1/f noise in bismuth consistent with defect motion

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The temperature dependence of the $1/f$ noise in bismuth films and whiskers was studied. Undamaged whiskers had less normalized noise power than films. Ion milling increased the normalized noise power in two whiskers to values close to those of the films. None of the whiskers showed the clear peak in the noise-versus-temperature curve found in the films. In the films, under some conditions low magnetic fields reduced the resistance fluctuations, despite the positive magnetoresistance. The frequency of the peak in the noise power shifted as a function of temperature, fitting an Arrhenius law with reasonable parameters for defect motion.

I. INTRODUCTION

$1/f$ noise in the semimetal Bi has been a somewhat puzzling anomaly.[1-9] While the noise levels in most metals increase as a function of residual resistance,[10] consistent with the standard picture of noise coming from moving defects,[1] in Bi the normalized noise level is greater in thick films with large grains than in thin films or films with more disorder.[1,5-7,9] Thus the noise seems to come from inside relatively good crystalline regions. That impression is supported by the unusually clear symmetry signature of the noise in Bi, which indicates that the noise comes from rotations of something in a relatively symmetrical environment.[1,5] Furthermore, the noise magnitude is unusually reproducible between different laboratories, hinting at the possibility of some intrinsic source. It is particularly intriguing that these three properties—high reproducibility in annealed samples, suppression of the main noise component in dirty samples, and a clear rotational symmetry—are shared only by the noise in Cr, which comes from antiferromagnetic spin-density-wave domain rotations.[1,11]

The spectral density of resistance fluctuations in evaporated or annealed Bi films shows a peak as a function of temperature T near 200 K. This peak is sharper than peaks in most metals, indicating a single noise source in a relatively homogeneous environment.[1,3,5,6-8] Table I shows the noise peak temperatures found by different investigators for evaporated Bi films. The peak is absent in sputtered films,[7] which are more disordered than evaporated films. The temperature dependence of the frequency of this peak allows an accurate measurement of an Arrhenius activation energy and attempt rate for the fluctuating objects. A prior measurement in films gave an unusually low attempt rate[5] for defect motion, encouraging the suspicion that something more exotic than defect motions might be involved.[1]

One possible way to check the role of defects in producing the noise would be to make samples with lower defect concentrations than can be obtained in films. Single-crystal whiskers should provide such samples. Previous temperature-dependence data[4] for whiskers showed large noise above 250 K, increasing as a function of T. This noise could not be explained by invoking defects with activated kinetics, again suggesting the possibility of something more interesting.

In this paper we report further investigations of noise in Bi in order to determine if the noise is consistent with ordinary defect motion sources[1] or if it can provide information about more exotic causes. We measured the noise in whiskers, finding that the spectral density can be significantly reduced by preparing a sufficiently defect-free crystal. We found in these good crystals a temperature dependence unlike that of either evaporated or sputtered films.

We also report the dependence of the noise spectral density on magnetic field, frequency, and temperature in annealed evaporated films. If the noise were not caused by defect motion, the temperature dependence of the characteristic frequency of the noise might be non-Arrhenius or have an anomalous activation energy or attempt rate. Likewise, because an exotic source might lead to distinct anomalies in the magnetic-field dependence of the overall noise magnitude, we checked that as well. All of these data are found to be consistent with a conventional defect-motion picture.

TABLE I. Summary of previous measurements of α's peak temperature [α defined in Eq. (1)]. We estimated uncertainties from the data plots of the various authors. f is the frequency at which the data was obtained. The last column is the peak temperature at 10 Hz calculated from the Arrhenius law (see text, Sec. IV) using $E_p = 0.43$ eV. This last column is shown in order to compare all the data at a common frequency.

Reference	T_p (K)	f (Hz)	T_p (10 Hz)
This paper	198(±5)	2	212
22	200(±15)	3	210
8	220(±20)	1	245
5	279(±10)	30	263
3	240	20	232
7	190(±20)	10	190

In order to describe the $1/f$ noise power independently of current and sample size for a given metal, we define

$$\alpha(f,T)\equiv\frac{N_a f S_R(f,T)}{R^2}\ , \tag{1}$$

where $S_R(f)$ is the power spectral density at frequency f of the fluctuations in resistance R and N_a is the number of atoms in the sample. α is often referred to as the normalized noise power.

II. EXPERIMENTAL METHODS

Films were evaporated at 6×10^{-6} Torr at a rate of 20 Å/sec onto a glass substrate at 230 °C, followed by 60 min of annealing at 230 °C. Films were then allowed to cool in vacuum to 100 °C in 30 min. Five-point bridge samples[12] (used to reduce the effects of temperature fluctuations and current source fluctuations) were made by photolithography. Indium was pressed onto bismuth contact areas, and wires were attached to the indium. The sample for which data is shown was 1000 μm long, 13.7 μm wide, and 280 nm thick.

Whiskers were grown by the squeeze technique at Clemson University.[13] Whisker dimensions varied, with lengths between 500 and 2000 μm and widths between 1.0 and 3.0 μm. We mounted the whiskers to glass substrates with small dabs of glue. After masking the sample length, we evaporated 500 nm of tin to make contact with the samples. Lines were scratched through the tin to separate current and voltage leads. Indium was pressed onto the tin and wire leads attached to the indium. Data are shown for samples with resistivity ρ between 500 and 700 $\mu\Omega$ cm. Since widths were not determined precisely, the uncertainty in ρ was large. Data from several samples were discarded. One sample was discarded because it had resistance nearly 3 times that expected, presumably from bad contacts, and was also unusually noisy. Several other discarded samples broke or burned out near contacts before the temperature dependence of the noise power could be measured.

A battery and a series resistor provided a constant current for whisker samples. Current densities ranged from 1×10^4 to 4×10^4 A/cm^2. An ac current was applied to the film sample, with a current density of 1.2×10^4 A/cm^2, which gave a sample voltage of 0.25 V (all ac voltages and currents will be given as rms) at 200 K. Noise was detected using a lock-in amplifier built in this laboratory for two-phase demodulation and cross correlation.[14] The magnetic-field dependence of the noise in films was measured using a dc current. Film data was obtained in a conventional cold-finger cryostat with the sample in a vacuum. A helium atmosphere was used to prevent whiskers from heating too much. Joule heating of the sample by the applied current was checked by measuring the nonlinear conductance of the sample, which results from the temperature dependence of R. In both whiskers and films the sample temperature was 1 K or less above the nominal cryostat temperature. One cryostat was equipped with a superconducting magnet, which in the "persistence mode" had a decay time of about a day, much longer than the time scale of our noise measurements.

III. WHISKERS

Figure 1 shows $\alpha(T)$ for three different samples. Sample No. 1 was ion milled before temperature-dependence data was taken, but α was measured at room temperature before milling. Sample Nos. 2 and 3 are ordinary whiskers. In contrast with previous whisker data,[4] α was not found to be unusually large at temperatures greater than 250 K. The α-vs-T data for whiskers show no clear peak. $\alpha(f)$ is plotted in Fig. 2; the spectral density very closely follows an f^{-1} dependence.

α of sample Nos. 2 and 3 is nearly an order of magnitude smaller than for films. Since the whiskers are expected to be nearly perfect crystals with few defects,[15] their lower α suggests that the film noise is caused by defect motion.

There is a complication, however, in concluding that the fluctuations in scattering cross sections in whiskers are smaller than in films. It is necessary to take into account the resistivity ρ as well as α in the whiskers. ρ of the whiskers was between 500 and 700 $\mu\Omega$ cm, compared with about 200 $\mu\Omega$ cm in films. The higher ρ in whiskers can be due to either a lower mobility μ or a lower carrier concentration. If it were due to a lower μ, i.e., an increased scattering rate, then the reduced α would simply result from having a higher net scattering rate, not from

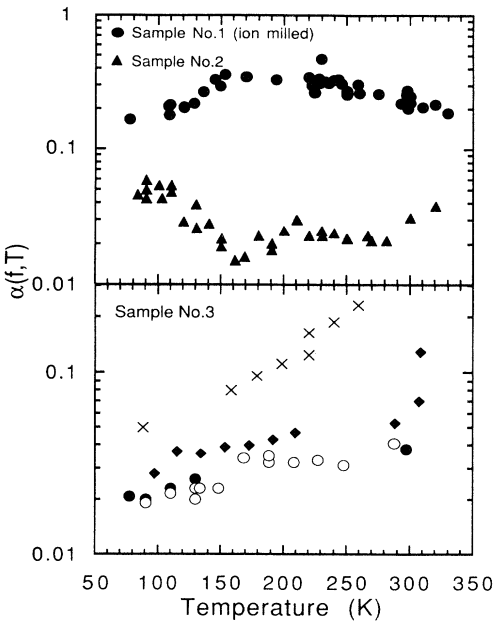

FIG. 1. α vs T for three whiskers. Whisker No. 1 was ion milled. Frequency f is 150 Hz for sample No. 1 data, 25 Hz for sample No. 2, and 12 Hz for sample No. 3. The different symbols on the sample No. 3 plot represent data taken with whisker No. 3 on different temperature cycles. ×'s represent the last thermal cycle made with this whisker; the higher α here probably reflects some damage done to the whisker as we finished taking the previous cycle's (diamonds) highest-temperature points.

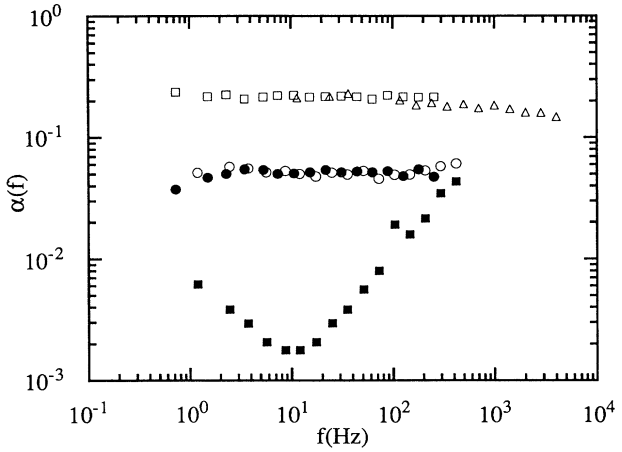

FIG. 2. Upper data points show $\alpha(f)$ of whisker No. 1 after ion milling; the middle data is noise before milling. Background noise due to amplifiers and temperature fluctuations, the lower set of data, is multiplied by f and scaled by the same factor as α for comparison. These data were taken at room temperature (298 K).

having smaller fluctuations in the scattering rate. Magnetoresistance data show that μ is not lower in whiskers. A lower μ in whiskers would give a lower magnetoresistance. Magnetoresistance $\Delta R/R$ at room temperature in both whiskers and films was measured with the magnetic field parallel to the current direction. $\Delta R/R$ was 0.21 at 17 kG in whiskers, slightly higher than $\Delta R/R$ of 0.17 in films. The higher ρ in whiskers therefore comes, not surprisingly, from a lower carrier concentration. Thus, if the scattering rate is fluctuating, it is appropriate to compare α to determine whether whiskers or films have larger fluctuations.

The lack of a sharp peak in the temperature dependence in relatively uniform crystals suggests that the particular mechanism producing noise in the evaporated films may be absent from the whiskers. In evaporated films, α is about 20 times lower at its minimum around 70 K ($f=1$ Hz) than at its peak.[8] This minimum falls between the peak and increase of noise at lower temperatures due to universal conductance fluctuations (UCF's).[8] In whiskers above 77 K, α remains approximately constant near this base value, again suggesting that the peak in films, absent from whiskers, comes from thermally activated motion of some particular defects absent from whiskers.

In whiskers that were ion milled, α at room temperature increased. Before sample No. 1 was ion milled, its $1/f$ noise was measured at room temperature. After milling, α was 4 times higher. Sample No. 2 was also milled after we measured its noise temperature dependence. If we assume that ρ was constant, so that the change in R can be used to calculate the change in whisker volume, α after milling was 0.15 at room temperature, which is a factor of 5 larger than α before milling. If the introduction of defects during milling increased ρ, our calculation of α would *underestimate* the increase in α by underestimating the final volume, according to Eq. (1). This is almost certainly the case. Estimating from the milling rate of Bi films, the cross-section decrease of the whiskers was somewhere between 10 and 30%, accounting for only a 10–40% increase in R. The actual resistance increase due to ion milling was a factor of 2.9 in sample No. 1 and 3.6 in sample No. 2. If we use the estimated reduction in volume due to milling, rather than assuming a constant ρ, we find that milling increased α by about an order of magnitude.

If the noise is due to fluctuations in scattering rates, these would be proportional to $\alpha\rho^2$. This means that the mean-square fluctuation in scattering rate after milling was a factor of 20–50 larger than before. This increase in the resistance noise in the milled whiskers is a rather clear indication that defect motion causes the noise in ion-milled whiskers.

The temperature dependence is not much different in the milled and unmilled whiskers; there is therefore no particular reason to think that the small noise in the unmilled whiskers comes from a different source than the larger noise in the milled whiskers. The fact that the shape of the temperature dependence is different from films suggests that the newly introduced defects are different from those in evaporated films. Since evaporated and sputtered films also show very different temperature dependences,[7] the idea that different detailed mechanisms are involved in whiskers and evaporated films is not surprising.

The increase in α in whiskers near and above 300 K may be due to local heating near contacts for higher currents and temperatures. Several samples blew out like a fuse, usually in a temperature range of 300–330 K with currents of 0.5 to 2 mA. α often increased dramatically just before these samples blew out. Large α previously measured near room temperature[4] might have come from a similar effect.

IV. FREQUENCY AND TEMPERATURE DEPENDENCE IN FILMS

We measured the temperature and frequency dependence of the noise in Bi films in detail in the neighborhood of the peak. The most detailed kinetic information can be obtained by studying the peak behavior, and the signal-to-background ratio is excellent in this regime. The general temperature dependence over a wider range has already been published.[8]

Figure 3 shows $\alpha(f,T)$ versus frequency at several temperatures. We fit the temperature dependence of f_p, the frequency of the peak in α, with an Arrhenius law $f_p=f_0e^{-E_p/kT}$, where k is Boltzmann's constant. Frequencies and normalized noise powers of Fig. 3 were multiplied by appropriate scaling factors so that α-vs-f plots for different temperatures would overlap (see Fig. 4). These scaling factors give the frequency shift of f_p as a function of T. Figure 5 shows $\ln[f_p(T)]$ as a function of $1/T$. The frequency shifts, in turn, yield E_p and f_0 and the overall scaling of the peak height with T. We found $E_p=0.43\pm0.07$ eV, giving an attempt rate $f_0=2\times10^{11\pm2}$ Hz. The attempt rate previously reported was very low

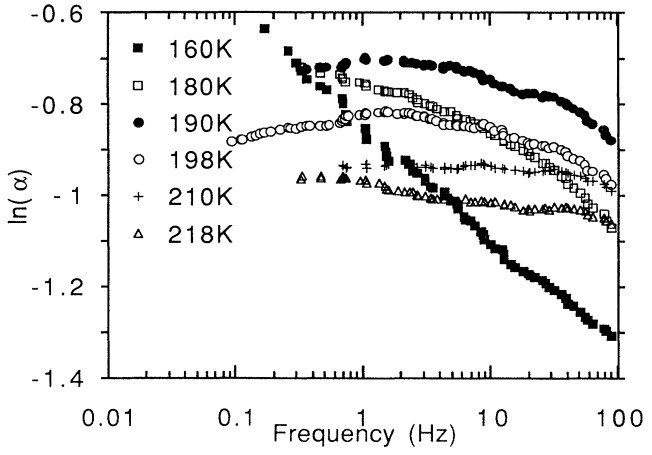

FIG. 3. α vs f in a 280-nm-thick film at six different temperatures.

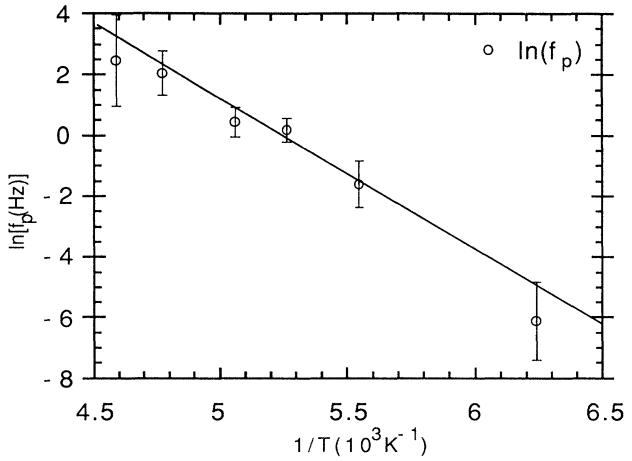

FIG. 5. Natural logarithm of the frequencies of the peaks, $\ln(f_p)$, is plotted against $10^3/T$. Fitting this plot to a straight line (solid line) gives E_a and f_0, 0.43 eV, and 10^{11} Hz, respectively.

compared with typical values for defect motion.[5] The E_p and f_0 found here, however, are reasonable for defect motion with activated kinetics.

An alternative way of analyzing our data is to fit it to the Dutta-Dimon-Horn (DDH) relation,[3] which relates temperature and frequency dependence of the noise:

$$-\frac{\partial \ln\alpha(f,T)}{\partial \ln f}=\frac{1}{\ln(f_0/f)}\left[\frac{\partial \ln\alpha(f,T)}{\partial \ln T}-1\right]. \quad (2)$$

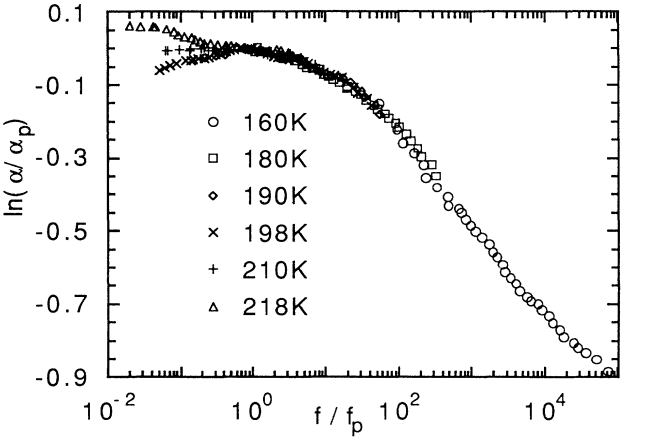

FIG. 4. Changes in temperature shifted the plots of α vs f (Fig. 3) in frequency, and the normalized noise power curves also exhibited an overall scaling with temperature. Here the plots of Fig. 3 were shifted until they overlapped. The shifts are plotted in Figs. 5 and 6. $\alpha_p(T)$ is the magnitude of α at the peak, and $f_p(T)$ is the peak frequency. If the peak is not seen in the data of Fig. 3, α_p and f_p are not the measured α and f at the peak but rather the fitting parameters. For temperatures of 210 K and especially 220 K, the higher noise power at low frequencies was likely due to temperature fluctuations, since we saw a corresponding increase at low frequencies in the feedback output of the temperature controller.

The DDH relation assumes that the integral over f of $\alpha(f,T)/f$ is independent of T and that the activation energies, not the attempt rates, are distributed. Changing the latter assumption can introduce an additional T dependence of between T^0 and T^{-1} to $\alpha(f,T)$. The former assumption can be incorrect for two reasons.[1] First, the number of active fluctuators depends on thermodynamics as well as kinetics. Often a factor of about T^1 is introduced by thermodynamic considerations, but much more dramatic effects can appear near phase transitions and in some other instances.[1] Second, the extent to which the underlying fluctuations couple to $\delta R/R$ can be strongly temperature sensitive, as in the UCF regime.[8] Since α represents the *fractional* fluctuations in ρ, if phonon scattering accounts for an increasing fraction of the resistance, α for defect noise will decrease, even though the fluctuations in defect scattering remain constant. Other effects can also influence this coupling, particularly in a semimetal such as Bi. Therefore, the DDH prediction that the peak in α scales as T^1 frequently fails.

We found that the peak α, plotted in Fig. 6, scaled about as $T^{-1.7}$, with some variation in the exponent. This temperature dependence was similar to but not the same as the $T^{-1.1}$ reported by Black *et al.*[5] Because it is not a T^1 dependence, plots of slope $\gamma = -\partial \ln S_R(f,T)/\partial \ln f$ and of its predicted value derived from $\partial \ln\alpha/\partial \ln T$ via Eq. (2) do not agree. Furthermore, because the temperature scaling did not follow the $T^{-1.7}$ dependence smoothly, plots of predicted and measured γ vs T are not uniformly offset, but show some independent scatter.

The origins of this detailed dependence of the noise magnitude on T are unclear, because neither the thermodynamics of the underlying motions nor the temperature dependence of its coupling to resistance are known ahead of time. Such complicating effects will generally be independent of f, however, leaving the *frequency* depen-

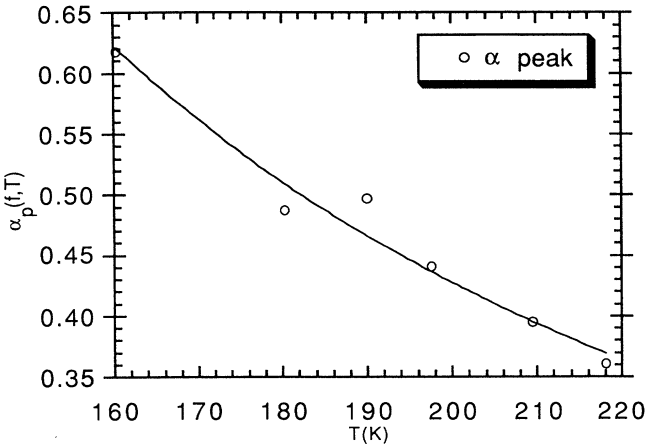

FIG. 6. Overall normalized noise power α falls off approximately as $T^{-1.7}$. This plot shows $\alpha_p(T)$, the peak in α, vs T. The solid curve is $T^{-1.7}$.

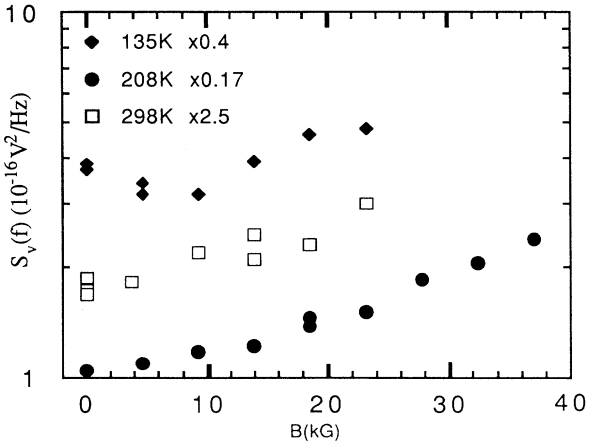

FIG. 7. Total noise power $S_v(f)$ increases much more slowly than R^2, and in one case ($T=135$ K) it actually decreases at low fields. $S_v(f)$ was scaled by the factors given in the legend in order to show all graphs together.

dence of γ in agreement with that predicted via Eq. (2). In general, an additional term $C(T)$, which is independent of frequency, can be added to the DDH relation:

$$\gamma-1=-\frac{\partial \ln\alpha(f,T)}{\partial \ln f}$$
$$=\frac{1}{\ln(f_0/f)}\left[\frac{\partial \ln\alpha(f,T)}{\partial \ln T}+C(T)\right]. \quad (3)$$

We take the derivative with respect to f of Eq. (3) in order to eliminate the adjustable parameter $C(T)$:

$$\frac{\partial\gamma}{\partial \ln f}=\frac{1}{\ln(f_0/f)}\left[\frac{\partial^2\ln\alpha}{\partial \ln T\,\partial \ln f}+\gamma-1\right]. \quad (4)$$

This procedure gives some information for Bi films because the feature is so sharp that higher derivatives are not negligible. We found $\partial\gamma/\partial \ln f=0.020\pm0.005$ from the data between 0.3 and 100 Hz at 190 K. Using Eq. (4), from the difference between the data at 198 and 180 K, we predicted $\partial\gamma/\partial \ln f=0.022\pm0.004$. Thus the data support a picture of activated defect kinetics, although with some minor complications in the overall T dependence of the noise magnitude.

V. MAGNETIC-FIELD DEPENDENCE

The most notable trend in the $1/f$ noise in the films as a function of magnetic field is that $S_R(f)$ at 135 K is reduced by the presence of a small magnetic field (Fig. 7) despite the positive magnetoresistance. The drop-off in α with magnetic field, plotted in Fig. 8, is therefore particularly sharp. In this temperature range the local interference model[16] applies; universal conductance fluctuations cannot account for the reduction in $S_R(f)$ at 135 K.[17]

A magnetic field can reduce $S_R(f)$ in a fluctuating defect-scattering model. A change in the scattering lifetime τ causes a change in ρ. $\partial\rho/\partial\tau$ changes sign as $\mathbf{B}$ increases from 0. In the simplest case, if only one charge carrier were present in an isotropic system with scalar scattering, we would find

$$\rho=\frac{m}{ne^2\tau}[1+(e\tau/m)^2\mathbf{B}^2], \quad (5)$$

where m is the effective mass. The sign change of $\partial\rho/\partial\tau$ at $|\mathbf{B}|=m/e\tau=\mu^{-1}$ can explain the reduction of $S_R(f)$ in a magnetic field (i.e., the dependence of resistivity on scattering must be zero when $\partial\rho/\partial\tau$ is zero).[18] (Vande Voorde and Love[19] may have seen this effect when α dropped by nearly an order of magnitude in a Cu-InSb contact in a magnetic field.)

In practice, however, it is unlikely that the sensitivity of all tensor components of ρ to fluctuating scattering rates would go to zero for a single value of $\mathbf{B}$, particularly in Bi films for which the fluctuations in ρ are far from scalar.[5] The noise is therefore not expected actually to vanish.[20] Additional complications arise from the differences between electrons and holes, making it uncer-

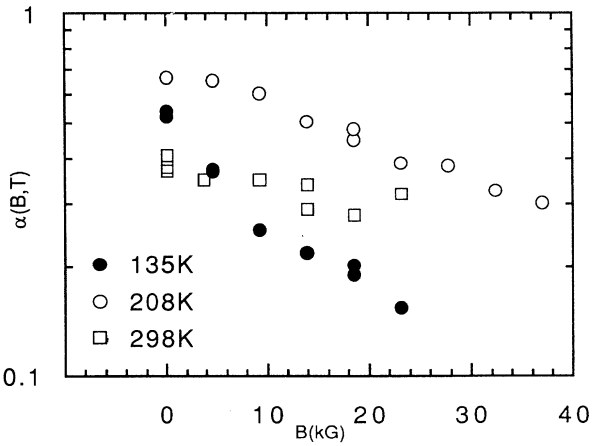

FIG. 8. α vs magnetic field at three different temperatures.

TABLE II. Electron and hole carrier concentrations n and p and electron and hole mobilities μ_n and μ_p are shown. n_k and μ_k are data from Komnik *et al.* (Ref. 22), included for comparison. Their data is only for electrons and assumes $n=p$. n_k was rapidly increasing with T near 300 K.

	$T=208$ K	$T=296$ K
n (10^{18} cm^{-3})	2.5±0.8	8±4
p (10^{18} cm^{-3})	6±2	5±2
n_k (10^{18} cm^{-3})	2.3	~6
μ_n (10^3 cm^2 V^{-1} sec^{-1})	4.2±0.5	12.0±0.3
μ_p (10^3 cm^2 V^{-1} sec^{-1})	3.2±0.4	3.0±0.5
μ_k (10^3 cm^2 V^{-1} sec^{-1})	5	3

tain whether $S_R(f)$ will reach a minimum in a nonzero magnetic field or simply show a slight anomalous reduction.

In order to estimate the field at which some anomalous reduction of the noise might be found for defect scattering fluctuations, we need a determination of μ. We measured the Hall effect at 208 and 297 K in the same thin-film sample as we used for the noise measurements. This, combined with magnetoresistance data and equations in Ref. 21, derived from the Ohm-Hall equations, gave us approximate values of the mobilities of electrons and holes and the carrier concentrations, which are given in Table II. Our results roughly agree with previous measurements[22] of mobility and carrier concentration in films. Allowing for nonscalar fluctuations and for the difference between electrons and holes, $S_R(f)$ should not increase as fast as R^2 between 15 and 30 kG in our temperature range.

Comparing the data with the above prediction, we see in Fig. 8 that α falls rapidly with increasing magnetic field, especially at temperatures significantly below room temperature, but still high enough to be in the local interference regime. Figure 7 shows that at 135 K even $S_R(f)$ diminishes at fields below 5 kG. Thus our data qualitatively agree with the arguments presented above. Although this result is not specific enough to strongly confirm a defect origin of the noise, it shows one more parameter which could be varied without finding any evidence against a defect origin.

VI. DISCUSSION

Results from all three parts of our experiment—temperature and milling dependence in whiskers, temperature and frequency dependence in films, and magnetic-field dependence in films—suggest that the $1/f$ noise in Bi is caused by the motion of defects. Most significantly, although Fleetwood *et al.*[7] have found evaporated films to be quieter than sputtered films above room temperature, our whisker data show that one can reduce the noise at and below room temperature in Bi by using a sufficiently good crystal. The distinctive peak found in evaporated films is simply absent in whiskers, indicating that its origin is not intrinsic to Bi. Furthermore, damage to very good crystals can increase the noise. These differences between whiskers, damaged whiskers, and evaporated films lead us to conclude that defects are the principal noise source in both damaged whiskers and evaporated films and, most likely, in "undamaged" whiskers, too, since the noise is much more sensitive to crystal quality than previous work had indicated. Furthermore, in films the activation energy and attempt rate are reasonable for motion of point defects. The magnetic-field dependence was also reasonable for defect noise, even showing a small predicted decrease at low fields and moderately low temperature.

Data from Alers and Weissman[23] show that the temperature dependences of the noise and the anelastic response in Bi films below about 250 K are quite similar, also consistent with a picture in which both effects arise from a rather uniform collection of thermally activated rotating defects. They found that the approximate magnitude of the ratio of the anelastic response to the noise was consistent with a model in which both effects arose from motion of small defects affecting resistance via local interference. They also found, by assuming a conventional value for the strain coupling of small defects, a mobile defect concentration of roughly 0.1 at. % and a fluctuating scattering cross section of about 10^{-16} cm^2.[16]

Previous measurements[1–9] on evaporated films produced the same α (allowing for reproducible dependence on thickness) to within the uncertainty of about a factor of 2 even though they were made in different laboratories, a surprising result if the defects were impurities. However, the lack of strong dependence of noise on annealing is surprising if the defects do not involve impurities. Secondary-ion-mass spectroscopy showed contamination of only around 0.01 at. % in films grown in this laboratory,[23] which is unlikely to be a high enough concentration of impurities to produce the peak in α.

Twinning boundaries which are very difficult to anneal out have been found in bulk.[24] Some local motion of these defects or of point defects in their vicinity might be implicated in the noise. Some other nonimpurity defects might be very difficult to anneal out as well. Our conclusion based on all available data for Bi suggests that the noise peak in evaporated annealed films is produced by thermally activated rotation of small defects, whose identity is still not known.

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