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Nucleation Theory for Yielding of Nearly Defect-Free Crystals: Understanding Rate Dependent Yield Points

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Experiments and simulations show that when an initially defect-free rigid crystal is subjected to deformation at a constant rate, irreversible plastic flow commences at the so-called yield point. The yield point is a weak function of the deformation rate, which is usually expressed as a power law with an extremely small nonuniversal exponent. We reanalyze a representative set of published data on nanometer sized, mostly defect-free Cu, Ni, and Au crystals in light of a recently proposed theory of yielding based on nucleation of stable stress-free regions inside the metastable rigid solid. The single relation derived here, which is *not* a power law, explains data covering 15 orders of magnitude in timescales.

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The phenomenon of yielding is possibly the most conspicuous, and therefore well studied, aspect of the mechanical response of materials upon external loading [1–4]. In a typical experiment, a sample of material is deformed at a constant rate and the point at which the stress suddenly drops, in a marked departure from reversible elastic behavior, is identified with the yield point [3]. No theory, starting from rigorous thermodynamic principles (e.g., shape independence of equilibrium free energy densities) [5], has been derived that predicts the yield point of a real solid. Detailed phenomenological models, on the other hand, have been extremely useful for this purpose [1,2]. All existing work on irreversible mechanical processes in crystalline solids ultimately connects to the static and dynamic properties of lattice defects viz. dislocations [6]. These are known to interact with each other as well as with impurities, grain boundaries, pinning sites, etc. [1,2]. However, a quantitative understanding of the collective behavior of large accumulations of dislocations, readily observed in large scale atomistic computer simulations [7] and expected to play a key role in yielding, remains elusive. Various models, such as the thermodynamic dislocation theory [8], based on depinning [9] or unjamming [10] of dislocations, have been proposed. Yielding is also viewed as a dynamical critical phenomenon [4,11]. Indeed, scale-free avalanches and intermittent phenomenon have been observed to accompany yielding [12]. A nonzero initial dislocation concentration is indispensable for most known models of yielding.

One of the most intriguing features of yield phenomena in solids is the history dependence of the yield point. It is known that the yield point stress σ_Y is a weak function of the rate $\dot{\varepsilon}$ by which the solid is deformed *prior* to the commencement of irreversibility [1,2,13–15]. This relation is expressed as a phenomenological power law $\sigma_Y \sim (\dot{\varepsilon})^m$. The rather nonuniversal, strain rate sensitivity (SRS) exponent m varies over a wide range 0.006–0.06 depending on many factors, such as the nature of the solid and the size of the specimen, as well as $\dot{\varepsilon}$ itself. The weak dependence on $\dot{\varepsilon}$ and the small value of m may be the reason why this aspect of yielding has not received the attention that it deserves. Usually very low (or very high) values of fitted exponents point to a gap in our understanding of the underlying physics, hinting that the actual relation may not, in fact, be a power law.

In this Letter, we show that, at least for one special class of materials, viz. nearly defect-free crystals, this apprehension is true. We derive and test a relation between the yield strain ε^* defined simply as σ_Y/K, where K is the appropriate elastic modulus, and $\dot{\varepsilon}$. This relation is not a power law and involves instead an essential singularity. It explains experimental and computer simulation data over a very wide range of temporal scales.

Our primary idea emerges from the following, somewhat surprising, fact. Even though σ_Y appears to decrease only slightly when $\dot{\varepsilon}$ decreases by a large amount, crystalline solids are guaranteed to yield at infinitesimal stresses when deformed at vanishing rates. This conclusion follows from very general considerations starting with the exact result that the free energy density of any material, made up of entities interacting with short ranged forces, cannot depend on the shape of the boundary [5,16]. This seemingly pedantic issue can be converted into a quantitative calculation by showing that, even at vanishingly small strains, an

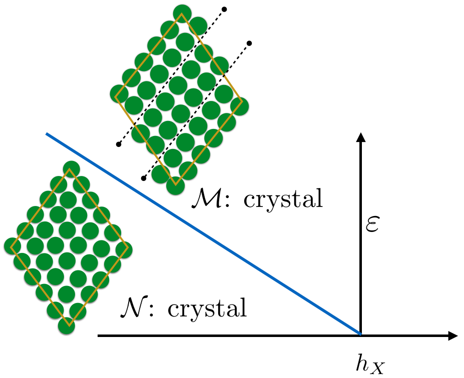

FIG. 1. Schematic phase diagram and $\mathcal{N}$, $\mathcal{M}$ phases [18] corresponding to the $d = 2$ square lattice, in the $h_X - \varepsilon$ plane (see text). The blue line is the first-order phase boundary. The dotted lines show slip planes in the $\mathcal{M}$ phase.

initially dislocation-free ideal crystal may nucleate regions, or bubbles, inside which atoms rearrange to eliminate stress [17]. These stress-free bubbles are embedded inside the deformed and stressed solid and bounded by an interface containing an array of defects. Since the volume energy contribution of the bubble is always negative, the surface energy cost from the defect array can be compensated as the size of the bubble increases. A rigid solid is therefore always metastable at infinitesimal deformation, although the relaxation time for bubble nucleation diverges as $\varepsilon \to 0$.

Recently, by introducing a fictitious field h_X, which penalizes atomic rearrangements, some of us showed that further insight may be obtained into this problem [18] (see also Supplemental Material [19]). One can then analyze the problem in the full $h_X - \varepsilon$ plane in the thermodynamic limit and let $h_X \to 0$ at the end. The analysis in Ref. [18] concludes that there exists an equilibrium first-order transition between two kinds of ideal crystals, say $\mathcal{N}$ and $\mathcal{M}$ in the $h_X - \varepsilon$ plane with a phase boundary that extrapolates to $\varepsilon = 0$ at $h_X = 0$ (Fig. 1). The two crystals have identical symmetry but differ in the way they respond to deformation. The $\mathcal{N}$ crystal produces an internal restoring stress upon deformation, while the $\mathcal{M}$ crystal undergoes spontaneous rearrangements (e.g., by slipping) to accommodate the deformation at zero stress. The latent heat released during the $\mathcal{N} \to \mathcal{M}$ transition is just the elastic energy stored in the rigid $\mathcal{N}$ phase. For any $h_X = 0$ and $\varepsilon \neq 0$, the $\mathcal{N}$ phase is metastable and decays to the stable $\mathcal{M}$ phase by bubble nucleation. Then, for a given ε, the free energy $\mathcal{F}$ of a spherical bubble of $\mathcal{M}$ phase, with radius R and in d dimensions, is given within a linear approximation by $\mathcal{F} = -\frac{1}{2}K\varepsilon^2 V_1 R^d + \gamma_s S_1 R^{d-1}$. Here, K is the elastic modulus and $V_1 = \pi^{(d/2)}/\Gamma[(d/2)+1]$ and $S_1 = dV_1$ are the volume and surface area of a d-dimensional unit sphere, respectively. The equilibrium surface energy γ_s is rigorously defined only at coexistence at the first-order boundary [28]. Extremizing $\mathcal{F}$ gives the free energy barrier $\Delta\mathcal{F}$, at a critical value of the radius $R = R_c$, as

$$\Delta\mathcal{F} = [2(d-1)]^{d-1} V_1 \gamma_s^d K^{-(d-1)} \varepsilon^{-2(d-1)}.$$

Up to now, we have considered the limit $\dot{\varepsilon} \to 0$. At a finite strain rate, the strain depends on time t, i.e., $\varepsilon = \dot{\varepsilon}t$, and thus the free energy barrier becomes time dependent, $\Delta\mathcal{F} = \Delta\mathcal{F}(\dot{\varepsilon}t)$. The time τ_{FP}, at which the system yields, can be understood as a (first passage) barrier crossing time and is associated with a critical strain $\varepsilon^* = \dot{\varepsilon}\tau_{\mathrm{FP}}$. Thus, it is given by [29] $\tau_{\mathrm{FP}} \equiv \varepsilon^*/\dot{\varepsilon} = \tau_0 \exp[\beta\Delta\mathcal{F}(\varepsilon^*)]$, where β is the inverse temperature and τ_0, the inverse "attempt frequency," is a microscopic timescale that is independent of the barrier. The equation for τ_0 is a self-consistency equation for ε^* [30]. Here, $\beta\Delta\mathcal{F} = \alpha(\varepsilon^*)^{-2(d-1)}$ with $\alpha = \beta[2(d-1)]^{d-1} V_1 \gamma_s^d K^{-(d-1)}$, and thus there is an essential singularity in the dependence of τ_{FP} on ε^*.

In Ref. [18], a similar equation derived for $d = 2$ was solved numerically. Here we show that it is possible to solve the relation analytically and obtain a closed form expression for ε^* as a function of $\dot{\varepsilon}$. Defining $x = 2(d-1)\alpha(\varepsilon^*)^{-2(d-1)} = 2(d-1)\beta\Delta\mathcal{F}$, we can write $2(d-1)\alpha(\dot{\varepsilon}\tau_0)^{-2(d-1)} = x\exp(x)$. This can now be inverted using the Lambert W function [31]: if $xe^x = f(x) = y$, then $x = f^{-1}(y) = W(y)$. We thus have finally

$$\varepsilon^* = \left(\frac{W[2(d-1)\alpha(\dot{\varepsilon}\tau_0)^{-2(d-1)}]}{2(d-1)\alpha}\right)^{-1/[2(d-1)]}. \qquad (1)$$

This is the desired closed form expression for the yield strain, a smooth and monotonically increasing function of $\dot{\varepsilon}$. Note that there are only two free parameters in Eq. (1): τ_0, a characteristic time and γ_s (or alternately α), a characteristic energy. Also, wherever Eq. (1) is valid, τ_0 is a constant. In Ref. [18], this was tested for a $d = 2$ perfect, initially defect-free Lennard-Jones solid [11]. First, γ_s was computed at phase coexistence using advanced Monte Carlo sampling methods for nonzero h_X and ε, with the final $h_X \to 0$ limit taken by numerical extrapolation. Equation (1) was then solved for τ_0 using ε^* obtained from molecular dynamics (MD) at various $\dot{\varepsilon}$. The results showed that this theory was excellent for $\beta\Delta\mathcal{F} \gtrsim 1$.

By construction, of course, Eq. (1) is thermodynamically consistent. Can it also explain real experimental data in $d = 3$? We check this here by reanalyzing a representative, though not exhaustive, set of available data [32–44]. The systems chosen consist of nearly defect-free ductile single crystals of high purity elemental metals like Cu [32,33] and Ni [34–37]. We include not only nanopillars [32] and nanowires [34] but also polycrystalline samples [33,35–37] with ultrafine grains. Finally, we also reanalyze the results of large scale MD simulations [38–43] of Au, Cu, and Ni single crystals within our framework. Ideally, τ_0 and γ_s should depend only on the material, but in practice factors like the experimental protocol, e.g., tensile or indentation, sample size, proximity to free surfaces, prestress, frozen-in

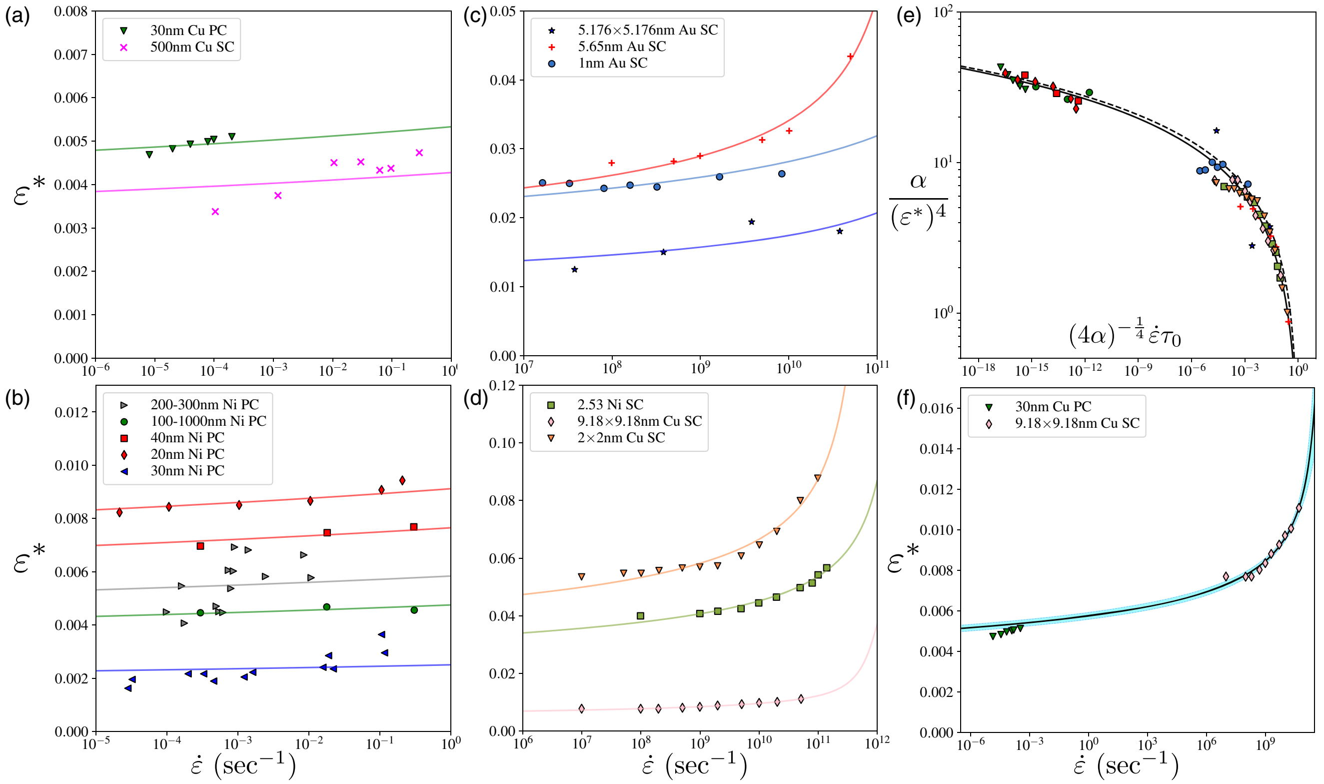

FIG. 2. (a),(b) Nonlinear least-square fits (solid lines) of Eq. (1) to experimental results (symbols) on Cu and Ni samples, both single crystal (SC) and ultrafine grained polycrystalline (PC); see text and Table I for details. (c),(d) Same as (a) and (b) but for MD simulations of Au, Cu, and Ni. (e) Plot of the data in (a)–(d) using scaled variables [with the same symbols as in (a)–(d)] showing collapse onto the single master curve (1) (solid black line). (Dashed) Approximate asymptotic form (2). Three experimental datasets with excessive (>50%) scatter have been omitted. (f) A log-linear plot of Eq. (1) shows that the parameters γ_s and τ_0, obtained from MD of Cu [43], are able to predict the results of experiments [33] performed 10^{-12} times more slowly. The light blue band on the solid line shows the uncertainty of our prediction at 95% confidence [from fit (2), see Table I].

defects, etc., may be important, causing variation in our fit parameters.

For all these systems (see Supplemental Material [19] for a summary), reasonably accurate yield point stresses σ_Y (under either compressive nanoindentation or tensile loads) have been reported as a function of $\dot{\varepsilon}$. The original authors of Refs. [32–44] had fitted the data to power laws and extracted widely different values of the SRS exponent m. We first digitize the available data and divide the quoted yield stress σ_Y by the appropriate elastic modulus of the bulk solid [45] to obtain ε^*; this is a dimensionless quantity that is easier to compare among the different datasets. Fits to Eq. (1) then give values of α and τ_0. The fits are shown in Figs. 2(a)–2(d); Table I lists the corresponding parameter values. Equation (1) describes the data rather well, with some of the experimental data showing, understandably, more scatter than the MD ones. The fitted values are also remarkably consistent across the different sets of data, apart from a few exceptions coming mainly from MD studies of very small crystals, where finite size effects may be strong. Most values for τ_0 (~10–100 fs) are in-line with typical dislocation nucleation times reported earlier [46], as well as our estimates based on MD simulations [18]. The interfacial energy γ_s (~10 mJ/m^2), computed from α, is also consistent with that of low energy solid interfaces (e.g., stacking faults) [1]. The assumption of homogeneous nucleation of the stress-free $\mathcal{M}$ state, implicit in Eq. (1), does not affect the qualitative nature of the solution as long as $\gamma_s > 0$. Systematic variations of γ_s are difficult to resolve, however, given the scatter in the original data.

We next look in more detail at the strain rate dependence of the yield strain predicted by Eq. (1). For small strain rates, where the argument of $W(y)$ becomes large, one can use the asymptotic behavior $W(y) = (\ln y) - \ln(\ln y) + \cdots$. The yield strain thus has a logarithmic rather than power law dependence on strain rate, which rationalizes the small effective SRS exponents found previously. Keeping only the leading term $W(y) \approx \ln y$ gives, in $d = 3$,

$$\alpha(\varepsilon^*)^{-4} = W[4\alpha(\dot{\varepsilon}\tau_0)^{-4}]/4 \approx \ln[(4\alpha)^{1/4}(\dot{\varepsilon}\tau_0)^{-1}]. \quad (2)$$

These relations (solid and dashed lines, respectively) are plotted along with experimental and simulation results in Fig. 2(e), showing collapse of datasets covering, altogether,

TABLE I. The results of nonlinear least-squares fits of Eq. (1) to τ_0 and α. Details of the method, i.e., experimental (Expt.) or MD, compressive or tensile loading and the materials considered, are also given along with the references (column 1). The values of K are taken from [45] (Cu, Ni) and [38] (Au). Except where mentioned otherwise, data correspond to room temperature (27 °C). The interfacial energy γ_s is computed from α and K. The last two rows give parameters obtained by fitting the same data using two different methods. Typical errors from MD are $\sim \pm 20\%$ for τ_0 and $\lesssim \pm 10\%$ for γ_s; error estimates for experimental data are higher due to more scatter and a smaller range of $\dot{\varepsilon}$ (see [19]).

Reference	Material	Method	Details	K (GPa)	τ_0 (fs)	γ_s (mJ/m^2)	$\alpha^{\frac{1}{4}}$
[32]	Cu	Expt.: compressive	≈500 nm diameter single crystal nanopillars	130	39.23	1.826	0.0096
[33]	Cu	Expt.: tensile	Polycrystal with ≈30 nm grains	130	37.59	2.801	0.0120
[34]	Ni	Expt.: tensile	≈200–300 nm diameter nanowires	200	57.55	4.207	0.0131
[35]	Ni	Expt.: compressive	Polycrystal with ≈100–1000 nm grains	200	86.82	3.138	0.0106
[35]	Ni	Expt.: compressive	Polycrystal with ≈40 nm grains	200	34.12	6.040	0.0173
[36]	Ni	Expt.: compressive	Polycrystal with ≈20 nm grains	200	44.42	7.662	0.0206
[37]	Ni	Expt.: tensile	Polycrystal with ≈30 nm grains	200	35.43	1.341	0.0056
[38]	Au	MD: tensile	5.176×5.176 nm nanowire	188	24.05	9.536	0.0251
[39]	Au	MD: tensile (0°C)	5.65 nm diameter nanowire	188	341.34	18.367	0.0420
[40]	Au	MD: tensile (-269°C)	1 nm diameter nanowire	188	11.02	4.658	0.0432
[41]	Ni	MD: tensile	2.53 nm diameter nanowire	200	63.87	35.141	0.0648
[42]	Cu	MD: tensile	2×2 nm nanowire	130	312.84	39.718	0.0880
[43]	Cu (1)	MD: tensile	9.18×9.18 nm nanowire	130	36.59	3.051	0.0128
[43]	Cu (2)	MD: tensile	9.18×9.18 nm nanowire	130	32.97	3.100	0.0129

15 orders of magnitude in timescale. Both forms, involving either $W(y)$ or $\ln(y)$, describe the data extremely well in this range. This makes sense as with $\tau_0 \sim 30$ fs (3×10^{-14} s) our data lie in the range $(\dot{\varepsilon}\tau_0)^{-1} \sim 10^3$–$10^{19}$ and $4\alpha(\dot{\varepsilon}\tau_0)^{-4} \sim 10^4$–$10^{68}$ is indeed typically large. The dimensionless nucleation barrier $\beta\Delta\mathcal{F}$ shown on the y axis is considerably larger for the experiments than for the MD data, where the barrier heights are at the limit of validity of our theory.

We find it remarkable that Eq. (1) is able to explain such a large and varied set of data. It is particularly surprising that even data from ultrafine grained polycrystalline samples appear to be describable within our picture. In these systems, there are no dislocations within the grains since the grain boundaries function as dislocation sinks [1,2]. The entire sample therefore behaves as an ensemble of nearly independent crystallites.

Defect-free nanometer sized crystals are an important group of materials that are fast gaining technological relevance [47]. Our idea, based on a simple, classical picture of nucleation of stress-free bubbles of $\mathcal{M}$ inside $\mathcal{N}$ describes yielding and SRS in these systems. In our picture, dislocations occur only at the interface between the $\mathcal{N}$ and $\mathcal{M}$ phases in order to match the two lattices. Their energy is *subsumed* into γ_s and they are constrained (by definition) to always move together with the interface, where the local stress gradient is large [48]. Interfaces exist either at equilibrium phase coexistence or when the growth of the $\mathcal{M}$ phase is arrested due to kinetic effects. In real situations ($h_X = 0$), phase coexistence does not occur for nonzero deformation in the thermodynamic limit [18]. Observed dislocations are associated, therefore, with nonequilibrium, or kinetically arrested, configurations. In experiments, dislocations are far easier to see directly, using electron microscopy [1], than the stress interface itself. The notion of a first-order transition has, therefore, escaped attention.

The two parameters of our theory, τ_0 and γ_s (or α), can be obtained by either fitting yielding data, as is done here, or from precise, finite size-scaled numerical computations as in Ref. [18]. We emphasize that our nucleation theory respects rigorous thermodynamic constraints [5] and at the same time offers a way to successfully predict "real-world" experimental data at low strain rates from simulations performed at much higher rates. We demonstrate this in Fig. 2(f) by using parameters obtained from the MD results of Ref. [43] to predict accurately the outcome of the experiments of Ref. [33], which are separated from the simulation data by 12 orders of magnitude in time. This is especially remarkable because an alternative form for $\Delta\mathcal{F}$ derived in Ref. [17], without considering a first-order phase transition, obtains qualitatively similar $\varepsilon^*(\dot{\varepsilon})$ but physically unreasonable yield points [19].

Finally, we believe that the work reported here allows us to reiterate an interesting perspective on the phenomenon of yielding. Despite the success of Eq. (1) in explaining such an extensive set of data, it is not a "theory for everything". Yielding in initially defect-free crystals is metastability controlled, with the large barrier $\Delta\mathcal{F} > k_B T$ between the distinct $\mathcal{N}$ and $\mathcal{M}$ phases playing a dominant role in determining the dynamical response of the solid to external load. On the other hand, in a solid with a large number of frozen-in defects, the barrier may be much smaller and may even vanish, $\Delta\mathcal{F} \to 0$. There is also a possibility of many coexisting, relatively shallow minima in $\mathcal{F}$ appearing [19]. If barriers disappear, the nucleation route could be replaced by spinodal decomposition [11], with accompanying critical-like behavior, avalanches, etc., as is observed [12].

In this regime, the yielding process would be instability controlled [49]. An actual dynamical critical point is also possible [4,9,10]. The situation in amorphous solids may be similar; here yielding is known to be preceded by the percolation of mobile clusters and critical behavior is often noticed [50]. Our theory cannot predict yielding behavior in this regime, where one commonly obtains either zero or even negative values of the SRS exponent m [51]. Is it possible to develop a theory of yielding with more general validity, capable of describing both the nucleation and critical regimes? We hope that our Letter generates enthusiasm for pursuing this direction of future research.

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