

1 **Validation of a phenomenological strain-gradient plasticity theory**

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7 **Validation of a phenomenological strain-gradient plasticity theory**

8 Strain-gradient plasticity theories have been developed to account for the size
9 effect in small-scale plasticity in metals. However, they remain of limited use in
10 engineering, for example in standards for nanoindentation, because of their
11 phenomenological nature. In particular, a key parameter, the characteristic length,
12 can only be determined by fitting to experiment. Here it is shown that the
13 characteristic length in one such theory derives directly from known quantities
14 through fundamental dislocation physics. This explains and validates the theory for
15 use in engineering.

16 Keywords: plasticity of metals; strengthening mechanisms; strained layers;
17 dislocations; strain-gradient theory; critical thickness theory.

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20 The increase in strength (the size effect) when dislocation-mediated plasticity is restricted
21 to small volumes has been extensively documented experimentally over the past 60 years
22 [see, e.g., 1–13]. It is an important effect in many technologies from metallurgy to
23 semiconductors, yet it is not fully understood [12, 14]. In micromechanics, many loading
24 conditions impose a plastic strain gradient, and so theories in which the strain gradient
25 plays a central role have been developed [3–6, 15–19]. In contrast, in semiconductor
26 technology, Matthews critical thickness theory has been largely accepted to explain and
27 predict the effect in terms simply of the size – stronger when smaller [20–22]. The
28 strain-gradient theories have not been comprehensively embraced [23], because of
29 ambiguities about the underlying physics and about the parameters – in particular, the
30 characteristic length – which enter into these theories. One consequence is that there are
31 no satisfactory international standards for comparing nanoindentation data, in which the

32 size effect plays an important role, with macroscopic indentation data. Here it is shown
33 that the Fleck-Hutchinson strain-gradient theory [4, 17–19, 23] follows mathematically
34 and physically directly from critical thickness theory [20–22]. The strain-gradient theory
35 fits experiment well, but with the characteristic length as a free fitting parameter. This
36 phenomenological parameter is here derived from known physical quantities *via* critical
37 thickness theory. The derivation and the associated re-interpretation validate the strain-
38 gradient theory for use in practical engineering contexts, as an approximation that
39 expresses a non-local property as a local property.

40 Increases in strength (the size effect) due to boundaries imposed on dislocation-
41 mediated plasticity on scales up to tens of microns have been presented for
42 nanoindentation [3,5], thin wires in torsion [4, 9, 10], thin foils in bending [6, 8], and for a
43 large variety of still smaller structures down to sub-micron sizes mostly created by
44 focused ion-beam (FIB) milling [e.g. 7, 11, 13]. Microstructural constraints giving rise to
45 the size effect include sub-grain boundaries [2] and grain boundaries (the Hall-Petch
46 effect) [1, 12]. Pseudomorphic (strained-layer) heteroepitaxial crystal growth is another
47 key example [20–22]. In many of these situations, plastic strain gradients are necessarily
48 or optionally present, and there is widespread agreement that in such situations the size
49 effect can be attributed to the strain gradient.

50 In formal continuum mechanics, to set up a strain-gradient plasticity theory
51 (SGP), the stress is not only a function of plastic strain ε_p , but also a function of its spatial
52 gradient $\ell \varepsilon'_p = \ell d\varepsilon_p / ds$ where s is position and the characteristic length ℓ is introduced
53 to give a dimensionless quantity [16–19]. Where a physical interpretation is called for,

54 appeal is made to the geometrically-necessary dislocations (GNDs) [3] which in a
55 crystalline material are necessarily associated with plastic strain gradients [15]. Values of
56 ℓ are found from fitting to experiment (see Fig.1). The major problem for such strain-
57 gradient theories is to give a reasonable physical interpretation of the values of ℓ that
58 result. There have been many proposals. See [24] for a recent discussion and a new
59 proposal.

60 Evans and Hutchinson [23] gave an appraisal of SGP theories, for brevity
61 confined to the Nix-Gao (NG) theory [3] and the Fleck-Hutchinson (FH) theory [4, 17,
62 19]. These two theories illustrate adequately both the successes of SGP theories in
63 general, and their difficulties. The successes lie in the good fits to experimental data that
64 these theories give. The major difficulty is that, fitting to experimental datasets for soft
65 metals, the NG theory gives characteristic lengths $\ell_{\text{NG}} \sim 25\text{mm}$, and the FH theory gives
66 $\ell_{\text{FH}} \sim 5\mu\text{m}$. **Neither is characteristic of any length scale experimentally observed in the**
67 **specimens, whether structural or microstructural.** For this reason, and because of the lack
68 of any explicit connection between the theories and dislocation dynamics, Evans and
69 Hutchinson noted that strain-gradient theories have not been comprehensively embraced
70 [23].

71 Here, the FH characteristic length is derived from critical thickness theory. This
72 reveals a previously unsuspected link between the two theories. In particular, it provides
73 the explicit connection between the FH theory and the physics of dislocation dynamics
74 that was previously lacking. It thereby validates the use of the FH theory for prediction in
75 engineering applications (with due attention to the approximations revealed in it).

76 It is not necessary to use a full derivation of strain-gradient theory. We take Evans
 77 and Hutchinson [23] as a starting-point. They define an effective stress σ which is a
 78 function of the yield stress and the plastic strain, $\sigma = \sigma_Y f(\varepsilon_P)$. For the FH theory, they
 79 state as a premise that the plastic work per unit volume may be written as

$$80 \quad U_P = \sigma_Y \int_0^{E_P} f(\varepsilon_P) d\varepsilon_P \quad (1)$$

81 The upper integral limit E_P brings in the effect of the strain gradient ε'_P by the definition

$$82 \quad E_P = \varepsilon_P + \ell_{FH} \varepsilon'_P \quad (2)$$

83 This is a specific form of the generalized effective plastic strain E_P [19]. Consider an
 84 object of size h , average plastic strain $\bar{\varepsilon}_P$ and average plastic strain gradient $\bar{\varepsilon}'_P = c\bar{\varepsilon}_P/h$
 85 with $c \sim 1$, and with perfect plasticity, $f(\varepsilon_P) = 1$. From equation (1), the average flow
 86 stress is

$$87 \quad \bar{\sigma} = \sigma_Y \left(1 + \frac{c\ell_{FH}}{h} \right) \quad (3)$$

88 This is equation (11) of Ref.23. Note that the strengthening is independent of ε_P . The
 89 strain gradient increases the yield strength but not the rate of strain-hardening. Using ℓ_{FH}
 90 = 5 μm and adding a work-hardening term, Evans and Hutchinson [23] obtain excellent
 91 fits to the data of Ehrler *et al.* [8] for nickel foils.

92 We apply equation (3) to simple and very well understood examples of the size
 93 effect. These are the plastic relaxation of non-lattice-matched epitaxial strained-layer

94 structures grown above their critical thicknesses. Growth is in the z direction to a
95 thickness h above the substrate at $z = 0$. At typical growth temperatures of 600°C for
96 GaAs-based structures (more than half the melting-point) the intrinsic yield strength is
97 very low. The ability to support elastic strains of 0.01 and more at thicknesses of tens of
98 nm comes from the size effect. In good-quality growth, there is little or no evidence of
99 work-hardening and the material may be taken to be perfectly plastic. Matthews critical
100 thickness theory [20–22] gives the critical thickness h_C at which misfit dislocations
101 (GNDs) may form at $z = 0$ to relieve the elastic strain in a simple layer with misfit strain
102 ε_0 . The result, for our purposes here, is best expressed by the geometrical version of
103 Matthew’s theory [25, 26], as $h_C \sim b/\varepsilon_0$ where b is the relevant (in-plane) component of
104 the Burgers vector of the misfit dislocations (the GNDs). This version agrees well with
105 experiment. Moreover, it omits unnecessary detail which is specific to single-crystal
106 cubic semiconductors and also it omits the ill-defined parameters, the inner and outer cut-
107 off radii, that appear in the calculation of the dislocation self-energy. The elastic strain ε_E
108 $= \varepsilon_0$ for $h < h_C$ and the plastic relaxation at greater thicknesses gives $\varepsilon_E \sim b/h$ for $h > h_C$.
109 The condition for plastic relaxation may be written in terms of the strain-thickness
110 product as $\varepsilon_E h \sim b$. The theory is readily generalised to more complicated structures
111 (graded layers with $\varepsilon_0 = gz$, multilayers and superlattices) by considering the strain-
112 thickness integral of $\varepsilon_E(z)dz$ over the thickness and introducing plastic relaxation during
113 growth as necessary to limit the integral to the value b [27]. Any intrinsic or bulk strength
114 simply adds to this size-effect strength. In all cases the size effect is due to the energy
115 required to create the length of GND needed to accommodate the misfit.

116 For significant plastic deformation (stress relaxation) when the initial dislocation
117 density is low, dislocation multiplication must take place – sources must operate.
118 Beanland showed that this requires a much greater thickness, $h_R \sim 5 h_C$ for simple layers
119 [28, 29]. In this case, the energy required to create the GNDs is small compared with the
120 energy dissipated in source operation. Then the strain-thickness product or integral during
121 plastic deformation is $\sim 5b$ for $h > h_R$. Experimentally, these predictions of the theory
122 have been confirmed extensively in simple layers, graded layers and in more complicated
123 structures [30–32]. The theory also predicts the spatial distribution of GNDs and of ε_P
124 [32], confirmed by discrete dislocation dynamics simulation [33].

125 We calculate the average plastic strain, the average plastic strain gradient, the
126 average stress, and the constant c for three standard epitaxial structures (Table I). For the
127 simple constant-composition strained layer with misfit strain ε_0 grown above its
128 relaxation critical thickness the plastic strain $\varepsilon_P(z)$ throughout the thickness of the layer is
129 constant and so this is also the average, $\bar{\varepsilon}_P = \varepsilon_P$. The average stress is $\bar{\sigma} = M\varepsilon_E$ where M
130 is the relevant elastic modulus. The plastic strain gradient is ideally infinite at the
131 substrate – layer interface and zero elsewhere, but the average comes just from the
132 change of plastic strain, from 0 at the substrate at $z = 0$ to ε_P at the top at $z = h$. The
133 constant $c = 1$ in this case by definition. Then the average stress (Table I), with a bulk
134 yield stress σ_Y added, may be set equal to the average stress predicted by the FH theory in
135 equation (3) giving,

136

$$\bar{\sigma} = \sigma_Y \left(1 + \frac{c \ell_{FH}}{h} \right) = M \varepsilon_E = \sigma_Y + \frac{5Mb}{h} \quad (4)$$

$$\ell_{FH} = \frac{5Mb}{\sigma_Y} = \frac{5b}{\varepsilon_Y}$$

137 where ε_Y is the yield strain.

138 In linearly-graded layers, with the misfit increasing as gz , the strain-thickness
 139 integral without plastic relaxation is $\frac{1}{2}gh^2$, and the critical thickness h_R is given by setting
 140 this equal to $5b$. When growth continues above h_R , the lower material relaxes completely.
 141 A top layer of thickness h_R has a uniform ε_P and stress increasing linearly with the slope
 142 Mg . We consider first a thin structure with growth to a thickness $h = h_R + \delta$ (δ small)
 143 giving constant plastic strain throughout the grade, except for the thin layer of thickness
 144 δh at the bottom (Table I) which we ignore. Again $c = 1$. The stress increases linearly so
 145 the average stress is half the surface stress (Table I). Again adding a bulk yield stress σ_Y
 146 and equating the average stress with the average stress of equation (3) we have

147

$$\bar{\sigma} = \sigma_Y \left(1 + \frac{c \ell_{FH}}{h} \right) = \sigma_Y + \frac{1}{2}Mgh_R \quad (5)$$

$$\ell_{FH} = \frac{\frac{1}{2}Mgh_R^2}{\sigma_Y} = \frac{5b}{\varepsilon_Y}$$

148 Graded-layer growth to a much greater thickness $h \gg h_R$ gives complete plastic
 149 relaxation to $\varepsilon_E = 0$, $\varepsilon_P = gz$ throughout the layer except for a thin region at the top of
 150 thickness h_R where ε_P is constant and the elastic strain ε_E rises from 0 to gh_R [27, 32].
 151 Neglecting the thin region at the top, the average plastic strain is $\frac{1}{2}gh$, while the average
 152 plastic strain gradient is just g , so that here $c = 2$. The stress is zero except in the thin

153 region at the top where it rises from zero to Mgh_R , so the stress-thickness integral is
 154 constant at $\frac{1}{2}Mgh_R$ and the average stress is obtained by multiplying by h_R/h . Again
 155 adding a bulk strength σ_Y and equating the average stress with the average stress of
 156 equation (3) we have,

$$\begin{aligned}
 \bar{\sigma} &= \sigma_Y \left(1 + \frac{c \ell_{FH}}{h} \right) = \sigma_Y + \frac{\frac{1}{2}Mgh_C^2}{h} \\
 \ell_{FH} &= \frac{\frac{1}{2}Mgh_C^2}{c\sigma_Y} = \frac{5b}{2\varepsilon_Y}
 \end{aligned}
 \tag{6}$$

158 All three examples, equations (4-6), give similar results, varying only because of
 159 the factor c , so we conclude that

$$\ell_{FH} = \frac{5b}{c\varepsilon_Y}
 \tag{7}$$

161 The problem of a linearly-graded layer maps perfectly onto half of the problem of a beam
 162 in bending, from the neutral plane to either free surface [33]. Taking typical numerical
 163 values for pure nickel and other soft metals, $M \sim 100$ GPa, $b \sim 0.25$ nm and yield
 164 strengths about 20 MPa, gives $\ell_{FH} = 3.125 \mu\text{m}$ from equation (6). This is in good
 165 agreement with the results from empirical fits (Fig.1).

166 Evans and Hutchinson [23] give values (but not error bars) of ℓ_{FH} obtained by
 167 fitting the FH theory to data from different authors for indentation of iridium, silver,
 168 copper and a superalloy, and to data for bending nickel foils. They note the inverse
 169 correlation between the values of ℓ_{FH} and the yield strain ε_Y of the material (figure 1), as
 170 in equations (4-7). Their tentative interpretation is that ℓ_{FH} represents the distance

171 moved by dislocations between e.g. cell walls or precipitates, which will be reduced as
172 σ_Y^{-1} in stronger materials. However, this interpretation overlooks the physical origin of
173 the size effect. Moreover, equation (7) *predicts* the absolute magnitudes of ℓ_{FH} very well
174 (figure 1).

175 The presence of c , the ratio of the peak value of ε_P to its average value, in the
176 denominator of equation (7) is interesting. Gradient theory fits DDD simulation results
177 better if the characteristic length is allowed to be a variable and to decrease with strain
178 [24]. The graded layers, equations (5, 6) show that c varies from 1 at low strain to 2 at
179 high strain, with a concomitant reduction of a factor of 2 in the characteristic length of
180 equation (7).

181 The phenomenological FH and similar strain-gradient theories express the
182 *outcomes* of the size effect accurately, but using a fitting parameter, the characteristic
183 length, which is not a true characteristic of the material. Evans and Hutchinson [23]
184 attribute equation (3) to the summation of the energy dissipation caused by the movement
185 of statistically-stored dislocations (SSDs) and that due to the movement of GNDs, the
186 second term.

187 Our interpretation of equation (3) is different. From figure 1 and equation (7), the
188 characteristic length is the Matthews critical thickness h_C or the relaxation critical
189 thickness h_R calculated using the elastic yield strain or flow stress of the material.
190 Equivalently, it is the thickness h at which the size effect doubles the strength of the
191 material. *Note that the σ_Y in the denominator of equation (7) permits rewriting equation*
192 *(3) as*

193
$$\bar{\sigma} = \sigma_Y + \frac{c\sigma_Y \ell_{FH}}{h} = \sigma_Y + \frac{5Mb}{h} \quad (8)$$

194 so that the inverse dependence of ℓ_{FH} on σ_Y is cancelled by the prefactor σ_Y . This is a
 195 very clear indication that the size effect is independent of the phenomena determining the
 196 yield strength, such as dislocation and defect densities. The first term does indeed
 197 represent whatever dissipative mechanism is responsible for the strength of bulk material
 198 without a size effect, such as the movements of SSDs. The second term, however, in the
 199 case that source operation is not required ($\varepsilon_E \sim b/h$), represents the energy stored (not
 200 dissipated) by the creation of GND length – the Matthews model [20–22]. In the case that
 201 source operation is required ($\varepsilon_E \sim 5b/h$), and this is generally the case for significant
 202 plastic deformation, the second term represents mostly the energy dissipated by source
 203 operation under the $\sim 5\times$ greater stress required to operate sources within a restricted size
 204 compared with the stress required merely to create extra GND length [29, 31]. In this
 205 interpretation, it is clear that neither the presence of GNDs nor the presence of a plastic
 206 strain gradient are directly responsible for the increased strength when they are present.
 207 The increased strength arises from the energy required to create the GNDs or to operate
 208 sources.

209 In this context, it is interesting to observe that the Matthews theory ($\varepsilon_E \sim b/h$) for
 210 simple strained layers requires the presence of a substrate, for otherwise misfit
 211 dislocations have nowhere to exist. But given the need for dislocation multiplication, the
 212 need to operate sources, the relationship $\varepsilon_E \sim 5b/h$ is independent of the presence or
 213 absence of a substrate, since two free surfaces with a separation h constrain the curvatures

214 of dislocations in a source (to more than $\sim h^{-1}$) in much the same way as one free surface
215 and a strained-layer – substrate interface or neutral plane does, or indeed the two
216 interfaces of a capped layer. Consequently, equation (7) applies as well to a stand-alone
217 thin foil, wire or micropillar under uniaxial tension or compression as it does to an
218 epitaxial layer on a substrate, or to a foil under bending or a wire under torsion, as long as
219 due attention is paid to the appropriate value of h in each case.

220 In the applications of equations (1–3) the primary unknown is the plastic strain
221 distribution. It can be obtained within the strain-gradient theory by analytic means for
222 very simple cases such as the beam in bending [23], or by numerical methods [19].
223 However, these methods rely upon the approximation that the stress-strain relationship
224 implied by equations (1–3) is local. This is an approximation that is severely in error for
225 the simple strained layer, since only the material at the substrate – layer interface
226 experiences a plastic strain gradient, yet the full thickness of the layer is capable of
227 sustaining the stress $M\epsilon_E \gg \sigma_Y$. Source operation and significant plastic deformation do
228 not depend upon conditions at a point, but upon conditions over an extended region
229 (source size) around the point, as recognised in nonlocal plasticity theories. Nevertheless,
230 the approximation can be good – this is best seen in the beam-bending or graded layer
231 problems. That is why, as observed by Liu *et al.* [10], the experimental data cannot test
232 between critical thickness theory and strain-gradient theory, for both will fit well.

233 It is worth commenting on the possible application of this analysis to other
234 gradient theories. Whenever the gradient term is multiplied by the yield or flow stress, as
235 in equation (3), and then the characteristic length turns out to vary as the inverse of the
236 yield or flow stress (or plastic strain), the separation we have done in equation (8) is

237 possible. This gives a gradient coefficient *unrelated* to yield or flow stress and then
238 interpretations in terms of dislocation or defect spacing become inappropriate. From the
239 review by Zhang and K. Aifantis [34], this seems to be the case for most gradient theories
240 including those based on, or equivalent to, the Aifantis theories [24, 35].

241 In conclusion, it is demonstrated that the characteristic length in the FH strain-
242 gradient theory can be obtained from known material and structural parameters,
243 $\ell_{FH} = 5b/c\varepsilon_Y$, $c \sim 1$. The derivation shows that this SGP corresponds physically to
244 critical thickness theory. It explains why SGP theories are capable of fitting experimental
245 data. It validates the use of this theory to obtain approximate constitutive laws for use in
246 finite-element calculations. It offers the prospect of understanding in general, on a secure
247 physical basis, why strong metals are strong, and how to include size effects in rigorous
248 engineering modelling and simulation.

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298

299 Table I. Parameters in the critical thickness calculations for strained layers with $\sigma_Y = 0$. Symbols are defined in the text.

Structure	$\varepsilon(z)$	H_R	h	$\varepsilon_E(z)$	$\bar{\sigma}$	$\varepsilon_P(z)$	$\bar{\varepsilon}_P$	$\bar{\varepsilon}'_P$	c
Simple layer	ε_0	$5b/\varepsilon_0$	$> h_R$	$5b/h$	$5Mb/h$	$\varepsilon_0 - \varepsilon_E$	ε_P	ε_P/h	1
Thin grade	gz	$\sqrt{10b/g}$	$h_R + \delta$	$z < \delta: 0$ else: $g(z-\delta)$	$\sim 1/2 Mgh_R$	$z < \delta: gz$ else: $g\delta$	$\sim g\delta$	$\sim g\delta/h$	~ 1
Thick grade	gz	$\sqrt{10b/g}$	$\gg h_R$	$z < (h-h_R): 0$ else: $g(z-h+h_R)$	$\sim 1/2 Mg \frac{h_R^2}{h}$	$z < (h-h_R): gz$ else: $g(h-h_R)$	$\sim 1/2 gh$	$\sim g$	~ 2

300

301 **Figure Caption**

302 Figure 1. Characteristic lengths ℓ_{FH} are plotted against the tensile yield strains ε_Y . The
303 length scales were found by fitting the FH theory to indentation data from the literature
304 for Ir, Ag, Cu and superalloy and to foil-bending data for Ni. *After figure 13 of reference*
305 *23.* The solid line is the prediction of equation (7), for a typical value of $b = 0.25$ nm and
306 with $c = 2$.
307