Dependence of $1/f$ Noise on Defects Induced in Copper Films by Electron Irradiation

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When 500-keV electron irradiation was used to induce defects in polycrystalline copper films maintained at 90 K, the spectral density of the $1/f$ noise voltage across the films increased by as much as 1 order of magnitude, while the resistivity increased by at most 10%. When the films were annealed at progressively higher temperatures, both the $1/f$ noise and the resistivity were reduced; however, at lower annealing temperatures, the fractional reduction in the added noise was substantially more than that in the added resistivity.

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Metal films, biased with a constant current, exhibit low-frequency voltage noise with a spectral density that scales approximately as $1/f$, where f is the frequency.[1,2] It is generally accepted that the noise arises from intrinsic resistance fluctuations.[3] Dutta *et al.*[1,4] showed that the noise could be explained by a random, thermally activated process with a broad distribution of activation rates, and suggested that crystalline defects might be responsible. There is growing experimental evidence that defects do indeed generate $1/f$ noise. Eberhard and Horn[5] showed that annealing reduced the $1/f$ noise, while Zhigal'skiy, Sokov, and Tomson[6] and Fleetwood and Giordano[7] showed that stress increased the level of $1/f$ noise. Black, Restle, and Weissman[8] deduced that defect motion could be responsible for the $1/f$ noise in metals from measurements of its nonscalar nature. There have been theoretical models attempting to explain the noise in terms of defect motion.[9,10] In this Letter, we show that defects induced in a controlled manner by 500-keV electron bombardment of Cu films maintained at 90 K increase both the average resistance and the level of $1/f$ noise. Subsequent annealing of the films at progressively higher temperatures reduces both the resistance and the noise very nearly to their initial values.

In our experiments, we evaporated 99.99% pure Cu to produce 100-nm-thick polycrystalline films on $\langle 100 \rangle$ Si wafers oxidized to generate a 350-nm layer of SiO_2. Photolithographic techniques were used to produce $90 \times 4\text{-}\mu m^2$ samples with five contact leads (inset of Fig. 1). The samples were annealed at 400 °C for 1 h at a pressure below 10^{-5} Torr, and had a typical crystallite size of about 200 nm. A small hole, roughly 300 μm in diameter, was etched through the Si (but not the SiO_2) from the reverse side, producing a window within 50 μm of each Cu sample to enable us to position the beam of an electron microscope on the sample. All of the resistance and noise measurements were made *in situ* at about 90 K on a custom-made cold stage of a Hitachi HU-650 electron microscope, with the sample, a heater, and a thermometer mounted at the optical axis of the microscope. From four-terminal measurements of the resistance, we obtained the relative resistivity to better than 0.1%; uncertainties in the sample dimensions produced a 20% error in the absolute resistivity, which was about 10^{-6} Ω cm at 90 K, compared with 3×10^{-6} Ω cm at room temperature. We measured the $1/f$ noise by using the sample as two arms of a Wheatstone bridge (inset of Fig. 2), the bias voltage of which was sinusoidally modulated at 2 kHz. The imbalance voltage was amplified with a liquid-nitrogen–cooled transformer and lock-in detected. The power spectrum of the demodulated signal was recorded with a spectrum analyzer in conjunction with a desk-top computer, and the power spectrum of the background noise without current modulation (predominantly the Nyquist noise of the sample) was subtracted. At least fifty data scans were averaged for each power spectrum, and least-squares fits were made to log-log plots of the data (with omission of the lowest and two highest frequency points) to obtain the magnitude and slope to within a few percent.

After cooling the sample in the microscope, we first

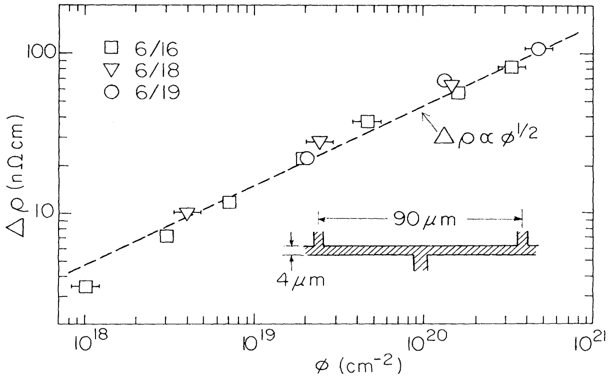

FIG. 1. Change in sample resistivity $\Delta\rho$ vs electron dosage ϕ, for three different dates. The dashed line $\Delta\rho \propto \phi^{1/2}$ is drawn for comparison. Inset: sample configuration.

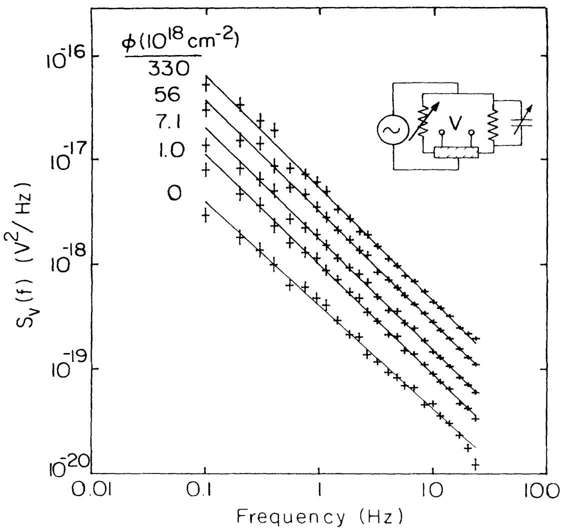

FIG. 2. Typical excess-voltage-noise power spectra and least-square fits for different electron dose ϕ. Data taken on 6/16. Inset: ac bridge configuration.

measured the average resistance and the noise. We then irradiated the sample with a beam of 500-keV electrons with a typical intensity of about $10^{17}\ \mathrm{cm}^2\ \mathrm{s}^{-1}$ for a specified time. By use of the sample resistance as a thermometer during the irradiation, we established that its temperature rose by at most 1 K. The electron-defect cross section in Cu is sufficiently small[11] to ensure uniform generation of spatially uncorrelated Frenkel (vacancy-interstitial) pairs; the cross section for the production of multiple defects is negligible. At 90 K, it is generally believed that in pure bulk copper the vacancy is frozen in place while the interstitial migrates freely until it recombines with a vacancy, is trapped at another defect such as an impurity, a dislocation, or a surface, or forms a cluster with other instabilities.[11,12] Thus, successive doses of electrons build up a defect concentration, which can be monitored by the change in resistivity: $\Delta\rho \approx 3\times10^{-4}\ \Omega$ cm/(fractional concentration of Frenkel pairs).[11] We emphasize here, however, that the complex nature of our samples (small crystallites, very thin films, probable oxidation at surfaces and grain boundaries, and the presence of a substrate) makes quantitative comparison with studies on freely suspended bulk materials uncertain, though qualitative similarities are evident. After each irradiation process, we measured the resistance and noise; typically 30 min elapsed between the irradiation and the noise measurements. At the end of this sequence, we annealed the film for 5 min at each of a series of progressively higher temperatures, remeasuring the resistance and noise at 90 K after each annealing step. The noise magnitude remained constant (within ±5%) for up to 2 h provided that the sample was maintained at 90 K. All of the data reported in this Letter are from a single Cu sample. Similar results were observed in two other Cu films.

Figure 1 shows $\Delta\rho$ vs total electron dose, ϕ, for three different runs on the same sample, each separated by a room-temperature annealing process of at least 12 h. We see that $\Delta\rho$ scales approximately as $\phi^{1/2}$, as has been observed in bulk materials.[12] In bulk materials, this behavior is generally explained by the "unsaturable trap" model,[12] which assumes that as the electron dose is increased, the interstitials have an increasing tendency to recombine with the more numerous frozen vacancies rather than to become trapped. The observed increase in resistivity for a given electron dose is, however, significantly larger than that observed in bulk materials under similar irradiation conditions.[11] We have also observed a significant amount of "subthreshold damage" for electron energies < 400 keV. We suspect that an anomalous damage mechanism is responsible for this behavior, perhaps arising from light-atom impurities, sample oxidation, or the presence of a substrate.

Figure 2 shows several voltage power spectra, $S_v(f)$ (with background noise subtracted), for the sample with progressively higher electron doses. The spectral density of the $1/f$ noise increases by more than an order of magnitude; in addition the slope steepens by about 9% ±2%, with most of the increase in slope occurring after the first irradiation. It is convenient to characterize the noise in terms of the parameters α and m, where $S_v(f)=\alpha\bar{V}^2/Nf^m(1\ \mathrm{Hz})^{1-m}$; here $\bar{V}$ is twice the rms voltage across half the sample, and $N \simeq 2.9\times10^{12}$ (±20%) is the estimated number of atoms in the sample. Before each irradiation sequence, the initial value of α at 90 K was within 10% of 5.5×10^{-4}. In Fig. 3, we plot $\Delta\alpha$ vs $\Delta\rho$ for the three data runs illustrated in Fig. 1; $\Delta\alpha$ and $\Delta\rho$ are the changes in α and ρ relative to the values before a particular irradiation sequence. The values of $\Delta\alpha$ obtained after successive irradiations fall approximately on the dashed line $\Delta\alpha\propto\Delta\rho^{0.6}$. With the assumption that $\Delta\rho$ is proportional to the added defect concentration, n_d, these data indicate that $\Delta\alpha$ scales as $n_d^{0.6}$.[13] We emphasize here that n_d is a measure of the total number of added defects, including many that are essentially frozen at 90 K. Existing defect-noise models,[9,10] however, relate $1/f$ noise only to mobile defects which change position in the same frequency range as the observed noise. Thus, the observed scaling law does not test directly the linear[9] or quadratic[10] dependence of the noise magnitude on defect concentration predicted by these models.

The dependence of $\Delta\alpha$ on $\Delta\rho$ after the samples were annealed is very different, as is shown by the dotted line in Fig. 3. The annealing process reduces the noise

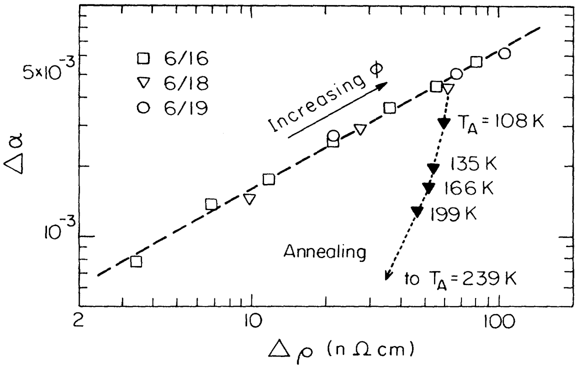

FIG. 3. Change in $1/f$ noise magnitude $\Delta\alpha$ vs change in sample resistivity $\Delta\rho$. The dashed line $\Delta\alpha \propto \Delta\rho^{0.6}$ is drawn as a guide to the eye. Points along this line correspond to increasing electron dose ϕ, while points along the dotted line correspond to annealing at successively higher temperatures. The datum point for $T_A = 239$ K (not shown) is $\Delta\rho \simeq 11.6$ nΩ cm, $\Delta\alpha \simeq 7\times10^{-5}$.

much more rapidly than the resistance, producing hysteresis in the plot of $\Delta\alpha$ vs $\Delta\rho$. This behavior is also illustrated in Fig. 4(a), which shows the annealing data plotted as *recovery curves,* namely $\Delta\rho/\Delta\rho_{\max}$ and $\Delta\alpha/\Delta\alpha_{\max}$ vs the annealing temperature T_A. Most of the resistivity recovery occurs in the temperature range 200 K $< T_A <$ 250 K, and is similar to the "stage-III-recovery" well documented for irradiated copper wires and foils,[11] although occurring at somewhat lower temperatures. The recovery in bulk materials has been attributed to the free migration of a defect obeying a second-order rate equation, with an activation energy in the range 0.60–0.72 eV.[14] The identity of the defect involved is still disputed,[15] although recent studies indicate that it is a monovacancy.[16] The noise magnitude, $\Delta\alpha$, recovers partially over the range 200 K $< T_A <$ 300 K in which the resistivity recovers, but, in addition, exhibits a strong recovery at temperatures below 135 K that is not readily apparent in the resistivity curve. We note here that a subpopulation of mobile defects presumably responsible for much of the added noise may represent only a small fraction of the total added defect density. The observed difference of the recovery of $\Delta\alpha$ and $\Delta\rho$ is readily explained if one assumes that these mobile defects are deactivated (via recombination or clustering, for example) at lower temperatures than the bulk of the defects.

The frequency exponent m of the noise also changed during the irradiation and annealing experiments. The annealing behavior, shown in Fig. 4(b), shows a striking dip at $T_A \approx 240$ K that is reproducible in all of the samples we have studied. We note that this annealing temperature falls within the range of the stage-III recovery of $\Delta\rho$ and $\Delta\alpha$.

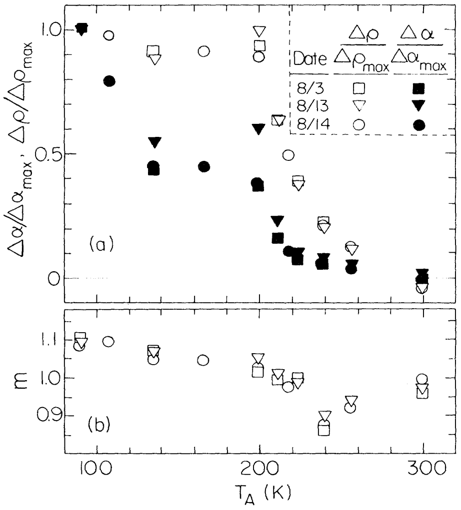

FIG. 4. Annealing behavior of the irradiated Cu film with $\Delta\rho_{\max} \approx 90$ nΩ m and $\Delta\alpha_{\max} \simeq 6\times10^{-3}$ prior to annealing. (a) Recovery of the $1/f$ noise magnitude ($\Delta\alpha/\Delta\alpha_{\max}$) and resistivity ($\Delta\rho/\Delta\rho_{\max}$) vs annealing temperature T_A; (b) the frequency exponent m vs annealing temperature T_A.

In summary, we have shown that the $1/f$ noise in polycrystalline Cu films increase in a systematic way with high-energy electron bombardment. The difference in the recovery of $\Delta\alpha$ and $\Delta\rho$ obtained after successive annealing steps suggests that a large fraction of the added noise may be generated by a small fraction of the added defects, presumed to be mobile, that are more readily annealed than the majority of the defects. Thus, experiments of this kind, in addition to demonstrating a direct connection between $1/f$ noise and defects in metals, may prove to be a valuable new tool for studying mobile defects in metals. We are currently using the Dutta-Dimon-Horn model[4] to test the predicted relation between the temperature dependence of the noise and the parameter m (not reported here), as well as extending our measurements to single-crystal films.

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[1] P. Dutta and P. M. Horn, Rev. Mod. Phys. **53**, 497 (1981).

[2] F. N. Hooge and A. M. F. Hoppenbrouwers, Physica (Utrecht) **45**, 386 (1969).

[3] R. F. Voss and J. Clarke, Phys. Rev. B **13**, 556 (1976).

[4] P. Dutta, P. Dimon, and P. M. Horn, Phys. Rev. Lett. **43**, 646 (1979).

[5] J. W. Eberhard and P. M. Horn, Phys. Rev. B **18**, 6681 (1978).

[6] G. P. Zhigal'sky, Yu. Ye. Sokov, and N. G. Tomson, Radiotekh. Elektron. **24**, 410 (1979) [Radio Eng. Electron. Phys. (USSR) **24**, 137 (1979)].

[7] D. M. Fleetwood and N. Giordano, Phys. Rev. B **28**, 3625 (1983).

[8] R. D. Black, P. J. Restle, and M. B. Weissman, Phys. Rev. Lett. **51**, 1476 (1983).

[9] F. N. H. Robinson, Phys. Lett. **97A**, 162 (1983).

[10] Sh. M. Kogan and K. E. Nagaev, Fiz. Tverd. Tela. (Leningrad) **24**, 3381 (1982) [Sov. Phys. Solid State **24**, 1921 (1982)].

[11] J. W. Corbett, *Electron Radiation Damage in Semiconductors and Metals* (Academic, New York, 1966), Chaps. 24 and 25.

[12] R. M. Walker, in *Radiation Damage in Solids,* edited by D. S. Billington (Academic, New York, 1962), p. 594.

[13] Deviations from Matthiessen's rule could make the relationship between $\Delta\rho$ and n_d nonlinear, and thus alter the exponent of the power law. For example, a 100% increase in $d(\Delta\rho)/dn_d$ over the range of $\Delta\rho$ in Fig. 3 would make $\Delta\alpha \propto n_d^{0.75}$.

[14] W. Schilling, G. Burger, K. Isebeck, and H. Wenzl, in *Vacancies and Interstitials in Metals,* edited by A. Seeger, D. Schumacher, W. Schilling, and J. Diehl (North-Holland, Amsterdam, 1970), p. 335, and references therein.

[15] W. Kienle, W. Frank, and A. Seeger, Radiat. Eff. **71**, 163 (1983).

[16] Th. Wichert, in *Point Defects and Defect Interactions in Metals,* edited by J. Takamuru, M. Doyama, and M. Kiritani (North-Holland, Amsterdam, 1982), p. 19; C. Minier and M. Minier, *ibid.*, p. 27.