From Mild to Wild Fluctuations in Crystal Plasticity

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(Received 12 October 2014; published 11 March 2015)

Macroscopic crystal plasticity is classically viewed as an outcome of uncorrelated dislocation motions producing Gaussian fluctuations. An apparently conflicting picture emerged in recent years emphasizing highly correlated dislocation dynamics characterized by power-law distributed fluctuations. We use acoustic emission measurements in crystals with different symmetries to show that intermittent and continuous visions of plastic flow are not incompatible. We demonstrate the existence of crossover regimes where strongly intermittent events coexist with a Gaussian quasiequilibrium background and propose a simple theoretical framework compatible with these observations.

DOI: 10.1103/PhysRevLett.114.105504 PACS numbers: 62.20.fq, 05.65.+b, 05.70.Jk, 81.40.Lm

Two contradicting pictures of dislocation-mediated plastic flow are discussed in the literature [1,2]. The classical paradigm assumes that correlations among individual dislocations are weak and fluctuations are roughly Gaussian, which makes the homogenized description adequate. A different point of view emerged from the analysis of high resolution acoustic emission (AE) data in plastically deforming hcp crystals which showed that temporal fluctuations may be power-law distributed in size and energy [3] and may be clustered in both space [4] and time [5]. These observations, suggesting that averages do not represent typical behavior, were corroborated by the study of statistics of slip events in micro- and nanopillars [6,7] for fcc and bcc metals and supported by numerical models [3,8–10].

In this Letter we provide the experimental evidence that intermittent and continuous visions of plastic flow are not incompatible and that in some crystalline materials mild (near Gaussian) and wild (infinite variance type) fluctuations can coexist. It has been long noticed that AE in plastically deformed crystals may include both continuous background and discrete bursts [11]. While the continuous AE was thoroughly studied, the bursts were generally simply counted [12], or omitted as spurious even though sudden slips at irregular intervals could be also observed directly [13]. In this Letter we show that mild fluctuations, revealing uncorrelated dislocation motions, prevail in crystals where highly constrained dislocation entanglements screen long-range interactions and prevent cooperative behavior. Instead, wild fluctuations, representing highly synchronized restructuring events, dominate in crystals where unconstrained long-range elastic interactions allow dislocations to self-organize. In the intermediate crossover regimes where strongly intermittent events coexist with a Gaussian quasiequilibrium background, the observed scaling exponents are nonuniversal.

To interpret these observations we propose a simple stochastic mean-field model where dislocation flow is represented by a Gibrat-type proportional dynamics [14]. Self-consistent single-site models of this type with other types of multiplicative noise have been used before to explain spatial scale invariance of plastic flows in the hardening regime [15] and to describe mean field interface depinning of dislocations [16]. However, none of these models was able to capture statistics of avalanches observed in our experiments, which is Gaussian for small events and power law for large events.

Experiment.—We studied the acoustic signature of plastic events during *monotonic* loading of hcp (ice, cadmium, Zn0.08%Al) and fcc (copper, aluminum, CuAl alloys) macroscopic (*cm* to *dm*) single and polycrystals. Additional *cyclic* tension-compression tests were performed on pure (99.95%) aluminum polycrystals with large (~5 mm) grain sizes. While fcc crystals have a large number of active slip planes, which facilitates formation of dislocation junctions and leads to significant isotropic hardening [17], hcp crystals have a small number of easy slip planes, only the basal one for the materials tested here. The absence of 3D entanglements in hcp crystals enables collective effects manifesting themselves through strong kinematic hardening induced by long-range elastic interactions. As we show in Fig. 1, the measured AE signals consistently substantiate these differences over a range of deformational regimes (compression creep, uniaxial monotonic tension, tension-compression cyclic loading). The details of the experimental method can be found in Ref. [18].

In ice crystals (hcp), the AE has a form of an intermittent signal with a negligible continuous background [Fig. 1(a)]; cadmium crystals (also hcp) show a similar picture [Fig. 1(b)]. Instead, in copper crystals (fcc), the measured

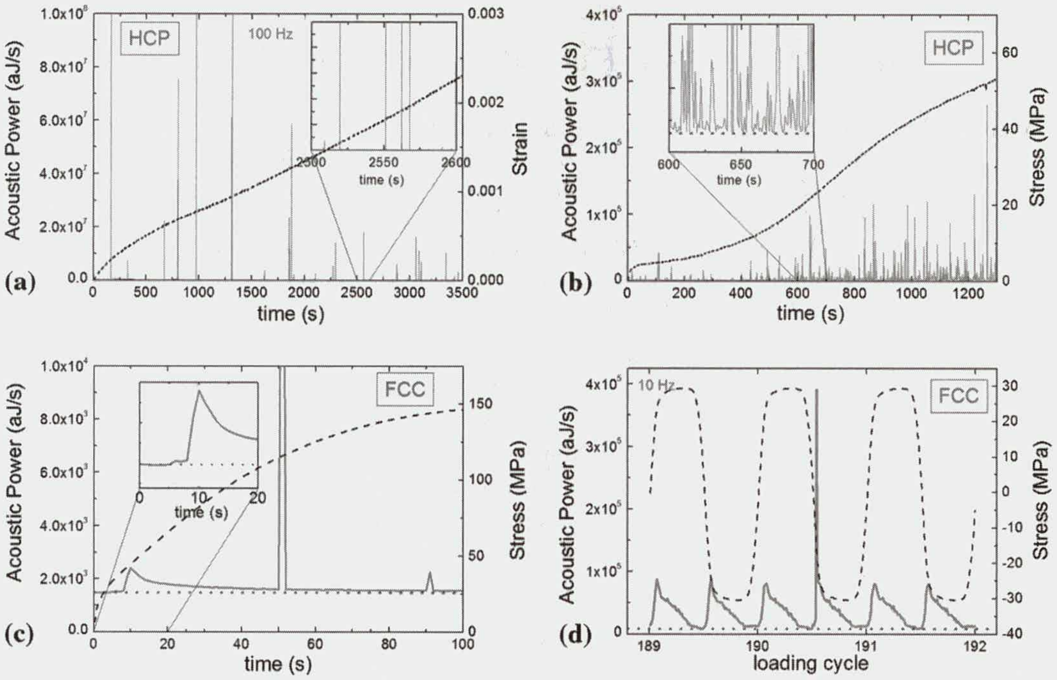

FIG. 1 (color online). Acoustic power recorded during plastic deformation. (a) Single crystal of ice (Ih) under uniaxial compression (creep at constant stress $\sigma = 0.56$ MPa, $T = -10\,^{\circ}\mathrm{C}$). (b) Cadmium single crystal under monotonic uniaxial tension ($\dot{\epsilon} = 1.3 \times 10^{-3}$ s^{-1}, $T = 20\,^{\circ}\mathrm{C}$). (c) Copper single crystal under monotonic uniaxial tension ($\dot{\epsilon}_I = 1.9 \times 10^{-2}$ s^{-1}, $T = 20\,^{\circ}\mathrm{C}$). (d) Aluminum polycrystal (grain size ~5 mm) under cyclic uniaxial strain control ($\epsilon_{\min}/\epsilon_{\max} = -1$, $\Delta t_{\mathrm{cycle}} = 10$ s, $\Delta\epsilon = 0.95\%$, $T = 20\,^{\circ}\mathrm{C}$). Red curves: acoustic power sampled at 1 Hz, unless noted otherwise. Black dashed curves: strain (a) or stress (b), (c), (d). Blue dotted lines indicate the level of instrumental noise.

acoustic signal is mainly continuous, reaching its maximum at plastic yield, with only occasional bursts above this background [Fig. 1(c)]. During cyclic loading of aluminum crystals (fcc), the acoustic signal is essentially continuous and symmetric in tension and compression, hence revealing its plastic origin. The continuous noise, however, is interrupted by bursts, on average less than 1 per loading cycle [Fig. 1(d)].

Remarkably, for both classes of crystals, the bursts are power-law distributed in maximum amplitude, $p(A_0) \sim A_0^{-\tau_A}$, and in dissipated energy, $p(E) \sim E^{-\tau_E}$ (Fig. 2). The exponents, estimated from a maximum likelihood method [20], are different for different types of crystals: for ice $\tau_E = 1.40 \pm 0.03$, for cadmium $\tau_E = 1.45 \pm 0.05$, for aluminum $\tau_E = 2.00 \pm 0.05$ and for copper and CuAl alloys $\tau_E = 1.54 \pm 0.08$. The average values and the associated standard deviations were obtained, for each material, over several tests; in the case of ice, our previous estimates of τ_E based on a least-squares fit of data [3] gave systematically larger values 1.5–1.6. The amplitude A_0, which is a proxy of the strain associated with the avalanche, scales as $A_0 \sim E^{1/2}$ [18], meaning that $\tau_A = 2\tau_E - 1$, i.e., $\tau_A = 1.8$ for ice and $\tau_A = 3.0$ for Al. Based on the value of τ_A, plastic fluctuations in ice can be qualified as wild with an undefined mean; for aluminum, with the variance diverging, we are just at the border between wild and mild fluctuations [21].

In contrast, the continuous AE signal sampled at 5 MHz is always near-Gaussian independently of the material and does not display any detectable intermittency or time clustering, see Fig. 3 and Ref. [18]. This is in agreement with the classical perspective where plasticity is viewed

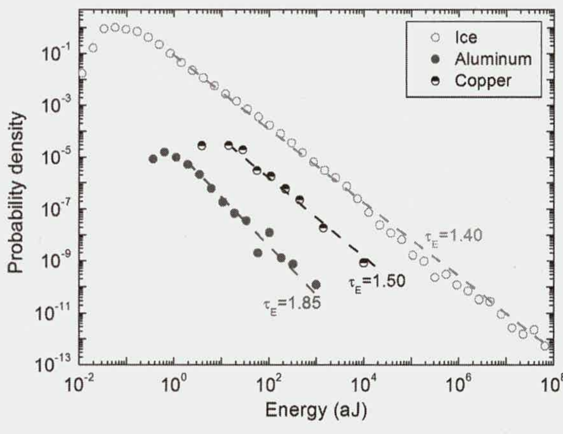

FIG. 2 (color online). AE energy probability density functions for bursts detected during: a uniaxial compression test on ice Ih [red open symbols; constant stress $\sigma = 0.56$ MPa, $T = -10\,^{\circ}\mathrm{C}$, see Fig. 1(a)], a monotonic tension test on copper (black semi-open symbols, $T = 20\,^{\circ}\mathrm{C}$, $\dot{\epsilon} =$ const), and a cycling loading test on aluminum under uniaxial tension compression [blue closed symbols, cycles 1 to 2000, see Fig. 1(d)]. The PDFs have been shifted vertically for clarity.

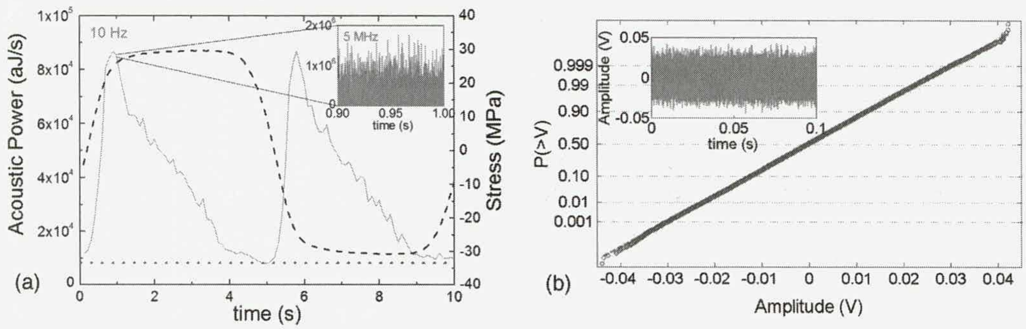

FIG. 3 (color online). Raw AE signal recorded during the cyclic loading of a polycrystal aluminum as in Fig. 1(a). (a) The red solid line shows the evolution of the AE power, sampled at 10 Hz. The stress is shown as a black dashed line. The inset shows the raw signal sampled at 5 MHz during 0.1 s near the plastic yield. (b) Distribution of local extrema of the acoustic signal (in V) shown as a normal probability plot (skewness $\zeta = 0.02$, excess kurtosis $\kappa = -0.27$).

as a sum of independent events similar in size and duration. The relative contribution of plastic avalanches responsible for bursts can be estimated by the amount of AE power recorded above the level of continuous signal with the acoustic power (in aJ/s) of the environmental noise first removed. Our measurements (see Table S1 in Ref. [18]) show that ice single and polycrystals represent a paradigmatic example of intermittent plasticity with nearly 100% of AE power released through AE bursts. In contrast, for aluminum, the contribution due to avalanches is small, reaching under cyclic loading at most a few percent during the first cycles, when the dislocation substructure has not yet fully developed. Copper and CuAl alloys stay in between, as it is also clear from the comparison of the exponents.

In summary, our observations show that hcp crystals with highly anisotropic slip (ice, Cd, Zn) exhibit correlated scale-free flows, facilitated by the dominance of long-range elastic interactions. Instead, in the studied fcc materials (Cu, Al, CuAl alloys), intermittent and continuous plastic flows coexist. The continuous component signifies the prevalence of small, uncorrelated dislocation motions taking place inside substructural units (cells, labyrinths, etc.) that effectively screen long-range interactions. The large bursts can be attributed to major autocatalytic cascades of unlocking events [17] leading to fundamental rearrangements of the transient dislocation substructures. This suggests that the commonly observed quasiequilibrium dislocation patterns in fcc crystals are only marginally stable and their restructuring can be triggered by insignificant changes in the global force balance. The intermediate behavior of Cu might be explained by a smaller stacking fault energy compared to Al, favoring the dissociation of dislocations and kinematic hardening.

Modeling.—A simple mean field type model, incorporating only the essentials of plausible mechanisms, provides a basic explanation for the coexistence of intermittent and continuous fluctuations. As a point of departure, we use a conventional mesoscopic framework and assume that the evolution of the spatially averaged density of *mobile* dislocations ρ is described (in a narrow window of stress variation) by a kinetic equation [22], $d\rho/d\gamma = a - c\rho$, where $a \geq 0$ accounts for the nucleation rate whereas $c \geq 0$ characterizes the prevalence of annihilation/immobilization over multiplication. Here we use the local shear strain γ as a parameter related to time through the Orowan relation. According to this model, in the steady state regime the average dislocation density is $\rho_c = a/c$, which introduces a characteristic scale.

A shortcoming of this coarse grained description is that it is fully deterministic. Stochastic models of plasticity with either additive [23] or multiplicative [15] noise have also been considered in the literature. The account of noise in the local kinetics of mobile dislocations is crucial because the yielding system is close to the state of marginal stability where fluctuations can be greatly enhanced and can interfere with the macroscopic evolution. If we make the simplest assumption that nucleation is deterministic but, due to environmental fluctuations, the annihilation rate c is randomly perturbed, we obtain the stochastic equation

$$d\rho/d\gamma = a - [c + \sqrt{2D}\xi(\gamma)]\rho, \qquad (1)$$

where $\langle\xi(\gamma)\rangle = 0$, $\langle\xi(\gamma_1), \xi(\gamma_2)\rangle = \delta(\gamma_1 - \gamma_2)$ and D is a constant parameter characterizing the intensity of fluctuations and introducing a second characteristic scale $\rho_D = a/D$. While Eq. (1) is linear, the nonlinearity of the microscopic dynamics is implicated through the randomness. In particular, the multiplicative noise describes the autocatalytic effect when dislocation clusters react to perturbations in a collective manner amplifying the effect of the noise proportionally to their size [24]. Such a cooperative response implies the presence of long-range fields that are not explicitly resolved in our zero-dimensional model; we also neglect quenched disorder and diffusion, whose account would allow one to model *spatial* intermittency [25] observed in microscopic models of crystal plasticity [8]. Multiplicative stochastic closure of the coarse grained models exemplified by Eq. (1) is rather common in the study of marginally stable driven systems [24,26] including turbulence [27], absorbing phase transitions [28], and depinning [29]. A direct link between the multiplicative random walks in the cluster size space and the emergence of criticality in systems with many degrees of freedom was established in Ref. [30].

To find the stationary probability distribution of the dislocation density $p = p_s(\rho)$ we need to solve the corresponding Fokker-Planck equation. We interpret it in the Stratonovich sense by assuming that $\xi(\gamma)$ is a colored noise with vanishing autocorrelation time [31]

$$\frac{dp}{d\gamma} = \frac{d}{d\rho}\left[[(c + D)\rho - a]p + D\rho^2 \frac{dp}{d\rho}\right]. \quad (2)$$

In the stationary regime [32]

$$p_s(\rho) \sim e^{-(a/D\rho)}\rho^{-[1+(c/D)]}. \quad (3)$$

At large values of ρ this distribution exhibits a power-law tail $\rho^{-\alpha}$ with exponent $\alpha = 1 + c/D$. Instead, around the maximum located at $\rho = a/(c + D)$ the distribution is Gaussian-like. When the noise is weak, $c/D \gg 1$, the fluctuations are mild, but as the strength of the noise increases, the system undergoes a noise-induced transition [33] with fluctuations becoming wild at $\rho_D/\rho_c = c/D \leq 2$. If we use the Ito interpretation, the power-law exponent in the tail changes to $\alpha = 2 + c/D$; however, the basic structure of the stationary distribution remains the same.

To link the proposed model with our AE measurements, we recall that the amplitude A_0 is proportional to the number of dislocations, involved in the avalanche, times their average length [18], hence, to ρ, thus giving $\alpha = \tau_A$. This identification, which we checked to be fully compatible with statistics of the dislocation density fluctuations in the microscopic model [8], allows one to interpret observed behaviors in terms of the values of the parameters a, c, D

First of all we note that to describe an idealized, single plane plastic flow without considerable nucleation and annihilation (modeled at the microlevel in Ref. [34]), we must consider the case when both a/D and c/D are small. Then Eq. (1) reduces to a logarithmic Brownian motion and $\alpha = \tau_A \to 1$ (Zipf law). In such systems dislocation dynamics is governed exclusively by elastic long-range interactions and this limit is approached by our hcp crystals where dislocation entanglements are minimal. In particular, our identification suggests that for ice $c/D = 0.8$ and also explains why in the corresponding experiments the Gaussian-like background was difficult to detect behind the experimental noise.

In materials characterized by stronger isotropic hardening, such as the fcc crystals tested here, short-range interactions are responsible for the formation of transient substructures that screen elastic interactions. Therefore, one can expect that $c/D \geq 1$, and accordingly, we obtain $c/D = 2.0$ for Al. In this case numerous independent nucleation events originating from cell walls would lead to continuous AE [35]. The observations also imply that the value of a is large enough to ensure a significant presence of the Gaussian plasticity. One can speculate that for bulk bcc materials in the low temperature regimes, where the Peierls stress is high, the appropriate scaling is $\rho_D \gg \rho_c$ and the statistics of fluctuations should be essentially Gaussian. This conjecture is supported by the fact that in bcc crystals, screw dislocation segments are not restricted to a single slip plane, thus favoring bulk multiplication [7], and by TEM *in situ* straining experiments showing parallel screws of both signs moving rather smoothly and experiencing quasicontinuous cross slip without any sudden bursts [36].

While these predictions are compatible with the difference between the fluctuation patterns in the *bulk* materials analyzed here, the situation is different for *nonbulk* systems such as nanopillars where power-law distribution of slip sizes was observed in both fcc and bcc crystals with an exponent of $\tau_A \sim 1.5$ [6,7], meaning $\tau_E \sim 1.25$. In these tests, however, the number of dislocations was small and their motion was limited to a single slip plane [6,37], thus precluding dislocation entanglements and short-range interactions (similar to bulk hexagonal crystals). The near critical behavior with low values of exponents in these nonbulk materials can be linked to the dominance of surface effects which limited nucleation and annihilation [38]. One can then argue that smaller is not only "stronger" but is also "wilder."

Despite the universally critical behavior at small sizes, one can expect for bcc and fcc crystals, a gradual transition from strongly intermittent to near Gaussian behavior of fluctuations as sample size increases. This is in full agreement with observations pointing towards smaller crossover lengths in bcc than in fcc nanopillars [7].

To conclude, we studied nonequilibrium steady state regimes of plastic flow, when a system continuously but unsuccessfully attempts to equilibrate by developing transient patterns with competing characteristic scales. The equilibration is never completely successful due to brutal rearrangements involving a broad range of scales. This picture is contained in our Eq. (1) which can serve as a stochastic rheological relation providing a closure for continuum plasticity [39]. The integration of intermittent and continuous regimes of plastic flow in a single computational framework will be an important step towards a reliable control of plastic deformation at micro- and nanoscales.

This work was supported by the French ANR-2008 grant EVOCRIT. J. W. and L. T. acknowledge the hospitality of the Aspen Center for Physics, supported by the NSF Grant No. PHY-1066293.

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