Direct correlation between strengthening mechanisms and electrical noise in strained copper wires

Natalia Bellido, Alain Pautrat,* Clement Keller, and Eric Hug
Laboratoire CRISMAT, UMR 6508 du CNRS, ENSICAEN et Université de Caen, 6 Boulevard Maréchal Juin, F-14050 Caen, France
(Received 27 May 2010; revised manuscript received 5 January 2011; published 25 March 2011)

We have measured the resistance noise of copper metallic wires during a tensile stress. The time variation of the main resistance is continuous up to the wire breakdown, but its fluctuations reveal the intermittent and heterogeneous character of plastic flow. We show in particular direct correlations between strengthening mechanisms and noise spectra characteristics.

DOI: 10.1103/PhysRevB.83.104108 PACS number(s): 72.70.+m, 72.15.Eb, 81.40.Jj

I. INTRODUCTION

Crystalline metals are subject to dislocation nucleation and motion when they are submitted to stress. This leads to strain hardening, and ultimately to the breakdown of the material. This mechanical aspect became a more fundamental one when it was realized that the dislocation kinetics exhibit avalanchelike, scale-free, properties,[1] opening a route to a generic, to some extent material-independent, physical approach to the process.[2] At a macroscopic scale, however, a typical stress-strain curve still appears continuous due to the statistical averaging effect. To reveal the motion of dislocations, microscopic samples can be used, and they display some characteristics of crackling noise, for example plastic strain bursts.[3] Acoustic emission is also a dedicated probe of dislocation kinetics, sensitive to the microscopic strain mechanism. The statistical analysis of the data allows the construction of a probability density function which displays power-law distribution, as expected for avalanches. Recently, small-scale self-organized structures of acoustic events were observed.[4] Concerning the electronic transport properties, it is known that the averaged resistivity can be influenced by the amount of dislocations which are present in the material.[5] When thermally activated, the dislocations move and lead to a resistance noise significantly larger than the thermal Johnson noise. It was proposed that $1/f^{\alpha}$ with $\alpha \approx 2$, observed in metallic films, is a characteristic of this dislocation noise, and is explained by a Poisson distribution of elementary voltage steps.[6] This is in contrast with the $1/f^{\alpha}$ with $\alpha \approx 1$ that generally arises in metals due to the quasiequilibrium motion of defects (for example from the Dutta-Dimon-Horn process[7]). Bertotti *et al.* also proposed that $1/f^{\alpha}$ noise with $\alpha \approx 4$ could appear for a frequency $f > f_0$ when the motion of dislocations is correlated.[8] In this model, f_0 is linked to the average duration of pulses due to dislocation motion. To generate the plastic deformation responsible for the dislocation noise, the thermally induced strain generated by the lattice mismatch between a metallic film and the substrate can be used. With this technique, $1/f^2$ noise was observed but without a $1/f^4$ contribution. This latter was supposed to be below the apparatus sensitivity.[9] Low-frequency electrical noise, but close to $1/f$, was measured in carbon fibers subjected to tensile stress[10] and in metal films submitted to different stresses with a choice of different substrates.[11] This $1/f$ noise spectrum can be attributed to a large distribution of activation energy in the range above $k_B T$ of elementary $1/f^2$ processes.[7] If one measures the electrical noise during a tensile stress test, one can expect to obtain data directly correlated with the nucleation and motion of dislocations in the metal. Electrical noise was studied in stainless steel wires under tensile stress, but the major contribution was a narrowband noise which was interpreted as a manifestation of fluctuations in the piezoresistivity.[12] To our knowledge, only a few works devoted to the correlation between electrical properties and dislocation behavior take into account the structural heterogeneity of the dislocation patterns which occurs progressively inside the material during the strengthening (see, for instance, Ref. 13).

In this paper, we report measurements of voltage noise in a wire of pure copper during a monotonic tensile test. These measurements show that the changes between different stages of strain hardening and the resistance noise are directly correlated. In particular, we report the observation of the f^{-2}-f^{-4} crossover typical of dislocation clustering, and some subtle effects which are hardly accessible by other techniques.

II. EXPERIMENT

Experiments were performed on a polycrystalline copper wire of radius $r = 150\ \mu$m and purity of 99.99% by weight. The averaged electrical resistivity was measured using a four-probe method with an Adret A-103 current supply and a Keithley 199 voltmeter. The measured value at $T = 300$ K was $\rho = 1.78\ \mu\Omega$ cm. Regarding the noise setup, the measured voltage was amplified by an ultralow-noise preamplifier (NF Electronics Instruments, model SA-400 F3, with an equivalent noise $\approx$0.5 nV/Hz$^{1/2}$) enclosed in a thick screening box. Details of the setup can be found in Ref. 14. Signals were recorded with a PCI-4551 (National Instruments) analog input channel, antialiased and Fourier transformed in real time. The quantity of interest here is the autopower spectrum $S_{vv}(f)$ (V^2/Hz) of the voltage noise measured under a constant and noise-free applied transport current of $I = 100$ mA. Monotonic tensile tests were carried out using an Instron 5569 tensile test machine with suitable grips for thin wires. The grips were covered with insulating tape in order to isolate the wire and the test machine. The top grip was displaced at a constant rate of 10 mm/min and the force was measured by an Instron static load cell of 50 kN. The total tested length was 250 mm, and voltage pads were separated by 187 mm. Special care was taken to ensure that the voltage spectrum was free from spurious contributions. In particular, a narrowband noise centered at a frequency of 25 Hz was observed, identified as coming from the vibration of the setup and subtracted from the final spectrum. The background electronic noise is

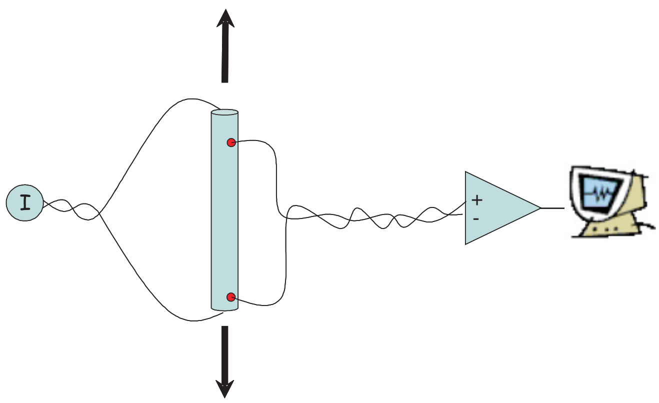

FIG. 1. (Color online) Schematic experimental setup showing the copper wire with the noise-free current supply, the contact pads, the preamplifier, and the computer for real-time data acquisition.

dominated by the preamplifier noise ($\approx$0.5 nV/Hz$^{1/2}$) which is higher than the Johnson noise of the copper wire at room temperature $[V_n = (4k_BTR)^{1/2} \approx 0.03$ nV/Hz$^{1/2}$ at $T =$ 300 K]. A schematic view of the setup is shown in Fig. 1. We made several measurements. Some of them were unsuccessful because the contact noise was dominant (Ohmic contacts were degraded during the tensile test). Two of them were free from such contact noise, and gave similar results that we report here.

III. HARDENING BEHAVIOR OF THIN COPPER WIRES

A typical rational stress (σ)–strain (ϵ) curve of Cu is shown in the inset of Fig. 2. The yield stress stands around

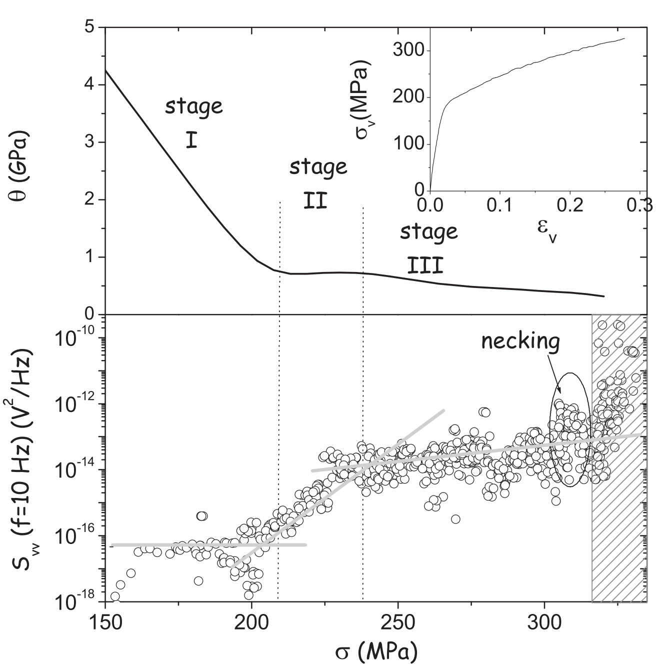

FIG. 2. Top: Kocks-Mecking plot(θ vs σ) for Cu polycrystalline thin wire plastically strained in tension. Also shown are the typical stages of plastic deformation (the inset shows the corresponding stress-strain curve of the material). Bottom: Voltage noise at $f =$ 10 Hz as function of the strain.

180 MPa. For higher stresses, the material exhibits three stages of strengthening typical of fcc metal behavior.[15] These stages can be conveniently studied by plotting the curve $\theta = f(\sigma)$, with $\theta = d\sigma/d\epsilon$ (top of Fig. 2). Stage I corresponds to microplasticity and appears for σ values close to the yield stress. During this stage, plasticity progressively develops from grain to grain until a homogeneous state is reached. During this first stage the well-favored slip systems $\{111\}\langle 110\rangle$ are activated in each grain. Stage II is characterized by a constant value of θ as a function of σ.[15] It is generally associated with the activation of less well-oriented systems and a few cross slips.[16] Activation of multiple gliding systems induces therefore the formation of heterogeneous dislocation structures like tangles, walls, or cells. Stage III begins with the decrease of $\theta = f(\sigma)$. It can be associated with a generalization of the cross slip, implying the annihilation of dislocations during dynamic recovery.[17] Transmission electron microscope observations performed on fcc metals, plastically strained at various strain levels, show that dislocation tangles develop at an early stage in the deformation.[18] Moreover, a well-defined cell structure is always observed when the hardening rate reaches the second stage (see, for instance, Ref. 19 for Cu or recently Ref. 20 for Ni). The dislocation cell size tends to decrease with an increase in strain, in a more pronounced way at the beginning of the second-stage hardening. The mean free path of mobile dislocations is therefore determined by the cell size.

The total dislocation density ρ^d grows monotonically whatever the cell structure from stage I to III. The stress σ is classically dependent on ρ^d following the relationship[21]

$$\sigma = \alpha\mu bM\sqrt{\rho^d}. \tag{1}$$

M is the Taylor factor, b the Burges vector, μ the shear modulus, and α a parameter taking into account the dislocation arrangement and the interactions between slip systems.

However, with the formation of cells, the local dislocation density may be significantly larger (ρ_w^d inside the cell walls) or smaller (ρ_c^d in the cell interior) than ρ^d. The latter is therefore a consequence of a mixture law, following a composite model previously proposed:[22]

$$\rho^d = f_w\rho_w^d + f_c\rho_c^d. \tag{2}$$

f_w and f_c are the area fractions of wall and cell interior, respectively, related through $f_w + f_c = 1$. In particular, mobile dislocations are encountered inside the cells and their density rises more slowly than that of stored dislocations inside the cell walls.

The copper wires experienced a strong necking before the final fracture, which begins for true stresses higher than 300 MPa. We observed by scanning electron microscopy a section reduction of about 2/3 at the fracture point.

IV. RESISTANCE NOISE DUE TO DISLOCATION KINETICS

We recall here the important points in the treatment of the dislocation noise, which was discussed, for example, in Ref. 9. The main idea is that abrupt changes in the dislocation density generate resistance pulses which can be described as Poisson processes, similar to a shot noise mechanism. In the simplest case, it is assumed that the time-dependent resistance is constituted of random and statistically independent (rectangular) pulses of height ΔR and duration τ. Some assumptions are required. τ is distributed according to an an exponential law $P(\tau) = \tau_0^{-1}\exp(-\tau/\tau_0)$ and the resistance change is proportional to the average number of dislocations (per unit time), N, in the sample. If the sample is biased with a constant current I_0, the voltage spectrum finally takes the form

$$S_{vv}^{(1)} = N(\Delta R)^2 I_0^2/\pi^2\left(1/\tau_0^2 + 4\pi^2 f^2\right). \quad (3)$$

This is a typical Lorentzian spectrum for Poisson-distributed elementary events.[23] The noise power is then proportional to f^{-2} for frequencies higher than $f_0 = 1/(2\pi\tau_0)$.

If clustering takes place, the resistance pulses are correlated, and the power spectrum changes accordingly. Note that the model is in several aspects similar to some models of the magnetic Barkhausen noise.[24] Considering bundles of dislocations, an averaged number of events per cluster, p, can be defined, with τ the averaged lifetime between such events in the cluster. This correlation between the pulses introduces a new term in the power spectrum, and an approximate expression for the noise spectrum can be worked out:[9]

$$S_{vv}^{(2)} = S_{vv}^{(1)}[1 + 2p(1 - N\tau_0)^2]/[1 + \omega^2\tau^2 p^2(1 - N\tau)^2]. \quad (4)$$

It can be simplified for an average number of pulses per clusters, $p \gg 1$,

$$S_{vv}^{(2)} = S_{vv}^{(1)}(1 + 2p)/(1 + \omega^2\tau^2 p^2). \quad (5)$$

According to Eqs. (3) and (5), the power spectrum can be rewritten in the compact form

$$S_{vv}^{(2)} = N(\Delta R)^2 I_0^2/\left[\pi^2\left(f_0^2 + f^2\right)\right] \times \{1 + 2p/[1 + (f/f^*)^2]\} \quad (6)$$

with $f^* = 1/(2\pi\tau p)$ the cutoff frequency of the clustered noise. Since $S_{vv}^{(1)} \propto f^{-2}$ above the cutoff frequency f_0, $S_{vv}^{(2)}$ tends to a f^{-4} behavior for large frequencies $f > f_0$. The characteristic frequencies follow the hierarchy $f^* < f_0$, i.e., the time constant of a cluster of dislocations (τp) is larger than the time constant of statistically independent dislocations.

V. RESULTS AND DISCUSSION

The voltage noise S_{vv} at a low frequency of 10 Hz is shown as function of the strain at the bottom of Fig. 2. Let us consider first the changes in the noise magnitude. A good correspondence between the variation of the voltage noise and the three stages of strengthening can be observed. As shown in Eq. (3), the noise magnitude is proportional to $N(\Delta R)^2$, i.e., to the rate of change of dislocation density in the sample and to the resistivity contribution of unit dislocation density.[9] For low plastic stress levels (stage I), the dislocation population mainly consists of isolated dislocations which slip along one crystallographic gliding system. During stage II multiple slip systems are activated leading to an increase in dislocation density and to the formation of cell walls. This change in the density of dislocations may be at the origin of the noise magnitude increase during stage II. When stage III is reached, the increase in S_{vv} is reduced; this phenomenon can be explained by the occurrence of dislocation annihilation processes, such as cross slips, which reduces the rate of dislocation accumulation.[17] Close to the necking, around 310 MPa, the noise magnitude at 10 Hz shows a bump. It is associated with a change of spectral shape which will be discussed later. Finally, just before the fracture of the wire, the noise increases strongly. This occurs for stresses higher than about 320 MPa. This last noise is essentially white and intermittent (not shown here). We propose that this increase in white noise is a resistance effect, revealed after the necking and explained by the fact that a local section of the wire decreases in diameter and that small cracks start to develop just before the fracture.

Let us consider the changes in the spectral shape during the tensile test. Some typical measured spectra are represented in Fig. 3. For low to moderate strains, the noise has a dominant $1/f$ character and a low magnitude Fig. 3(b)]. This behavior changes when $\sigma \geqslant 200$ MPa, close to the beginning of stage II, with a progressive increase of the noise. At this stage, the noise spectrum changes to a $1/f^2$ form Fig. 3(c)]. The noise value tends to saturate for $\sigma \approx 225$ MPa, apparently corresponding to the transition between stages II and III. Finally, close to the necking ($\sigma \approx 310$ MPa), the noise amount increases again. This time, both f^{-4} and f^{-2} components, separated by the characteristic frequency f_0, can be observed [Fig. 3(d)].

Regarding these spectral shapes, since stage II is defined by the formation of heterogeneous dislocation structures, two processes are expected: a slow one related to the clusters and a fast one related to dislocations inside clusters, so that according to Bertotti *et al.*[9] both f^{-2} and f^{-4} behavior should appear with a characteristic frequency of crossover of f_0. This frequency corresponds to an average duration $\tau_0 \approx 2\pi/f_0$. This duration is that of the independent pulses due to the dislocation motion. At the beginning, f_0 is not observed in the experimental window and only f^{-2} behavior is seen. But at the end of stage III, f_0 decreases and reaches the experimental window, so that both f^{-2} and f^{-4} are observed. The evolution of f_0 as function of stress is shown in Fig. 4. A clear maximum can be observed around

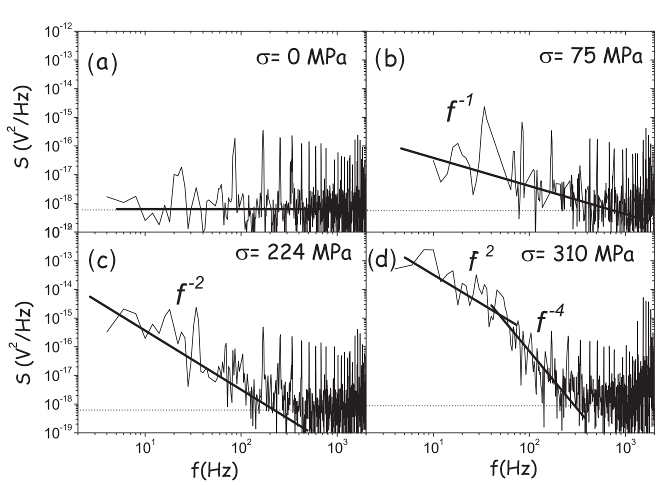

FIG. 3. Typical spectra observed during the wire deformation, for different strain values. The appearance of an f^{-4} dependence is consistent with dislocation clustering, observed here for strain values close to the necking.

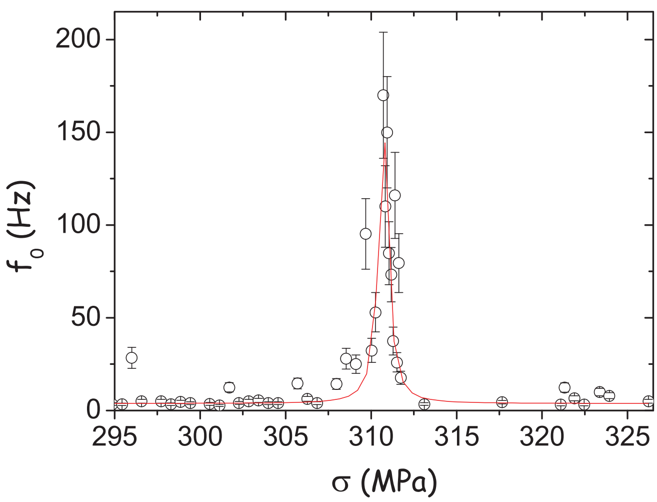

FIG. 4. (Color online) Variation of the crossover frequency f_0 as a function of the strain close to the necking. Note the maximum of f_0 at $\sigma \approx 311$ MPa.

311 MPa, i.e., in the regime where the necking is thought to occur following the strain-stress behavior (Fig. 2). Remembering that the necking is characterized by concentration of dislocations which suddenly localize in particular places, we think that the maximum of the frequency f_0 can be used as a precise probe of the necking of the wire. We show here that a high level of stress is necessary to reveal the f^{-2}-f^{-4} crossover, likely associated with a large concentration of dislocations. This fact might explain why it was not observed in Ref. 9, where the stress was achieved by changing the temperature of an aluminum thin film clamped over a silicon substrate. In addition, in our case the study of dislocation noise is performed isothermally, avoiding the fact that dislocation dynamics under stress is temperature dependent, and thus allowing a more direct analysis.

An interesting aspect of plastic deformation is that the rearrangement of correlated dislocations shows similarities with an avalanche process.[1,2] Scaling laws typical of crackling noise can then be expected, and are actually observed by the acoustic emission experiments.[25] After plotting the number of noisy events as a function of the noise values on a log-log scale, we observe a non-Gaussian probability distribution (Fig. 5). More precisely, a power-law dependence $P(\delta V) \propto \delta V^{-\alpha}$ reasonably fits our data with $\alpha = 1.5 \pm 0.1$. This exponent is close to the one measured when analyzing the intermittent behavior of the plastic response with acoustic emission, a result which indicates that dislocations likely move in a scale-free

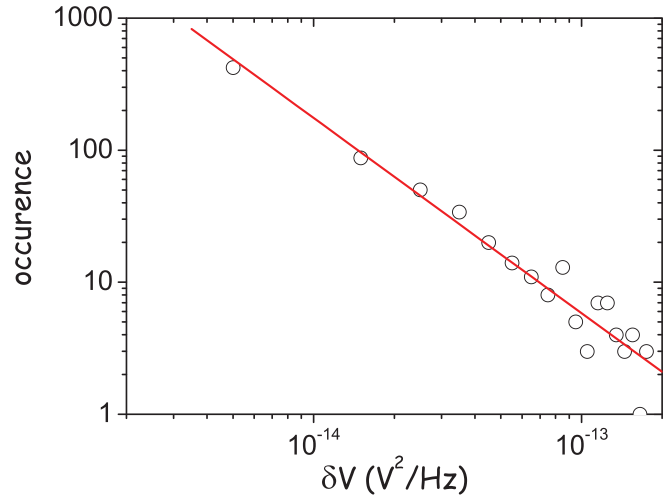

FIG. 5. (Color online) Histogram of the noise values on a log-log scale. The fit gives a slope $\alpha = -1.5 \pm 0.1$.

fashion.[1] The data scale whatever the strengthening stage (in stages II and III), as observed for the acoustic emission experiments performed to study plastic activity.[25] Note, however, that we have limited statistics with our data, and a power-law behavior cannot be rigorously proved. It remains that this result gives a good indication that our noise measurements and the acoustic emission experiments are observing similar statistical events.

VI. CONCLUSIONS

In summary, we have observed resistance noise due to dislocation motion in a metallic wire submitted to stress. It was shown that different stages of the strengthening can be related to a specific behavior of the voltage noise arising from the heterogeneity in the dislocation motion. Signatures of dislocation clustering are revealed. In addition to the technical interest of providing a nondestructive characterization of the strengthening stages and of the kinetics of plastic deformation, this technique is an interesting tool to reveal hidden processes and to study avalanchelike dislocation kinetics, and is certainly complementary to acoustic emission experiments.

ACKNOWLEDGMENTS

N.B. and A.P. thank Julien Aubril for his invaluable help during the data analysis.

*alain.pautrat@ensicaen.fr

[1] M. C. Miguel, A. Vespignani, S. Zapperi, J. Weiss, and J. R. Grasso, Nature (London) 410, 667 (2001).
[2] M. Zaiser, Adv. Phys. 55, 185 (2006).
[3] D. M. Dimiduk, C. Woodward, R. LeSar, and M. D. Uchic, Science 312, 1188 (2006).
[4] M. A. Lebyodkin, T. A. Lebedkina, F. Chmelík, T. T. Lamark, Y. Estrin, C. Fressengeas, and J. Weiss, Phys. Rev. B 79, 174114 (2009).
[5] P. V. Andrews, M. B. West, and C. R. Roheson, Philos. Mag. 19, 887 (1969).
[6] G. Bertotti, A. Ferro, F. Fiorillo, and P. Mazetti, in *Electrical Noise Associated with Dislocations and Plastic Flow in Metals*, Dislocations in Solids, Vol. 7, edited by F. R. N. Nabarro (North-Holland, Amsterdam, 1986), p. 1.
[7] P. Dutta, P. Dimon, and P. M. Horn, Phys. Rev. Lett. 43, 646 (1979).
[8] G. Bertotti, F. Fiorillo, and P. Mazetti, Phys. Scr. T 1, 134 (1982).

[9]G. Bertotti, M. Celasco, F. Fiorillo, and P. Mazzetti, J. Appl. Phys. **50**, 6948 (1979).
[10]Dinesh Patel, Yves Dumont, and I. L. Spain, J. Appl. Phys. **72**, 1901 (1992).
[11]D. M. Fleetwood and N. Giordano, Phys. Rev. B **28**, 3625 (1983).
[12]Lyndon D. Segales, James R. Gaines, Anupam K. Misra, and Richard E. Rocheleau, J. Appl. Phys. **88**, 4146 (2000).
[13]T. Narutani and J. Takamura, Acta Metall. Mater. **39**, 2037 (1991).
[14]J. Scola, A. Pautrat, C. Goupil, and Ch. Simon, Phys. Rev. B **71**, 104507 (2005).
[15]H. Mecking, in *Work Hardening in Tension and Fatigue*, edited by A. W. Thompson (TMS-AIME, New York, 1977), p. 67.
[16]J. E. Flinn, D. P. Field, G. E. Korth, T. M. Lillo, and J. Macheret, Acta Mater. **49**, 2065 (2001).
[17]U. Essmann and H. Mughrabi, Philos. Mag. A **40**, 731 (1979).
[18]K. Sumino, Y. Kawasaki, M. Yamamoto, and M. P. Sumino, Acta Metall. **11**, 1235 (1963).
[19]X. Huang, Scr. Mater. **38**, 1697 (1998).
[20]C. Keller, E. Hug, R. Retoux, and X. Feaugas, Mech. Mater. **42**, 44 (2010).
[21]H. Mecking and U. F. Kocks, Acta Metall. **29**, 1865 (1981).
[22]H. Mughrabi, Mater. Sci. Eng. A **85**, 15 (1987).
[23]A. Van der Ziel, Physica (Utrecht) **19**, 742 (1953).
[24]G. Montalenti, Rev. Phys. Appl. **5**, 87 (1970).
[25]J. Weiss, T. Richeton, F. Louchet, F. Chmelik, P. Dobron, D. Entemeyer, M. Lebyodkin, T. Lebedkina, C. Fressengeas, and R. J. McDonald, Phys. Rev. B **76**, 224110 (2007).