# $1/f$ Noise and Grain-Boundary Diffusion in Aluminum and Aluminum Alloys

R. H. Koch
*IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598*

J. R. Lloyd
*IBM East Fishkill, Hopewell Junction, New York 12533*

and

J. Cronin
*IBM Burlington, Essex Junction, Vermont 05452*
(Received 24 April 1985)

We have measured the $1/f$ noise in polycrystalline films of Al, Al-Si(1%), and Al-Cu(4%) in the temperature range of 300 to 600 K. The temperature dependence indicated activation energies of 0.69, 0.80, and 0.89 eV, respectively. These energies are similar to the activation energies found for Al diffusion along grain boundaries for films of the same size and composition measured in the same temperature range. Measurements of samples with identical compositions but differing widths and thicknesses revealed significant departures from the usual inverse volume dependence of the $1/f$ noise.

PACS numbers: 72.70.+m, 73.60.Dt

The origin of $1/f$ noise in metal films remains one of the longest-standing unanswered questions of solid-state physics. The details of the microscopic source of the noise remain elusive despite its almost universal presence in all metal films. An empirical expression for the $1/f$-noise power, $S_V$, of a current-biased metal film obtained by Hooge[1] in 1969 is

$$S_V/V^2=\alpha/\Omega f, \quad (1)$$

where $V$ is voltage across the sample, $\Omega$ is the sample volume, $f$ is the frequency of measurement, and $\alpha$ is an empirical parameter. Since the $1/f$ noise in metal films has been shown to be a fluctuation of the sample resistance,[2] the quantity $S_V/V^2$ is independent of the current bias. Hooge originally postulated that the parameter $\alpha$ was constant for all samples; however, many recent experiments have shown this parameter to be temperature and sample dependent.[3,4] Detailed measurements of the frequency exponent of the noise have verified that the exact exponent is seldom exactly $-1$ (or $1/f$) but is also a function of temperature and sample.[5] Dutta and Horn proposed that the $1/f$ noise in a metal film results from thermally activated processes and demonstrated that the noise and frequency exponent when measured as a function of temperature can be used to measure the average activation energy and the width of the distribution of activation energies.[5] The questions of what is activating and where still remain unanswered.

We report in this paper the first systematic measurements of the temperature dependence of the $1/f$ noise in polycrystalline thin films of Al, Al-Cu, and Al-Si alloy. Additions of Cu or Si to an Al film increase the activation energy for Al atomic motion along grain boundaries[6] in the films but not the activation energy for Al motion in the bulk of a grain.[7] We measured a corresponding increase in the activation energy of the $1/f$ noise. Measurements were also made of the area and thickness dependence of the noise, and the results departed from the predicted $1/\Omega$ dependence that is usually observed. Both observations are evidence that the $1/f$ noise in polycrystalline Al films is created on the grain boundaries.

The samples were evaporated onto oxidized-silicon substrates. The sample dimensions and compositions varied and are listed below in the text. The measured resistivities at 290 K were 3.8 $\mu\Omega$-cm for the Al samples, 3.9 $\mu\Omega$-cm for the Al-Si(1%) samples, and 4.3 $\mu\Omega$-cm for the Al-Cu(4%) samples. Each sample was mounted on a header and placed in an oven with a thermocouple mounted on the sampler holder. All samples were measured with use of a four-terminal setup and a dc current bias density of $2\times10^6$ A/cm$^2$ unless otherwise noted. Measurements of the sample resistance versus temperature indicated that Joule heating raised the temperature of the sample stripe 10 to 15 K above the thermocouple temperature. This small offset in the sample temperature was not added to the thermocouple temperature in the data reported here. The voltage across each sample was ac coupled into two PAR 1900 transformers and into two PAR 113 amplifiers. The voltage output of each amplifier was digitized by an IBM PC microcomputer and Fourier transformed. After sufficient averaging, the cross product of the two transformed signals contained noise power only from the Johnson noise of the sample and the $1/f$ noise from the sample resistance fluctuations. The Johnson noise of the sample at each

 

temperature was measured without current bias and subtracted from the final data. The magnitude and frequency exponent of the spectra were averaged at 10 Hz. This technique allowed accurate measurements of noise magnitudes 10 times smaller than the individual amplifier-noise contributions after averaging 1000 times.

Figure 1 plots the normalized magnitude of the $1/f$ noise, $S_V\Omega/V^2$, versus temperature for three films of the indicated compositions. These films were $0.1\times4.5\times450$ $\mu m^3$ in dimension, unannealed, and unpassivated. The noise magnitude peaked at a temperature $T_{max}$ in all cases. The values for $T_{max}$ in the Al, Al-Si(1%), and Al-Cu(4%) samples were 325, 360, and 440 K, respectively. The average grain sizes, measured with use of a transmission electron microscope, for the three compositions were 50, 130, and 160 nm, respectively. Measurements made on identical samples with 2%, 4%, or 6% copper added had the same value of $T_{max}$. The measured frequency exponent, $d(\log S_V)/d(\log f)$, of the noise as a function of frequency was less than $-1$ below $T_{max}$ (typically $-1.15\pm0.05$ for Al at 290 K) and increased to about $-1$ at $T_{max}$. At higher temperatures, well beyond $T_{max}$, the exponent decreased as the noise magnitude increased. Samples with identical compositions but 11 times thicker were fabricated with the same mask pattern and evaporation equipment. These samples had approximately the same temperature dependence and values of $T_{max}$ as the thinner samples except that the magnitude of the normalized noise power, $S_V\Omega/V^2$, was about half as large, independent of composition. The average grain sizes for these samples were 540 and 810 nm for the Al and Al-Cu(4%) compositions.

We also measured the $1/f$ noise in annealed and passivated films of Al-Cu(4%) that were $0.85\times1\times250$ $\mu m^3$ in dimension. These films had a 0.15-$\mu$m Cr-$CrO_2$-Cermet underlay, and the grain sizes were typically 1.0 to 1.5 $\mu$m. The normalized noise magnitudes for three films are plotted in Fig. 2 (lower lines). The noise magnitude peaks at 420 K, compared to 440 K for the unannealed and unpassivated Al-Cu(4%) samples. At temperatures above 500 K the noise increased rapidly with increasing temperature. At all the temperatures measured the normalized noise magnitude, $S_V\Omega/V^2$, was independent of current density. $1/f$-noise measurements made on samples with wider stripes fabricated simultaneously on the same chips as the previous samples are also plotted in Fig. 2 (upper lines). These stripes were $0.85\times10\times250$ $\mu m^3$ or 10 times wider than the previous samples. The normalized noise magnitude is reproducibly larger by a factor of about 1.6 times. The temperature of the peak in the noise magnitude is about the same as for the narrower stripes, but the temperature dependence is much weaker in the wider samples. These samples also displayed a rapidly increasing noise magnitude with increasing temperature. The temperature dependence of the frequency exponent was similar to that of the narrow samples.

To explore the relation between the processes of electromigration and $1/f$ noise we have measured the noise at high current densities, $(0.6–4)\times10^6$ A/cm$^2$, and at temperatures over 500 K where electromigration is prevalent. Under these conditions the films typically passed current for 4 to 48 h before a macroscopic void in the film enlarged enough to stop the current flow, i.e., an electromigration failure of the film. The sample resistance and $1/f$ noise as a func-

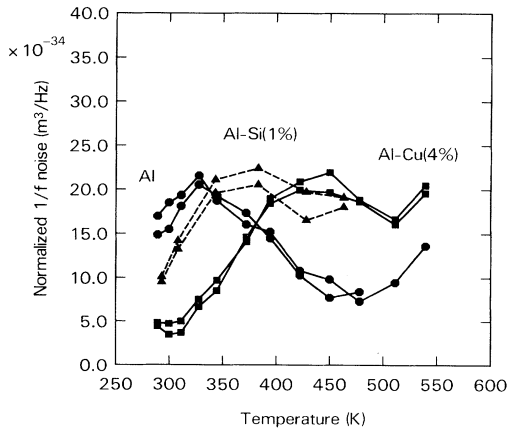


FIG. 1. Normalized $1/f$ noise ($S_V\Omega/V^2$) vs temperature for six samples of three different compositions. The samples were $0.1\times4.5\times450$ $\mu m^3$ in dimension, unannealed, unpassivated, and measured with a current density of $2\times10^6$ A/cm$^2$ at a frequency of 10 Hz.

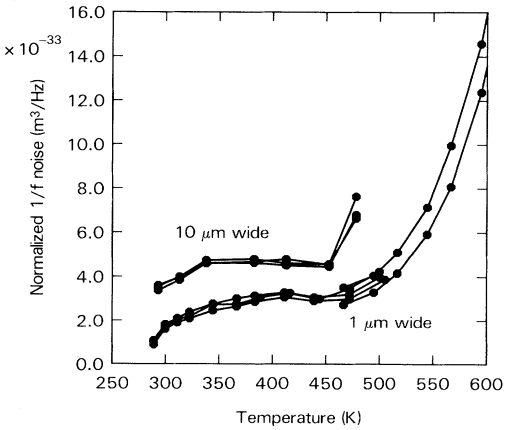


FIG. 2. Normalized $1/f$ noise ($S_V\Omega/V^2$) vs temperature for six samples of identical composition [0.15 $\mu$m of Cr-CrO-Cermet and 0.85 $\mu$m of Al-Cu(4%)] and length (250 $\mu$m), but differing widths. The samples were passivated, annealed, and measured with a current density of $2\times10^6$ A/cm$^2$ at a frequency of 10 Hz.

tion of time is plotted for one such film from the time of the first application of the current until the sample stripe opened (Fig. 3). Each independent point on this plot represents the average of 500 independent Fourier transforms or about 1500 s. This is a typical result of about twenty such experiments all done with use of the $0.1\times4.5\times450$-$\mu m^3$ unannealed and unpassivated samples. The behavior usually can be separated into three distinct stages. (1) At first the $1/f$ noise decreases slowly. During this time the normalized noise magnitude was independent of current. This was verified at a current density of $4\times10^6$ A/cm$^2$ and at a temperature of 610 K in several samples. The frequency exponent was typically $-1$ to $-1.5$. The decrease occurred for all compositions and for films annealed for several days at 250 °C. (2) The $1/f$ noise becomes several times larger. Often the second stage starts abruptly, and unlike the first stage, the individual transforms making up the average show large variations. The frequency exponent tended to decrease. The normalized $1/f$ noise was still independent of current for averages formed during the "quiet" time of the film. The second stage usually starts several hours before the final breakdown. (3) The final failure is a very fast resistance increase of the sample and is usually accompanied by a very large increase in the noise displayed on the spectrum analyzer. The analyzer was now recording the drift in the sample resistance, and the frequency exponent was close to $-2$. The size of the resistance change up until this stage is relatively small compared to the change in $1/f$ noise.

The temperature dependence of the $1/f$ noise in many types of materials has been shown to be characteristic of thermally activated behavior.[5] Aluminum is

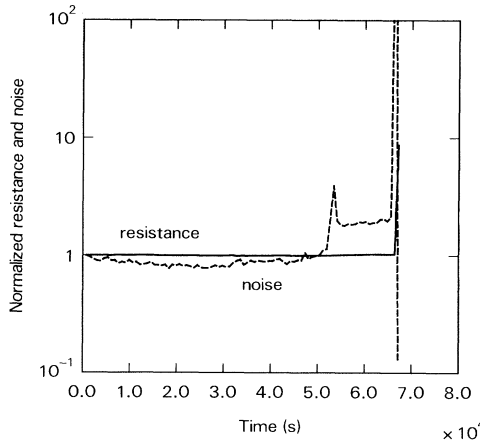


FIG. 3. $1/f$ noise and resistance vs time for an Al-Cu(4%) sample of Fig. 1. The noise magnitude, $S_V/V^2$, at 10 Hz and the resistance were normalized to the initial values at $t=0$. The oven temperature was 500 K, and the nominal current density was $2\times10^6$ A/cm$^2$.

no exception. The activation energy for the noise process, $E^n_{act}$, can be evaluated with use of the relationship of Dutta and Horn,

$$E^n_{act} = -k_B T_{max} \ln(2\pi f \tau_0), \quad (2)$$

where $\tau_0$ is the attempt time for the activated process. From the temperature dependence of the $1/f$ noise and a value for $\tau_0$ of $10^{-3}$ s, we have used the inversion methods of Dutta and Horn[5] to estimate that the $1/f$-noise activation energies for the Al, Al-Si(1%), and Al-Cu(4%) films were 0.69, 0.80, and 0.89 eV ($\pm0.05$ eV), respectively. This value of $\tau_0$ is typical for atomic diffusion in a metal lattice.[8] The attempt time for diffusion on a grain boundary is not well known, but it is expected to be longer, which would reduce the inferred activation energies. The uncertainty in the attempt time, $\tau_0$, used in the inversion represents the major source of error in the values of the inferred activation energies.

An independent measure of these activation energies can be obtained from electromigration life tests. The average lifetime of a film, $t_f$, biased at high current densities and high temperatures is usually fitted with the empirical form[9]

$$t_f \propto j^{-2} \exp(E_{act}/k_B T), \quad (3)$$

where $j$ is the applied current density, and $E_{act}$ is the activation energy for grain-boundary diffusion of the solvent (in this case Al). The measured values of the activation energies obtained by use of Eq. (3) are 0.55, 0.60, and 0.73 eV for the three compositions.[6] However, recently, Shatzkes and Lloyd[10] have formally derived the lifetime equation where vacancy diffusion in a concentration gradient and that due to the electromigration driving force are treated concurrently and they find the correct expression to be

$$t_f \propto j^{-2} T^2 \exp(E_{act}/k_B T). \quad (4)$$

If the existing life-test data are fitted by Eq. (4), the correct activation energies are 0.65, 0.70, and 0.83 eV, respectively, in reasonable agreement with the $1/f$-noise activation energies. The systematic increase in the activation energies of the $1/f$ noise as Cu or Si is added to the aluminum is excellent evidence that the $1/f$ noise in aluminum originates from the diffusion of Al atoms or conversely vacancies along grain boundaries. Motion internal to the grains can be excluded as a possibility since the activation energy for aluminum-atom lattice diffusion (internal to the grain) is not increased by the addition of most solutes, including copper and silicon.[7]

The failure of the simple volume scaling law [Eq. (1)] to predict the noise in the samples can be partially explained if the noise originates at the grain boundaries. A $1/f$-noise source localized on the grain boundaries would have a normalized noise magnitude

that depended inversely on the average grain size, $a$, as

$$S_V \Omega / V^2 \propto 1/a. \quad (5)$$

This equation assumes that the grains are small compared to the dimensions of the film, and the microscopic noise source is not dependent directly on the grain size or angle, or the angle of the grain boundary with respect to the current flow. An extension of Eq. (5) having predictive value would probably not incorporate all of these simplifying assumptions. In the samples where the width was increased from 1 to 10 $\mu$m the average grain size was about 1 to 1.5 $\mu$m, so that the grain boundaries formed a "bamboo"-type structure on the narrow lines. The measured differences in normalized noise magnitude cannot be understood if noise originated internal to the grains. The direction of current flow was primarily perpendicular to the grain-boundary surfaces in the narrower films, which could account for the differences in noise magnitude and temperature dependence. Also the connectivity of the grain boundaries is lower in the "bamboo"-type films. In the samples where the thickness was changed, the average grain size increased by 5 to 10 times when the thickness was increased by 10 times. The normalized noise decreased by about a factor of 2, which is less than predicted by Eq. (5), but more than predicted by Eq. (1).

We found no evidence that the normalized $1/f$ noise was current dependent, as might be speculated if the noise had originated from the nonequilibrium motion of the atoms induced by the current bias or the rate of vacancy and/or void formation in the film. Even at these high temperatures and currents, equilibrium fluctuations are the dominant source of the noise. The noise does depend, of course, on the number of vacancies or voids in the film. The equilibrium motion can be biased in a particular direction by the applied current, but the net difference in directional diffusion rates is small compared to the overall diffusion rate. The large increase in the noise above 500 K observed on all the films is not related to an increase in grain-boundary vacancy concentration or diffusion rate with temperature, but is more likely a result of a different process with a higher activation energy, possibly vacancy diffusion internal to the grain.

The changes in film microstructure during the lifetest experiments were reflected in the $1/f$ noise. The decrease in $1/f$ noise during the first stage of the lifetest experiments was probably associated with the increase in grain size that was induced by the current.[11] The increase in noise during stage 2 most likely resulted from small cracks or voids forming in the film and the resultant current crowding.[12] The final failure was always a large crack or void across all the area of the film. A more detailed description will be published elsewhere.

We believe that this work represents the first strong evidence that the $1/f$ noise in Al comes from the equilibrium motion of Al atoms near or along grain boundaries. This type of noise source is probably dominant in many other metals. $1/f$ noise can also be useful tool in the understanding of the microstructure of thin metal films once the source is well characterized. We thank K. Holland and J. Ryan of IBM Burlington for sample fabrication.

[1]F. N. Hooge, Phys. Lett. **29A**, 139 (1969).

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

[3]J. W. Eberhard and P. M. Horn, Phys. Rev. Lett. **39**, 643 (1977).

[4]J. H. Scofield, J. V. Mantese, and W. W. Webb, Phys. Rev. B (to be published).

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

[6]J. Towner, Solid-State Technol. **27**, No. 10, 197 (1984); A. J. Learn, J. Electron. Mater. **3**, 531 (1974); J. R. Black, in *Proceedings of the Sixteenth Annual International Reliability Physics Symposium, San Diego, 1978* (IEEE, New York, 1978), p. 233.

[7]S. Fujikawa, K. Hirano, and Y. Fukushima, Metall. Trans. A **9**, 1811 (1978); R. V. Penney, J. Phys. Chem. Solids **25**, 335 (1964).

[8]P. G. Shewmon, *Diffusion in Solids* (McGraw-Hill, New York, 1963).

[9]J. R. Black, IEEE Trans. Electron Devices **16**, 338 (1969).

[10]M. Shatzkes and J. R. Lloyd, to be published.

[11]E. Levine, unpublished.

[12]J. L. Vossen, Appl. Phys. Lett. **23**, 287 (1973).