Low-frequency electrical noise in Ni: The effects of magnetic fluctuations

N. Giordano
Department of Physics, Purdue University, West Lafayette, Indiana 47907-1396
(Received 9 August 1995)

We report measurements of the low-frequency electrical noise of Ni films at temperatures from 300 K to above the Curie temperature ($T_c \sim 625$ K). The noise magnitude is found to exhibit peaks at ~ 450 K and at T_c. This behavior and the associated noise spectra are distinctly different from those of other metals in this temperature range, and we argue that they are caused by magnetic fluctuations. The form of the density of states of the responsible excitations is inferred and found to have some interesting features. [S0163-1829(96)05922-X]

I. INTRODUCTION AND BACKGROUND

The origin of low-frequency, often referred to as $1/f$, noise in metals has been a matter of much, sometimes heated, debate for many years.[1–3] It is now widely believed that in the vast majority of cases this noise is due to the thermally activated motion of (or changes in) electron scattering centers, usually structural defects, and there is a well-developed theoretical understanding of how the thermally activated kinetics of such objects leads in a very natural way to an approximately $1/f$ spectrum.[4,1,3,5] One of the features which made $1/f$ noise so difficult to understand is its ''scale-invariant'' spectrum, i.e., a noise power which varies as $S \sim 1/f$, since a purely $1/f$ spectrum does not have a characteristic frequency which might provide clues as to its origin. The Dutta-Horn model[1] explains why a spectrum which closely approximates $1/f$ is so ubiquitous, and shows how a process which is not scale invariant, i.e., thermally activated kinetics, can give rise to a spectrum which is approximately invariant. Moreover, the model makes some very accessible predictions with regards to measurable quantities.

While the simplest Dutta-Horn framework works well for a great many metals,[1,3] there are some well-documented exceptions, such as Nb doped with H,[6] Cr near its magnetic transition,[7,3] and several spin glasses and related random magnetic systems.[8] In these cases the unusual behavior of the noise magnitude or spectrum, or both, has led to insights with regards to the excitations responsible for the noise, which would have been very difficult to obtain through other types of measurements. Another interesting case in which deviations from the simplest Dutta-Horn picture have been observed is the ferromagnet Ni. Eberhard and Horn[9] reported that the noise magnitude in Ni exhibits a small ($\sim 15\%$) but distinct peak at T_c. This result is especially interesting since it suggests that critical fluctuations near T_c, which are scale invariant, can give rise to scale invariant noise. Such a connection had been been hypothesized some time ago,[1] but so far as we are aware, no detailed theory based on this mechanism has ever been developed.

To the best of our knowledge, the connection between critical fluctuations and $1/f$ noise in Ni discovered by Eberhard and Horn has remained both unexplained and unexamined for the past 15 years.[10] This is unfortunate, since in many respects Ni is much simpler from a magnetic standpoint than the other magnetic systems in which the noise has been investigated more recently (see, for example, Ref. 8). For this reason we have taken a new look at the noise in Ni. We also find a peak in the magnitude of the noise at T_c, but the behavior we observe is somewhat different, and much richer, than reported by Eberhard and Horn. Our results suggest that the critical fluctuations near T_c have a pronounced effect on both the magnitude and frequency dependence of the noise. In addition, the noise exhibits novel behavior well below T_c.

II. EXPERIMENTAL TECHNIQUE

Our samples were made from thermally evaporated Ni films. Using photolithography and etching with dilute HNO_3, they were patterned into a ''two-leg,'' six-lead geometry of the type described by Scofield.[11] The films varied in thickness from 125 to 1000 Å, and sample dimensions were typically 4×35 μm^2 per leg for the thicker samples and 10×35 μm^2 per leg for the thinner ones. Resistivities at room temperature were initially ~ 25 $\mu\Omega$ cm; after annealing at temperatures above 650 K they dropped to ~ 12 $\mu\Omega$ cm at 300 K and ~ 33 $\mu\Omega$ cm at 600 K. Their magnetization, anisotropy, and anisotropic magnetoresistance were all measured at room temperature, and were typical of those reported by previous workers.[12]

For the noise measurements the samples were mounted on a Cu block in a small vacuum chamber. A Pt resistance thermometer was positioned near (a few mm from) the sample, and a resistive heating element somewhat farther (a few cm) away. Electrical leads were attached with Pb-In-Ag solder together with Ag paint. The noise was measured with a standard ac technique[11] using a carrier frequency of 160 Hz and a PAR 126 lock-in amplifier as the demodulator. The usual tests were made to check for contact noise, and none was found. The background noise level was just the Johnson noise of the sample, and was typically 1×10^{-18} V^2/Hz at room temperature, and a factor of 2 higher near T_c.

After patterning, the samples were annealed in vacuum at temperatures between 650 and 700 K; this was above T_c, which was ≈ 625 K for the thickest samples, and somewhat lower, due to finite-size effects, for the thinner ones. Most of the annealing occurred within a few hours (as determined by monitoring the resistance), but the noise measurements indi-

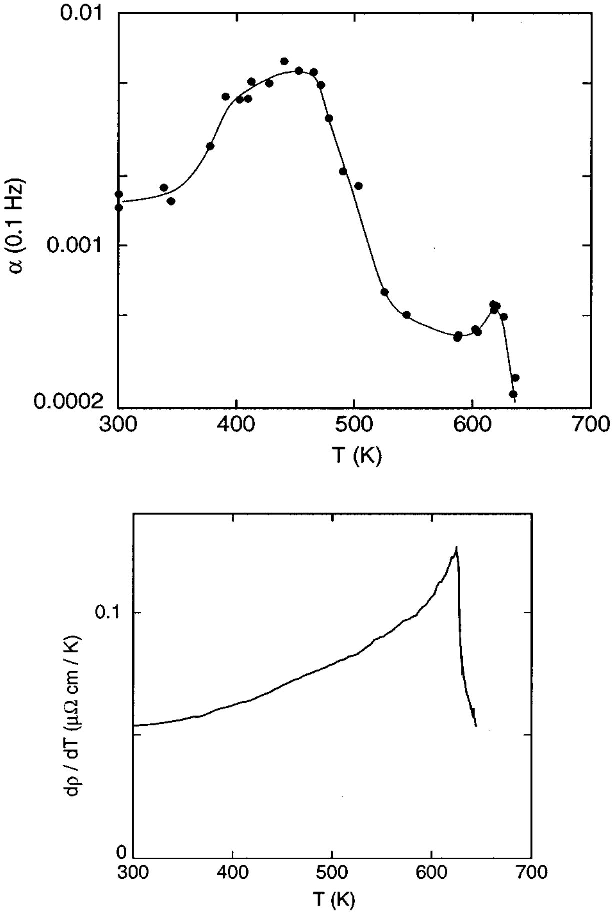

FIG. 1. Top: normalized noise power α measured at 0.1 Hz as a function of temperature. The smooth curve is a guide to the eye. Bottom: $d\rho/dT$ as a function of temperature. The ''noise'' in the results for $d\rho/dT$ is due to the measurement uncertainties.

cated that ''complete'' annealing took much longer. Stable, reproducible results for the noise could only be obtained after a sample had been maintained at temperatures near or above 600 K for several days. The noise level dropped substantially and monotonically during the annealing period. In this paper we will focus on the behavior of one particular thick (1200 Å) sample; the same behavior was found for several other samples with thicknesses near 1000 Å. The behavior of the thinner samples was basically similar.

III. RESULTS AND DISCUSSION

Figure 1 shows the normalized noise power (in excess of the Johnson noise), $\alpha \equiv S_V N f/V^2$, at 0.1 Hz as a function of temperature. Here S_V is the voltage power spectral density, N is the number of atoms in the sample, and V is the sample voltage (note that α is also equal to $S_R N f/R^2$, where R is the sample resistance). This normalization removes the usual dependences on sample dimensions, measuring current, and frequency, making it possible to directly compare different samples.[3] These results were completely reproducible during repeated thermal cycling between 300 K and 650 K, the highest temperature we employed, over the course of several weeks (the extent of the experiment); α was also independent of the measuring current. Two peaks were observed in $\alpha(T)$, a rather broad one at ~ 450 K and a narrower one at T_c. This behavior is quite different from that reported by Eberhard and Horn.[9] They found that α increased monotonically by a factor of 40 from 300 K to T_c. Moreover, they measured $\alpha \sim 0.2$ at 300 K, which is more than two orders of magnitude larger than we find. So far as we know, there has been only one other study on the noise in Ni;[13] it was limited to 300 K, and reported a value of α in agreement with our results. It thus seems that the samples of Eberhard and Horn contained a substantial amount of ''extra'' (extrinsic) noise, perhaps due to insufficient annealing. Figure 1 also shows the temperature dependence of $d\rho/dT$, where ρ is the resistivity; the behavior is very similar to that previously reported for bulk Ni,[14] with a specific-heat-like peak at T_c,[15] and shows that while the peak in α at 620 K is clearly linked with T_c, the peak at 450 K is not associated with any obvious feature in ρ. That is, there did not appear to be any transition or anomaly near 450 K.

Returning to the noise data in Fig. 1, to the best of our knowledge, the behavior of $\alpha(T)$ is unlike that found for *any* other metal,[1,3] even spin-glass-type systems possessing one or more magnetic transitions.[8]. One of the peaks in α occurs at T_c while the other is at a much lower temperature, far from the transition (see the bottom part of Fig. 1), and so we will consider the behavior near these two peaks separately.

As first found by Horn and co-workers, a number of metals exhibits broad peaks in α above room temperature, and the peak at ~ 450 K in Fig. 1 is somewhat reminiscent of the behavior of both Ag and Cu.[4] The peaks in those cases are well accounted for by the Dutta-Horn theory, which attributes it to the thermally activated motion of electron scattering centers,[4,1,3] with a broadly peaked distribution of activation energies. While this distribution cannot be measured independently, the theory also relates α to the spectral exponent, $\gamma \equiv -\partial \ln S/\partial \ln f$. In the simplest case, which has been shown to be applicable for most metals (including Ag, Au, and Cu), γ and α are predicted to be connected by the so-called Dutta-Horn relation[4,1,3,16]

$$\gamma = 1 + \frac{1}{\ln(f_0/f)} \left(\frac{\partial \ln \alpha}{\partial \ln T} - 1 \right), \tag{1}$$

where f_0 is a characteristic microscopic frequency which is generally taken to be of order $\sim 10^{14}$ Hz [the quantitative values derived from (1) are extremely insensitive to the choice of f_0]. We can thus test the applicability of the theory in our case by comparing the measured values of γ with the results calculated from (1) using the results for α in Fig. 1. Figure 2 shows spectra at several temperatures in the neighborhood of the 450 K peak. Since $\alpha \sim Sf$, power law behavior of the noise power would give a straight line with a slope $1-\gamma$ in Fig. 2, and a ''pure'' $1/f$ variation (i.e., $\gamma = 1$) would give a horizontal line. We see that at all of the temperatures considered in Fig. 2 there is some upward curvature at the lowest frequencies (below about 0.1 Hz), so that none of the spectra can be strictly characterized by a simple power law. However, this normalization of the spectrum emphasizes deviations from a simple power law; the magnitude of the deviations seen in Fig. 2 are in fact typical of ''$1/f$'' spectra in metals,[3,8] and are commonly accounted for within the Dutta-Horn framework.[17] Nevertheless, if we restrict our-

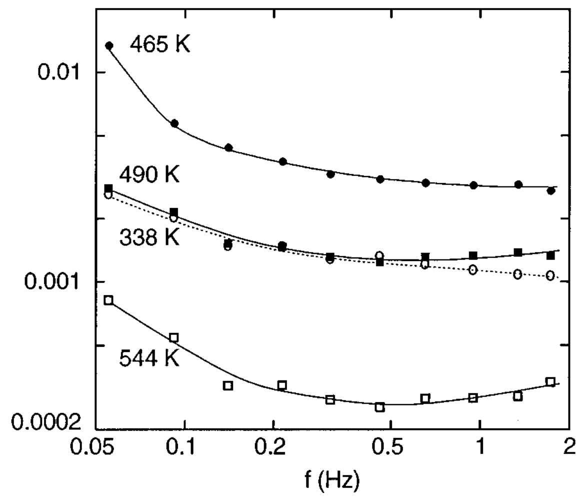

FIG. 2. Normalized noise power α as a function of frequency at several temperatures. Note that ''pure'' $1/f$ noise, i.e., $\gamma=1$, would give a horizontal line in this figure. The smooth curves are guides to the eye.

selves to frequencies above 0.1 Hz, a power law gives a reasonable description of the spectra. The best-fit values of γ are then 1.14, 1.14, 1.04, and 1.01, at $T=338$, 465, 490, and 544 K, respectively (we estimate the uncertainties in γ to be about ± 0.05). The values calculated from Fig. 1 using (1) are 1.20, 0.97, 0.57, and 0.97, respectively. We thus have substantial disagreement with the theory at 465 K, where $\partial\alpha/\partial T$ vanishes, and at 490 K, where $\partial\alpha/\partial T$ is much less than zero.[18] At this point we should note again that these spectra (and also the ones shown below) were completely reproducible over the course of the measurements (several weeks); there was no ''spectral wandering'' (i.e., drift) of the type found in spin glasses.[8] In addition, examination of the noise signal in the time domain did not reveal any two-state fluctuators like those commonly found in (the generally much smaller) systems whose spectra exhibit deviations from a simple power law.[8]

Figure 3 shows results for spectra near T_c. These are very different in form from those at lower temperatures. Near T_c the spectra cannot be described, even approximately, by a simple power law. At high frequencies the spectral slope is less than unity ($\gamma\sim$ 0.6–0.8 for the data in Fig. 3), while at lower frequencies it is as large as 1.5. The frequency $f_{\min}$, at which the crossover from a negative to positive spectral slope occurs, is plotted in the inset, and is also seen to move to higher frequencies as $T\rightarrow T_c$, from both above and below.

So far as we know, the behavior shown in Figs. 1–3 is quite unlike that found in any other metal, especially above room temperature. The simplest Dutta-Horn relation (1) generally works well for metals, but it does involve several assumptions; it is natural to attribute the deviations from (1) that we have observed to violations of these assumptions. The two that we believe may be violated are (a) that the number of fluctuators that give rise to the noise is independent of T, and (b) that the coupling strength of each fluctuator is independent of T. It is straightforward to generalize the Dutta-Horn relation to include the temperatures dependences of these quantities, and one finds[8]

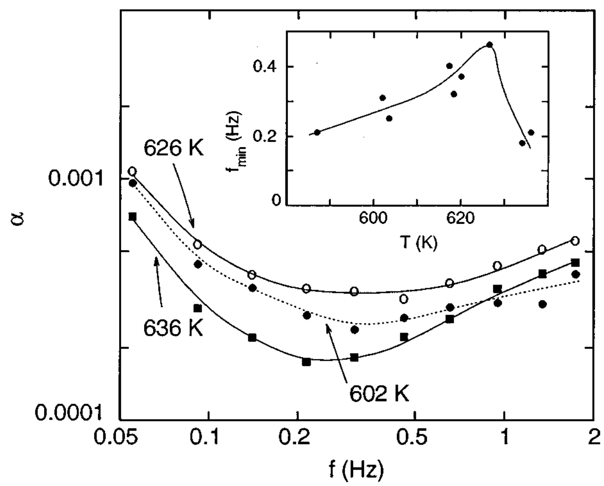

FIG. 3. Normalized noise power α as a function of frequency at several temperatures near $T_c\sim 625$ K. The inset shows the variation of $f_{\min}$, the frequency at which α displays a minimum, with T. The uncertainties in $f_{\min}$ are difficult to estimate, and are probably best inferred from the scatter in the data. The smooth curves are guides to the eye.

$$\gamma=1+\frac{1}{\ln(f_0/f)}\left(\frac{\partial\ln\alpha}{\partial\ln T}-1-\frac{\partial\ln n_f}{\partial\ln T}-\frac{\partial\ln c_f}{\partial\ln T}\right), \qquad (2)$$

where n_f is the number of fluctuators and c_f their coupling strength. Our results for γ then imply that $(\partial\ln n_f/\partial\ln T+\partial\ln c_f/\partial\ln T)$ has a value of ~ 0 at 338 and 544 K, ~ -6 at 465 K, and ~ -16 at 490 K. Hence, the number of fluctuators or their coupling strength, or both, must be decreasing rapidly with increasing T to account for the peak in α at 450 K. This decrease can be understood, at least qualitatively, in terms of a model in which the scattering of electrons by correlated clusters of spins dominates the noise. We would expect such clusters to have a typical size of ξ, the correlation length,[19] but also a very broad distribution of sizes, extending down to clusters of only a few spins. The direction of the magnetization of a cluster will fluctuate in time, making these the basic fluctuators in the Dutta-Horn picture. They could couple to the resistivity in several different ways, including the anisotropic magnetoresistance[20] and boundary (i.e., ''domain wall'') scattering. We would expect the strength of these (and other) coupling mechanisms to be temperature dependent, which seems necessary to produce the strongly nonmonotonic behavior that is observed.

The activation energy for a cluster fluctuation, E_a, will be proportional to the volume of the cluster. Calculations using a simple spin model indicate that at 300 K, E_a for a cluster of size ξ is such that it will contribute strongly to the noise. However, as T_c is approached, the correlation length will grow, and if we assume that it determines the cluster size, then the average size will increase, which will tend to make the associated E_a also grow. We estimate that above about 400–500 K this energy for clusters of size ξ will be so large that, according to the usual Dutta-Horn formulation, it will no longer contribute to the noise. In this case only smaller clusters which will have smaller values of E_a will be important. These smaller fluctuators will still give rise to $\approx 1/f$ noise, but the number of spins involved, and hence the number of fluctuators, will be reduced since an ever-increasing number of spins will be taken up in the larger clusters. A reduced number of fluctuators is just what is needed to account for the peak in α near 450 K (a reduced coupling strength could, of course, also play a role). We believe that this picture can account for the behavior of ($\partial \ln n_f / \partial \ln T + \partial \ln c_f / \partial \ln T$) inferred above at temperatures in the range 300–500 K. The result at 544 K does not fit neatly into this picture, probably because the effects which give rise to the peak in the noise at T_c are then becoming important.

To understand the behavior near T_c, it is not enough to consider the temperature dependence of n_f and c_f. Within the Dutta-Horn framework, the deviations from a power law spectrum seen in Fig. 3 imply significant structure in the distribution of activation energies, $D(E_a)$, of the fluctuators. Model calculations of $D(E_a)$ based on these measured spectra are straightforward, and imply that $D(E_a)$ cannot have a simple, single peak as a function of E_a. Instead, the spectra are consistent with $D(E_a)$ possessing a double-peaked structure. The frequency at which α has its minima, $f_{\min}$, is then determined by the energy of the minimum in $D(E_a)$, $E_{\min}$, and T. Model calculations show that for a fixed $D(E_a)$ there will be monotonic increase of $f_{\min}$ with T, and the magnitude of this increase is consistent with the behavior observed below T_c (see the inset to Fig. 3). However, the abrupt decrease of $f_{\min}$ seen above T_c implies that $D(E_a)$ itself changes rapidly with T in this range. Such behavior would be in accordance with a model of scattering from spin fluctuations like that described above, since the energies of such fluctuations and their kinetics should be strongly affected by the correlations in the critical region.

In summary, we have observed the low-frequency electrical noise in Ni to be quite unlike that found in other metals, as it deviates strongly from the expectations of the simplest Dutta-Horn model, Eq. (1). These deviations appear to be due to the importance of magnetic fluctuations, and an analysis of the temperature and frequency dependence of spectra yields a semiquantitative picture of the behavior of the fluctuators responsible for the noise. Our explanation of the noise treats the behavior near T_c separately from that well below T_c. It is not clear at present if this distinction is artificial or real. In any event, we hope that these measurements will point the way to a fully quantitative theory. We find it intriguing that such unusual noise behavior in such a simple system has gone unnoticed for so long. Further study of the noise near T_c may provide an interesting new avenue for the study of critical fluctuations.

ACKNOWLEDGMENTS

I thank J. D. Monnier for help in the early stages of the experiment, and P. F. Muzikar, M. B. Weissman, R. E. Bartolo, M. A. Blachly, and K. Hong for helpful discussions. This work was supported by the NSF through Grant No. DMR-9220455.

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

[2] M. B. Weissman, in *Proceedings of the Sixth International Conference on Noise in Physical Systems,* edited by P. H. E. Meijer, R. H. Mountain, and R. J. Soulen, Natl. Bur. Stand. Spec. Publ. No. 619 (U.S. GPO, Washington, D.C., 1981), p. 133.

[3] M. B. Weissman, Rev. Mod. Phys. **60**, 537 (1988).

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

[5] N. Giordano, Rev. Solid State Sci. **3**, 27 (1989).

[6] J. H. Scofield and W. W. Webb, Phys. Rev. Lett. **54**, 353 (1985).

[7] N. E. Israeloff, M. B. Weissman, G. A. Garfunkel, D. J. Van Harlingen, J. H. Scofield, and A. J. Lucero, Phys. Rev. Lett. **60**, 152 (1988).

[8] M. B. Weissman, Rev. Mod. Phys. **65**, 829 (1993); R. P. Michel and M. B. Weissman, Phys. Rev. B **47**, 574 (1993); C. D. Keener and M. B. Weissman, *ibid.* **49**, 3944 (1994).

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

[10] The only other work on the noise near T_c in a ferromagnet is that by B. M. Lebed', I. I. Marchik, V. A. Noskin, V. G. Shirko, and Yu. S. Erov, Fiz. Tverd. Tela. **27**, 757 (1985) [Sov. Phys. Solid State **27**, 466 (1985)]. The noise they observed appeared to be due to hysteretic domain motion, and was quite different from what we find in Ni.

[11] J. H. Scofield, Rev. Sci. Instrumn. **58**, 985 (1987).

[12] Our films were similar in all respects to those described and characterized in K. Hong and N. Giordano, Phys. Rev. B **51**, 9855 (1995).

[13] J. H. Scofield, J. V. Mantese, and W. W. Webb, Phys. Rev. **32**, 736 (1985).

[14] F. C. Zumsteg and R. D. Parks, Phys. Rev. Lett. **24**, 520 (1970).

[15] M. E. Fisher and J. S. Langer, Phys. Rev. Lett. **20**, 665 (1968).

[16] The Dutta-Horn relation is usually written in terms of $\partial \ln S/\partial \ln T$ instead of $\partial \ln\alpha/\partial \ln T$ as we have done here. In our case the difference between these two derivatives is negligible.

[17] From measurements over long periods, we have been able to establish that these deviations from a pure power law are not due to slow drifts, etc. Measurements at higher frequencies were not feasible, due to limitations caused by Johnson noise and the maximum allowable sample current.

[18] In the much noisier samples of Eberhard and Horn, γ varied monotonically from $\sim$1.05 at room temperature to $\sim$0.90 near T_c.

[19] In our estimates involving the correlation length, we follow previous work [see, for example, H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford University Press, Oxford, 1971)] and assume that ξ diverges with the usual critical exponent ν, and has a magnitude of order one lattice spacing well below T_c.

[20] See, for example, T. R. McGuire and R. I. Potter, IEEE Trans. Magn. **MAG-11**, 1018 (1975).