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# Yielding and tensile behavior of nanocrystalline copper

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ABSTRACT

Two theoretical frameworks are calibrated to capture the inverse Hall–Petch phenomenon, as well as the stress–strain response of nanocrystalline Cu during plastic flow. The first framework employed is gradient plasticity enhanced with an interface energy term, the use of which is dictated by the fact that at the nanoscale interfaces play a dominant role in the mechanical behavior of nanocrystalline materials. The second formulation involves a simplified gradient plasticity model without an interface energy term coupled with wavelet analysis.



## 1. Introduction

Hall [1] and Petch [2] observed in the early 1950s that the yield stress ($\sigma_y$) required for continuous plastic deformation increased with decreasing grain size ($d$) leading to the well-known Hall–Petch relation ($\sigma_y \sim 1/\sqrt{d}$). Various dislocation models were used to predict that the hardness of nanomaterials would be higher due to the fact that grain boundaries are obstacles to intragranular dislocation motion (since at the nanoscale ~1/3 of the total volume fraction is attributed to grain boundary area, a high strength was expected). Experiments, however, involving very small grain sizes (~10 nm) established the inverse or "anomalous", Hall–Petch behavior [3–5]. This phenomenon is of great interest, especially in the micro-electronics industry, since in order to achieve further miniaturization of devices, the deformation at the nanoscale must be understood.

## 2. Outline of the two gradient plasticity models

### 2.1. Gradient plasticity model with interface energy term

In order to explicitly account for interfaces, Aifantis and Willis [6,7] added an interface energy term $\varphi$ in gradient plasticity [8,9], as

$$\Psi(\varepsilon_{ij}, \varepsilon^p_{ij}) = \int_\Omega [L_{ijkl}(\varepsilon_{ij} - \varepsilon^{\mathrm{p}}_{ij})(\varepsilon_{kl} - \varepsilon^{\mathrm{p}}_{kl}) + V(\varepsilon^{\mathrm{p}}_{ij}, \varepsilon^{\mathrm{p}}_{ij,k})]\ \mathrm{d}\Omega + \int_\Gamma \varphi(\varepsilon^{\mathrm{p}}_{ij})\,\mathrm{d}\Gamma, \quad (1)$$

where $L_{ijkl}$ are the components of the elastic stiffness tensor, $\varepsilon$ is the total strain, $\varepsilon^{\mathrm{p}}$ is the plastic strain, and $V$ is the plastic potential of the grains. By performing the principle of virtual work ($\delta\Psi = 0$), for prescribed displacements, and defining the conjugate variables $\sigma_{ij} = \partial U/\partial\varepsilon_{ij}$, $s_{ij} = \partial U/\partial\varepsilon^{\mathrm{p}}_{ij}$ and $\tau_{ijk} = \partial U/\partial\varepsilon^{\mathrm{p}}_{ij,k} = \partial V/\partial\varepsilon^{\mathrm{p}}_{ij,k}$ the following equilibrium, boundary, and interface conditions are obtained [7]:

$$\sigma_{ij,j} = 0 \quad \text{and} \quad s_{ij} - \tau_{ijk,k} = 0, \quad \text{in } \Omega; \quad (2a)$$

$$\tau_{ijk} n_k = 0 \quad \text{and} \quad u_i = u^o_i, \quad \text{on the outer boundary } \partial\Omega; \quad (2b)$$

$$\left[\sigma_{ij} n_j\right] = 0 \quad \text{and} \quad \left[\tau_{ijk} n_k\right] = \partial\varphi/\partial\varepsilon^{\mathrm{p}}_{ij}, \quad \text{across the grain boundary } \Gamma. \quad (2c)$$

The brackets [..] denote the jump of the enclosed quantity across the interface. It should be noted that the second expression in Eq. (2c) suggests that the gradient of the plastic strain is discontinuous across interfaces and, hence, allows interfaces to follow their own yield behavior.

Various material types can be modeled by the above formulation, by calibrating appropriately the interface ($\varphi$) and plastic ($V$)

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potentials which can be identified as [10]:

$$\varphi(\varepsilon^{\mathrm{p}}) \equiv \gamma\varepsilon^{\mathrm{p}}, \tag{3}$$

$$V(\varepsilon^{\mathrm{p}}, \varepsilon^{\mathrm{p}}_{,x}) = \sigma_0|\varepsilon^{\mathrm{p}}| + \frac{1}{2}\beta\ell^2(\varepsilon^{\mathrm{p}}_{,x})^2, \tag{4}$$

where $\gamma$ is an interface parameter indicating in a way the strength of the interface, $\sigma_0$ is the grain interior yield stress, $\beta$ is the hardening modulus, and $\ell$ is the internal length, a characteristic material parameter required for dimensional consistency coming into play in all gradient theories. It should be noted that the expressions for $(\varphi)$ and $(V)$ defined above allow for perfect plasticity to take place in the grain boundary and boundary layer, since these expressions allow for perfect plasticity to be attained, once yielding occurs.

Deformation at the nanoscale is governed through grain boundary rotation and sliding, while deformation in the grain interior is very difficult to initiate (the dislocation core may be comparable to the grain size) and, hence, dislocation formation and motion in grains may be impossible. Therefore, grains are assumed to experience no plastic strain and upon deformation the material is split into three regions: a plastically deforming grain boundary (located at $x=0$), a plastically deforming boundary layer of length $2L^*$, since the grain material adjacent to the grain boundary also yields, and an elastic grain interior.

Therefore, to obtain the plastic response in the material we must consider the plastic zone that lies between $-L^* \le x \le L^*$. Inserting Eq. (4) in Eq. $(2a)_2$ allows the differential equation in the perfectly plastic nanocrystal to be obtained as

$$\ell^2\frac{\mathrm{d}^2\varepsilon^{\mathrm{p}}_i}{\mathrm{d}x^2} + \frac{\bar{\sigma}-\sigma_0}{\beta} = 0 \Rightarrow \varepsilon^{\mathrm{p}}_i = -\frac{1}{2}\frac{\bar{\sigma}-\sigma_0}{\beta\ell^2}x^2 + A_i x + B_i, \tag{5}$$

where $i=1,2$ denotes the areas to the right and left of the interface, respectively. In order, therefore, to obtain the plastic strain distribution in the boundary layer we need to find the constants $A_1$, $B_1$, $A_2$, $B_2$, as well as the length of the boundary layer $L^*$. The following boundary layer ($x=\pm L^*$) and grain boundary ($x=0$) conditions are used:

$$\varepsilon^{\mathrm{p}}_1(L^*) = 0, \qquad \varepsilon^{\mathrm{p}}_2(L^*) = 0, \qquad \varepsilon^{\mathrm{p}}_1(0) = \varepsilon^{\mathrm{p}}_2(0),$$

$$[\tau] = \gamma \Rightarrow \left.\frac{\mathrm{d}\varepsilon^{\mathrm{p}}_1}{\mathrm{d}x}\right|_{x=0^-} - \left.\frac{\mathrm{d}\varepsilon^{\mathrm{p}}_2}{\mathrm{d}x}\right|_{x=0^+} = \frac{\gamma}{\beta\ell^2},$$

$$\tau(L^*) = \beta\ell^2\left.\frac{\mathrm{d}\varepsilon^{\mathrm{p}}_2}{\mathrm{d}x}\right|_{x=L^*} = 0. \tag{6}$$

Eq. $(6)_a$ and Eq. $(6)_b$ ensure continuity of the plastic strain between the purely elastic grain and the plastic boundary layer; Eq. $(6)_c$ ensures continuity of the plastic strain across the grain boundary; Eqs. $(6)_d$ and $(6)_e$ correspond to the boundary conditions given in Eqs. $(2c)_2$ and $(2b)_1$.

Solving Eq. (6) with $\varepsilon^{\mathrm{p}}$ given by Eq. (5) allows the constants of the plastic strain, as well as the thickness of the boundary layer that deforms along with the grain boundary to be obtained:

$$L^* = \frac{\gamma}{2(\sigma_0-\sigma)}, \qquad A_1 = \frac{\gamma}{2\beta\ell^2}, \qquad B_1 = \frac{\gamma^2}{8\beta\ell^2(\sigma_0-\sigma)},$$

$$A_2 = -A_1, \qquad B_2 = -B_1. \tag{7}$$

The boundary layer thickness $L^*$, however, is a fraction of the grain size $d$, hence $L^* = ad$, where $a$ is a constant. According to Hall–Petch, the yield stress $\sigma_0$ of the grain can be set equal to $k/\sqrt{d}$, where $k$ is the Hall–Petch slope, hence Eq. (7) takes the form:

$$L^* = \frac{\gamma}{2(\sigma_0-\sigma)} \Rightarrow \sigma = \frac{k}{\sqrt{d}} - \frac{\gamma}{2ad}. \tag{8}$$

Eq. (8) depends on the deforming boundary layer thickness and on the interface parameter $\gamma$, hence it gives the stress required in order to initiate deformation in the grain boundaries. But since deformation at the nanoscale is initiated and governed through grain boundary deformation, the stress given by Eq. (8) can be identified with the yield stress of the nanocrystal. Eq. (8) can, therefore, be used to model experimental data which show how the yield stress in nanomaterials depends on their grain sizes.

Furthermore, the above gradient plasticity model can be used to obtain the stress required for continuous deformation within the boundary layer. This is done by substituting the various constants of Eq. (7) in Eq. (5) and integrating between $-L^* \le x \le L^*$. The mean plastic strain in the grain boundaries is then given by

$$\bar{\varepsilon}^{\mathrm{p}} = \frac{(\sigma_0-\sigma)(L^*)^2}{6\beta\ell^2} + \frac{\gamma^2}{8\beta\ell^2(\sigma_0-\sigma)} - \frac{\gamma L^*}{4}, \tag{9}$$

which in turn can be solved for $\sigma$ providing the following stress–strain relation in the boundary layer (for convenience we let $\sigma \equiv \sigma_{\mathrm{b}}$).

$$\sigma_{\mathrm{b}} = \sigma_0 - \frac{3}{L^*}\left(\frac{\gamma}{4} + \frac{\beta\ell^2}{L^*}\bar{\varepsilon}^{\mathrm{p}}\right) + \frac{\sqrt{3}}{(L^*)^2}\sqrt{48(\beta\ell^2\bar{\varepsilon}^{\mathrm{p}})^2 + \gamma L^*(24\beta\ell^2\bar{\varepsilon}^{\mathrm{p}} - \gamma L^*)}. \tag{10}$$

To obtain the overall stress–strain response, one must also account for the deformation taking place in the grains, since it is assumed that with continuous deformation the grain interior will eventually also yield. A rule of mixtures is used, by which the overall yield stress $\bar{\sigma}_y$ depends on the flow stress in the grains $(\sigma_{\mathrm{g}})$ and the flow stress in the boundary layer $(\sigma_{\mathrm{b}})$, as $\bar{\sigma}_y = (1-f)\sigma_{\mathrm{g}} + f\sigma_{\mathrm{b}}$, where $f = 2L^*/d$ is the volume fraction of the boundary layer. To allow for eventual perfect plasticity a Voce-type model is used $\sigma_{\mathrm{g}} = \sigma_{\mathrm{f}} + (\sigma_{\mathrm{s}} - \sigma_{\mathrm{f}})\tanh[h_0\bar{\varepsilon}/(\sigma_{\mathrm{s}} - \sigma_{\mathrm{f}})]$ with $\sigma_{\mathrm{s}} = \sigma_{0\mathrm{s}} + b/\sqrt{d}$ [11], where $\sigma_{\mathrm{f}}$ denotes the friction stress, $\sigma_{\mathrm{s}}$ the saturation stress and $h_0$ the initial hardening modulus. Therefore, the stress–strain relation for the deforming nanocrystal is given as

$$\bar{\sigma} = (1-f)\sigma_{\mathrm{g}} + f\sigma_{\mathrm{b}} = (1-2a)\left[\sigma_{\mathrm{f}} + (\sigma_{\mathrm{s}} - \sigma_{\mathrm{f}})\tanh\left(\frac{h_0\bar{\varepsilon}}{\sigma_{\mathrm{s}} - \sigma_{\mathrm{f}}}\right)\right] + 2a\sigma_{\mathrm{b}}, \tag{11}$$

where $\sigma_{\mathrm{b}}$ is given in Eq. (10). Therefore, Eq. (11) can be used to predict how grain size affects the overall flow stress in materials.

### 2.2. Gradient plasticity model utilizing wavelet analysis

Although the model presented in the previous subsection is rather efficient in capturing interfacial effects, the key constants have not yet yielded to precise physical interpretation. The higher-order $(\tau)$ boundary condition requirements, arising from the use of the strain gradient, is another physical drawback, which was circumvented in recent works of Aifantis and his co-workers [12] through the use of wavelet analysis as follows. The initial gradient constitutive equation for the flow stress has the form [12–14]:

$$\sigma = \kappa(\varepsilon^{\mathrm{p}}) - c\nabla^2\varepsilon^{\mathrm{p}}, \tag{12}$$

with $\kappa(\varepsilon^{\mathrm{p}})$ denoting the "homogeneous" part of the flow stress and $c$ the so-called gradient coefficient related to the internal length of the previous gradient theory as $c \sim \ell^2$. As proposed in [12–14], the wavelet representation $\delta_{\mathrm{s}}$ of the $\delta$-function multiplied by a factor $s_0$, given by $s_0\delta_{\mathrm{s}}(x) = (s_0/2s\sqrt{\pi})\exp(-x^2/4s^2)$, can be used to express a shear band solution [8,9] or localized distribution of strain $\varepsilon$ of the form $\varepsilon = \varepsilon_\infty + s_0\delta_{\mathrm{s}}(x)$, with $\varepsilon_\infty$ denoting the uniform strain