Title

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Polycrystalline copper films were maintained at 90K on the cold stage of an electron microscope and irradiated with 500keV electrons to induce defects. With an electron dose of about $5 \times 10^{20}\mathrm{cm}^{-2}$, the spectral density of the noise voltage across the films increased by an order of magnitude while the electrical resistivity increased by at most 10%. The films were annealed at progressively higher temperatures; after each annealing process the 1/f noise and resistivity were remeasured at 90K. Both the 1/f noise and resistivity were reduced, but at the lower annealing temperatures the fractional reduction in the added noise was substantially more than in the added resistivity. These results suggest that a large fraction of the added noise may be generated by a small mobile fraction of the added defects that are more readily annealed than the majority of the defects. After a room temperature annealing process, both the noise and resistivity returned nearly to their initial values. The temperature dependence of the noise after irradiation and partial annealing was consistant with the Dutta-Dimon-Horn thermal activation model.

The origin of 1/f noise in thin metal films has been a puzzle for many years[1,2]. Dutta, Dimon and Horn[3] (DDH) proposed a general model that explained many features of the noise but did not identify the particular microscopic process responsible. Recently there has been growing evidence that crystal defects are involved in the noise[4,5,6,7], and theoretcial models have been proposed explaining the noise in terms of defect motion[8,9].

It is well known that bombardment by electrons with kinetic energy > 400 keV will create defects within pure bulk copper, mostly in the form of Frenkel Pairs (FP), i.e. vacancy-interstitial pairs[10]. These defects are mobile at room temperature and anneal via recombination; thus, an enhanced FP population is retained only at lower temperatres. In this paper, we show that 500 keV electron irradiation of Cu films maintained at 90K increases both the resistance and the level of 1/f noise. Subsequent annealing at progressively higher temperatures reduces both the resistance and the noise very nearly to their initial values. The temperature dependence of the noise depends strongly on the history of irradiation and annealing, and is consistant with the DDH thermal activation model[3].

In our experiments, 90μm × 4μm × 100nm polycrystalline Cu films were mounted on a custom-built cold stage of an Hitachi HU-650 electron microscope, and the noise (over a 0.1Hz-25Hz frequency band) and resistance were measured in-situ. The sample preparation and experimental apparatus have been described elsewhere[11]. All irradiations were performed with the samples held at 90K. The samples were considered fully annealed or "unirradiated" after an extended (>12hr) room temperature annealing process, since both the resistance and noise returned nearly to their initial values. All data reported in this paper are from a single Cu sample; similar results were obtained from two other Cu samples.

For pure, bulk-like Cu at 90K it is generally believed that a radiation-induced vacancy is frozen in place, while an interstitial migrates freely until it recombines with a vacancy (removing a FP), is trapped at another defect (such as a grain boundary, surface, or impurity), or clusters with other interstitials[10,12]. The defect concentration thus builds up in the form of frozen vacancies and trapped or clustered interstitials, which can be monitored by the change in sample resistivity[10]: $\Delta\rho = 3 \times 10^{-6}$ Ωcm/at.% FP. This type of trapping dynamics, in the form of the "unsaturable trap model", has been used to explain the observation that $\Delta\rho$ scales as $\phi^{1/2}$ in bulk materials irradiated near 90K, where ϕ is the electron dose[12].

In our first series of measurements, the sample was cooled to 90K inside the microscope, and the resistance and noise were measured. We then irradited the sample with a known dose of electrons, and remeasured the resistance and noise. For three separate irradiation sequences, each starting with the sample in the unirradiated state, we found that $\Delta\rho$ scales approximately as $\phi^{1/2}$ for $3.5\text{n}\Omega\text{cm} < \Delta\rho < 100\text{n}\Omega\text{cm}$ and $10^{18}\text{cm}^{-2} < \phi < 5 \times 10^{20}\text{cm}^{-2}$. This scaling is similar to the behavior seen in bulk copper; however the magnitude of $\Delta\rho$ we measure for a given dose ϕ is significantly larger than expected in bulk copper. We also measure significant "subthreshold damage" for incident electron energy < 400keV. We note here that our samples (with small crystallites, probable oxidation at surfaces and grain boundaries, and the presence of a substrate) are quite different from the freely suspended bulk-like materials used in previous studies. We suspect that these different sample conditions are responsible for the anomalous behavior.

It is convenient to characterize the measured 1/f noise in terms of the parameters m and α, where the frequency exponent m is the fitted slope of a log-log plot of the noise spectral density, and

$$\alpha = f_o S_V(f_o) N/\overline{V}^2 = f_o S_R(f_o) N/R^2.$$

Here, $S_V(f)$ and $S_R(f)$ are the spectral densities of the sample voltage and resistance fluctuations, respectively, $\overline{V}$ is twice the rms voltage across half the sample, R is the sample resistance, $N = 2.9 \times 10^{12}$ (±20%) is the estimated number of atoms in the sample, and f_o = 1Hz. Before each irradiation sequence, the initial value of α at 90K was within 10% of 5.5×10^{-4}, and m was within 3% of 0.98. In Fig. 1, we plot $\Delta\alpha$ vs. $\Delta\rho$ (along the dashed line) for the three irradiation sequences described in the previous paragraph. We see that α increases by an order of magnitude. Simultaneously, m increases by about 10%, with most of the increase occurring after the first irradiation (not shown). The data fall approximately on the dashed line $\Delta\alpha \propto \Delta\rho^{0.6}$. Assuming $\Delta\rho$ to be proportional to the added defect concentration n_d, these data indicate that $\Delta\alpha$ scales as $n_d^{0.6}$. We note here that n_d is a measure of the total number of added defects, including many which are essentially frozen at 90K. Existing defect-noise models[8,9] however, relate 1/f noise only to mobile defects which change position or orientation in the same frequency range as the observed noise. Thus the observed scaling law does not test directly the dependence of the noise magnitude on mobile defect concentration predicted by these models.

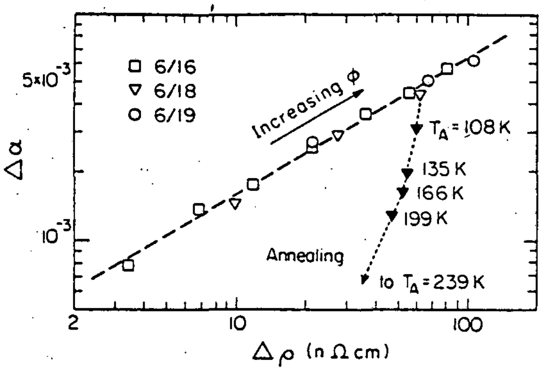

Fig. 1. Change in 1/f noise magnitude $\Delta\alpha$ vs. change in sample resistivity $\Delta\rho$. The dashed line $\Delta\alpha \propto \Delta\rho^{0.6}$ is drawn for comparison. Points along this line correspond to increasing electron dose ϕ, while points along the dotted line correspond to annealing at successively higher temperatures. The data point for T_A = 239K (not shown) is $\Delta\rho$ = 11.6nΩcm, $\Delta\alpha = 7 \times 10^{-5}$.

In our second series of measurements, we

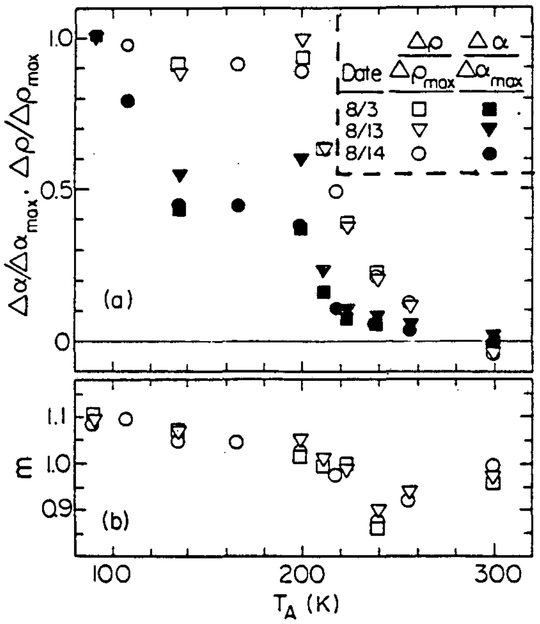

Fig. 2. Annealing behavior of the irradiated Cu film with $\Delta\rho_{max} = 90n\Omega cm$ and $\Delta\alpha_{max} = 6 \times 10^{-3}$ prior to annealing. (a) Recovery of the 1/f noise magnitude ($\Delta\alpha/\Delta\alpha_{max}$) and resistivity ($\Delta\rho/\Delta\rho_{max}$) vs. annealing temperature T_A; (b) the frequency exponent m vs. T_A.

heated the sample (after irradiation) to a temperature T_A for five minutes, then cooled it to 90K to remeasure the noise and resistance. The sequence is repeated for higher T_A. The dependence of $\Delta\alpha$ on $\Delta\rho$ after annealing is shown by the dotted line in Fig. 1. The annealing reduces the noise much more rapidly than the resistance, producing hysteresis in the plot of $\Delta\alpha$ vs. $\Delta\rho$. This behavior is also illustrated in Fig. 2(a), which shows annealing behavior in the form of recovery curves. Most of the resistivity recovery occurs in the range 200K < T_A < 250K, and is similar to the "stage III recovery" well documented for irradiated bulk copper[10] although occurring at somewhat lower temperatures. Recent studies indicate that this recovery step in bulk copper is connected to the free migration of a monovacancy[13]. The noise magnitude, $\Delta\alpha$, recovers partially over the range 200K < T_A < 300K in which the resistivity recovers, but also exhibits a strong recovery at temperatures below 135K that is not readily apparent in the resistivity curve. We note here that a small fraction (presumed to be mobile) of the added defects may be responsible for much of the added noise. The observed difference in the recovery of $\Delta\alpha$ and $\Delta\rho$ is readily explained if one assumes that these "noisy" defects anneal at lower temperatures than the bulk of the added defects.

The frequency exponent m of the noise also changes during the annealing experiments. The annealing behavior, shown in Fig. 2(b), shows a striking dip at $T_A = 240K$. We note that this annealing temperature falls within the "stage III" recovery of $\Delta\alpha$ and $\Delta\rho$.

In a third set of measurements, we first irradiated the sample to $\Delta\rho = 85n\Omega cm$ and annealed it at a temperature T_A before measuring the noise as a function of temperature for T < T_A. In Fig. 3 the noise magnitude $NS_R(1Hz)$ is plotted as a function of T, for $T_A = 201K$, $T_A = 239K$, and for the sample in the "unirradiated" (i.e. fully annealed) state. We see that the temperature dependence of the noise magnitude is a strong function of the annealing state of the sample. The three curves in Fig. 3 are quadratic fits to the three sets of data points. In Fig. 4, the frequency exponent m is plotted for the same data series shown in Fig. 3. Shown also as curves are the predictions for the temperature dependence of m, where we have used the fitted curves from Fig. 3 and the DDH model[3]:

$$m(\omega,T) = 1 - [1/\ln(\omega\tau_0)][\partial\ln S_R(\omega,T)/\partial\ln T - 1].$$

Here, $\omega/2\pi = 1Hz$ and τ_0 is taken to be $10^{-14}s$. The curves precict the general trends of the data points rather well, indicating that our results are consistent with the DDH model.

In summary, we have shown that the 1/f noise in polycrystalline Cu films increases in a systematic way with 500 keV electron bombardment, and thus have demonstrated a direct connection

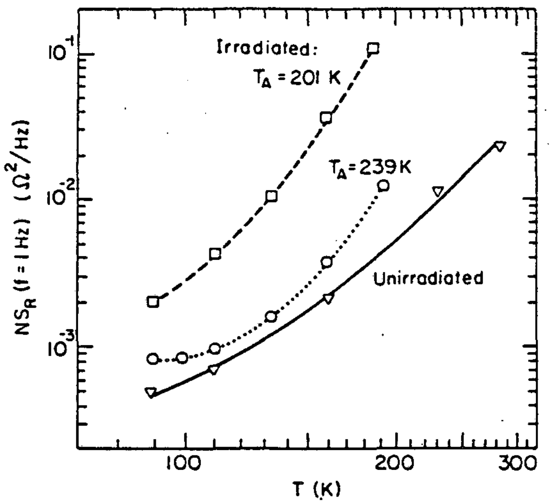

Fig. 3. Dependence of NS_R(1Hz) on temperature T for the sample in the unirradated state, and for $\Delta\rho_{max}$ = 85nΩcm followed by a 5 min anneal at T_A. Curves are quadratic fits to the data points.

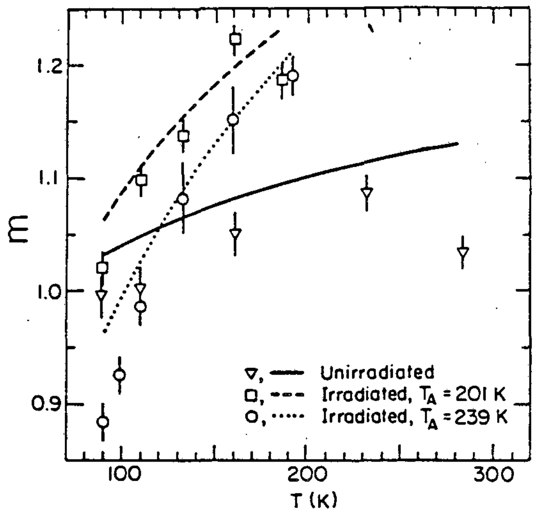

Fig. 4. Dependence of the frequency exponent m on T for the same data series as Fig. 3. Curves are predictions of the DDH model.

between 1/f noise and defects in metals. The difference in the recovery of Δα and Δρ obtained after successive annealing steps suggests that a large fraction of the added noise is generated by a subpopulation of "mobile" defects that are more readily annealed than the majority of added defects. The temperature dependence of the noise magnitude and frequency exponent m after irradiation is consistent with the Dutta-Dimon-Horn model, indicating that thermally activated kinetics govern the added 1/f noise.

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