "local-interference" model, since the principal effect arises only from interference of electrons scattered by centers within a few lattice constants of each other. In the remainder of the paper, we focus on the *anisotropy* introduced by the interference and the resulting effect on the resistivity when defects reorient. In considering more general defect rearrangements, one should also include effects of changing the separation of the scattering centers. We note here that Martin assumes the Fermi surface to be spherical, which is approximately correct for a nearly-free-electron metal, and single-scatterer matrix elements which are independent of position of the center relative to the lattice. Only the positions of the scattering center relative *to each other* enter these resistivity calculations.

Martin calculated the tensor components ρ_{xx}, ρ_{yy}, and ρ_{zz} for several defect configurations likely to occur in fcc metals after high-energy electron, neutron, or ion irradiation. Here the x, y, and z directions correspond to the fcc $\langle 100 \rangle$ axes. For some of these defects[16] one can use Martin's calculated values, the defect symmetry, and the properties of tensor rotations to determine the principal moments ρ_1, ρ_2, and ρ_3 of the resistivity tensor $\rho_{\alpha\beta}$. If one measures the resistivity in a particular direction (say the z

direction) and assumes random defect orientation, one can show that

$$\langle \rho_{zz} \rangle \equiv \langle \rho \rangle = (\rho_1 + \rho_2 + \rho_3)/3 \ , \tag{1}$$

$$\langle (\rho_{zz} - \langle \rho \rangle)^2 \rangle \equiv \langle (\delta\rho)^2 \rangle = \tfrac{4}{45} (\rho_1^2 + \rho_2^2 + \rho_3^2 - \rho_1\rho_2 - \rho_1\rho_3 - \rho_2\rho_3) \ . \tag{2}$$

We define a root-mean-square anisotropy parameter β by

$$\beta^2 \equiv \langle (\delta\rho)^2 \rangle / \langle \rho \rangle^2 = \langle (\delta\sigma)^2 \rangle / \langle \sigma \rangle^2 \ . \tag{3}$$

This parameter expresses the average fractional change of the scattering cross section of a defect when it moves.

In Table I we list several defects considered by Martin, along with β and the relevant tensor components. We see that values of β are typically a factor of 5 to 10 smaller than the estimate $\delta\sigma \sim \sigma$ given by Kogan and Nagaev. Comparison of the first- and second-nearest-neighbor divacancy cases in Table I demonstrates that β decreases rapidly with increasing separation of the scattering centers.

We now express the resistivity noise magnitude in terms of the parameter β and the concentration of mobile defects. Similar expressions have been derived previously.[6,12,17] We define:

$$\rho = K l^{-1} \ , \tag{4}$$

$$l^{-1} = l_{\text{in}}^{-1} + l_e^{-1} \ , \tag{5}$$

and

$$l_e^{-1} = \sum_i \sigma_i / V \ . \tag{6}$$

Here Eq. (4) is the free-electron expression for the resistivity ρ in terms of the electron scattering length l, and K is a material-dependent parameter. Equation (5) is Matthiessen's law, which expresses l in terms of the inelastic length l_{in} and the elastic (defect) length l_e. In Eq. (6) the sum extends over all defect cross sections σ_i in the sample volume V.

If a particular defect moves and its cross section changes by an average amount $\langle \delta\sigma_i \rangle = \beta\sigma$, we have

$$\delta\rho_i = K(\beta\sigma/V) \ . \tag{7}$$

If we now assume a concentration n_m of mobile defects with uncorrelated motion, then the total normalized fluctuation in the sample is

$$N\langle (\delta\rho)^2 \rangle / \rho^2 = (nl\beta\sigma)^2 (n_m/n) \ , \tag{8}$$

where N is the number of atoms in the sample and $n = N/V$.

To test this model rigorously, one should measure the noise and the concentration of *mobile* defects in a particular sample. To date this has not been done. However, we have measured radiation-induced $1/f$ noise in Cu films at 90 K as a function of the *total* added defect concentration.[7] We thus have an upper limit on the mobile defect concentration, and can check the consistency of the LI model with these measurements.

One often characterizes $1/f$ noise in terms of the Hooge parameter[18] α as $NS_\rho(f)/\rho^2 \approx \alpha/f$, where $S_\rho(f)$ is the spectral density of the resistivity fluctuations and f is the frequency. Integrating this expression over the experimental bandwidth (0.1 to 25 Hz) we obtain $N\langle (\delta\rho)^2 \rangle / \rho^2 \approx \alpha \ln(25/0.1) \approx 5.5\alpha$. For an added resistivity $\Delta\rho \simeq 10$ nΩ cm, we found that[7] the parameter α changed by $\Delta\alpha \simeq 1.6 \times 10^{-3}$. Using Eq. (8) and $n \simeq 8.5 \times 10^{22}$ cm^{-3}, $l \simeq 800$ Å, $\beta \simeq 0.15$, and $\sigma \simeq 4\pi/k_F^2 \simeq 6.5 \times 10^{-16}$ cm^2, one can account for the added noise with $(n_m/n) \simeq 2 \times 10^{-6}$. An added resistivity $\Delta\rho \simeq 10$ nΩ cm corresponds to a fractional concentration of Frenkel pairs[19] $(n_{\text{FP}}/n) \simeq 4 \times 10^{-5}$, and we thus require about 5% of the added defects to be mobile within the experimental bandwidth to account for the added noise. If we assume the $1/f$ spectrum extends over a larger bandwidth, we require larger numbers of moving defects: For example, a bandwidth of 10 decades would require 20% of the added defects to be mobile. These numbers are quite reasonable, especially

TABLE I. Resistivity tensor components and anisotropy parameter β for six types of defects. The components ρ_{xx}, ρ_{yy}, and ρ_{zz} are from Ref. 15; ρ_1, ρ_2, and ρ_3 are the corresponding principal moments. For each defect type, the upper row lists values for the bare defect, while the lower row includes effects from surrounding lattice relaxation. All resistivities are normalized to that of the bare monovacancy.

Defect type	ρ_{xx}	ρ_{yy}	ρ_{zz}	ρ_1, ρ_2	ρ_3	β
Monovacancy	1.00	1.00	1.00	1.00	1.00	0.00
	0.92	0.92	0.92	0.92	0.92	0.00
First-neighbor divacancy	1.80	1.80	2.27	2.27	1.33	0.14
	1.65	1.65	2.13	2.13	1.17	0.16
Second-neighbor divacancy	2.04	1.85	2.04	2.04	1.85	0.03
	1.73	1.70	1.73	1.73	1.70	0.01
180° trivacancy	3.54	2.58	2.58	3.54	1.62	0.20
	3.35	2.48	2.45	3.35	1.58	0.19
⟨100⟩ split interstitial	1.08	1.08	2.33	1.08	2.33	0.25
	2.19	2.17	3.63	2.18	3.63	0.16
⟨110⟩ split interstitial	1.70	1.70	1.07	1.07	2.33	0.25
	3.48	3.47	2.44	2.44	4.51	0.20

when one considers that all the added defects are metastable and hence inherently mobile. Most of the defects anneal at temperatures below 300 K, via thermally activated free migration and defect trap release.[19] Precisely how the type of motion which produces noise relates to the motion involved in annealing has yet to be determined.

When the temperature of the films is raised above 90 K, the induced $1/f$ noise increases.[8] In particular, the films become extremely noisy at temperatures near 200 K, just below the temperature at which large-scale annealing of the defects occurs via vacancy migration.[7] To account for the high level of noise that we observe at 185 K after annealing the film at 200 K for 5 min, the LI model would require approximately 50% of the remaining defects to be mobile within the experimental bandwidth, although uncertainties in the values of β and σ make this estimate somewhat rough. In fact, such a large fraction may not be unreasonable since most of the defects are on the verge of annealing and a high degree of thermal motion is likely. Furthermore, in the case in question a measurable decrease in the average resistivity of the sample occurred *during* the noise measurement, confirming that there was indeed substantial defect motion. This also suggests that nonstationary processes not specifically included in the LI model (e.g., defect dissociation, free migration, and annihilation) may have added substantial amounts of noise.

Measurements of $1/f$ noise have been made on a wide variety of metal films at room temperature, with α in the range[6] $10^{-5} < \alpha < 10^{-1}$. In relatively clean metals ($l \approx l_{\text{in}} \approx 400$ Å) the LI model would require mobile defect concentrations in the range $2\times10^{-7} < n_m/n < 2\times10^{-3}$ to account for this range of $1/f$ noise if one assumes a 10-decade bandwidth. These concentrations of mobile defects would seem reasonable, especially if we include atoms at grain boundaries and surfaces[9,10] as "lattice defects."

In concluding this discussion of the LI model we note that it requires defects which are each composed of multiple scattering centers within one or two lattice constants of each other, and not a system composed of randomly placed point scattering centers with no spatial correlation. In such a random system with average scattering-center separation $R \gg 7/k_F$ one expects less noise than outlined above.[20] However, in a (mostly) crystalline real metal, anisotropic agglomerates of near-neighbor scattering centers are in fact the norm,[19] in the form of "split" interstitials, defect clusters, dislocations, grain boundaries, surfaces, etc., with the monovacancy and substitutional impurities notable exceptions. One therefore expects large interference effects in these structures, with corresponding resistivity fluctuations when scattering centers move.

We now turn to a discussion of the $1/f$ noise produced by the LI and UCF models, both of which are based on scattered electron interference effects, but which differ significantly in several ways. The LI model considers *single* scattering events by defects in relatively clean systems. Only interference from nearby scattering centers produces a resistivity change when a defect moves. The UCF model,[13] on the other hand, considers interference effects from *multiple* elastic-scattering events in disordered systems, and is nonlocal in the sense that the motion of any scattering center within an inelastic diffusion length L_{in} can affect the interference. The theory was originally formulated for the limit $k_F l_e \sim 1$, but should remain applicable when $k_F l_e > 1$ provided $l_{\text{in}} \gg l_e$. To obtain a quantitative estimate of the $1/f$ noise predicted by the UCF model, we consider the three-dimensional case in which all sample dimensions are larger than L_{in} and assume that the conductance fluctuations from all mobile defects within a cube of side L_{in} add incoherently. We take $\rho = m_e v_F/ne^2 l$ (v_F is the Fermi velocity), assume one conduction electron per atom, and assume sufficient disorder to make $l \approx l_e$. From Eqs. (3) and (6) of Ref. 13, assuming $\alpha(k_F \delta r) = f(\hbar/k_B T \tau_{\text{in}}) = 1$, we obtain

$$N\langle(\delta\rho)^2\rangle/\rho^2 = (m_e v_F/h)^2 (L_{\text{in}}/l)^3 (n_m/n)\sigma = (m_e/h)^2 (v_F^7 \tau_{\text{in}}^3/27 l^3)^{1/2} (n_m/n)\sigma \ , \quad (9)$$

where we have set $L_{\text{in}} = (v_F l \tau_{\text{in}}/3)^{1/2}$ with τ_{in} the inelastic scattering time. Equation (9) is valid provided $(n_m/n) \leq (l/n\sigma L_{\text{in}}^2)$. The noise saturates for larger (n_m/n) since the conductance fluctuation per cubic volume L_{in}^3 cannot exceed[13] e^2/h. We see that at a given temperature and mobile defect concentration the UCF model predicts that $N\langle(\delta\rho)^2\rangle/\rho^2$ scales as $l^{-3/2}$ while the LI model predicts a scaling with l^2. Thus, in sufficiently dirty metals the UCF mechanism is expected to dominate while in sufficiently clean metals the LI mechanism takes over.

Taking the ratio of Eqs. (8) and (9) we find

$$\frac{\langle(\delta\rho)^2\rangle_{\text{LI}}}{\langle(\delta\rho)^2\rangle_{\text{UCF}}} = \left(\frac{n\beta h}{m_e}\right)^2 \sigma \left(\frac{27 l^7}{v_F^7 \tau_{\text{in}}^3}\right)^{1/2} . \quad (10)$$

Using the values of n, β, and σ listed above with $v_F = 1.6\times10^8$ cm s^{-1} and $\tau_{\text{in}} \approx \hbar/k_B T$ at $T = 300$ K, we find that the two mechanisms produce equal levels of $1/f$ noise when $l \approx l_e \approx 25$ Å $\equiv l_0$. In metals with $l_e > l_0$ we thus expect the LI mechanism to dominate, while in clean metal films with $l_e > l_{\text{in}}$ the UCF theory does not apply,[13] leaving only the LI mechanism. Equation (10) also demonstrates that for a given mean free path the UCF mechanism becomes progressively more important as the temperature is lowered to produce an increase in τ_{in}.

In summary, we have used Martin's calculations of defect resistivity to make quantitative predictions of $1/f$ noise generated by moving defects. Martin's work emphasizes the importance of local interference effects, which can produce rms fractional asymmetries in the cross-section of defects ranging from 0 to 0.25. These calculations can account for the $1/f$ noise generated by radiation-induced defects in Cu films at 90 K provided one assumes that approximately 5% of the *added* defects move at frequencies within the experimental bandwidth, though a larger percentage is required when the irradiated films are at a higher temperature. In the case of metal films at room temperature, the measured range in the level of $1/f$ noise over a similar bandwidth requires a fractional concentration of mobile defects (n_m/n) between 10^{-7} and 10^{-3}. At room temperature, the $1/f$ noise in metal films with mean free paths greater than about 25 Å should be dominated by the LI mechanism rather than by the UCF mechanism; at lower temperatures and/or shorter mean free paths the UCF mechanism is expected to dominate.

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