Excess (1/f) noise in metals

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We have measured the temperature dependence of the excess (1/f) voltage noise in thin films of the group-IB transition metals and in Ni. In the group-IB metals, the magnitude of the noise increases rapidly with temperature below room temperature and reaches a peak in Ag and Cu at $T_p \simeq 410$ K and $T_p \simeq 490$ K, respectively. In Ag, the noise dips through a valley at approximately 520 K, and increases again up to 600 K, while in Au there is flattening but no peak up to 525 K. In Ni, a somewhat slower increase in noise magnitude with temperature is observed, but in contrast to model predictions, which suggest a sharp peak in the noise at the Curie point, only peak structure is observed near T_c. Measurements of the power spectrum $S_v(f)$ ($\propto f^{-\alpha}$) show that α decreases slowly with increasing temperature for all four metals. The role of the substrate is examined by measuring the noise in films on substrates whose thermal properties differ greatly (sapphire and fused silica). A comparison of the results leads to the identification of two types of noise in metal films and the explanation of some apparent contradictions in existing data.

I. INTRODUCTION

Voltage noise with a 1/f frequency spectrum is present in a tremendous variety of physical systems. One of the earliest observations of this phenomenon was by Johnson,[1] who observed excessive low-frequency noise with a 1/f spectrum while measuring shot noise in vacuum tubes. 1/f noise has also been seen in ionic solutions,[2] where the low-frequency voltage fluctuations have been shown to be due to carrier mobility fluctuations rather than fluctuations in carrier number, and in the organic conductor tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ), where the magnitude of the noise shows critical behavior in the neighborhood of the 53-K metal-insulator transition.[3] However, the phenomenon of 1/f noise has perhaps been most widely studied in semiconductors,[4-7] where the magnitude of the noise is much larger for a given sample volume than in metals. In semiconductors, the dependence of the noise on temperature, impurity concentration, and other material parameters has been extensively examined. Of particular interest have been the studies directed at determining the limits of 1/f spectral region. Caloyannides[8] has recently extended the low-frequency limit to less than 10^{-6} HZ and found no significant deviation from the 1/f law.

In spite of the vast accumulation of experimental data, the origin of 1/f noise, even in semiconductors, remains enigmatic. Several models have been proposed to explain the origin of the noise in semiconductors,[9-11] the most widely accepted of which is probably that due to McWhorter.[12] In McWhorter's model, the 1/f noise arises from fluctuations in the occupancy of surface states with a distribution of relaxation times. While this model is well verified in certain cases, it appears experimentally that 1/f noise in semiconductors can also arise from bulk effects.[13]

The study of 1/f noise in metals began with measurements on discontinuous films, i.e., very thin films which no longer exhibit bulk properties. Discontinuous films of gold and platinum were measured by Williams and co-workers[14,15] in order to correlate the noise with the film structure in an attempt to characterize the conduction process. The origin of the 1/f noise in these systems appears to depend directly on the island structure of the films and is thought to have little direct bearing on noise in bulklike metals.

The first measurements of 1/f noise in continuous (bulklike) metal films are due to Hooge and Hoppenbrouwers (HH), who studied gold films on glass substrates.[16] They found that the room-temperature noise in these systems could be well characterized by the phenomenological expression

$$S_v(f)/V^2 = 2.4 \times 10^{-3}/N_c f , \tag{1}$$

where $S_v(f)$ is the frequency dependent spectral density of the voltage noise, V is the dc voltage across the sample, and N_c is the number of charge carriers in the sample. Note that the spectral density is assumed to be independent of temperature and of material parameters. Indeed, Hooge[13] has tabulated a vast amount of experimental data on both metals and semiconductors which is in order of magnitude agreement with Eq. (1). Furthermore, Hooge has suggested that Eq. (1) is valid for all uniform homogeneous materials regardless of the dominating conduction processes.

Taken at face value, the universal validity of Eq. (1) is a striking statement providing a powerful

key to the understanding of the origin of $1/f$ noise. First, the dependence of $S_v(f)$ on the total number of charge carriers N_c implies that $1/f$ noise is strictly a bulk, rather than a surface, phenomenon. Moreover, the universal value of the proportionality constant, 2.4×10^{-3}, implies that the origin of the noise is universal, encompassing metals, ionic solutions, and semiconductors. Unfortunately, the strong dependence of the noise in semiconductors on the oxidation state of the surface[12] and the magnitude of the noise in the semimetal bismuth[17] are in strong disagreement with Eq. (1). Furthermore, the noise in semiconductors can vary by orders of magnitude depending on the details of the sample preparation. Thus, the Hooge formula cannot completely characterize $1/f$ noise in semiconductors and surface effects as exemplified by the McWhorter mechanism must be playing some role.

In this study, we focus attention on metal films where there is widespread agreement on the magnitude of the voltage noise at room temperature. Moreover, the noise in metals appears to be independent of the state of the surface[17] and probably is, therefore, a bulk phenomenon. Clarke and co-workers have made extensive observations on a variety of metals at room temperature[17,18] and near the superconducting transition in Sn and Pb.[19,20] The most striking of their results are the scaling of the noise with $(dR/dT)^2$, the slope of the resistance vs temperature curve, near the superconducting transition temperature, and the observance of spatial correlations in the noise over distances comparable to thermal diffusion lengths. In light of these observations, they have proposed a thermal diffusion model to account for the origin of $1/f$ noise in metal films.

Use of diffusion-type models in the $1/f$ noise problem is well known. Richardson[21] has proposed a rather general model in which sample resistance is linearly coupled to a diffusing quantity and noise is generated by the fluctuating resistance. The model allows either energy or particle concentration to be used as the diffusing variable and is general enough to handle both metals and semiconductors. But Richardson found that, except in very special cases, the resulting spectrum is not $1/f$. Lundstrom *et al.*[22] have reexamined one of these special cases and have shown that if the diffusing variable is spatially correlated (instead of uncorrelated as in the standard Langevin form), an extended $1/f$ region can result. The physical origin of this spatial correlation is unclear, however.

Voss and Clarke[17] (VC) use a diffusion approach in which temperature is the fluctuating quantity. Temperature fluctuations cause resistance fluctuations which in turn cause voltage fluctuations according to the equation

$$S_v(f)/V^2=\beta^2 k_B T^2/C_v[3+2\ln(l/\omega)]f, \tag{2}$$

where $\beta=(1/R)(dR/dT)$, the temperature coefficient of resistance, C_v is the total heat capacity of the sample ($C_v=3Nk_B$ for metals at room temperature, where N is the number of atoms in the sample), and l and ω are the sample length and width, respectively. The $1/f$ frequency dependence does not, of course, follow directly from this standard diffusion equation approach, but is put in *ad hoc*. Nevertheless, coupled with an appropriate normalization, this approach gives noise magnitudes in good agreement with experimental values at room temperature. In a second approach using the model of Lundstrom *et al.*,[22] VC were able to calculate a power spectrum quite similar to Eq. (2) whose frequency dependence is, in fact, $1/f$ over a certain frequency range. Voss and Clarke argue that this model may account in some manner for the effects of the substrate, but a detailed justification of this approach is not given. Nevertheless, in both cases the scaling with $(dR/dT)^2$ near the superconducting transition and the existence of thermal diffusion lengths are explained. Equation (2) also predicts the temperature dependence of the noise which will be examined in detail below.

In this article, we present measurements of the temperature dependence of $1/f$ noise in metals of fused silica and sapphire substrates. Previously, no systematic study has been made. Hooge and Hoppenbrouwers[16] observed a weak temperature dependence upon direct immersion of gold films on glass substrates in liquid-nitrogen and liquid-helium baths. Voss and Clarke[17] found similar results for Au and Bi films on glass substrates. Unfortunately, during the course of measuring the noise, driving current densities are used which are sufficiently high to cause substantial local heating of the sample. Therefore, as we will discuss in Sec. II, conventional thermometry is unacceptable and these measurements must be considered approximate. This work examines the low-temperature behavior in detail and extends the measurements above room temperature. Some of these results have been reported previously.[23,24] For purposes of comparison in the course of the work, the following expression for the spectral density will prove useful:

$$S_v(f)\propto V^\beta/Nf^\alpha, \tag{3}$$

where N is the number of atoms in the sample. At room temperature, generic $1/f$ noise includes

all α in the range $0.9 \lesssim \alpha \lesssim 1.4$. We will show that α is temperature dependent. Linear response theory predicts that $\beta = 2.0$, but experimentally, β can deviate somewhat from that value.[15,25]

In Sec. II we describe the experimental details and techniques. In Sec. III we present the temperature dependence of the noise in the group IB transition metals on sapphire substrates, above and below room temperature, and examine the dependence of the noise on applied dc voltage. Section IV contains the temperature dependence of the noise in Ni, with emphasis on noise near the Curie point. In Sec. V, we examine the role of the substrate by presenting results of noise measurements on Cu and Ag films on fused silica substrates, and in Sec. VI, the rapid temperature dependence of the noise is reexamined at low temperature, and the effects of annealing and thickness are considered. Section VII contains a discussion of our results along with our conclusions.

II. MEASUREMENTS OF NOISE IN METAL FILMS

Measurements of the noise power magnitude and frequency spectrum of a variety of metal films were made using the circuit shown in Fig. 1. A dc current is supplied to the sample by a battery pack in series with a limiting resistor. The noise signal is passed to an impedance matching transformer (PAR 190), a low-noise preamplifier (PAR 113), and finally to a frequency spectrum analyzer (SAICOR 51B). For several intermediate resistance samples, the impedence matching transformer (PAR 190) was replaced by a second preamplifier (PAR 113) with identical results. The SAICOR 51B allows spectral analysis to frequencies as low as 0.1 Hz and as high as 1 MHz, and provides for the possibility of spectral averaging. In general, spectra were taken in the range $0.25 \leq f \leq 50$ Hz as a function of temperature. This allows for a point by point determination of the exponents in Eq. (3) as well as the noise magnitude by examining the signal at say 20 Hz. The magnitude of the measured noise is reduced, however, from the actual voltage fluctuation magnitude by the presence of the finite

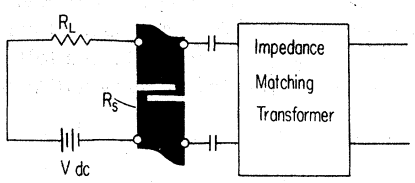

FIG. 1. Circuit configuration for noise measurement showing typical sample geometry.

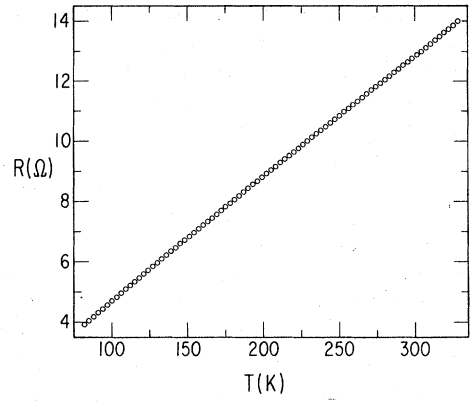

FIG. 2. Resistance vs temperature for an annealed, 1400-Å-thick Ag film.

limiting resistor R_L in Fig. 1, since R_L and the sample (R_s) effectively serve as a voltage divider for the noise signal. In order to correct for this, the measured noise power is multiplied by the factor $[(R_L + R_s)/R_L]^2$ to arrive at the total noise signal.[26] Typically, this correction is less than 20% even at the highest applied voltages. The Nyquist noise background is measured in the absence of current and subtracted from the total noise power to arrive at the excess noise spectrum. Measurements with a wire-wound resistor in place of the sample assured that noise was not generated in the measuring circuitry.

Thin films of the various metals were prepared by thermal evaporation onto sapphire or fused silica substrates in a vacuum of 1×10^{-7} Torr. Typical sample dimensions are: length: 200–2500 μ; width: 7–60 μ; thickness: 400–1600 Å. The resistivities and temperature coefficients of resistance of the metals varied in the as prepared films. However, after typical annealing in a partial pressure of helium (1 h at 625 K for Ag, Cu, Au; 5 min at 925 K for Ni) both these quantities varied from bulk values by less than 15%. The dc bias voltage applied to the films ranged from 0.25 to 3.0 V. A typical R vs T curve for an annealed 1400-Å Ag film is shown in Fig. 2.

The standard four-probe measurement technique was used. Contacts were made of various materials for different temperature ranges. Pressed indium, 60–40 Sn-Pb solder, and Dupont No. 8032 conductive silver composition were used. All gave identical results in the temperature regions where comparison could be made. In addition, measurements of the noise in air, helium, and vacuum gave identical results.

In order to maximize the ratio of excess noise to Nyquist noise, current densities in excess of 2×10^6 A/cm^2 are commonly used.[17] Current densities this large cause substantial local heating

of the film over the ambient substrate temperature, which makes the use of conventional thermometry impossible. To circumvent this problem, we first determine the resistance of the sample as a function of temperature at low current densities. The noise is then measured (in the presence of a large driving current) as a function of sample resistance. This technique allows the sample to serve as its own thermometer.

III. TEMPERATURE DEPENDENCE OF NOISE IN NOBLE METALS

A typical frequency spectrum is shown in Fig. 3 for an annealed 800-Å Ag film at 390 K. α, defined by Eq. (3), is 1.03 ± 0.06. Note especially the absence of knees or bends in the spectrum. The temperature dependence of α is shown in Fig. 4. For all four metals, α is greater than 1.0 at room temperature. In Ag, α changes by about 30% between 200 and 525 K, while in Cu, α changes by 25%, and in Au by 50% over the same temperature range. Most of the points in Fig. 4 are derived from frequency spectra taken over the range $0.25 \leq f \leq 50$ Hz, and represent an average over several samples. This accounts for the slight variation (within experimental error) between the particular value shown for α in Fig. 3 and the value shown in Fig. 4. α was found to be independent of film thickness and also independent of the annealing state of the sample. Probably most important is the general trend shown in Fig. 4: α decreases with increasing temperature in each metal studied independent of whether or not there is detailed structure in the magnitude of the noise (see below).

The temperature dependence of the magnitude of the noise shows striking features in the noble metals. In this section, we present the T dependence of the noise in Ag, Cu, and Au on sapphire substrates. In Sec. V, we will discuss the results for Au and Cu films on fused silica substrates in the context of examining the role of the substrate on $1/f$ noise. The temperature dependence of the magnitude of the noise at 20 Hz in an 800-Å Ag film on a sapphire substrate is shown in Fig. 5. Note the rapid decrease of the noise below room temperature, the peak at $T_p \cong 410$ K, the valley at approximately 520 K, and finally the upturn in the noise at the highest temperatures. These features are all qualitatively reproduced in a 1400-Å Ag film, the only quantitative difference being a somewhat weaker dip in the region around 500 K, a slight upward shift (to about 430 K) in the position of the peak, and a slightly stronger decrease at low temperatures. In gen-

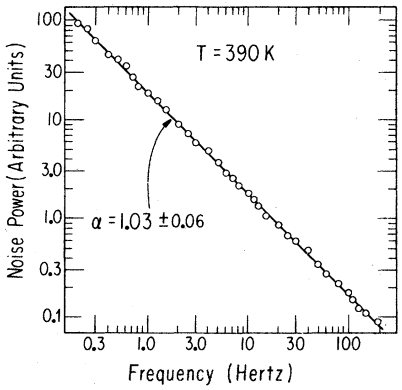

FIG. 3. Noise power frequency spectrum in Ag at 390 K.

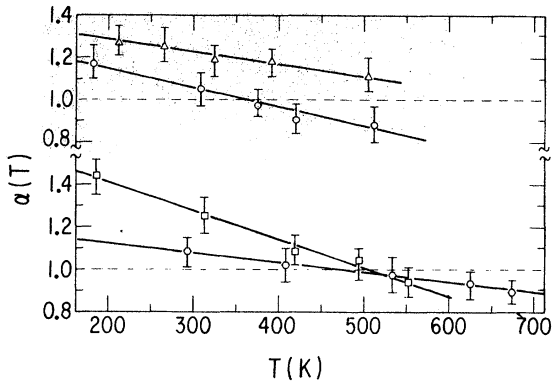

FIG. 4. Frequency exponent α [$S_v(f) \propto f^{-\alpha}$] vs temperature for Ag (0), Cu (△), Au (□), and Ni (o).

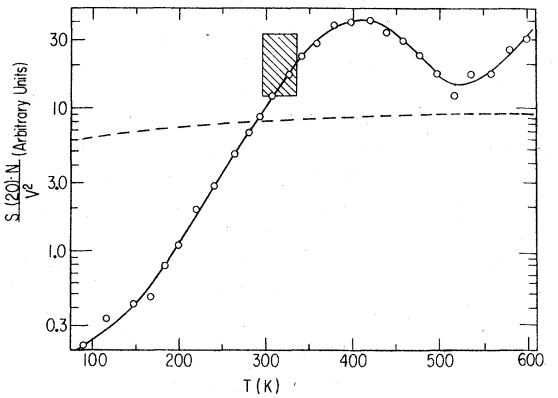

FIG. 5. Voltage noise measured at 20 Hz, $[S_v(20)/V^2]$ N vs temperature for 800-Å Ag film on sapphire substrate. The dashed line is the prediction of the temperature fluctuation model [Eq. (2)]; the hatched box represents a summary of measurements on Ag by VC (Ref. 17).

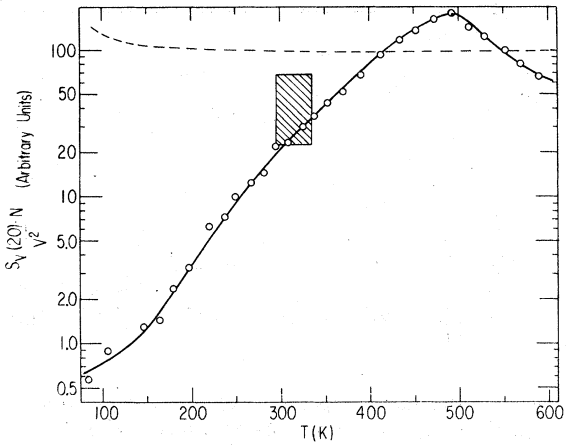

FIG. 6. $[S_v(20)/V^2]N$ vs temperature for 800-Å Cu film on sapphire substrate. The dashed line is the prediction of the temperature fluctuation model [Eq. (2)]; the hatched box represents a summary of measurements on Cu by VC (Ref. 17).

eral, however, the temperature-dependence curves for films of different thicknesses fall virtually on top of one another when properly scaled according to Eq. (3) for differences in absolute magnitude. The hatched box at room temperature represents a summary of measurements on Ag films by VC. The room-temperature agreement between the two results is clear. The dashed curve represents the prediction of the temperature-fluctuation model of Ref. 17 and Eq. (2). While the agreement at room temperature is satisfactory, it is quite clear that the temperature-fluctuation model completely fails to describe the temperature dependence of the noise in Ag films. Similar disagreement between experiment and theory was observed in all materials. A more detailed discussion of this model is found in Sec. V.

The experimentally determined absolute magnitude of the noise and the comparison to model predictions require careful discussion. First of all, annealing decreases the room-temperature magnitude of the noise by a factor of 2 to 6, depending on film thickness. After a single anneal, however, data runs are reproducible and data taken on decreasing temperature runs confirm the structure taken on increasing temperature runs, thus ruling out annealing during measurement as the origin of the structure in the data. Secondly, changes in α with temperature cause slight difficulty in using $[S_v(20)/V^2]N$ [see Eq. (3)] as a measure of the noise magnitude. A plot of $[S_v(20)/V^2]Nf^\alpha$ would remove the effects of the temperature dependence of α from the change in noise magnitude with temperature. But the precision of the noise magnitude data is rather higher than the determination of α, so significant scatter would be added by this procedure. In addition, f^α changes much more slowly with temperature than $[S_v(20)/V^2]N$; only approximately 10% of the total change of noise magnitude with temperature can be attributed to the temperature dependence of α. The general shape and structure of the temperature dependence of the noise are unaffected by this effect. Finally, comparison of theory to experiment is difficult because of difficulty in determining N, the number of atoms in the sample, and even for seemingly identical samples, noise can vary by as much as a factor of 2 in absolute magnitude.[17] Nevertheless, $[S_v(20)/V^2]N$ is plotted to allow for comparison of noise between samples of different volumes. To within the above mentioned uncertainty, the absolute magnitude of the noise at $T_p = 410$ K in Ag films is $[S_v(20)/V^2]N = 3.4 \times 10^{-3}$ Hz.$^{-1}$

Figure 6 shows the temperature dependence of the noise at 20 Hz in an 800-Å Cu film on a sapphire substrate. The hatched box and dashed line again represent a summary of the measurements of VC and the prediction of the temperature-fluctuation model, respectively. In Cu the observed noise is slightly smaller than the model prediction at room temperature, and as in Ag, there is no similarity between the temperature dependence predicted by the model and the experimental data. In this film, the noise peaks at $T_p = 490$ K and continues to decrease above that to the highest temperatures observed. Rapid decrease in the noise below T_p is again evident. The

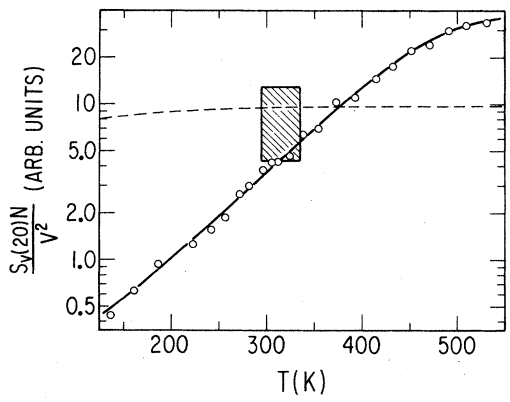

FIG. 7. $[S_v(20)/V^2]\,N$ vs temperature for 800-Å Au film on sapphire substrate. The dashed line is the prediction of the temperature fluctuation model [Eq. (2)]; the hatched box represents a summary of measurements on Au by VC (Ref. 17).

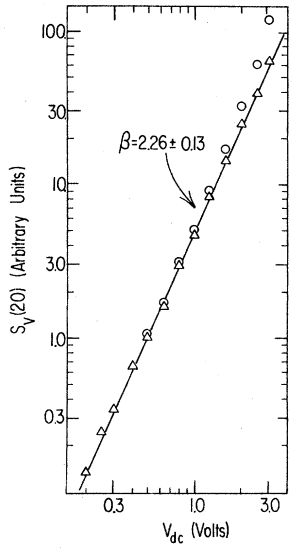

FIG. 8. $S_v(20)$ vs V_{dc} for 800-Å Ag film on sapphire substrate. 0—direct measurement. △—corrected for temperature dependence of noise.

noise in this film changes by nearly three orders of magnitude between 100 and 490 K. The absolute magnitude of the noise at $T_p = 490$ K is $[S_v(20)/V^2]N = 2.4 \times 10^{-3}\,\mathrm{Hz}^{-1}$.

Figure 7 shows our results for Au. Significant flattening is observed in the noise above about 450 K, but no peak was observed up to 525 K. For temperatures above 525 K, our data on Au were not totally reproducible and hence are not shown. The magnitude of the noise at room temperature is again below the temperature-fluctuation model prediction but slightly above the prediction of the phenomenological expression of HH [Eq. (1)]. At $T = 308$ K, $[S_v(20)/V^2]N = 2.0 \times 10^{-4}\ \mathrm{Hz}^{-1}$. We observe rapid temperature dependence of the noise below room temperature, while HH report[16] very little change in noise in gold films on glass substrates upon direct immersion in liquid-nitrogen and liquid-helium baths (T dependence no faster than $T^{1/2}$, or activation energy less than 0.001 eV). We believe much of this difference can be accounted for by differences in substrates as will be discussed in detail in Sec. V.

In addition to the temperature dependence of the noise, we have examined the dependence of the noise on the applied dc voltage. The results are shown in Fig. 8 for an 800-Å Ag film on a sapphire substrate. At dc bias voltages above 0.5 V, significant heating begins to manifest itself. Through the use of the sample as its own thermometer, and using the noise vs temperature data of Fig. 5, we also plot in Fig. 8 the voltage dependence of the noise corrected to remove the effects of sample heating. β at room temperature in Eq. (3), determined from the corrected data, is $\beta = 2.26 \pm 0.13$. The bending of the uncorrected data above this line in Fig. 8 shows that sample heating plays a significant role in the voltage dependence of the noise. The deviation of β from 2.0 is discussed in Sec. VII. We should point out, however, that it has very little effect on the measured dependence of the noise, since this effect is very nearly temperature independent and is completely overwhelmed by the huge intrinsic change in noise magnitude with temperature.

The temperature dependence of the noise in noble metals shows several striking features. α in Eq. (3) changes with temperature and a correct determination of β also requires taking the temperature dependence into account. The peak structure in the noise magnitude varies from metal to metal, but below about 350 K, a rapid decrease in the noise with decreasing temperature is evident in all three metals. In Sec. VI, we will attempt to characterize this low-temperature universal behavior in more detail.

IV. TEMPERATURE DEPENDENCE OF NOISE IN NICKEL

Measurements of $1/f$ noise near critical points should, in principle, provide a stringent test of models attempting to explain the origin of the noise. In particular, the temperature-fluctuation model of VC [Eq. (2)] has been examined in some detail near the superconducting transition in Sn and Pb,[20] where dR/dT has sharp temperature dependence, and the noise has been found to scale with $(dR/dT)^2$ over two decades. In contrast, our measurements on noble metals in the range 100 to 600 K show significant deviations from the temperature-fluctuation model predictions (Figs. 5–7).

One might argue that the dependence of the noise on absolute temperature represented in Eq. (2) needs modification, perhaps to include the thermal boundary effects at the sample-substrate interface, but the dependence of the noise on $(dR/dT)^2$ at fixed absolute temperature is essentially correctly given by Eq. (2). As a result, we have undertaken measurements on Ni near the Curie point where the temperature coefficient of resistance $\beta = (1/R)(dR/dT)$ peaks with a specific heatlike exponent[27] leading to a sharp peak in the predicted noise magnitude. Even if some other mechanism is responsible for the noise it is not unreasonable to expect the behavior in the neighborhood of a critical point could shed considerable light on its origin.

The temperature dependence of the magnitude

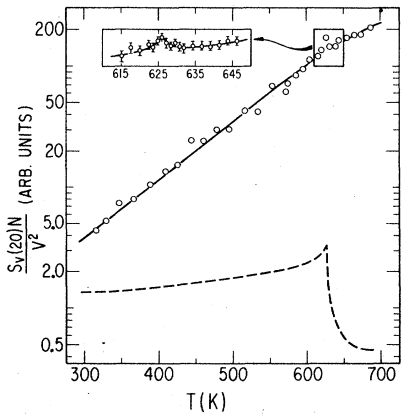

FIG. 9. $[S_v(20)/V^2]\,N$ vs temperature for 800-Å Ni film on sapphire substrate. The dashed line is the prediction of the temperature-fluctuation model [Eq. (2)]; the inset represents the results of detailed measurements very near the Curie point.

of the noise at 20 Hz in an 800-Å Ni film on a sapphire substrate is shown in Fig. 9. The general features are a rather rapid increase in magnitude above room temperature (though slower than in the noble metals) followed by a weak flattening at the highest temperatures. Specific features very near T_c (see inset) will be discussed. The temperature-fluctuation model prediction is also shown in Fig. 9. The temperature dependence of the model is given by $[S_v(f)/V^2] \propto \beta^2 T^2/C_v$ [see Eq. (2)]. To determine $\beta^2 T^2/C_v$, the value of β is derived from R vs T measurements on the same film for which the noise is measured. β shows a sharp peak at a temperature only slightly (~5 K) below the Curie temperature for highly annealed bulk samples,[28] indicating a film with good bulklike properties. Values for the specific heat, which is also sharply peaked near the Curie point, are taken from the measurements of Connelly, Loomis, and Mapother,[28] with the bulk Curie temperature matched to the film Curie temperature. As is clear from Fig. 9, both the magnitude and general structure of the data are in disagreement with the temperature-fluctuation model prediction. The absolute magnitude of the noise at $T = 320$ K is given by $[S_v(20)/V^2]N = 1.03 \times 10^{-2}$ Hz^{-1}, approximately a factor of 3 greater than the model at room temperature, and agreement becomes less satisfactory as temperature increases.

The results of careful measurements near the Curie point are shown in the inset of Fig. 9. The noise increases in magnitude by approximately 15% to a peak at $T_c \cong 626$ K. This peak coincides with the Curie temperature of our Ni film and one can therefore assume that the peak in the noise is a manifestation of the fluctuations associated with this second-order phase transition. The noise predicted by the temperature-fluctuation model, which also exhibits an anomaly at the Curie point due to the $\beta^2 T^2/C_v$ dependence, is nearly two orders of magnitude below the measured noise. In addition, direct measurements of the temperature fluctuations indicate that the effect of the substrate causes the model spectrum to be decreased, compared to the value shown in Fig. 9, by a factor of about 60 for sapphire substrates.[24] These two factors suggest that it is quite unlikely that temperature fluctuations are the cause of the noise peak and that $1/f$ noise is affected by critical fluctuations in a nontrivial way, thus opening a promising path for further investigation.

V. TEMPERATURE FLUCTUATIONS AND THE ROLE OF THE SUBSTRATE

In spite of its failure to correctly predict the temperature dependence of $1/f$ noise in various metal films on sapphire substrates (Figs. 5–7 and 9), the temperature-fluctuation model presented[17] by VC has several attractive features. The scaling of the noise magnitude with $(dR/dT)^2$ near the superconducting transition in Sn and Pb, and the observance of correlations in the noise over length scales comparable to thermal diffusion lengths in Bi films at room temperature are strong evidence that temperature fluctuations play a significant role in the origin of the noise. Therefore, it is useful to examine this model more closely in order to understand just how the expression Eq. (2) arises.

In the canonical ensemble of statistical mechanics the temperature is fixed and defined by the thermal reservoir. The energy of a system is contact with this reservoir can fluctuate, although its average value is fixed by the bath. In terms of these energy fluctuations, it is convenient to define the temperature *of the sample* as a fluctuating variable through the relationship $\Delta E = C_v \Delta T$. Temperature fluctuations can lead to resistance fluctuations, which in the presence of a constant current cause voltage fluctuations (voltage noise) according to

$$S_v(f) \propto I^2 \left(\frac{dR}{dT}\right)^2 \langle (\Delta T)^2 \rangle . \tag{4}$$

Multiplying and dividing by R^2 and using the thermodynamic result

$$\langle (\Delta T)^2 \rangle = (1/C_v^2) \langle (\Delta E)^2 \rangle = k_B T^2/C_v$$

yields

$$S_v(f) \propto V^2 \beta^2 k_B T^2/C_v , \tag{5}$$

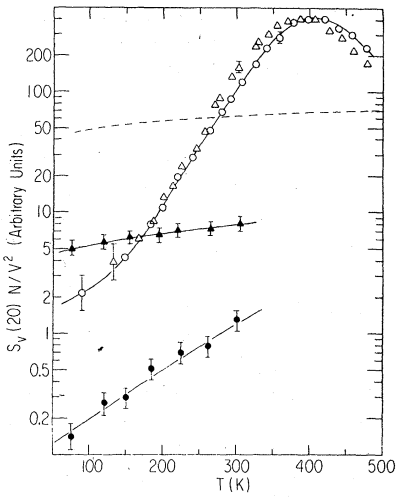

FIG. 10. Voltage noise, $[S_v(20)/V^2]\,N$ vs temperature for 800-Å Ag films on sapphire (0) and fused silica (△) substrates. Error bars do not reflect calibration errors due to uncertainty in N. Temperature-fluctuation induced noise, $[S_{v,\mathrm{TF}}(20)/V^2]\,N$ vs temperature from direct measurement of temperature fluctuations on 800-Å Ag films for sapphire (●) and fused silica (▲) substrates by Dutta, Eberhard, and Horn (Ref. 24).

which clearly explains the scaling of the noise with β^2. The use of a diffusion equation to determine the frequency spectrum accounts for the voltage correlations over thermal diffusion lengths, but unfortunately, a $1/f$ spectrum does not result naturally. Introducing the $1/f$ dependence *ad hoc* with the appropriate normalization gives the expression in Eq. (2) which is rather successful in predicting the room-temperature magnitude of the noise in various metals. Voss and Clarke do not, however, consider the effects of coupling to the substrate in detail in this model and this could account for its inability to describe the results of Sec. III, IV, and VI. A film on a substrate is a configuration not well approximated by the VC assumption of a regular subvolume of a uniform medium. In particular, the presence of the substrate could affect: (a) the noise magnitude in a given frequency range, if temperature fluctuations decay into the substrate instead of being confined to the film, (b) the frequency spectrum, as suggested by VC, and (c) the temperature dependence of the noise, due to thermal boundary conditions or other features.

In order to investigate experimentally the effects of the substrate on the noise, the noise was measured in Ag and Cu films on fused silica substrates. Thermal conductivities of fused silica and sapphire differ by an order of magnitude at room temperature; fused silica decreases by a factor of two at 100 K, while sapphire increases 100 times by 100 K. Thus, differences in the noise brought about by differences in the thermal properties of the substrate should be amply evident in the measurements of films on fused silica substrates.

The results for 800-Å Ag films on the two substrates are compared in Fig. 10. The absolute magnitude of the noise $[S_v(20)/V^2]N$ in the two films agrees to within the factor of 2 uncertainty discussed earlier; therefore, for quantitative comparison, the magnitudes have been matched at 400 K. Note that at room temperature, the measured noise is larger than the model prediction by about a factor of 2. The two results are virtually identical in all features over the entire 450-K range, except for a slight shift (~15 K) in the peak position.

Figure 11 is a comparison of the results for 800-Å Cu films on the two substrates. Again the magnitudes agree to within a factor of 2 at high temperatures and again they have been matched at 400 K. In Cu at room temperature, the measured noise is smaller than the model prediction by roughly a factor of 6. The most significant feature of Fig. 11 is the flattening of the noise on fused silica below 250 K. At these low temperatures, the shape of the noise begins to approach the *shape* of the temperature-fluctuation model prediction rather closely, though it falls below the model in magnitude by roughly a factor of 12 at 200 K. The shape of the data on fused silica is significantly different from the

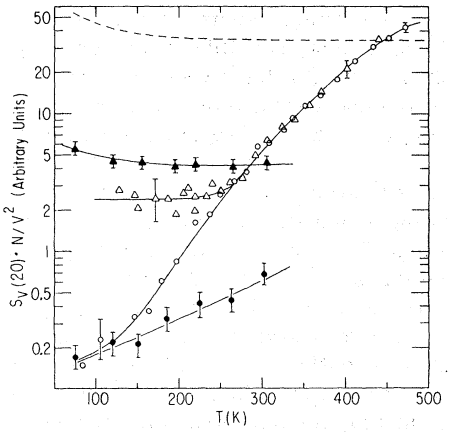

FIG. 11. Voltage noise, $[S_v(20)/V^2]\,N$ vs temperature for 800-Å Cu film on sapphire (○) and fused silica (△) substrates. Error bars do not reflect calibration errors due to uncertainty in N. Temperature-fluctuation induced noise, $[S_{v,\mathrm{TF}}(20)/V^2]\,N$ vs temperature from direct measurement of temperature fluctuations on 800-Å Cu films for sapphire (●) and fused silica (▲) substrates by Dutta, Eberhard, and Horn (Ref. 24).

rapid temperature dependence found on sapphire below 250 K.

In order to determine whether this difference in low-temperature behavior of the noise in Cu films on different substrates could indeed be attributed to the substrate, especially when measurements on Ag films apparently show no substrate dependence, direct measurements of the temperature fluctuation spectrum, as suggested by VC,[17] and reported by Dutta, Eberhard, and Horn in Ref. 24 were carried out. These measurements essentially record the thermal response of metal films to step function and delta function power inputs. The spectral density of the thermal response is identical (under conditions described in detail in Ref. 17 and 24) to the spectral density of temperature fluctuations in the film. Hence, by measuring this thermal response, we can determine the spectral density of the temperature fluctuations and [by use of Eq. (4)] of the noise.

The results are shown as solid circles and triangles in Figs. 10 and 11. These measurements will not be discussed here except to note that temperature fluctuations in metal films in contact with a substrate show roughly the same temperature dependence as the model, but that the magnitude of the fluctuations is decreased by approximately a factor of 8 for metals on fused silica and by approximately a factor of 60 for metals on sapphire, independent of metal. This result, in combination with the flattening of the noise in Cu films on fused silica at low temperatures, suggests the existence of two types of voltage noise in metal films.[24,29] (a) Noise which is weakly temperature dependent and strongly substrate dependent, possibly caused by temperature fluctuations. (b) Noise which is strongly temperature dependent and only very weakly substrate dependent, whose origin is presently unknown.

This identification of two types of noise provides the following explanation of our results in Cu: Noise of type B is dominant at room temperature and above for both types of substrate. For Cu on sapphire, noise type B is dominant except perhaps at the very lowest temperature measured. The temperature-fluctuation induced type A noise is a factor of 60 in magnitude less at room temperature than the temperature-fluctuation model curve (dashed line) in Fig. 11, and this is simply too small to become the dominant contribution except perhaps at 100 K. For Cu on fused silica, however, type A noise is only a factor of 8 below the temperature-fluctuation model curve at 200 K, and this agrees quite well (to within the uncertainty in the absolute magnitude of the noise discussed earlier) with the measured noise which is a factor of 12 below the temperature-fluctuation model curve at 200 K.

Note that in Cu at room temperature (Fig. 11), the measured noise is below the temperature-fluctuation model curve, while in Ag at room temperature (Fig. 10) measured noise is larger than the model. This implies that type B noise is a larger fraction of total noise in Ag than in Cu at room temperature, which explains why type A noise is not seen in Ag. This agrees quantitatively with direct measurements of the temperature fluctuations in fused silica substrates.[24] The proximity of the closed triangles to the open triangles and open circles (within experimental error), however, suggests that at somewhat lower temperatures for Ag films on fused silica substrates, type A noise could dominate. Our results in Ni suggest that it is type B noise which is dominant near the Curie point and undergoes a 15% anomaly. A more complete discussion of these results is given in Ref. 29.

The identification of two types of noise in metal films makes possible the explanation of a considerable amount of experimental data, some of it seemingly contradictory. First, it explains the difference between the results of Figs. 5–7 and 9, which are dominated by type B noise, and the noise near the superconducting transition[20] where temperature fluctuations and hence presumably type A noise play the dominant role. Second, it explains the apparent discrepancy between the weak temperature dependence of the noise in Au on glass seen by HH at low temperatures[16] and the much more rapid temperature dependence of noise in Au on sapphire in Fig. 7 at low temperatures. The thermal conductivity of glass is quite similar to that of fused silica in this temperature range, hence the measurements of HH were probably predominantly on type A noise, while the results in Fig. 7 are for type B noise. Third, it suggests that noise in Cu whiskers[29,30] which is significantly larger in magnitude than temperature-fluctuation model predictions and has very rapid temperature dependence, is dominated by type B noise. Finally, it explains the partial spatial correlations observed in the noise in Bi films at room temperature, since a fraction of the noise may be assumed to have temperature fluctuations as its origin.

One note of caution should be raised. Experimental measurements of type B noise disagree strongly in both magnitude and temperature dependence from the prediction of known models for noise originating from temperature fluctuations. In addition, as pointed out above, type B noise is very weakly substrate dependent while

direct measurements of temperature fluctuations show that temperature-fluctuation induced noise described by current models is strongly substrate dependent. Hence, known models for noise originating from temperature fluctuations can be rather convincingly ruled out as the origin of type B noise. It is impossible, however, to strictly rule out the possibility that type B noise could be produced by temperature fluctuations in some form. Other origins of type B noise appear at least as likely, and one possibility is presented in Sec. VII.

VI. RE-EXAMINATION OF RAPID TEMPERATURE DEPENDENCE OF THE NOISE

It is quite clear from the preceding discussion that for $T > 100$ K, essentially all of the noise observed in metal films is what we have called type B noise. In order to investigate the origin of this type of noise, it is useful to reexamine the rapid temperature dependence of the measured noise on sapphire substrates at low temperatures. The logarithm of the noise vs temperature for 800-Å annealed films of the group-IB metals is shown in Fig. 12. To good approximation, the temperature dependence for $T \lesssim 350$ K can be described by

$$S_v(20) = N_0 + N' \exp(-E_g/k_B T), \tag{6}$$

when E_g, N_0, and N' depend on the detailed characteristics of the particular film, including its thickness. In all samples studied, at room temperature $N_0 \ll N' e^{-E_g/k_B T}$. The activation energies E_g for the films in Fig. 12 are: Ag: $E_g = 1750$ K; Au: $E_g = 1400$ K; Cu: $E_g = 1250$ K. It is important to point out that only the noise shows activation. The sample resistance is characteristic of a metal and shows the expected linear R vs T behavior at all temperatures examined (Fig. 2).

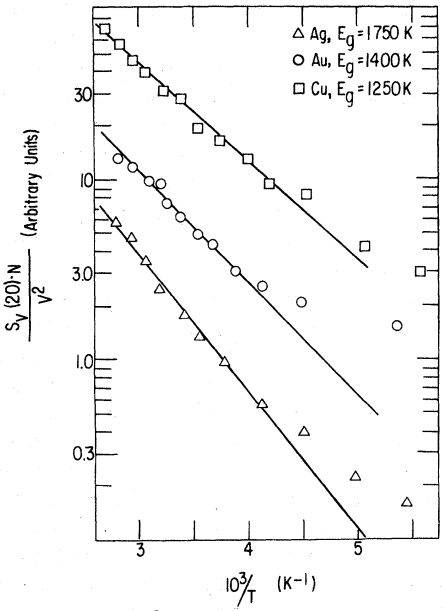

FIG. 12. $[S_v(20)/V^2]\,N$ vs inverse temperature at low temperatures for 800-Å annealed films of Ag(△), Au(○), and Cu(□) on sapphire substrates.

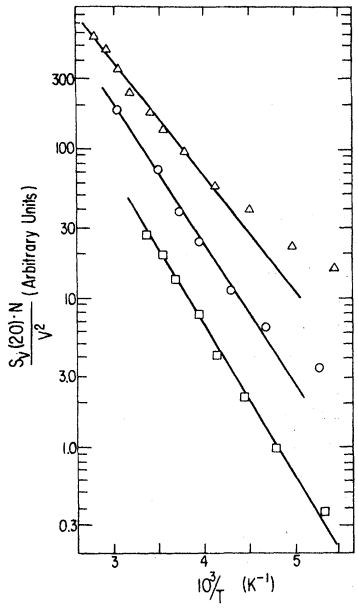

FIG. 13. $[S_v(20)/V^2]\,N$ vs inverse temperature at low temperature for annealed 800-Å Ag film ($E_g = 1750$ K) (△), annealed 1400-Å Ag film ($E_g = 2200$ K) (○), and unannealed 800-Å Ag film ($E_g = 2300$ K) (□) on sapphire substrates.

No systematic correlation could be found between the temperature of the noise peak T_p (if there was one), the low-temperature activation energy E_g, and the magnitude of the noise in the various metals. Perhaps this is an indication that an activated expression for the noise has no fundamental significance. Nevertheless, the activated form certainly highlights the rapid temperature dependence of the noise at low temperatures, and Eq. (6) represents a convenient parameterization of the noise data in different systems.

The dependence of the noise on both annealing and film thickness is illustrated in Fig. 13. We should stress that all of the qualitative features of the noise vs temperature curve, including the high-temperature peak structure, are independent of film thickness. The low-temperature activation energy, however, increases slightly as the film thickness increases. In particular, the 1400-Å

Ag film shown in Fig. 13 has $E_g = 2200$ K, approximately 25% larger than the result for an 800-Å film. The dependence of the noise on the state of anneal is also illustrated in Fig. 13. At room temperature the noise magnitude $[S_v(20)/V^2]N$ in 800-Å films decreases by a factor of 2 to 6 with annealing. In terms of the parameters of Eq. (6), the annealing effects can be characterized as follows: E_g decreases by approximately 25%, N' decreases by a factor of 15-20, and N_0 remains approximately constant. In order to further document the effects of the annealing procedure, scanning electron micrographs were taken on both as prepared and annealed films. We found that the annealing procedure increases the diameter of the grains in the film by roughly a factor of 4, up to approximately 2000 Å.

Finally, a check was made to determine whether details of the film surface might affect the noise. To prevent surface oxidation, an 800-Å Ag film was prepared with a 1000-Å SiO overlay. The SiO overlay was found to have no effect on either the magnitude of the noise or the frequency exponent α.

VII. DISCUSSION

The temperature dependence of the magnitude of $1/f$ noise in bulklike metals contains several striking features. Perhaps the most difficult to understand are the high-temperature peaks in Ag and Cu. Physical processes which occur at energy scales of 400–500 K are simply not familiar in bulk metals, thus explanation of the peak structure appears to be a formidable task. The data presented above certainly cannot be adequately explained by any of the existing models for the origin of voltage noise in metals. For example, the phenomenological (HH) formula, Eq. (1), which is in order of magnitude agreement with the noise data in various metals at room temperature, neglects temperature completely. The observation of strikingly different temperature dependences in different materials implies that the HH formula cannot be simply corrected by allowing the proportionality constant to be a universal function of temperature. Furthermore, the change in the low-temperature behavior of the noise as the film thickness increases implies that the $1/N$ dependence of Eq. (1) is at best approximate.

The temperature-fluctuation model of VC similarly fails to describe the temperature dependence of the noise in metal films. On the other hand, there is some evidence that temperature fluctuations lead to a small, weakly temperature dependent fraction of the noise. We have called this type A noise. In our experiments, type A noise is only important for Cu films on quartz substrates at temperatures below 250 K. However, at sufficiently low temperatures, or near superconducting transitions, one might expect this to be the dominant type of noise in all metals. At room temperature and above, and for metals on high thermal conductivity substrates at all temperatures studied, the noise appears to be generated by a mechanism other than temperature fluctuations. We have called noise from this second origin type B noise. The primary accomplishment of the present work is the characterization of type B noise. Type B noise is strongly temperature dependent and independent of the substrate used to support the films. Furthermore, type B noise displays a weak anomaly at the Ni Curie point suggesting that it is weakly coupled to the critical magnetic fluctuations.

Perhaps the most distrubing feature of our results is the deviation of the voltage exponent β from the value 2.0 predicted by linear response theory. Values of β greater than two seem to be present even after the effects of sample heating are taken into account. Furthermore, the experimental values of β are somewhat larger for films on fused silica substrates where sample heating by the applied current is significant even at the smallest dc biasing voltages. One might expect that values of β greater than two are a manifestation of the effects of terms of higher order than V^2 in a perturbation expansion for the noise. If this were the case, the voltage dependence of the noise would be represented more properly by a power series in V^2 than by a power law, V^β, with noninteger power. In the former case, $\lim_{V\sim 0}[dS_v(f)/dV^2]$ is constant, while in the latter $\lim_{V\sim 0}[dS_v(f)/dV^2] = 0$. Unfortunately, our data at small voltages are not accurate enough to distinguish between these two possibilities. Clearly, further experimental work is called for.

It is interesting to speculate regarding physical mechanisms which could account for this strongly temperature-dependent noise, and at the same time show a $1/f$ spectrum. Let us therefore speculate on whether the present experimental results could somehow be fitted into a trapping model similar to the one suggested by McWhorter[12] for $1/f$ noise in the semiconductors. The necessary ingredients for achieving a $1/f$ spectrum in such a model are the presence of shot noiselike spectra of the form $\tau/[1+(2\pi f)^2\tau^2]$ with a distribution of characteristic times inversely proportional to τ. For time constants arising from a process involving an activation energy, i.e., $\tau = \tau_0 \exp(E/k_B T)$, this second criterion is equivalent to a uniform distribution of activation energies. The

range of energies over which the distribution is uniform determines the limits of the $1/f$ region.

Now consider the creation and diffusion of vacancies in thin metallic films. The creation of vacancies is an activated process occurring preferentially near a surface or crystal deformation, with the equilibrium number of vacancies given by

$$n \propto \exp(-E_v/k_B T), \tag{7}$$

where E_v is the energy required to create the vacancy. Once created, these vacancies diffuse through the crystal in a process characterized by a diffusion constant D given (in very simple models) by

$$D = D_0 \exp(-E_D/k_B T), \tag{8}$$

where E_D is the energy barrier presented to the diffusing vacancy. If E_D is uniformly distributed in energy, then exactly as in the McWhorter model, we get a range of time constants from an activated process, in this case the diffusion of vacancies, and $1/f$ spectrum results. The temperature dependence of the magnitude of the noise on the other hand, could be due to the creation of vacancies, the magnitude of the noise being proportional to the *number* of diffusing vacancies. The observed frequency spectrum (Fig. 3) requires that the barrier heights E_D for vacancy diffusion be uniformly distributed at least over the range $0.15 \leq E_D \leq 1.1$ eV, a range consistent with observations on bulk metals. On the other hand, the observed activation energy for creation of vacancies E_v/k_B is found to be roughly 12 000 K in bulk metals while the noise activation energies in Fig. 12 are more nearly 1500 K. But vacancies are created preferentially at surfaces and deformations (e.g., at grain boundaries) so a decrease in bulk activation energy by roughly a factor of 8 is not unrealistic for films. Indeed as film thickness is increased to 1400 Å in Ag films, the activation energy increases to 2200 K (Fig. 13).

Our annealing studies are also roughly consistent with a vacancy diffusion model. Naively, one would expect N' in Eq. (6) to be proportional to the number of sites for easy formation of a vacancy-interstitial pair. If such pairs are formed at the grain boundary surfaces, then N' should be inversely proportional to the square of the diameter of the grains. Empirically, we have found that N' decreases by a factor of 15–20 with annealing, consistent with the observed factor of 4 increase in the grain diameter. On the other hand, the decrease in E_g with annealing would suggest, within such a model, that it is somehow easier to form a vacancy-interstitial pair within the grain boundary itself after the anneal. Certainly such a suggestion, along with the entire vacancy diffusion model, is highly speculative. Furthermore, the model does not relate to perhaps the most interesting feature of our data, the peak at high temperatures. On the other hand, a two-step process of creation-diffusion of vacancies does provide a strong temperature dependence not inconsistent with the low-temperature experimental data.

Perhaps the most interesting feature of the suggested vacancy diffusion model is the existence of a two-step process; first the creation, then the diffusion of vacancies. Almost certainly, any model which describes data of the type shown in Figs. 12 and 13 will have this feature. That is, two energy scales are required, one to create the $1/f$ frequency spectrum and one to obtain the temperature dependence. It is difficult to imagine a process based on one energy scale which would maintain a $1/f$ noise spectrum coincidentally with a strongly temperature-dependent magnitude.

At this stage it is of some interest to compare the results presented here on the temperature dependence of noise in bulklike metals, with measurements made on other systems. Of particular note are the measurements of noise in discontinuous metal films by Williams and coworkers.[14,15] The temperature dependence of the noise magnitude in discontinuous gold films[14] in the region just below room temperature shows that the noise is activated with activation energy in the range 300–600 K. Later measurements on discontinuous platinum films[15] also show activated behavior below room temperature with activation energy in the range 100–300 K, and a *peak* in noise magnitude near room temperature. It should be pointed out that the films measured by Williams and coworkers all had activated conductivity and in fact, the noise magnitude can be explained adequately by a model based on electron tunneling between the island of the film. Furthermore, the absolute magnitude of the noise is of orders of magnitude larger in these discontinuous films than in bulklike metals. Nevertheless, the similarity in the temperature dependence of the magnitude of the noise in bulklike and discontinuous metal films is certainly striking.

In semiconductors, where the noise depends strongly on the preparation of the surface, the situation is more complicated.[31] Still, there is some evidence that rapid temperature dependence of the noise magnitude may be present in these systems as well.[7,32] At this stage it would be inappropriate to suggest that the similarity in these temperature dependences implies any un-

animity of origin. Nevertheless, the similarities in the temperature dependence of the noise magnitude in these significantly different systems are certainly worthy of note.

In conclusion, we have measured the temperature dependence of excess noise in bulklike metal films. We have observed a striking high-temperature peak structure in the noise magnitude in thin films of noble metals, and rather weak structure in the noise magnitude near the Curie point in Ni. In addition, we have measured the substrate dependence of the noise and we have identified two types of noise in metal films. To our knowledge no existing model can adequately describe the data presented. Hopefully, however, these observations will shed enough light on the nature of noise in metals that a comprehensive model for its origin will soon be forthcoming.

ACKNOWLEDGMENTS

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