

Resistance noise in amorphous Ni-Zr: Hydrogen diffusion and universal conductance fluctuations

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Resistance fluctuations with a power spectrum of approximately $1/f$ have been studied in amorphous thin films of $\text{Ni}_{30}\text{Zr}_{70}$ and $\text{Ni}_{50}\text{Zr}_{50}$ in the temperature range $4 < T < 350$ K. The primary noise-generating source was found to be hydrogen contamination with concentrations up to roughly 30 at. %. Exceptionally large peaks in the spectral density versus T corresponding to short-range hopping of hydrogen atoms were found between 90 and 200 K. The rms resistivity change due to short-range hopping of a hydrogen atom was found to be larger than the average resistivity increase per atom, indicating the importance of the random local environment. Below 50 K the noise increased with decreasing temperature and was reduced by magnetic fields, indicating a universal conductance-fluctuation-coupling mechanism.

INTRODUCTION

Properties of metallic glasses can be completely different from those of the corresponding crystalline states. The electrical resistivity of a metallic glass often has a negative temperature coefficient for reasons that are still controversial.¹ Atomic diffusion in amorphous systems can occur with much faster rates than the corresponding crystalline states. In this paper we present measurements of electrical noise due to motion of dissolved hydrogen in an amorphous metal and examine their implications for the electrical properties and diffusion dynamics.

Dissolved hydrogen has been used extensively as a probe of metallic glass structure.² The effects of hydrogen on the electrical properties of amorphous metals have not been studied as much. However, it has been shown that diffusion of hydrogen through a metal can cause resistance fluctuations³ from both long-range diffusion⁴ corresponding to anelastic mechanical relaxation from the Gorsky effect or short-range hopping⁵ corresponding to internal friction mechanisms. The resistance noise can therefore be a very useful tool to study the electronic properties and atomic diffusion characteristics of amorphous thin metal films. The material chosen for this study was Ni-Zr, which has an amorphous state stable to temperatures well above room temperature.

Intrinsic resistance fluctuations of metallic resistors typically have a roughly $1/f$ spectrum with resistance and volume scalings given by the Hooge relation:

$$S_R(f)/R^2 = \frac{\alpha_A}{N_a f}, \quad (1)$$

where $S_R(f)$ is the spectral power density of resistance fluctuations, N_a is the number of atoms in the sample,

and α_A is a material-dependent constant used to indicate the magnitude of the fluctuations.³

For most metals the resistance fluctuations arise from thermally activated defect motions^{3,6,7} that cause changes in electron scattering. The spectrum of the fluctuations is determined by the distribution of activation energies and attempt rates for the defect motions. Typically, a single site with activation energy E_a and attempt frequency f_0 would give a Lorentzian contribution with characteristic frequency $f = f_0 e^{-E_a/kT}$. Values for f_0 are usually $10^{12 \pm 2}$ Hz. A flat distribution of activation energies would give a pure $1/f$ noise spectrum.

The temperature dependence of the spectral density is then determined by the distribution of activation energies (and, to a lesser extent, of attempt frequencies), by thermodynamic factors which determine how many sites are allowed to fluctuate at any rate and by any temperature dependence of the sensitivity of the resistance to atomic motion. Since the same distribution of activation energies appears in the the temperature dependence and in the spectral slope, one contribution to the temperature dependence may be inferred approximately from the spectral slope using the Dutta-Dimon-Horn⁷ relation:

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

where $S_k(T, f)$ is the spectral density at a frequency f normalized to have an integral of one. Typically $fS_k(T, f)$ is in the range 0.01–0.1, on the order of $1/\ln(f_0/f)$.

The coupling of defect motion to resistance fluctuations is a complicated problem. In crystalline materials with dilute defects the rotation of defects with symmetry less than the crystal will cause tensor resistance fluctua-

tions.⁸ Other defect configuration changes⁶ can appear as noise only if there are inequivalent configurations with nearly equal energies. In materials with less dilute defects, relative motion of defects may change scattering rates. Quantitatively, the fluctuating contribution of a defect to the resistance via single-scattering events in moderately clean systems can be calculated⁹ and is typically about an order of magnitude smaller than its net contribution to the resistance.

For amorphous materials the idea of a defect and its electron scattering cross section is less clear. One expects that without crystal symmetry any atomic motion can contribute to the noise. At low temperatures in disordered systems atomic motions can lead to large fluctuations in the conductance through universal conductance fluctuations¹⁰ (UCF) from random interference between multiple elastic scattering paths within an inelastic scattering length. Feng, Lee, and Stone proposed the significance of UCF's for $1/f$ noise in the limit that $L_i \gg l_e$ and have shown that the magnitude is given to within a dimensionless constant by¹⁰⁻¹²

$$a_A \approx (e^2/h)^2 s^{-2} n_a n_s(T) f S_K(f, T) L_i^{4-d} L_{\min}^2 k_f^{-1}, \quad (3)$$

where n_a is the density of atoms, l_e is the elastic scattering length, L_i is the inelastic scattering length, L_{\min} is the shorter of the inelastic scattering length and the thermal length $L_T = (Dh/k_B T)^{1/2}$, and d is the dimensionality. $n_s(T)$ is a temperature-dependent density of scattering sites which have thermodynamically allowed motions. By extrapolation of typical heat-capacity measurements on glassy materials, we can estimate $n_s(T)$ to be proportional to $T^{1.3 \pm 0.3}$ at low temperatures.¹³ The factor $L_i^{4-d} L_{\min}^2$ contains the temperature dependence of the sensitivity of the resistance to defect motion in the UCF regime. This last factor is absent when $l_e \gg L_{\min}$ and single-scattering events become more important.¹⁴ Therefore, the factors of L_i and L_T in UCF noise give a distinctive negative temperature dependence at low temperatures which is difficult to account for by any other theory. Another distinctive feature of UCF noise is that an applied magnetic field can reduce the noise by up to a factor of 2 by breaking time reversal symmetry.¹⁵ At a magnetic field of $B_c \approx (0.15hc/eL_i^2)$ the noise should be reduced by 25%.^{12,15} UCF $1/f$ noise from atomic motions has been observed only recently in metal-insulator thin films of C-Cu,¹¹ Ge-Au,¹¹ Si-Au,¹⁶ and metallic films of Bi.¹²

By looking at the noise in amorphous systems over a broad temperature range it can be possible to explore the coupling of defect motion to resistivity through universal conductance fluctuations at low temperatures and lower-order scattering effects at high temperatures.

When atoms with a very high mobility, such as hydrogen, are present it is possible to see resistance fluctuations from long-range atomic diffusion through the sample.⁴ The hydrogen atoms act as additional scattering sites that on the average increase the resistance. Fluctuations in the resistance then occur from random diffusion in and out of a fixed volume. The spectral distribution of this type of noise has been calculated before⁴ and has a well-

defined power spectrum of $f^{-3/2}$ for $f \gg f_c$ and $f^{-1/2}$ for $f \ll f_c$, where f_c^{-1} is the characteristic time for the atom to diffuse across the sample given by $f_c = D/\pi L^2$ where D is the diffusion constant and L is the length of the sample parallel to the current flow for a line-shape resistor. The total magnitude of the fluctuations is then given by the relative concentration of atoms diffusing through the sample, C_0 times the squared resistance change per unit hydrogen

$$\int_0^\infty S_R(f) df = \frac{C_0}{N_a} \left[\frac{\partial R}{\partial C} \right]^2, \quad (4)$$

where N_a is the number of atoms in the resistor. This assumes a noninteracting ideal gas of hydrogen. If interactions are included, the magnitude might be reduced by the nonideal dependence of the chemical potential on concentration. Pressure-concentration isotherms¹⁷ for amorphous Ni₆₄Zr₃₆ have shown only minor deviations from Sieverts' law for the concentrations and temperatures involved here so that Eq. (4) should be accurate to within a factor of 2. The dependence of resistance on concentration can be found for bulk samples by standard hydrogen sorption.¹⁸ For thin films C_0 and $(\partial R/\partial C)$ can both be calculated if the noise and resistance are measured at two different H concentrations, even if those concentrations are not independently known. The frequency range over which the long-range diffusion characteristics can be observed is of course finite, but the spectral integral converges rapidly enough so that only the region around f_c need be measured.

At frequencies much higher than f_c interstitial hopping of the diffusing atom can cause $1/f$ noise that is intrinsic to the bulk and does not depend on sample geometry.⁵ Interstitial hopping is a thermally activated process in most of the temperature range of our experiments so that the Dutta-Dimon-Horn relation may be used. If the activation energy distribution is sufficiently peaked, the spectral density should show a peak when $T = E_a/k_B \ln(f_0/f)$, where E_a is the most common activation energy for the hopping. Similar peaks due to the motion of defects and interstitial hopping can be observed in internal friction and ultrasonic attenuation studies. The connection between internal friction and $1/f$ noise has been noted before.^{3,19} However, internal friction measurements are difficult to perform on thin films²⁰ and reveal nothing about the electrical properties.

EXPERIMENT

Thin films of amorphous Ni-Zr were prepared by co-evaporation in a vacuum of 10^{-8} torr with substrates either of Si with a $1\text{-}\mu\text{m}$ SiO₂ layer on the surface or of sapphire. Films were known to be amorphous from transmission electron microscopy (TEM) studies and x-ray diffraction. Samples were patterned by conventional photolithography techniques and etched by Ar ion milling in a background pressure of 10^{-6} torr. It is not known at what point the hydrogen was entering into the samples. Ni-Zr is a known hydrogen getter with absorption properties very dependent on the condition of the

surface.²¹ Most likely, hydrogen entered in at some stage of photolithography or etching because samples prepared from the same substrate were found to have different hydrogen concentrations. The thin films had a typical thickness of 35 nm with a sheet resistance of $70 \Omega/\square$ and resistance ratio $[R(300 \text{ K})/R(4.2 \text{ K})]$ of about 0.8 with a nearly constant negative temperature coefficient of resistance.

All noise measurements reported in this paper were performed using an ac measurement technique that usually used an eight-point double-bridge sample pattern and lock-in amplifier as shown in Fig. 1. This pattern, like a more conventional five-point bridge,⁴ is immune to fluctuations in the current source and, approximately, to uniform temperature fluctuations. Unlike a conventional bridge, it is also approximately immune to uniform temperature gradients and to contact noise. This last property allows the use of much less powerful current sources, since large current-limiting resistors are not needed. Sample dimensions were typically $2 \mu\text{m}$ wide and between 40 to $640 \mu\text{m}$ long on each leg of the bridge and 35 nm thick. To observe long-range diffusion a five-point bridge pattern with two legs of dimension $1 \times 3 \mu\text{m}^2$ separated by 1 mm was used. Contact was made by evaporating silver onto the Ni-Zr after a brief milling in Ar to remove any surface contamination. Indium pads were then used to attach the leads. Possible noise from the contacts was checked for by using the sample in a five-point bridge circuit and correlating the noise on the current leads to the noise on the voltage leads. Temperature was controlled using a flow-through cryostat supplied by either liquid nitrogen or liquid helium. The largest uncertainty for the absolute noise magnitudes was sample size which was known to an accuracy of only about 25%. For a single sample the relative noise magnitudes versus T could be found to better than 1%. Some discarded samples showed exceptionally large noise magnitudes that depended on the ac reference frequency used. We believe this exceptional noise came from noisy shorts to the Si substrate. No such exceptional noise was found using sapphire substrates.

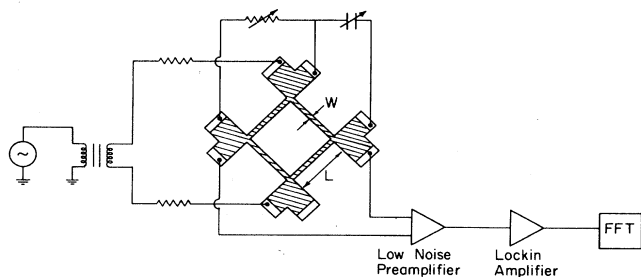


FIG. 1. Experimental apparatus for measuring the excess noise using an eight-probe double-bridge sample geometry. The variable resistor and capacitor are used to balance the bridge. The series resistors are optional for constant voltage instead of constant current. This method is less sensitive to contact noise or uniform temperature fluctuations than other techniques.

Small changes in hydrogen concentration were made on some samples by exposure to an atmosphere of H_2 gas at room temperature without any surface preparation. Changes in hydrogen concentration could be observed by changes in the resistance and noise magnitude. Resistance changes of 20% could be obtained after several days of exposure.

RESULTS

The temperature dependences of the noise were measured for eight samples listed in Table I and are shown for three typical samples in Fig. 2. Above about 250 K sample geometry became important because of long-range diffusion. Spectra for long-range diffusion were measured on samples with lengths of $3 \mu\text{m}$. A typical spectrum is shown in Fig. 3 with a region of $f^{-3/2}$ frequency dependency turning over into $f^{-1/2}$ as expected. Each sample had a characteristic frequency of 0.06 Hz (within a factor of 1.5) at room temperature which implies a diffusion constant of about $2 \times 10^{-8} \text{ cm}^2/\text{sec}$. From the observed shift in frequency with raising temperature an activation energy for the long-range diffusion could be calculated to be $450 \pm 50 \text{ meV}$ in fair agreement with values obtained for bulk samples of $\text{Ni}_{64}\text{Zr}_{36}$.²² Long-range diffusion data on samples 7 and 8 was used to estimate hydrogen concentrations which were found to be 0.20 and 0.30 H/M, respectively (within about a factor of 1.5) using $(\partial R / \partial C) = 100 (\mu\Omega \text{ cm}/\text{fraction H})$ from previous measurements.¹⁸

The most pronounced feature of the noise temperature dependence is the large peak between 90 and 200 K. These peaks are similar to internal friction peaks observed before²² that were attributed to short-range hydrogen hopping. Figure 3 shows the spectrum at the noise

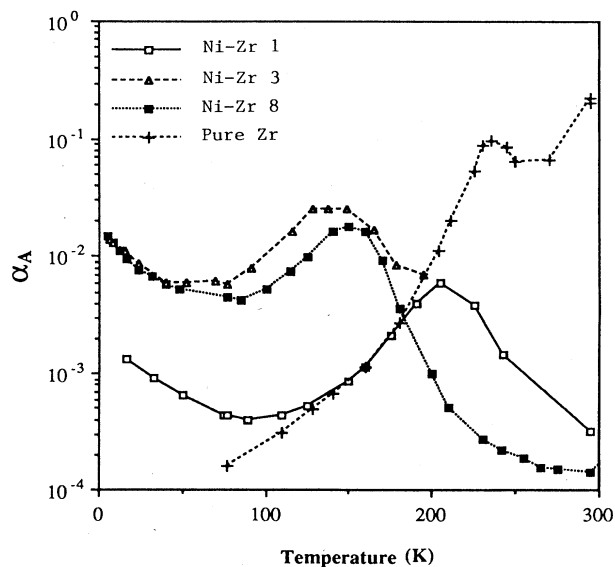


FIG. 2. The temperature dependence of the noise in one octave centered at 1 Hz for three typical samples of Ni-Zr and one sample of polycrystalline Zr. Noise magnitude is expressed in terms of the Hooge parameter defined in the text.

TABLE I. Summary of noise data for amorphous Ni-Zr thin films. Most samples came from different evaporations.

Sample number	Composition	Sample length (μm)	Peak temperature (K)	Activation energy (meV)	Noise at peak ($10^3 \alpha_A$)
1	Ni ₅₀ Zr ₅₀	40	200	500 \pm 50	6.0
2	Ni ₃₀ Zr ₇₀	60	135	350 \pm 90	23
3	Ni ₅₀ Zr ₅₀	60	140	230 \pm 60	26
4	Ni ₃₀ Zr ₇₀	60	110	140 \pm 35	23
5	Ni ₃₀ Zr ₇₀	40	90	76 \pm 15	38
6	Ni ₃₀ Zr ₇₀	640	140	230 \pm 80	19
7	Ni ₅₀ Zr ₅₀	3	145	300 \pm 50	20
8	Ni ₃₀ Zr ₇₀	3	150	380 \pm 50	18

peak. It appears that the distribution of activation energies is fairly flat at lower energies (high frequencies) with a sharp high-energy (low frequency) cutoff. Because the spectrum itself shows a sharp feature, it is possible to measure an activation energy for this short-range hopping as well. Activation energy, peak temperature, and approximate peak magnitude are shown in Table I. The noise magnitudes at the peak are only qualitatively indicative of the hydrogen concentration because of uncertainties in sample size and variations in the peak shape. There does appear to be a trend of decreased activation energy with increased hydrogen concentration as would be expected.²³

On sample 8 the activation energy for both the long-range diffusion (requiring short samples) and short-range diffusion (easier with long samples) was found. The long-range diffusion activation energy of 450 \pm 50 meV agrees within experimental uncertainty to the short-range hopping activation energy of 380 \pm 50 meV. Furthermore, if it is assumed that these two peaks are due to the same diffusive motions, but with relevant distance scales

of about 0.3 nm and 3 μm , respectively, one obtains another independent estimate of the activation energy of 410 \pm 50 meV, with the sources of error in this last estimate being mainly the estimates of the relevant characteristic distances.

These peaks are as much as two orders of magnitude above the "baseline" $1/f$ noise and are at least as prominent as any other peaks observed previously in noise studies in metals. They allow an accurate fit of the temperature dependence and the spectral slope to the model of thermally activated noise as shown in Fig. 4. Deviation from the Dutta-Dimon-Horn prediction occurs in the high-temperature region where long-range diffusion is important. At low temperatures there is deviation that occurs from the extra temperature dependence of the sensitivity of resistance to defect motion from the UCF coupling. In the region of the peak the agreement is very good, confirming that this peak is indeed due to a thermally activated process approximately meeting the criteria for the Dutta-Dimon-Horn relation. In particular, the agreement shows that the integral of the spectral

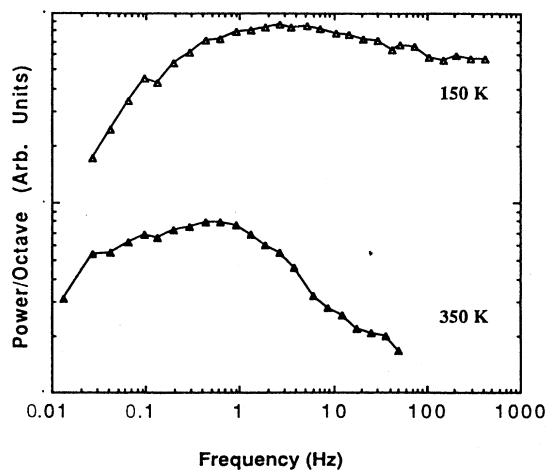


FIG. 3. Spectra of the noise measured on sample 8 for long-range diffusion noise at 350 K (lower spectrum) and short-range hopping noise at 150 K (upper spectrum). Each spectrum has a different scale. In this format f^{-1} appears flat.

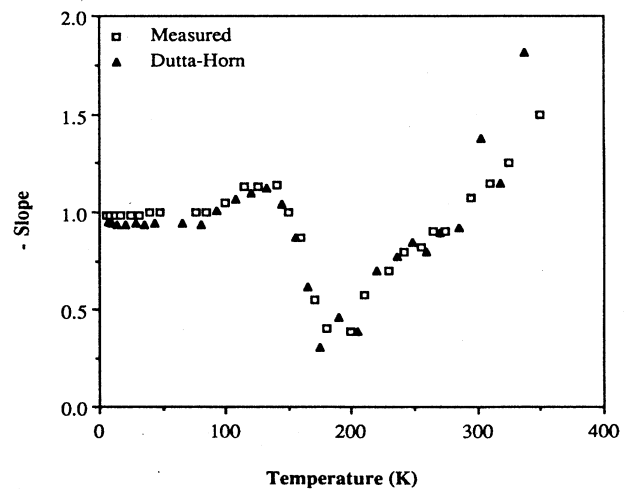


FIG. 4. Comparison of the measured slope for the spectral density $f^{-\alpha}$ at 1 Hz with the exponent obtained from the temperature dependence and the Dutta-Dimon-Horn relation of Eq. (2).

density is not strongly temperature dependent in this regime, ruling out the possibility that the sharp temperature dependence could be associated with any sort of phase transition. The frequency dependence of the noise data can therefore give direct information on the distribution of the activation energies for H hops.

On sample 6 increasing the hydrogen concentration by exposure to a hydrogen atmosphere changed the noise magnitude at the peak by a factor of 2.0 with a corresponding resistance change of 12% and a shift in the frequency of the peak at fixed temperature by roughly a factor of 3. From this it was possible to estimate $(\partial R/\partial C)$ and C_0 using long-range diffusion data of other samples. We obtained an $(\partial R/\partial C)$ of 80 ± 40 ($\mu\Omega$ cm/fraction H) with a concentration of about 0.3 H/M (within a factor of 2) before H_2 exposure. (The large error bars reflect the sensitivity to uncertainties in the sample size.) Previous measurements¹⁸ of resistivity change with hydrogen concentration on bulk samples of $Ni_{64}Zr_{36}$ yielded values of 70–130 ($\mu\Omega$ cm/fraction H) depending on hydrogen concentration in fair agreement with our values.

At lower temperatures the spectral density began to rise as roughly $T^{-1/2}$. The spectral slope of $f^{-0.98}$ implies that the distribution of activation energies in this region is nearly flat and precludes any possible thermally activated feature at these temperatures. The expected kinetic temperature dependence of the spectral density for thermally activated processes is $T^{0.4 \pm 0.4}$ obtained from the slope and the Dutta-Dimon-Horn relation. Including the density of mobile scatterers $n_s(T) \sim T^{1.3 \pm 0.3}$, then the expected temperature dependence of the spectral density becomes $T^{1.6 \pm 0.5}$ neglecting any temperature dependence of the sensitivity of the resistance to atomic motion. Some such dependence is obviously needed to fit the data and can be obtained if we assume UCF coupling to the fluctuations.

Reduction of the noise magnitude in a magnetic field can provide a definite sign of UCF noise. In a magnetic field of 30 kG at 4.2 K a 15% reduction of the noise was observed with essentially no effect on the resistance (<0.1%). Significant reduction of the noise in a metal by such a small magnetic field is very difficult to account for with any model of $1/f$ noise except UCF. A 25% reduction was extrapolated to occur at 70 kG from which, assuming a UCF model, we may infer an inelastic scattering length of about 10 nm. This is comparable to results obtained from magnetoresistance data on similar materials without hydrogen.²⁴

The prominent UCF noise at low temperatures and the prominent noise peak at high temperatures seem to be unique to the amorphous state. As a comparison, the temperature dependence of the $1/f$ noise was measured in polycrystalline Zr films as shown in Fig. 2. The noise showed no rise from UCF effects at low temperatures. A peak was observed at 235 K with a measured activation energy of 570 ± 100 meV. From the literature, the activation energy for long-range H diffusion in crystalline bulk Zr has been measured to be 490 meV (Ref. 25) which is within the large uncertainty of our value. These results are very similar to those obtained by Zimmerman⁵ on hydrogen in palladium films who also found a peak that was

attributed to interstitial hopping of the hydrogen. No evidence of UCF noise was found in that system either down to 20 K.

DISCUSSION

Several features of the peaks in the noise magnitude are significant. The agreement of the activation energy and diffusion constant with previous measurements for hydrogen diffusion supports the idea of a hydrogen diffusion mechanism for the noise. It is very unlikely that anything else could have such a large diffusion constant at room temperature. Furthermore the agreement of the inferred concentration dependence of the resistance and the systematic variation of the noise magnitude with H-induced resistance changes in single samples leaves little doubt that H motions are the dominant noise source in these experiments.

The existence of rather sharp structure in the spectra, as seen in Fig. 3 is itself rather remarkable for an amorphous system. Because the structure showed up as both a function of frequency and as a function of temperature, it requires sharp features in the distributions both of activation energies and of attempt rates. With the prominence of the features, we were able to confirm the Dutta-Dimon-Horn relation with much greater accuracy than is customary.

Since the noise-peak temperature depended on H concentration a highly homogeneous set of hopping sites cannot be invoked to account for the sharpness of the feature in the distribution of activation energies. Instead, it seems necessary to make a description of the sites for hydrogen hopping in which the bonding energy, attempt rate, and hopping activation energy are all highly correlated.²³ Thus the sites whose occupancy fluctuates at a particular H concentration do not have the full range of kinetic parameters. If the potential saddles between binding sites were all identical, then the characteristic frequency would be determined by the energy depth of the fluctuating site. While our data do not require a uniform saddle-point energy, they do strongly point to a rather sharp maximum saddle-point energy. It would be particularly useful in a future study to combine noise measurements, which give a good picture of the distribution of hopping parameters, with other techniques which give more accurate thermodynamic characterization.

These noise peaks provide a unique probe into the effects of the hydrogen on the local electronic structure. The relative size of the random effect of an H atom on the resistance and the average effect is quite different in amorphous Ni-Zr than in Zr or Pd. From the integrated noise magnitudes of the long-range diffusion peak and the short-range hopping peak one can compare the quantity $\langle \text{var}(\Delta R_H) \rangle$, the mean-square resistivity change per hop from short-range diffusion with $\langle (\Delta R_H) \rangle^2$, the average resistivity change per hydrogen atom determined from long-range diffusion noise. The quantity $\langle \text{var}(\Delta R_H) \rangle$ reflects the different relative effects that the occupation sites have on the resistance, whereas $\langle (\Delta R_H) \rangle^2$ measures the resistance change averaged over all occupation sites. For the short-range hopping peak the entire frequency

range cannot be taken, but a fairly close lower limit on the average resistance change per H hop can be calculated by integrating over the experimental bandwidth of five decades. The finite bandwidth is not a problem for the long-range diffusion, because the functional form of the spectrum is known and converges fairly rapidly around f_c .

This comparison was made on sample 8 where it was possible to measure the noise of both long- and short-range diffusion. We found $\langle \text{var}(\Delta R_H) \rangle / \langle (\Delta R_H) \rangle^2 \approx 3.0$, with an uncertainty of about a factor of 2 from estimating the net short-range noise and from possible deviations from ideal behavior in the long-range noise. This means that the rms resistance change due to hydrogen hopping is as large or larger than the average effect of introducing a hydrogen atom into the lattice. Treating hydrogen as a simple scatterer with constant cross section is therefore very inadequate without considering the random effect of the local amorphous environment on the hydrogen scattering. In contrast, for crystalline palladium Zimmerman⁵ found $\langle \text{var}(\Delta R_H) \rangle / \langle (\Delta R_H) \rangle^2 \approx 0.1$. We obtained a similar small value (about 0.2) for H diffusion in Zr.

We believe that the ratio $\langle \text{var}(\Delta R_H) \rangle / \langle (\Delta R_H) \rangle^2$ may prove to be a useful parameter in understanding the electronic properties of amorphous metals. The comparison of the random and systematic effects of a small probe scatterer introduced interstitially is obviously sensitive to how close the electronic eigenstates are to simple Bloch waves. It should be a calculable quantity within the context of different theories that attempt to account for the temperature dependence of the conductivity in disordered metals. Qualitatively, it certainly makes sense for the value of the ratio to be high in disordered metals. Even in a simple, and inappropriate, Born-approximation treatment, the net scattering cross section introduced by a single scatterer involves interference terms with all other scatterers located within about a wavelength. Since in an amorphous metal, essentially every atom can be viewed as a scatterer, it is not surprising that the random interference terms exceed the scattering introduced by a single H. The relatively small scattering amplitude of an H atom, which enters quadratically into the average scattering cross section but only linearly into the random interference terms, contributes to the relatively large magnitude of the random effects. The small average scattering does not, however, contribute to the contrast between amorphous and crystalline materials.

The strong kinetic structure of the data in the 100-to-200 K range makes it difficult to pick out any nonkinetic temperature dependence of the spectral density. Thus the temperature dependence alone does not tell us if the noise is in the UCF-dominated regime within that range. We did not have access to large enough quiet magnets to test for magnetic UCF effects at high temperature. By comparison with other disordered materials in which the nonkinetic temperature dependence could be accurately determined,¹¹ we think that it is unlikely that UCF effects are important in this temperature range. What does be-

come apparent is the importance of medium-range interference effects even at these high temperatures.

In the low-temperature range the noise was consistent with UCF coupling. From the reduction of the noise in a magnetic field we find an inelastic scattering length of about 10 nm at 4.2 K which is smaller than $L_T = 50$ nm and the thickness so that the sample is three dimensional with $L_{\min} = L_i$. The exact exponent for the temperature dependence of the noise depends on the dimensionality of the system, the temperature dependence of the inelastic scattering length, and the distribution of thermodynamic energies of the two-level systems or other mobile sites. Using Eq. (2) in three dimensions with $n_s(T) \propto T^{1.3 \pm 3}$, $S_k(T) \propto T^{0.5 \pm 0.4}$, we can infer a very reasonable $L_i \propto T^{-0.77 \pm 0.2}$ from the observed temperature dependence of $T^{-0.5 \pm 0.1}$ for the noise. The scaling of L_i is nearly identical to that found in other similar materials.^{11,12,24}

The magnitude of the UCF noise appears to scale with hydrogen concentration implying that locally mobile hydrogen dominates the noise-generating mechanism even at these temperatures. It should not be expected that all hydrogen atoms that contribute to the noise at 300 K should contribute at 4 K. Using Eq. (3) as a very rough estimate of the noise magnitude (within about an order of magnitude) we can make an estimate of the density of active hydrogen using $L_i = 10$ nm, $k_f = 10^8$ cm⁻¹, and a noise magnitude of $a_A = 0.01$. We obtain a density of active hydrogen/octave at 1 Hz of about 3×10^{18} cm⁻³, or about 10^{20} cm⁻³ total mobile H concentration. This is more than two orders of magnitude larger than typical densities of two level systems in glasses without hydrogen, but is still a factor of about 10^3 less than the total concentration of hydrogen. While these numbers should not be taken too seriously, they do show that the low-temperature results are nicely self-consistent and fit a reasonable physical picture of noise from mobile hydrogen coupled via UCF.

The conductance fluctuations at 4 K for a coherence volume of 10 nm on a side can be calculated to be $(\partial G)^2 \approx 10^{-12}$ Ω^{-2} per octave which is about three orders of magnitude less than the saturation value of $(e^2/h)^2$. Even integrating over a large range of about 40 octaves would still leave the noise substantially less than the UCF saturation value. Thus the observed frequency dependence of $f^{-0.98}$ should closely reflect the actual distribution of characteristic frequencies of H motions.²⁶

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- ¹D. G. Naugle, *J. Phys. Chem. Solids* **45**, 367 (1984); P. A. Lee and T. V. Ramakrishnan, *Rev. Mod. Phys.* **57**, 287 (1985).
- ²R. C. Bowman, in *Hydrogen in Disordered and Amorphous Solids*, Vol. 136 of *NATO Advanced Study Institute, Series B: Physics*, edited by G. Bambakidis and R. C. Bowman, Jr. (Plenum, New York, 1986), pp. 237–262.
- ³M. B. Weissman, *Rev. Mod. Phys.* **60**, 537 (1988).
- ⁴J. H. Schofield and Watt W. Webb, *Phys. Rev. Lett.* **54**, 353 (1985); J. H. Schofield and W. W. Webb, in *Noise in Physical Systems and 1/f Noise*, edited by M. Savelli, G. Lecoy, and J. P. Nougier (North-Holland, Amsterdam, 1983), p. 405.
- ⁵Neil Zimmerman and Watt W. Webb, *Phys. Rev. Lett.* **61**, 889 (1988).
- ⁶Jonathan Pelz, John Clarke, and Wayne E. King, *Phys. Rev. B* **38**, 10 371 (1988); Jonathan Pelz and John Clarke, *Phys. Rev. Lett.* **55**, 738 (1985).
- ⁷P. Dutta, P. Dimon, and P. M. Horn, *Phys. Rev. Lett.* **43**, 646 (1979); P. Dutta and P. M. Horn, *Rev. Mod. Phys.* **53**, 497 (1981).
- ⁸R. D. Black, P. J. Restle, and M. B. Weissman, *Phys. Rev. Lett.* **51**, 1476 (1983).
- ⁹J. Pelz and J. Clarke, *Phys. Rev. B* **36**, 4479 (1987).
- ¹⁰S. Feng, P. A. Lee, and A. D. Stone, *Phys. Rev. Lett.* **56**, 1960 (1986); P. A. Lee and A. D. Stone, *ibid.* **55**, 1622 (1985); P. A. Lee, A. D. Stone, and H. Fukuyama, *Phys. Rev. B* **35**, 1039 (1987).
- ¹¹G. A. Garfunkel, G. B. Alers, M. B. Weissman, J. M. Mochel, and D. J. Vanharlingen, *Phys. Rev. Lett.* **60**, 2773 (1988).
- ¹²Norman O. Birge, Brage Golding, and W. H. Haemmerle, *Phys. Rev. Lett.* **62**, 195 (1989).
- ¹³W. A. Phillips, *Rep. Prog. Phys.* **50**, 1657 (1987); S. Hunklinger and M. V. Schickfus, in *Amorphous Solids: Low Temperature Properties*, edited by W. A. Phillips (Springer-Verlag, Berlin, 1981), p. 81.
- ¹⁴Selman Hershfield, *Phys. Rev. B* **37**, 8557 (1988).
- ¹⁵A. D. Stone, *Bull. Am. Phys. Soc.* **33**, 632 (1988); B. L. Altshuler and B. I. Shklovski, *Zh. Eksp. Teor. Fiz.* **91**, 220 (1986) [*Sov. Phys.—JETP* **64**, 127 (1986)].
- ¹⁶G. A. Garfunkel (private communication).
- ¹⁷Kiyoshi Aoki, Makota Kamachi, and Tsuyoshi Masumoto, *J. Non-Cryst. Solids* **61**, 679 (1984).
- ¹⁸B. Chelluri and R. Kircheim, *J. Non-Cryst. Solids* **54**, 107 (1983).
- ¹⁹Sh. M. Kogan and K. E. Nagaev, *Solid State Commun.* **49**, 387 (1984).
- ²⁰B. S. Berry and W. C. Pritchett, *J. Phys. (Paris)* **42**, 1111 (1981).
- ²¹F. Spit, K. Blok, E. Hendriks, G. Winkels, W. Turkenburg, J. W. Drijver, and S. Radelaar, *Proceedings of the 4th International Conference on Rapidly Quenched Metals, 1981*, edited by T. Masumoto and K. Suzuki (The Japan Institute of Metals, Sendai, 1982), Vol. II, p. 1635.
- ²²B. S. Berry and W. C. Pritchett, *J. Phys. (Paris)* **46**, 457 (1985).
- ²³R. C. Brouwer, E. Salomons, and R. Griessen, *Phys. Rev. B* **38**, 10 217 (1988), and references therein.
- ²⁴J. B. Beri, A. Fert, G. Creuzet, and J. C. Ousset, *Solid State Commun.* **49**, 849 (1984); B. Leontic, J. Lukatela, P. Dubeek, and J. Kokanovic, *Phys. Rev. Lett.* **58**, 1479 (1987).
- ²⁵E. A. Gulbransen and K. F. Andrew, *J. Electrochem. Soc.* **101**, 565 (1954).
- ²⁶M. B. Weissman, *Phys. Rev. Lett.* **59**, 1772 (1987).