Self-Organized Criticality of the Fracture Processes Associated with Hydrogen Precipitation in Niobium by Acoustic Emission

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The statistical analysis of the acoustic signals, emitted by hydrogenated Nb samples during hydrogen precipitation from solid solution, reveals that the distribution of their amplitudes follows the Gutenberg-Richter power law, $N(A' > A) \sim A^{-c}$ with $c = 0.9$ over more than 2 orders of magnitude. This behavior can be considered as evidence that fracture processes due to hydride precipitation in the Nb-H system determine a self-organized critical state.

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The notion of self-organized criticality (SOC) has been recently introduced by Bak, Tang, and Wiesenfeld [1,2] to explain the behavior exhibited in nature by dynamic composite systems having numerous interacting elements. According to Refs. [1,2] such systems organize themselves into a stationary critical state in which a minor event starts off a chain reaction that can affect any number of elements. The critical stationary state is characterized by spatial and temporal correlation functions following power laws. The importance of the SOC theory derives from the fact that it can serve as a framework to explain a variety of seemingly disconnected phenomena such as the dynamics of earthquakes [3,4], the current fluctuations through resistors (flicker noise) [1], the random steplike changes of the magnetization of ferromagnetic samples [5], and the behavior of traffic and market movements [2].

The study of the SOC systems has to a great extent been based on simple physical models and computer simulations which should capture the essential features of the real systems. An example of these models is a sandpile [1,6] to which sand grains are continuously added until the pile reaches a critical stationary slope, beyond which a single grain can provoke an avalanche of any size. Other cellular automata [4,7,8], which constitute an evolution of the Burridge-Knopoff model of earthquakes [9], are systems of blocks interconnected by springs and put on a frictional plate; an external force can provoke the sliding of a particular block if the spring force exceeds a critical value, and the model evolves to a critical state in which both small and large earthquakelike phenomena are generated.

Recently, an interesting manifestation of SOC has been revealed by recording the underground ultrasonic signals due to microfractures and structural movements following the primary activity of the Stromboli volcano [10]. The continuous volcanic tremors present the opportunity to observe a large number of seisms in a relatively short period of time.

As earthquakes are associated with fracture phenomena, the question arises of whether fractures in materials subjected to externally applied stress also display SOC. Indeed, little attention has been devoted to this aspect up to now; to our knowledge the only report of this kind is a reference by Chen, Bak, and Obukhov [4] to an unpublished work showing a power-law distribution of fracture events in stressed Al and Nb rods.

Plastic deformation and fracture processes occur also in metal-hydrogen systems during precipitation of hydrogen from solid solution (α phase), due to the accommodation of the hydride domains (β phase), which generally have a density different from that of the host lattice (12% volume increase in Nb-H) [11,12]. It has been found [13] that the nucleation and propagation of microcracks during hydride precipitation is accompanied by intense ultrasound emission caused by sudden releases of elastic energy.

In the present paper the acoustic emission (AE) signals detected during hydride formation in Nb-H are analyzed to see whether this system shows a SOC behavior. The hydride formation was stimulated by decreasing temperature from 330 to 220 K. The samples were five circular plates (30–36 mm in diameter, 3–6 mm thick) of 99.9% pure polycrystalline niobium from different manufacturers, hereafter labeled as Hr2, Hr4, K1, K2, and Hy, having masses 17.8, 17, 29.5, 45, and 38.5 g, respectively. The AE signals were detected while the temperature (measured by an iron-Constantan thermocouple directly attached to the specimen) was decreased at a constant rate of 1 or 4 K/min.

The AE setup consisted of a PZT transducer coupled to the specimen by silicone grease, a low-noise preamplifier (40 dB gain, 6 μV input noise), and a high-pass filter at 100 kHz. From the filter the signals were sensed by an amplitude detector (with adjustable threshold put at 30 dB and with 100 dB of dynamic range) giving an output proportional to the log of the peak amplitude of the burst; then they were sent to a distribution analyzer providing the total number N_e of events (bursts) and their distribution in 100 levels. A burst signal occurring more than 10 ms after the level of the preceding burst had decayed below the threshold level was counted as a

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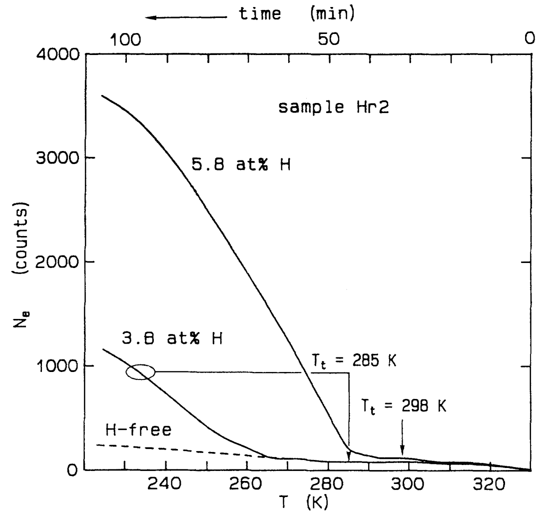

FIG. 1. Total count of acoustic signals (bursts) recorded from sample Hr2 at two different H concentrations during the first cooling after the charge; the sample was cooled at a constant rate of 1 K/min. The temperatures T_t of the onset of H precipitation are indicated by the arrows.

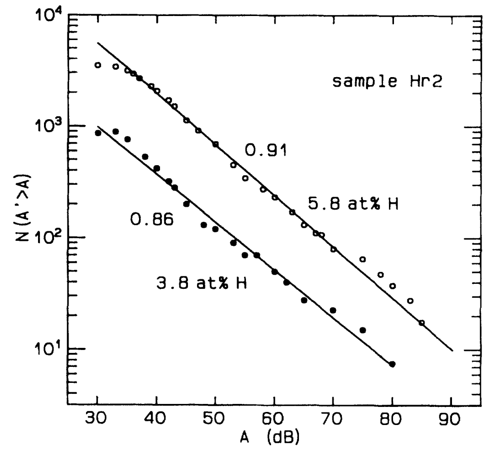

FIG. 2. Number of bursts with amplitudes higher than that given by the corresponding abscissa, during the two cooling runs of Fig. 1. The continuous line is the least squares fit of the power law $N(A'>A) \sim A^{-c}$; the derived exponent is indicated for each curve. The amplitude is expressed in dB above 1 μV at the PZT output.

separate event.

Hydrogen doping of the samples was conducted at 550 °C in a controlled pure H_2 atmosphere. The H contents were determined by the mass variation of the samples, both after the H charge and after vacuum extraction subsequent to the AE experiments; the concentrations were chosen so that the hydride precipitation occurred in the investigated temperature range. The H precipitation temperature T_t was determined either by the solvus line of the Nb-H system [14] or by the sharp increase of the elastic energy dissipation and modulus occurring at the onset of the α-β phase transformation [15,16]. The AE observed in all the samples during hydride formation had the character of isolated, intense bursts having peak amplitudes distributed in the voltage range 0.1–30 mV.

Figure 1 displays the total number of events recorded in sample Hr2 during the first cooling after two different H charges, at 3.8 and 5.8 at.% H. The cumulative counts of events from the beginning of the experiment are reported as a function of both temperature and time, because the sample was cooled at a constant rate. The background of the H-free sample is reported in the same figure. The H charges were preceded by a thermal treatment at 800 °C for 2 h in a vacuum of 5×10^{-4} Pa.

Acoustic emission starts at a temperature somewhat lower than that of the onset of hydride precipitation. It has been demonstrated [13] that the observed emission is due to the nucleation and growth of microcracks and not to the dislocation multiplication accompanying the hydride formation, which is characterized by signals of much lower amplitude. When the local stress field around the growing precipitates reaches a value such that elastic and plastic accommodation is no longer possible, microcracks nucleate and subsequently multiply and propagate, giving rise to the stress wave emission.

Figure 2 presents the amplitude distribution of the events recorded during the two cooling runs of Fig. 1. The ordinates correspond to the number of events (after subtraction of the background) whose peak amplitude exceeds the level given by the corresponding abscissa. The amplitude is expressed in dB above 1 μV at the transducer output.

The background consisted of the number of events recorded in each channel with an H-free sample cooled under the same conditions; it had little influence on the plots even in samples displaying low acoustic emission.

Similar amplitude distributions of events were also observed during the first cooling runs of the samples Hr4, K1, K2, and Hy conducted at two different cooling rates, 1 or 4 K/min, as shown in Fig. 3. If reference to the first cooling is made [17], at a given cooling rate the total number of events generally increases with increasing H concentration and sample mass. High cooling rates may produce low AE or no activity at all; this is due to a finer dispersion of precipitates, whose growth is diffusion controlled.

The cumulative distribution functions of the amplitudes of the transient elastic waves follow the power law:

$$N(A'>A) \sim A^{-c}, \quad c=0.9\pm0.1\,,$$

over more than 2 orders of magnitude (Figs. 2 and 3). This law is expected to be valid for signals with amplitude higher than a threshold connected with some minimal event, e.g., the minimum energy for crack propagation. Here we limit its validity to signals higher than the exper-

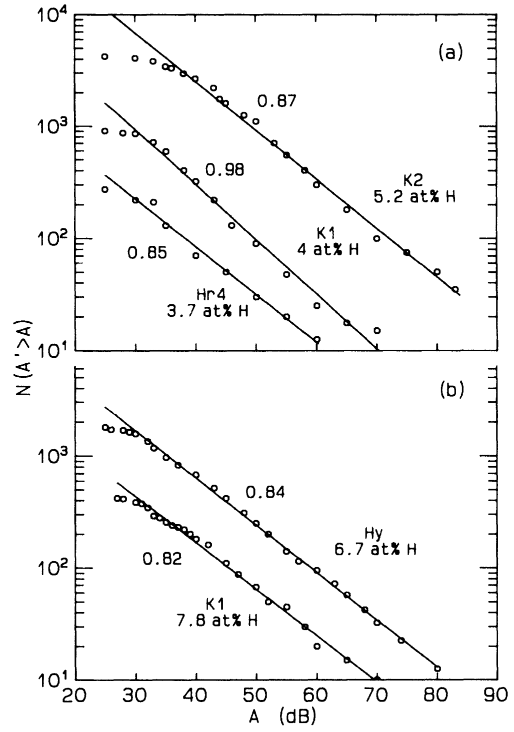

FIG. 3. Number of bursts with amplitudes higher than that given by the corresponding abscissa during the first run after the H charge of various samples at two cooling rates: (a) 1 K/min; (b) 4 K/min. The continuous line is the least squares fit of the power law $N(A'>A) \sim A^{-c}$; the derived exponent is indicated for each curve. The amplitude is expressed in dB above 1 μV at the PZT output.

imental noise (30 dB corresponding to $\sim 30\ \mu$V).

Remembering that the squared amplitude of the events is proportional to their energy, we can express the distribution function also in terms of the number of events with energy between E and $E+dE$,

$$N(E) \sim E^{-(1+c/2)}, \quad c/2 = 0.45 \pm 0.05 ,$$

where $c/2$ corresponds to the exponent b of Ref. [4].

The occurrence of power-law distribution functions suggests the absence of characteristic length and time scales, as in critical phenomena near a second order phase transformation, in fractals and in SOC systems. We exclude an explanation of our data in terms of a critical phenomenon because hydride precipitation is a first order transformation, which does not imply fluctuations over all scales. Indeed, the shapes and dimensions of the precipitate particles are rather regular and depend on microstructure and cooling rate, because of the diffusion controlled nature of the process. Moreover, as each sample had a homogeneous grain texture, we can also exclude that the observed power law may arise from a possible preexistent fractal structure of grains or domains. Therefore we conclude that during the H precipitation the system goes into a SOC state, as far as the AE is concerned.

The observed power-law distribution of the events during the hydride precipitation gives a value for $c/2$ very close to that estimated [18] by the data of the Harvard earthquakes catalog $c/2 = 0.55$. This coincidence in phenomena involving remarkably different scales and mechanisms should be considered as experimental evidence of what is suggested by numerical simulations [1,2,4,7–9]; i.e., fracture processes evolve into a SOC state.

In conclusion, we have shown that hydride precipitation stimulated in Nb by decreasing temperature produces microseism activity, revealed by recording the impulsive stress waves at the surface of the specimens. The AE signal amplitudes follow a power-law distribution which is indicative of a SOC state. The derived exponent is very close to that of the Gutenberg-Richter law of earthquakes, suggesting that fracture processes represent a category of SOC states.

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[13] that the AE in metal-hydrogen systems decreases with thermal cycling until it disappears. The AE can be restimulated either partially by thermal treatments at 150 °C or totally by ultrahigh vacuum treatments at higher temperatures (2000 °C). The former treatment activates the migration of interstitial impurities (O,N,C) which pin dislocations, while the latter one produces annealing of dislocations but cannot eliminate the cracks introduced by precipitation. The AE of the samples annealed at 2000 °C, which had undergone severe damage due to multiple precipitations, exhibited very intense acoustic signals whose amplitude distribution no longer followed a power law.

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