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We demonstrate the gradual shift from scale-free intermittent microplasticity to a scale-dependent behavior via the introduction of a variety of microstructural features within the Al-Cu binary alloy system. As long as the obstacles to dislocation motion remain shearable, the statistics of intermittent microplasticity has fat-tailed contributions. First the introduction of incoherent precipitates leads to a complete transition from scale-free power-law scaling to an exponential and scale-dependent distribution. These results demonstrate how non-Gaussian interactions survive across different microstructures and suggest further that characteristic microstructural length scales and obstacle pinning strengths are of secondary importance for the intermittency statistics as long as dislocations can shear their local environment.

I. Introduction

That plastic flow of crystals can proceed both smoothly and intermittently is a long-known realization in materials science and materials physics. Almost hundred years ago, when the foundations for today's crystal plasticity were laid by Orowan, Becker, Valouch, Schmid, and many others, it was already recognized that pure single crystals can, under certain conditions, exhibit an overall flow response that was composed of smooth continuous stress-strain segments, intermixed with abrupt stochastic strain jumps [1-3]. This bimodal mechanical response originates from strain localization where, in addition to homogeneous flow, large plastic events intermittently occur and lead to discrete plastic strain events. Despite the coexistence of smooth and intermittent plasticity in one and the same materials, approaches that homogenize plastic flow were successfully developed and are still used today. A prominent and simple example is the Orowan equation, $\dot{\gamma} = b\rho\bar{v}$, where $\bar{v}$ and ρ are related to average quantities that implicitly assume some Gaussian statistics over a large representative volume element, or simply the tested material. Exceptions to the norm of homogenous plastic (or viscous flow [4]), of pure metals have sporadically been reported in the literature, where either remarkable instrumentation with very high strain-resolution was used [5], or sensitive acoustic emission methods [6] revealed a fundamentally intermittent dislocation process that must underly the stochastic strain increments or pulse-energy excursions.

Today, the spatiotemporal nature of intermittent plastic flow is an established concept and in fact seen in a variety of bulk-scale experiments [7-9], and essentially the norm in small-scale mechanical testing [10-14]. Progress in understanding the intermittent rearrangement of the mediating dislocation network was mainly made via a statistical assessment of either acoustic emission pulses [15] or stress-strain increments [16], revealing non-Gaussian behavior in the form of perfect power-laws, $P \sim S^{-\tau}$ [17] or exponentially truncated power-laws, $P \sim S^{-\tau} e^{-L/s}$ [18,19], where S is the event size, P the probability of the event occurring, and L a material-specific length-scale. Such statistical signatures of the occurring events S represent a paradigm shift away from classical homogenization schemes [20] and indicate divergent length-scales over which the rearranging dislocation network couples and collectively responds to relax internal stresses. Both mean-field modelling of avalanches near the depinning transition in the presence

of a static pinning field [21] and dislocation dynamics simulations interpreting the stochastic evolution of a line-defect population as a jamming-unjamming [22,23] transition have given analytical insights into the scale-free statistics of intermittent plasticity.

Intriguingly, statistical divergence in the form of power-law scaling can persist for microstructures with internal length-scales [3], across grain boundaries [7], but it has also been shown that the introduction of deformation sub-structures can decrease S, which truncates $P(S)$ without affecting τ [19], or that both the truncation and the scaling exponents are affected by obstacles [24,25] or grain boundaries [26]. Very recent work on $P(S)$ further demonstrated how power-law truncation of micron-sized crystals is not linked to a finite size-effect, but rather to the degree of how collective dislocation activity localizes spatially [27]. In other words, L/S is linked to an internal length scale instead of the finite crystal dimension. Similarly, the scaling exponent τ can be sensitive to the presence of strong obstacles, as revealed with discrete dislocation dynamics, where the strength and distribution of static pinning fields [22,28] or the ratio of average precipitate size to sample diameter can be varied [25,29]. Being convoluted with a variety of other effects, such as external size-variation, strain-rate effects, and different microstructural obstacles, direct comparisons are difficult. These scattered and partially inconsistent observations therefore prompt the question what relationship there may be between an internal microstructural length-scale on one side, and the scaling exponent or the truncation length scale on the other side.

To shed more light onto this question, we probe here intermittent plasticity of pure Al and a binary Al-Cu alloy, which serve as model materials, where the alloy offers a tunable microstructure depending on its annealing history, spanning from a solid solution to a population of un-shearable θ'/θ-precipitates. From a materials physics perspective, this choice of crystals encompasses a structure admitting jamming-unjamming transitions in the case of pure Al and a variety of microstructures in which pinning-depinning dynamics is expected due to the different populations of shearable and unshearable obstacles in the binary alloy. Across the different microstructures but identical deformation rates and external sample sizes, a gradual truncation of the avalanche statistics from a truncated power-law to a finite valued distribution is revealed when transitioning from pure Al to a θ'/θ-precipitate-containing structure. The introduction of a

solid solution causes a pronounced increase of small avalanches, but the avalanche statistics continues to have a power-law like tail, albeit of lower probability than for the pure Al crystal. Quantifiable length-scales of the sampled microstructures do not yield any immediate rationale for the changes in intermittency statistics, and the loss of the extreme-value tail emerges due to the formation of un-shearable θ'/θ-precipitates.

II. Experimental Methods

Microcrystals were produced from a pure bulk Al polycrystal (99.995%, provided by Teck Products) and from a bulk polycrystalline Al-4.85wt%Cu alloy produced by Goodfellow. Hereafter we will refer to AlCu samples for simplicity instead of Al-4.85wt%Cu. All samples were mechanically polished to a mirror finish prior to microcrystal extraction. To ensure full dissolution of Cu in the Al matrix, the binary alloy was homogenized for 90 h at 813 K. Subsequent to homogenization, different bulk pieces of the AlCu underwent specific heat treatments to generate precipitation microstructures, as summarized in Table I.

Microstructure	Heat treatment
Solid Solution (AlCuSS)	Room temperature water quench (WQ) $\rightarrow$ stored at $273 - 278\,K$
GP zones (AlCuGP)	WQ $\rightarrow$ Naturally aged (NA) at room temperature ($293\,K$)
θ'' precipitates (AlCuθ'')	WQ $\rightarrow$ Artificially aged for $30\,h$ at $453\,K$
θ'/θ precipitates (AlCuθ'/θ)	WQ $\rightarrow$ Artificially aged for $200\,h$ at $453\,K$

Table 1: Heat treatments applied to each sample to obtain the desired microstructure. These microstructures were all confirmed via TEM analysis.

All microstructures (including pure Al) were characterized via EBSD in a JEOL 7000M scanning-electron microscope (SEM). Grains were found to be equiaxed and randomly oriented, with average diameters ranging between $50 - 500\,\mu m$. For each microstructure, a grain oriented as close as possible to the [001] direction (with misorientations $\leq 5°$ and accounted for when calculating Schmid factors) was selected for transmission-electron microscope (TEM) analysis and microcrystal preparation. TEM lamellae were extracted either in a FEI Helios 600i Dual Beam SEM/FIB or a Thermo Scios2 Dual-Beam SEM/FIB. The lamellae were analyzed in a JEOL 2010 LAB6 TEM to study dislocation and precipitate structures. Additional lamellae were

extracted *ad hoc* and subsequently analyzed to ensure statistical significance when determining precipitate size and spacing, as well as dislocation density.

  Microcrystals of cylindrical shape, with a height of $6\,\mu m$ and a diameter of $2\,\mu m$, were carved into the identified grains via FIB milling. The final tapering angle along the length of each microcrystal was evaluated to less than $1°$. A crater-like zone, $35\,\mu m$ in diameter, was left around each microcrystal to allow mechanical loading of the microcrystal without contacting the bulk of the crystal. The microcrystals were mechanically stressed under uniaxial compression with the use of a $11\,\mu m$ flat punch indentation tip affixed to a TI-950 Bruker-Hysitron nanoindenter. The compression experiments were displacement controlled at a constant rate of $0.6\,nm.s^{-1}$ over a total displacement per experiment of $150\,nm$, which corresponds to a strain rate of $10^{-4}s^{-1}$. The total compression displacement for each microcrystal was set as $1500\,nm$, i.e., ten individual loading experiments per microcrystal. The data acquisition rate was limited by the indenter's internal memory but was at least $800\,Hz$ for all experiments.

  The data output of the experiments are simple arrays containing time, depth, load, and corresponding voltages. All data files were processed via a series of Matlab routines. First, the slope of depth over time is output as an approximation for the quasistatic deformation response, where intermittent plastic events are detected by an abrupt increase of the moving slope. Second, selected event segments are manually reviewed and either kept as intermittent plasticity events or discarded as noise, the definition of which will be addressed later. Third, each event is bound in time and its depth/force trace is denoised by Wiener filtering. Once filtered, the peak velocity of the event is established, and new time bounds are set up where the momentary deformation velocity reaches 10% of the peak velocity on each side of the peak. The resulting events have a filtered depth, timespan, and velocity profile, and are statistically analyzed using the Python maximum likelihood estimation (MLE) method that is part of the `powerlaw` package developed by Jeff Alstott [30].

  Given the relatively small magnitude of some of the resulting intermittent plasticity events (less than $0.5\,nm$, see Fig. 5), particular care went into establishing a noise threshold. To this end, the AlCuθ'/θ sample was placed in the TI-950 under a load of $200\,\mu N$ (well within the

elastic domain of the sample) for a duration of $5\ min$, so as to obtain recordings of the background noise during experimentation. The background noise was found to follow a normal distribution with $\mu = 2 \times 10^{-5}\ nm$ (which can be approximated as zero) and a standard deviation $\sigma = 0.23\ nm$. However, this distribution returns the absolute displacement between two consecutive points, which is not necessarily applicable to intermittent plastic behavior, which is expected to consist in a sustained increase in depth over several data points, as highlighted in Figure 1. Here, the data points before and after the event have been separated, their respective noise distributions were calculated, and a data "envelope" corresponding to a distance of $\pm 2\sigma$ from the data mean was determined. If the envelopes before and after the event show a shift of at least half their width, it is considered that a plastic event has occurred beyond a reasonable doubt. Following this approach, the detection threshold for events was determined to $0.17\ nm$. Consequently, only events whose size S exceeds the threshold have been considered in the following.
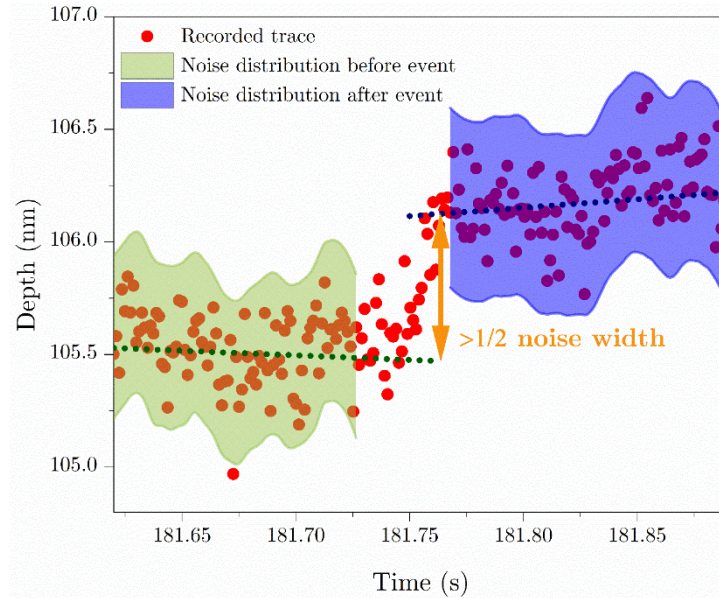
Figure 1: An example of a real data trace of an event for AlCuθ'/θ. An envelope of two standard deviations indicates the data distribution on both sides of the event. Since the change in the data distribution overall shifts to higher depth by more than half of the enveloped standard deviations, the event is detected as real. Here the example event as a final magnitude of ca. 0.6 nm.
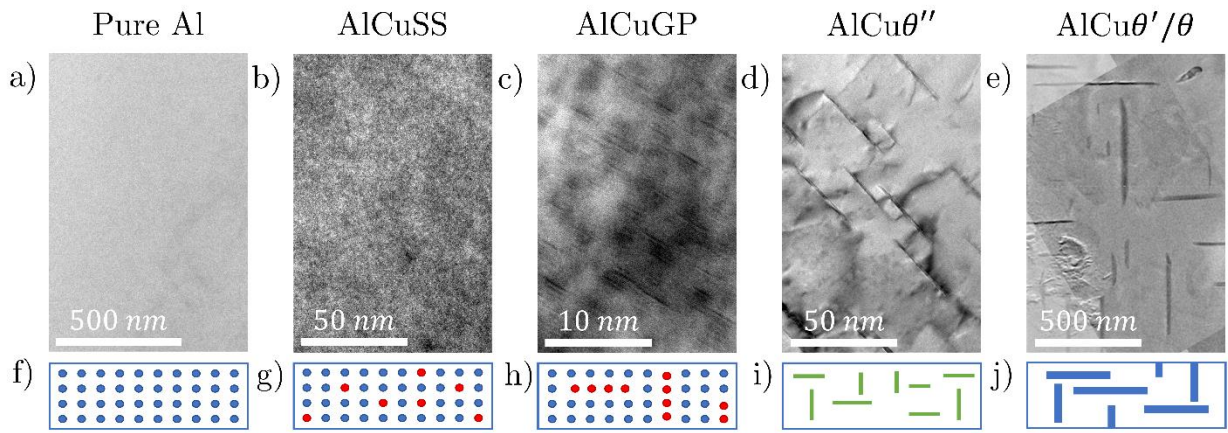
III. Results & Discussion

Figure 2: The top row (a-e) displays representative TEM micrographs of all probed microstructures and the bottom row (f-j) schematically visualizes the structures.

Figure 2a-e shows the TEM micrographs of the actual microstructures, while Fig. 2f-j shows a schematic of the expected idealized microstructures of each structural state. The microstructures are described as follows: a,f) pure and well annealed aluminum with a dislocation density of ca. $5 \times 10^{12} \, m^{-2}$. No precipitates or dislocation braids could be identified. b,g) AlCuSS, a substitutional solid-solution of copper in an aluminum matrix. The nominal composition of the alloy is 4.85 wt%Cu, which is below the maximum solubility of Cu into Al near the eutectic temperature ($821.4 \, K$), but results in a supersaturated solid solution once quenched to room temperature. In order to avoid room temperature aging, all sample of this microstructure were stored after quenching at a temperature of 273-278 K. c,h) AlCuGP containing Guinier-Preston (GP) zones in an AlCu matrix. GP zones are a well-known transitional structure encountered during precipitate growth in AlCu samples at room temperature. They consist of local concentrations of copper atoms that segregate into energetically favorable configurations following random diffusion. These configurations are AlCuθ'', small discs aligned on {001}-planes with a diameter approx. $1 - 10 \, nm$, and a thickness of up to a few atomic layers. Seen edge-on as is the case in Fig 1c), they appear as thin line contrasts only visible once atomic resolution is reached. d,i) a structure of θ''-precipitates that correspond to the next precipitation stage in AlCu alloys. θ''-precipitates (also known as GP II) are fully coherent with the matrix; literature shows that they are formed by Cu substitution on the FCC lattice sites at a distance of

three atomic planes and create a tetragonal structure [31]. They are the continuation of the Cu agglomeration process initiated with GP zones and are discs with a diameter of $10 - 100 \, nm$, a thickness of $5 - 10 \, nm$, and again visible viewed edge on, although circular regions of darker contrast could also be identified in the structure as the θ''-phase seen from their face side. e,j) AlCuθ'/θ is a structure containing the final stages of precipitation in AlCu alloys, namely θ'- and θ-precipitates. Unlike θ''-precipitates which grow directly out of GP zones, θ' precipitation can occur not only on GP/θ'' structures, but also on defects like dislocations and low-angle grain boundaries. θ'-precipitates are metastable and associated with a loss of coherency. The precipitates are still disc-shaped on the {001}-planes, with diameters ranging between $100 - 500 \, \mu m$ and having a thickness of $15 - 40 \, nm$. θ-precipitates are seen once the aging results in primarily incoherent precipitates that departs from the previous disc shape. Their size may also exceed that of the θ'-phase. While previous structures contribute to strengthening via shear mechanisms, the presence of θ'/θ induces a change from precipitate-shearing to Orowan looping. These precipitates have been extensively described in the literature [32-35], but their description of a relatively continuous growth process as separate categories brings some ambiguity when considering transition structures that don't fall squarely into one category. It should be noted that the precipitates are not expected to entirely exhaust the supply of Cu of the system, leaving dissolved copper in the Al matrix in all the AlCu-microstructures.

Key to the here aimed quantification of how internal length-scale may affect the statistical signature of plastic fluctuations, is a careful initial determination of both precipitate dimensions and spacing. To this end, a series of TEM lamellas were extracted from the GP, θ'', and θ'/θ-microstructure and evaluated.
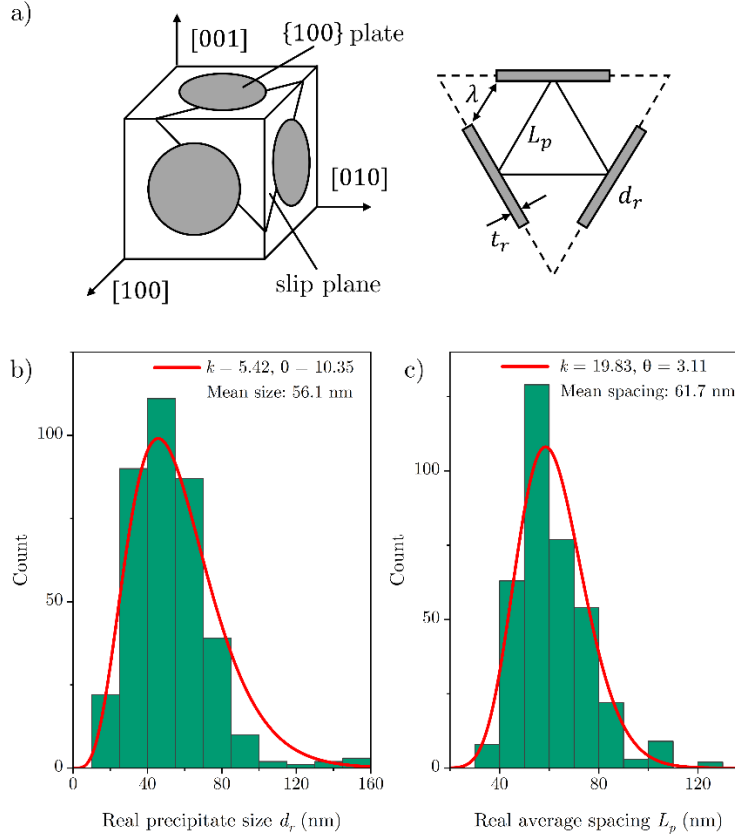
Figure 3: a) Schematic describing the relationship between the measured quantities on {001}-planes and the quantities as seen on the {111}-slip plane family relevant for dislocation-precipitate interaction. b) Histogram of precipitate size d_r for AlCuθ''. c) Histogram of precipitate spacing L_p for AlCuθ'' as defined in 3a. For both distributions, the best fit is given by the gamma distribution and allows us to output a mean value.

Before any quantitative analysis of the length-scales derived with TEM, it is imperative to realize that they originate from a 2D-projection. Figure 3a shows the real shape and distribution of the precipitates, as well as their center-to-center (L_p) and edge-to-edge (λ) spacings, which are relevant when discussing dislocation motion on the {111}-plane. The translation of measured dimensions into real length-scales of the 3D microstructure has been established before by Nie [36,37], and proven accurate [38-40]. Following Ref. [25], the real average diameter of the precipitates d_r is given by

$$d_r = \frac{2}{\pi}\left(d_m - t_f + \sqrt{\left(d_m - t_f\right)^2 + \pi d_m t_f}\,\right), \tag{1}$$

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where d_m is the average measured diameter, and t_f the TEM foil thickness. Determining the precipitate thickness is straightforward since the TEM micrographs are obtained using a [001] zone axis. The real thickness t_r can be shown to be $t_r = t_m \times \sin 54.74°$, with t_m the mean measured thickness and the 54.74° angle corresponding to the angle between the {001}- and the {111}-planes. Finally, we determine the average distance between precipitates that dislocations would encounter on the {111}-planes, being L_p and λ. As per the calculations detailed in Refs. [25,36], both distances can be related to precipitate size according to

$$\lambda = L_p - \frac{d_m}{2} - \frac{\sqrt{3}}{2}t_m = \frac{1.030}{\sqrt{N_v d_r}} - \frac{\pi d_r}{8} - \frac{\sqrt{3}}{2 \sin 54.74°}t_d, \qquad (2)$$

with N_v the precipitate density. The results for d_r, t_r, L_p and λ were compiled in histograms if applicable for the studied microstructures. For all histograms, the best fit to the data was found to be a gamma distribution (example fits are displayed in Figs. 3b and 3c), whose probability distribution function is of the form: $f_x = \frac{x^{k-1} e^{-\frac{x}{\Theta}}}{\Theta^k \Gamma(k)}$ with k and Θ being fitting parameters. The mean values, $x_m = k\Theta$, for the quantified length-scales are listed in Table 2. In the case of AlCuSS, λ and L_p are replaced by the average spacing between nearest-neighbor copper atoms based on the assumption of a homogeneous distribution of the atoms within the matrix. The values of Table 2 are consistent with similar studies [32,38,41], with roughly one order of magnitude difference in precipitate size and spacing between AlCuGP, AlCuθ'' and AlCuθ'/θ.

Structure	d_r (nm)	t_r (nm)	L_p (nm)	λ (nm)	τ_{pin} (MPa)	l (nm)
Pure Al	-	-	-	-	-	-
AlCuSS	-	-	2.1	2.1	106.7	67.0
AlCuGP	3.5	0.3	4.3	2.6	68.6	104.2
AlCuθ''	56.1	6.1	61.7	30.4	92.1	77.6
AlCuθ'/θ	437	19.1	365	173	58.0	123.2

Table 2: Real precipitates dimensions based on measured values during TEM analysis, as well as pinning strength τ_{pin} and corresponding characteristic length l as defined in Ref. [24]

An engineering stress-strain curve of each microstructure type is shown Fig. 4a. The strong variation in initial loading slope is a convolution of alignment imperfections and drift rates and should not be used for modulus estimations [42]. However, the flow regime is a robust plastic response of the material. Each flow curve exhibits stress-strain instabilities of different degrees that are separated by smooth deformation-curve segments. The abrupt strain increments are caused by collective dislocation rearrangements or dislocation avalanches, which are known to be dominated by the activation of single-armed dislocation sources that lead to the formation of visible slip offsets (inset in Fig. 4a) [43]. Qualitatively, the stress-strain curves reveal a gradual reduction of intermittency as the precipitate size increases. Such a change in intermittency has experimentally been discussed in the context of microcrystal size [44] and precipitate-microcrystal size ratio [25], whereas the effect of a varying disorder strength of a static pinning field – here represented by the microstructural deviations away from pure Al – has so far been limited to two-dimensional dislocation dynamics simulations [45]. Before quantifying the change in intermittency due to different types of internal pinning sites and characteristic length-scales, we turn our attention to the strength variation across microstructures, which is summarized and compared to corresponding bulk experiments [38] in Fig. 4b.
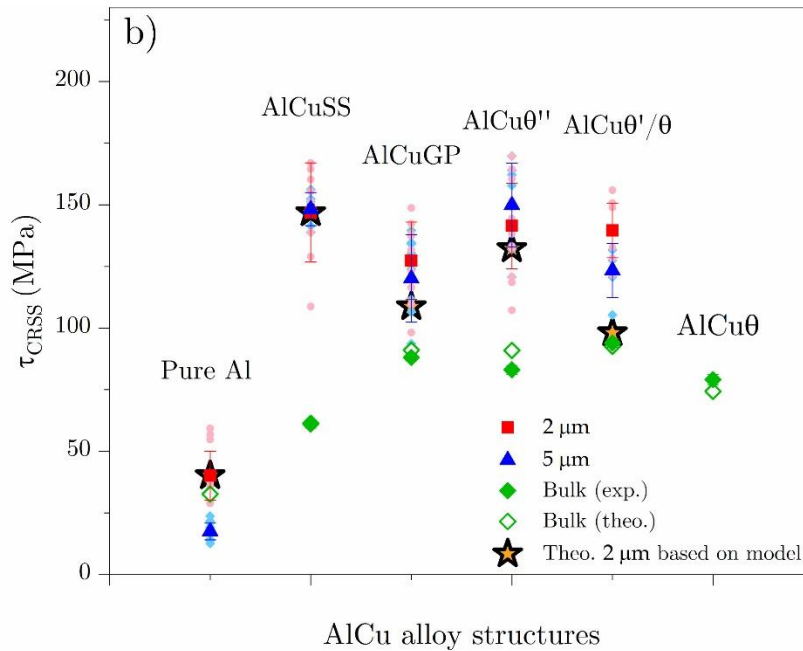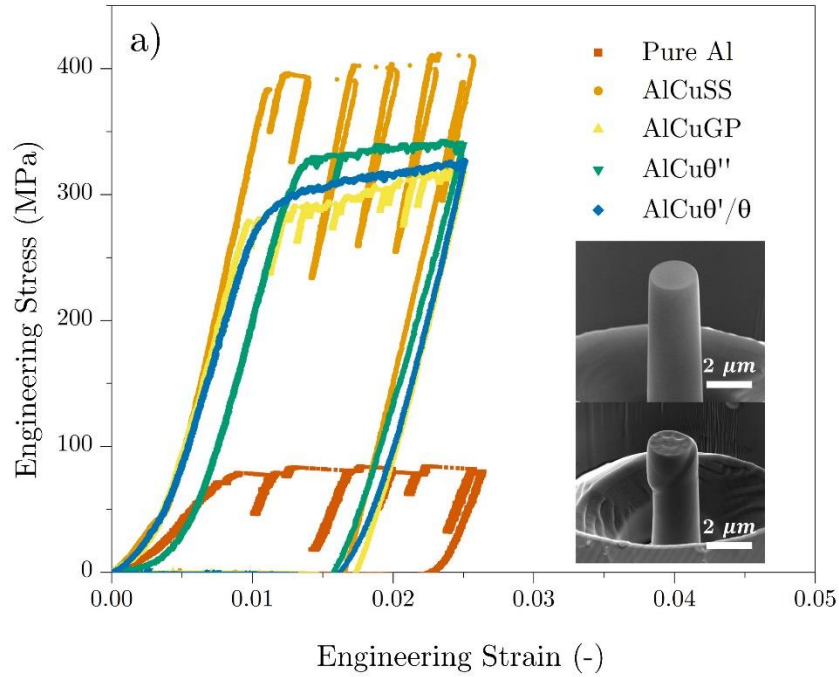
Figure 4: a) Measured engineering-stress strain curves for each sample microstructure. The addition of Cu results in a noticeable increase in flow stress. Inset: SEM micrographs of a pure Al microcrystal before and after microcompression. The sharp slip trace is the expected behavior for an FCC structure. b) Critical resolve shear stress, τ_{CRSS}, for each different microstructure, measured on microcrystals of diameter 2 μm and 5 μm. Unlike the tendency observed in the bulk in literature, the highest τ_{CRSS} are

measured for the AlCuSS structure. The bulk theoretical and experimental values of Ref. [38] are included for comparison purposes.

Given the typically large scatter in strength at the micron-scale, the measured critical resolved shear stress, τ_{CRSS}, during the flow regime of more than ten microcrystals has been averaged for each alloy. Figure 4b displays both data points for individual microcrystals and their average values for both 2 μm and 5 μm diameter samples. The reason for also testing larger microcrystals will be addressed later. As expected, the pure Al crystals exhibit the well-known finite sample-size effect due to dislocation sampling statistics, source hardening and exhaustion hardening [46,47]. Introducing Cu to Al, a substantial jump in τ_{CRSS} is seen for all AlCu-microstructures, but within the spread of individual measurements, no justifiable difference in strength between them can be derived. Irrespective of AlCu-microstructure, the average shear flow stress attains a level of 130-150 MPa with a spread of around 50 MPa indicating no gradual increase in strength as known for the bulk microstructures that continuously become stronger towards the AlCuθ'/θ and weaken with the dominance of θ due to increasing Orowan looping. This classically known behavior of peak-aging [35,48,49] is captured by the bulk data contained in Fig. 4b for an AlCu4.2% binary.

To better compare the strength difference between the microcrystals and the expected bulk values, we use the same theoretical strength model underlying the bulk data in Fig. 4b [38]. This model relies on summing the different strengthening contributions of each microstructural feature to a base strength of pure Al. For the solid solution, the strengthening contribution is given by $\tau_{ss} = HX_{Cu}^n$, with X_{Cu} being the weight fraction of Cu dissolved as a solid solution in the Al (here $4.85\ wt\%$), and H and n constants found to be $22\ MPa$ and 1 respectively for Al-Cu binary systems [50]. The contribution of precipitates is a bit more complex and depends largely on their geometry, orientation and shearability. GP zones, in particular, have proven hard to be modelled and instead rely on a simplification, where GP zones are assumed to be spheres of diameter d, according to

$$\tau_{GP} = \frac{1}{b}\sqrt{\frac{3f}{2\pi}}(0.72Gb^2)\left(\frac{2}{d_c}\right)^{1.5}\left(\frac{d}{2}\right)^{0.5}, \tag{3}$$

where $d_c = 20\ nm$ and $f = \frac{d_r^2 N_v \pi}{4} t_d$ [37]. The above model is only valid if the GP zones' diameter $d < d_c$, which as per Fig. 2 is the case. θ''-precipitates are treated as circular plates on {001}-planes and we expect them to be weak enough to be sheared on the {111}-planes, in which case the corresponding strengthening contribution has been described by [51]:

$$\tau_{\theta''} = \frac{0.908 d_r}{t_d^2} \left(\frac{2\pi b f}{G b^2\ ln \sqrt{\frac{d_r^2}{2b^2 f}}} \right)^{0.5} \gamma^{1.5}. \tag{4}$$

Here, γ is the interfacial energy between the matrix and precipitates, evaluated to be $154\ mJ.m^{-2}$ [38]. Finally, θ' and θ precipitates are too large and incoherent to allow shearing; instead, dislocations interact with them through Orowan bowing. The resulting strength increase is modelled as [36]:

$$\tau_{\theta'} = \frac{2}{\lambda} \left(\frac{Gb}{4\pi\sqrt{1-\nu}} \right) \ln \left(\frac{1.225 t_d}{b} \right). \tag{5}$$

Using this approach, the tested microstructures can thus be given an estimated bulk strength of value $\tau_{CRSS} = \tau_{CRSS,Al} + \tau_{SS} + \tau_{GP} + \tau_{\theta''} + \tau_{\theta'}$, where pure Al, the dominant obstacle type, and the remaining solid-solution contribution is used. The thereby obtained values are included in Fig. 4b and show good agreement with the experimental data for all but the AlCuθ'/θ microstructure. We understand this discrepancy by the fact that the sampled volume is not sufficiently large to truly represent an average strengthening due to a θ'/θ precipitate structure. Now much of the deforming volume has a free surface instead of being confined by non-shearable obstacles.

With these bulk values at hands, it becomes evident, that the characteristic length-scales listed in Table 2 are sufficiently averaged over in the case of the AlCuSS, the AlCuGP, and the AlCuθ'' microstructures as to be in a size-independent regime. In other words, the here tested AlCu-microcrystals are small volumes which's flow response is representative of bulk plasticity. To

test this idea further, we conducted a test series of 5 µm diameter crystals of each microstructure and the averaged data is also shown in Fig. 4b. Only in the case of pure Al, a clear size-dependent reduction in the average flow shear stress is observed, whereas the binary microstructures exhibit identical flow stresses for both sample dimensions, supporting the view that we probe a size-independent flow behavior.

Returning our attention to the pronounced strain excursions seen in Fig. 4a. These abrupt strain increments are in addition to the finite-size strengthening the second size-effect at this sample scale [52]. We now construct the size statistics of all extracted dislocation avalanches from each microstructure and all stresses. To this end, we rely on the stress-integrated Complementary Cumulative Distribution Function (CCDF), which is defined as $C(S) = P(S \geq S_{set}) = D \int_{S}^{\infty} P(S) dS$, or in other words the probability of a recorded event size S to be larger or equal in magnitude to a given size S_{set}, with D being a normalization prefactor. The CCDF is preferred over the PDF because it is defined for every value and does not require any binning of discrete data that can have subtle but important effects on subsequently performed distribution fitting. The here collected data is known to follow a power-law or truncated power-law (TPL), $P(S) \propto S^{-\alpha} e^{-\delta S}$, where α is the scaling exponent of the power-law and $e^{-\delta S}$ an exponential cutoff function described by the parameter δ. The corresponding CCDF follows mathematically the same trend, being $C(S) \propto S^{-\tau} e^{-\mu S}$, with $\tau = \alpha - 1$ and μ is yet a different non-universal parameter that describes the cutoff. Figure 5 summarizes the CCDF of the experimental data.
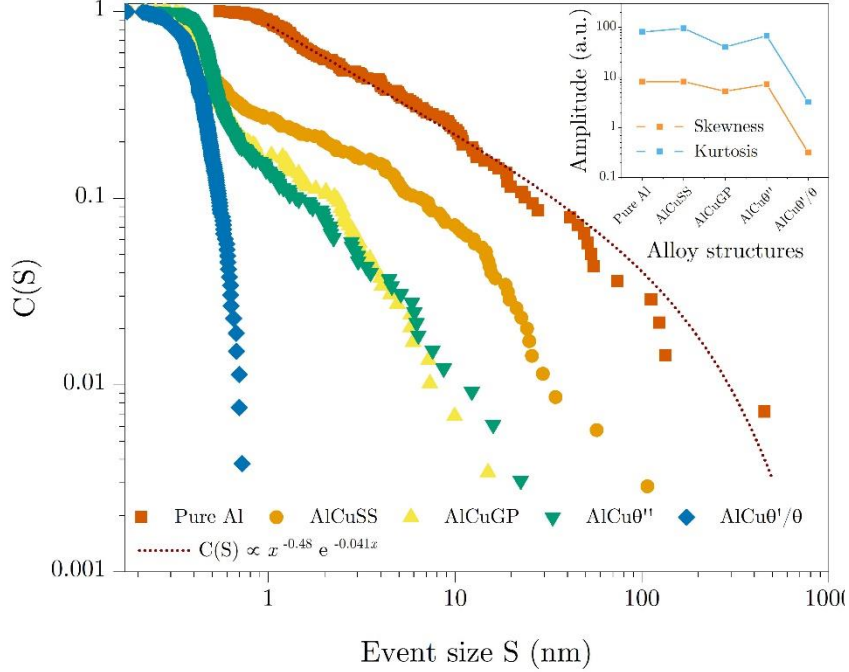
Figure 5: CCDF of event sizes S for all considered structures. The evolution of event size distribution is non-trivial and except for pure Al, they cannot be captured robustly with a given statistical function. Inset: Statistical moments (skewness, kurtosis) for all considered structures.

A strong variation of event-size statistics can be seen, admitting a TPL distribution for the case of pure Al and an exponential distribution describes the data for the AlCuθ'/θ-microstructure. Roughly, a gradual suppression of long-range correlated and scale-free like dislocation activity is observed in order from pure Al, AlCuSS, AlCuGP, AlCuθ'', to AlCuθ'/θ. Whilst the scaling exponent $\tau \approx 0.5$ (or $\alpha \approx 1.5$) would be in agreement with mean-field depinning [53], we emphasize here that this must be more a coincidence than anything else, as both experiments [39] and modelling [54] have demonstrated the large variety of scaling exponents in the range of approximately $1 < \alpha < 2$. This range seems to be a result of a variety of boundary conditions, as well as the dynamical internal dislocation network evolution, and is more compatible with the jamming-unjamming framework [23]. Introducing a population of Cu atoms into the Al matrix clearly causes a significant drop of $C(S)$ for $S > 1$ nm. In other words, a substantial number of resolvable dislocation avalanches are now suppressed to the part of the distribution containing the smallest displacement magnitudes. Similarly, the largest events reduce their scale to ca. 100 nm, which is about an order of magnitude smaller than the truncation length-scale for pure Al. This observation remains somewhat qualitative due to the inability of a meaningful fitting of the

distribution. Attempts of fitting the overall data set for AlCuSS with a weighted exponential and TPL-distribution for the different size regimes did not yield robust results, which is due to the insufficient total range of S. This decrease of the truncation length scale is much less than reported for cluster formation in an Al0.3wt%Sc-alloy with a majority of clusters in the size of 2-20 nm ($L_p \approx 20$), for which the event-size distribution truncates below 10 nm [24]. The contribution of the exponential shoulder at low value of S increases for AlCuGP and AlCuθ'', but the fat-tail avalanche part at large S remains statistically indistinguishable for both cases. We note that the tail for these two microstructures approximates to a slop of unity that would be an exponent of 2 in the case of the PDF, which indeed is in good agreement with a prediction by Brown [55]. With the emergence of the θ'/θ-precipitates, all correlated-long range interaction that would emerge via a strong PL or TPL contribution is lost, and avalanches now follow scale-dependent statistics best described by an exponential distribution. Essentially, the here seen transition in statistical behavior is much like a so-called mild-to-wild transition discussed earlier [56]. We note that the underlying dislocation activity for the θ'/θ-microstructure still is weakly correlated, since the avalanche statistics does not transition to a pure Gaussian form [57,58]. A moment analysis for the different distributions captures this change from scale-free to scale-dependent statistics clearly via a marked change in both kurtosis (4th normalized moment) and skewness (3rd normalized moment) once fully coherent precipitates are present (inset in Fig. 5). This trend is qualitatively compatible with earlier work focusing on the finite-sample size variation across an AlCuθ'-microstructure with 2.5wt% ($L_p \approx 1000$ nm) and 4.0wt% Cu ($L_p \approx 700$ nm) and therefore significantly larger L_p-values [25] that are approaching the sample dimension. In Ref. [25] the tail structure of the distributions was approximated with a PL, of which the scaling exponent increased strongly with decreasing L_p for this particular precipitate type.

How can this transition to a finite avalanche size scale be understood? Long-range correlated collective dislocation dynamics with scale-free avalanche statistics in pure metallic systems is now an established phenomenon in this type of microplasticity [3]. A specific microstructural length-scale correlating with either the scaling exponent or the truncation length-scale has so far not been identified for pure metallic systems that only contain an evolutionary dislocation network. This is also agreeing with work proposing a model that links the fraction of

intermittently admitted strain to the scaling exponent for a variety of AlSc- and AlCu-alloys [24,25]. Without the presence of quenched disorder (static pinning sites), it continues to remain unclear what property of the existing and evolving microstructure determines α or the truncation term μS. Indeed, TEM investigations yield a typical picture of a well-established dislocation network for the deformed microcrystals, as displayed in Fig. 6a. Estimating the dislocation density for the undeformed and deformed Al yields ca. $5 \times 10^{12} \; m^{-2}$ (equivalent to a mean spacing of 445 nm) and ca. $8 \times 10^{13} m^{-2}$ (mean spacing of 110 nm), respectively.

The introduction of quenched disorder via the solution of Cu does alter this behavior, but clearly long-range coupling and avalanching over length scales orders of magnitude larger than the solute spacing still remains a strong part of the network evolution. Now TEM micrographs reveal the expected pinned dislocation lines in the deformed microcrystals (Fig. 6b). Those events underlying the emerging shoulder at low S for AlCuSS are just above the resolution limit and admit slip equivalent to $0.4 - 0.8$ nm in net magnitude; a length scale that in fact is in good agreement with the average spacing of the introduced Cu solutes. Depleting the matrix of Cu and forming AlCuGP and AlCuθ'' introduces microstructural length scales of very different magnitude (Table 2). Despite the order of magnitude larger precipitates and their distances, both microstructures exhibit the same avalanche-size distribution $C(S)$ in Fig. 5. Post-mortem TEM micrographs (Figs. 6c and 6d) reveal dislocation lines crossing the GP- and θ''-precipitate structure and no particular change of $C(S)$ is observed. Both the size and the distances of the θ'' structure are at or beyond the truncation of the distribution. In both AlCuGP and AlCuθ'', however the shoulder at low S remains, which is understandable as sufficient Cu remains solved in the matrix. Given this similar avalanche distribution, one may ask if the AlCuθ'' still contains a population of GP-zones, which could be the dominant obstacle population controlling $C(S)$. To verify this, TEM micrographs from both initial microstructures are examined, paying attention to any possible signature of GP-zones in the AlCuθ''-alloy. Figures 6e and 6f demonstrate that the tested AlCuθ''-alloy was sufficiently aged as to dissolve all GP-zones. This leads to the conclusion that $C(S)$ is experimentally indistinguishable for both microstructures due to the shearability of their precipitates. In view of the result by Sun and co-workers [24,25], where the fraction of plasticity due to intermittent events is linked to the ratio between the sample diameter and a length scale $l = Gb/\tau_{pin}$, with G being the shear modulus, b the Burgers vector and τ_{pin}

the effective pinning strength of the obstacles, $C(S)$ for AlCuGP and AlCuθ'' would be expected to differ due to different τ_{pin} (Table 2 lists both τ_{pin} and l for the here investigated alloys). This is not the case, which however may be drowned out in the large scatter of the data underlying the proposed model.
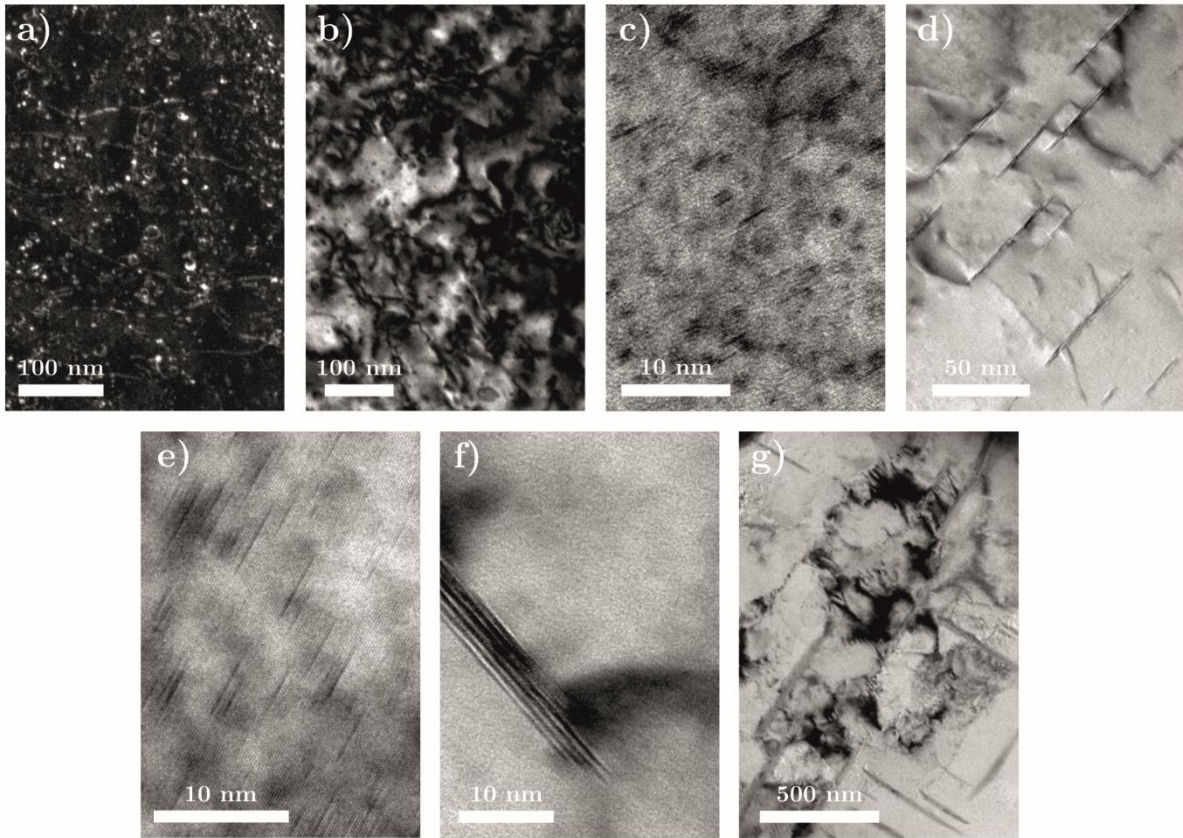
Figure 6: TEM micrographs of dislocation behavior in each sample microstructure. a) Dislocation density in pure Al after deformation (seen here in DF). b) Dislocations in AlCuSS, where some amount of pinning is observed. c) Dislocation lines being pinned at some GP zones. d) Dislocations interacting with shearable θ'' precipitates in AlCuθ''. e) Micrograph of AlCuGP structure showing the presence of GP zones throughout the entire sample. f) High-resolution micrograph of AlCuθ'' featuring the edge of a precipitate but no GP zones are observed. g) Dislocation-precipitate interaction in AlCuθ'/θ. Impenetrable precipitates cause pockets of high dislocation densities and pile up, suppressing correlated long-range interaction.

Once incoherent, the precipitates in AlCuθ'/θ can no longer be sheared anymore and significant pile-up can be observed in the inter-precipitate regions. This leads to confined regions of

significant plastic activity, whereas other inter-precipitate regions show no dislocation accumulation in post-mortem TEM micrographs (Fig. 6g). With a center-to-center distance still being smaller than the microcrystal diameter, selected sample sub-volumes accommodate the far-field stress via intense dislocation activity without being able to develop sufficient collective behavior as to admit large instantaneous slip events. We note that the fraction of strain (or displacement) admitted by resolvable discrete plastic events is still between 10-20%, even though such discrete events have no scale-free like statistics. As such, the gradual reduction in truncation length and the change for scale-free to scale-dependent avalanche statistics seen in Fig. 5 seems to be dictated by the nature of the microstructural obstacles (pinning strength, shearable vs. not shearable), whereas their introduced length-scale matters little.

IV. Conclusion

How microstructural length-scales and specific obstacle types affect the correlated-collective dislocation motion that underlies intermittent plastic events in form of dislocation avalanches continues to remain unclear. Here we observe a systematic transition from a correlated scale-free like to a still correlated but scale-dependent statistical signature of intermittency when probing the microplastic response of Al and a variety of microstructures produced via controlled annealing of an Al-4.85wt%Cu binary alloy. The simplest deviation from pure Al is the addition of Cu as a supersaturated solid solution, for which the avalanche distribution continues to have a strong power-law like tail admitting avalanche sizes orders of magnitude beyond the characteristic length scale of the solid solution. In co-existence with this continued long-range correlated-collective dislocation activity, a marked contribution of short-range interactions emerges in the size distribution. In this case, being an ideal model of a static-pinning field, the strong signature of small avalanche sizes manifests itself at the same length-scale as characteristic for the solid solution. For all other microstructures that contain different types of precipitates and characteristic length scales, such a correspondence cannot be observed. Instead, a continued suppression of the high-value tail (power-law signature) seems to be governed by the shearability of the microstructural obstacles rather than their related length scales or pinning strength. This is in qualitative agreement with dislocation dynamics simulations, in which only the introduction of a sufficiently high pinning strength eventually suppresses the critical (scale-

free) dynamics [59]. Whilst the present work cannot precisely account for the pinning strength distribution of the complex microstructures, in which a combination of a solid solution and a precipitate type always prevails, we conclude that scale-free dislocation avalanche dynamics in similar fcc systems with clear microstructural length-scales occurs simultaneously with weakly correlated and scale-dependent non-Gaussian activity as long as penetrable interfaces (lattice coherency) exist. The same must apply to hexagonal closed packed polycrystalline metals containing a boundary network, for which signatures of intermittent avalanche activity still can be revealed [7,26]. These collected experimental observations add to reports demonstrating changes from scale-free to scale-dependent intermittency due to the stress state [12], as well as a temperature-dependent suppression of critical avalanche dynamics due to an increasing dominance of the lattice friction in bcc metals [60], all of which reinforce the notion of intricate non-universal and microstructural-specific avalanche dynamics. Overall, it becomes increasingly clear that Gaussian and non-Gaussian dislocation activity co-exists, and that the latter includes the extreme dislocation events that may be rare but that can trigger significant local stress singularities. Even though dislocation self-organization and patterning can suppress the non-Gaussian correlated activity during deformation, it emerges again prior to failure, giving strong indications for its critical role in triggering failure [61,62]. These recent insights and the here observed statistical transition due to the microstructure in complex engineering alloys urge for continued fundamental efforts that take into account the co-existence of Gaussian and non-Gaussian defect behavior such that metallurgical failure prediction eventually embraces both statistical components.

Acknowledgements

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