Chapter 2
Gradient Plasticity Formulations

A theoretical framework that has potential to cover a large range of gradient plasticity effects in isotropic materials is presented. Four classes of SGP theories are considered, aiming to span a wide domain of gradient plasticity formulations. The models considered as most representative of the state-of-the-art are the Mechanism-based Strain Gradient (MSG) plasticity theory [1], the phenomenological SGP theory reformulated by Fleck and Hutchinson [2], the advanced model by Gudmundson [3], incorporating dissipative and energetic terms, and the more general theory by Gurtin [4], which is generally referred to as Distortion Gradient Plasticity (DGP). The compatibility, balance and constitutive equations of the aforementioned SGP models are respectively described in Sects. 2.1, 2.2, 2.3 and 2.4.

Gradient plasticity formulations introduce one or several length parameters to capture the experimentally-observed size effects. Despite the efficiency of SGP theories, a unified interpretation of these internal length scales is still lacking and its relationship with the material microstructure has not been clearly understood [5]. By fitting experimental measurements of micro-tests through a specific SGP theory (in a way that resembles the fitting of the strain hardening exponent by means of a specific power law) the observed range of the magnitude of these length parameters roughly falls within 1–10 μm, as depicted in Table 2.1.

Finally, it is necessary to remark that SGP theories are continuously being enriched and modified; as the field evolves the role of novel formulations on crack tip mechanics must be assessed.

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E. Martínez Pañeda, *Strain Gradient Plasticity-Based Modeling of Damage and Fracture*, Springer Theses, DOI 10.1007/978-3-319-63384-8_2

Table 2.1 Compilation of experimentally reported length scales with their associated SGP formulation

Work	Material	Experiment	Length scale—SGP model
Fleck et al. (1994) [6]	Cu	Torsion	3.7 μm—Fleck and Hutchinson (1993) [7]
Nix and Gao (1998) [8]	Cu (cold worked)	Indentation	5.84 μm—Nix and Gao (1998) [8]
	Single crystal Cu (annealed)	Indentation	12 μm—Nix and Gao (1998) [8]
Stölken and Evans (1998) [9]	Ni	Bending	5.2 μm—Fleck and Hutchinson (1993) [7]
Shrotriya et al. (2003) [10]	Ni	Bending	5.6 μm—Fleck and Hutchinson (1993) [7]
Haque and Saif (2003) [11]	Al	Bending	4.5 μm—Gao et al. (1999) [1]
Ro et al. (2006) [12]	Al2024	Indentation	0.2 μm—Nix and Gao (1998) [8]
Qian et al. (2014) [13]	Steel S355	Indentation	7 μm—Gao et al. (1999) [1]
	Steel S690	Indentation	7 μm—Gao et al. (1999) [1]
Guo et al. (2017) [14]	Cu	Torsion	3 μm—Fleck and Hutchinson (1993) [7]
Iliev et al. (2017) [15]	In	Indentation	85.21 μm—Nix and Gao (1998) [8]
	In	Bending	93.34 μm—Fleck and Hutchinson (1993) [7]

2.1 Mechanism-Based Gradient Plasticity

The mechanism-based theory of strain gradient plasticity was proposed by Gao and co-workers [1, 16] based on a multiscale framework linking the microscale concept of SSDs and GNDs to the mesoscale notion of plastic strains and strain gradients. Unlike other SGP formulations, MSG plasticity introduces a linear dependence of the square of plastic flow stress on strain gradient. This linear dependence was largely motivated by the nano-indentation experiments of Nix and Gao [8] and comes out naturally from Taylor's dislocation model [17], on which MSG plasticity is built. Therefore, while all continuum formulations have a strong phenomenological component, MSG plasticity differs from all existing phenomenological theories in its mechanism-based guiding principles. The constitutive equations common to mechanism-based theories are summarized below, more details can be found in the original articles [1, 16].

In MSG plasticity, since the Taylor model is adopted as a founding principle, the shear flow stress τ is formulated in terms of the dislocation density ρ as

$$\tau = \alpha \mu b \sqrt{\rho} \tag{2.1}$$

Here, μ is the shear modulus, b is the magnitude of the Burgers vector and α is an empirical coefficient which takes values between 0.3 and 0.5. The dislocation density is composed of the sum of the density ρ_S for SSDs and the density ρ_G for GNDs as

$$\rho = \rho_S + \rho_G \tag{2.2}$$

The GND density ρ_G is related to the effective plastic strain gradient η^p by:

$$\rho_G = \overline{r} \frac{\eta^p}{b} \tag{2.3}$$

where $\overline{r}$ is the Nye-factor which is assumed to be 1.90 for face-centered-cubic (fcc) polycrystals. Following Fleck and Hutchinson [18], Gao et al. [1] used three quadratic invariants of the plastic strain gradient tensor to represent the effective plastic strain gradient η^p as

$$\eta^p = \sqrt{c_1 \eta^p_{iik} \eta^p_{jjk} + c_2 \eta^p_{ijk} \eta^p_{ijk} + c_3 \eta^p_{ijk} \eta^p_{kji}} \tag{2.4}$$

The coefficients were determined to be equal to $c_1 = 0, c_2 = 1/4$ and $c_3 = 0$ from three dislocation models for bending, torsion and void growth, leading to

$$\eta^p = \sqrt{\frac{1}{4} \eta^p_{ijk} \eta^p_{ijk}} \tag{2.5}$$

where the components of the strain gradient tensor are obtained by $\eta^p_{ijk} = \varepsilon^p_{ik,j} + \varepsilon^p_{jk,i} - \varepsilon^p_{ij,k}$. The tensile flow stress σ_{flow} is related to the shear flow stress τ by:

$$\sigma_{flow} = M \tau \tag{2.6}$$

where M is the Taylor factor, taken to be 3.06 for fcc metals. Rearranging Eqs. (2.1–2.3) and Eq. (2.6) yields

$$\sigma_{flow} = M \alpha \mu b \sqrt{\rho_S + \overline{r} \frac{\eta^p}{b}} \tag{2.7}$$

The SSD density ρ_S can be determined from (2.7) knowing the relation in uniaxial tension between the flow stress and the material stress-strain curve as follows

$$\rho_S = [\sigma_{ref} f(\varepsilon^p) / (M \alpha \mu b)]^2 \tag{2.8}$$

Here σ_{ref} is a reference stress and f is a non-dimensional function of the plastic strain ε^p determined from the uniaxial stress-strain curve. Substituting back into (2.7), σ_{flow} yields:

$$\sigma_{flow} = \sigma_{ref}\sqrt{f^2(\varepsilon^p) + l\eta^p} \tag{2.9}$$

where l is the intrinsic material length based on parameters of elasticity (μ), plasticity (σ_{ref}) and atomic spacing (b). Such that, for fcc metals,

$$l = M^2\bar{r}\alpha^2\left(\frac{\mu}{\sigma_{ref}}\right)^2 b = 18\alpha^2\left(\frac{\mu}{\sigma_{ref}}\right)^2 b \tag{2.10}$$

Several observations on the flow stress (2.9) must be remarked: [19]

(i) If the characteristic length of plastic deformation is much larger than the intrinsic material length l, the GNDs-related term $l\eta^p$ becomes negligible, such that the flow stress degenerates to $\sigma_{ref} f(\varepsilon^p)$, as in conventional plasticity.
(ii) The flow stress in Eq. (2.9) is based on the Taylor dislocation model, which represents an average of dislocation activities and is therefore only applicable at a scale much larger than the average dislocation spacing. For a typical dislocation density of $10^{15}/\text{m}^2$, the average dislocation spacing is around 30 nm such that the flow stress in (2.9) holds at a scale above 100 nm.
(iii) Even though the intrinsic material length l in (2.10) depends on the choice of the reference stress σ_{ref}, the flow stress in (2.9) is, in fact, independent of σ_{ref}. This is because both terms inside the square root in (2.9) are independent of σ_{ref}.

2.2 Fleck-Hutchinson 2001 Theory

The 2001 re-formulation by Fleck and Hutchinson [2] of their earlier phenomenological higher order SGP model [18] is one of the most widely used gradient plasticity theories. Of similar form to that proposed by Aifantis [20, 21], the novel formulation enjoys distinct computational advantages that makes it attractive for applications requiring the generation of numerical solutions. There are two important differences with its predecessors: (i) plastic strains are treated as primary variables, placing them on a footing similar to the displacements and (ii) length parameters are only present in the plastic range.

2.2.1 Infinitesimal Deformation Framework

Within a small strain setup, where the elastic and plastic strains are respectively denoted as ε^e_{ij} and ε^p_{ij}, the total strain is given by

$$\varepsilon_{ij} = \varepsilon^e_{ij} + \varepsilon^p_{ij} \tag{2.11}$$

The conventional effective plastic strain rate is $\dot{\varepsilon}_p = \sqrt{\frac{2}{3}\dot{\varepsilon}^p_{ij}\dot{\varepsilon}^p_{ij}}$, with $\varepsilon_p = \int \dot{\varepsilon}_p dt$. In Fleck-Hutchinson [2] theory hardening effects due to plastic strain gradients are included through the gradient of the plastic strain rate

$$\dot{\varepsilon}^p_{ij,k} = \left(m_{ij}\dot{\varepsilon}_p\right)_{,k} \tag{2.12}$$

where $m_{ij} = \frac{3}{2}\sigma'_{ij}/\sigma_e$ is the direction of the plastic strain increment, with σ'_{ij} denoting the stress deviator, and σ_e the von Mises effective stress. Variations of the displacements u_i, the effective plastic strain ε_p and the gradient of the effective plastic strain $\varepsilon_{p,i}$ appear in the variational principles underlying the formulation and a higher order stress vector quantity τ_i arises naturally as work conjugate to $\varepsilon_{p,i}$. The internal virtual work increment takes the form

$$\delta W_i = \int_V \left(\sigma_{ij}\delta\varepsilon^e_{ij} + Q\delta\varepsilon_p + \tau_i\delta\varepsilon_{p,i}\right) dV \tag{2.13}$$

Here σ_{ij} denotes the Cauchy stress tensor, Q is defined as the work conjugate to ε_p and $\tau_i\delta\varepsilon_{p,i}$ is the contribution due to the plastic strain gradients. Since $\dot{\varepsilon}^p_{ij} = \dot{\varepsilon}_p m_{ij}$, and consequently $\sigma_{ij}\delta\varepsilon^p_{ij} = \sigma_e\delta\varepsilon_p$, integrating by parts (2.13) renders

$$\begin{aligned}\delta W_i = &\int_V \left[-\sigma_{ij,j}\delta u_i + \left(Q - \sigma_e - \tau_{i,i}\right)\delta\varepsilon_p\right] dV \\ &+ \int_S \left(\sigma_{ij}n_j\delta u_i + \tau_i n_i\delta\varepsilon_p\right) dS\end{aligned} \tag{2.14}$$

where n_i is the unit outward normal to S. In the absence of body forces, the volume integral on the right-hand side of (2.14) vanishes, resulting in the conventional equilibrium relation

$$\sigma_{ij,j} = 0 \ \text{ on } V \tag{2.15}$$

and its higher order counterpart

$$Q = \sigma_e + \tau_{i,i} \ \text{ on } V \tag{2.16}$$

Consequently, the principle of virtual work follows as

$$\int_V \left(\sigma_{ij}\delta\varepsilon_{ij} + \tau_{i,i}\delta\varepsilon_p + \tau_i\delta\varepsilon_{p,i}\right) dV = \int_S \left(T_i\delta u_i + t\delta\varepsilon_p\right) dS \tag{2.17}$$

with the traction quantities T_i and t satisfying the natural boundary conditions

$$T_i = \sigma_{ij} n_j \quad \text{on } S \tag{2.18}$$

$$t = \tau_i n_i \quad \text{on } S \tag{2.19}$$

The third order plastic strain gradient tensor $\dot{\varepsilon}^p_{ij,k}$ can be decomposed, via orthogonal decomposition, into three independent tensors $\dot{\varepsilon}^{p(G)}_{ij,k}$, with $G = 1, 3$ [22, 23]. Such that a gradient-enhanced effective plastic strain rate, $\dot{E}^p$ can be defined in terms of three unique, non-negative invariants of $\dot{\varepsilon}^p_{ij,k}$, which are homogeneous of degree two:

$$\dot{E}^p = \sqrt{\dot{\varepsilon}^2_p + l_1^2 \dot{\varepsilon}^{p(1)}_{ij,k} \dot{\varepsilon}^{p(1)}_{ij,k} + 4 l_2^2 \dot{\varepsilon}^{p(2)}_{ij,k} \dot{\varepsilon}^{p(2)}_{ij,k} + \frac{8}{3} l_3^2 \dot{\varepsilon}^{p(3)}_{ij,k} \dot{\varepsilon}^{p(3)}_{ij,k}} \tag{2.20}$$

where, l_1, l_2 and l_3 are material length parameters. The effective plastic strain rate can be expressed explicitly in terms of $\dot{\varepsilon}_p$ and $\dot{\varepsilon}_{p,i}$:

$$\dot{E}^p = \sqrt{\dot{\varepsilon}^2_p + A_{ij} \dot{\varepsilon}_{p,i} \dot{\varepsilon}_{p,j} + B_i \dot{\varepsilon}_{p,i} \dot{\varepsilon}_p + C \dot{\varepsilon}^2_p} \tag{2.21}$$

where the coefficients A_{ij}, B_i and C depend on the three material length parameters as well as on the spatial gradients of the plastic strain increment direction,

$$\begin{aligned} A_{ij} &= l_1^2 \left(\frac{1}{2} \delta_{ij} + \frac{2}{5} m_{ip} m_{jp} \right) \\ &\quad + \epsilon_{pir} m_{qr} \left(L_2^2 \epsilon_{pjv} m_{qv} + L_3^2 \epsilon_{qjv} m_{pv} \right) \end{aligned} \tag{2.22}$$

$$\begin{aligned} B_i &= l_1^2 \left(\frac{4}{3} m_{pq} m_{pi,q} - \frac{8}{15} m_{ip} m_{pq,q} \right) \\ &\quad + 2 \epsilon_{pir} m_{qr} \left(L_2^2 \epsilon_{puv} m_{qv,u} + L_3^2 \epsilon_{quv} m_{pv,u} \right) \end{aligned} \tag{2.23}$$

$$\begin{aligned} C &= l_1^2 \left[\frac{1}{3} m_{ij,k} \left(m_{ij,k} + 2 m_{jk,i} \right) - \frac{4}{15} m_{ki,i} m_{kj,j} \right] \\ &\quad + \epsilon_{pir} m_{qr,i} \left(L_2^2 \epsilon_{puv} m_{qv,u} + L_3^2 \epsilon_{quv} m_{pv,u} \right) \end{aligned} \tag{2.24}$$

with $L_2^2 = \frac{4}{3} l_2^2 + \frac{8}{5} l_3^2$ and $L_3^2 = \frac{4}{3} l_2^2 - \frac{8}{5} l_3^2$. Here, δ_{ij} is the Kronecker delta and ϵ_{ijk} is the permutation tensor.

By the alternative definitions $A_{ij} = l^{*^2} \delta_{ij}$, $B_i = 0$ and $C = 0$ a single length scale parameter theory closely related to the strain gradient theory of Aifantis [20] can be formulated using a new length parameter l^* with

$$\dot{E}^p = \sqrt{\dot{\varepsilon}^2_p + l^{*^2} \dot{\varepsilon}_{p,i} \dot{\varepsilon}_{p,i}} \tag{2.25}$$

The stresses are related to the strains through the following incremental relations

$$\dot{\sigma}_{ij} = C_{ijkl}\left(\dot{\varepsilon}_{kl} - \dot{\varepsilon}_p m_{kl}\right) \tag{2.26}$$

$$\dot{Q} = h\left(E^p\right)\left(\dot{\varepsilon}_p + \frac{1}{2}B_i\dot{\varepsilon}_{p,i} + C\dot{\varepsilon}_p\right) \tag{2.27}$$

$$\dot{\tau}_i = h\left(E^p\right)\left(A_{ij}\dot{\varepsilon}_{p,j} + \frac{1}{2}B_i\dot{\varepsilon}_p\right) \tag{2.28}$$

where C_{ijkl} is the elastic stiffness tensor in an isotropic solid and $h\left(E^p\right) = d\sigma/d\varepsilon_p$ is the tangent hardening quantity, evaluated from uniaxial tensile data of stress σ versus plastic strain ε_p, as in the case of classic plasticity. For the one-parameter version (2.25) the constitutive relations yield

$$\dot{Q} = h\left(E^p\right)\dot{\varepsilon}_p \tag{2.29}$$

$$\dot{\tau}_i = l^{*^2}h\left(E^p\right)\dot{\varepsilon}_{p,i} \tag{2.30}$$

The yield condition is specified in terms of the generalized effective stress $Q_y = Q$, with the evolution of Q_y being respectively given by $Q_y = h\left(\dot{\varepsilon}_p + \frac{1}{2}B_i\dot{\varepsilon}_{p,i} + C\dot{\varepsilon}_p\right)$ and $Q_y = h\left(\dot{\varepsilon}_p\right)$ for the multiple and single length parameter versions.

2.2.2 *Finite Deformation Framework*

The phenomenological SGP theory by Fleck and Hutchinson [2] is generalized following the work by Niordson and Redanz [24], where a thorough description can be found (see also the work by Niordson and Tvergaard [25]). An updated Lagrangian configuration is adopted and by means of Kirchhoff stress measures the incremental principle of virtual work, Eq. (2.17), can be expressed as:

$$\int_V \Big(\tilde{\varsigma}_{ij}\delta\dot{\varepsilon}_{ij} - \sigma_{ij}\left(2\dot{\varepsilon}_{ik}\delta\dot{\varepsilon}_{kj} - \dot{e}_{kj}\delta\dot{e}_{ki}\right) + \left(\dot{q} - \dot{\sigma}_e^{\varsigma}\right)\delta\dot{\varepsilon}^p + \breve{\varrho}_i\delta\dot{\varepsilon}^p_{,i}\Big)\mathrm{dV} = \int_S \left(\dot{T}_{0i}\delta\dot{u}_i + \dot{t}_0\delta\dot{\varepsilon}^p\right)\mathrm{dS} \tag{2.31}$$

Here, $\tilde{\varsigma}_{ij}$ is the Jaumann rate of the Kirchhoff stress, $\dot{q}$ is the rate of the Kirchhoff variant of the effective stress, $\breve{\varrho}_i$ is the convected derivative of the higher order Kirchhoff stress and the velocity gradient is denoted by $\dot{e}_{ij}$. $\dot{T}_{0i}$ and $\dot{t}_0$ are the nominal traction and the nominal higher order traction, respectively, with the subscript 0 referring to the reference configuration. The Kirchhoff quantities are related to their Cauchy counterparts in Eq. (2.17) by the determinant, J, of the deformation gradient: $\varsigma_{ij} = J\sigma_{ij}$, $\varrho_i = J\zeta_i$, $q = JQ$ and $\sigma_e^{\varsigma} = J\sigma_e$. The finite strain generalization, for a hardening modulus $h\left[E^p\right]$, of the constitutive equations for the stress measures corresponding to the total strain, the plastic strain, and the plastic strain gradient,

respectively, are given by:

$$\tilde{\varsigma}_{ij} = C_{ijkl}\left(\dot{\varepsilon}_{kl} - \dot{\varepsilon}^{p} m_{kl}\right) = \dot{\varsigma}_{ij} - \dot{\omega}_{ik}\sigma_{kj} - \sigma_{ik}\dot{\omega}_{jk} \tag{2.32}$$

$$\dot{q} - \dot{\sigma}^{\varsigma}_{(e)} = h\left(\dot{\varepsilon}^{p} + \frac{1}{2}B_i\dot{\varepsilon}^{p}_{,i} + C\dot{\varepsilon}^{p}\right) - m_{ij}\tilde{\varsigma}_{ij} \tag{2.33}$$

$$\breve{\varrho}_i = h\left(A_{ij}\dot{\varepsilon}^{p}_{,j} + \frac{1}{2}B_i\dot{\varepsilon}^{p}\right) = \dot{\varrho}_i - \dot{e}_{ik}\varrho_k \tag{2.34}$$

where the elastic stiffness tensor is given by

$$C_{ijkl} = \frac{E}{1+\nu}\left(\frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) + \frac{\nu}{1-2\nu}\delta_{ij}\delta_{kl}\right) \tag{2.35}$$

and $\dot{\omega}_{ij} = \frac{1}{2}\left(\dot{e}_{ij} - \dot{e}_{ji}\right)$ is the anti-symmetric part of the velocity gradient. Here δ_{ij} is the Kronecker delta while E and ν denote Young's modulus and the Poisson ratio, respectively.

Fleck-Hutchinson (2001) [2] higher order phenomenological SGP theory has been consistently employed to model size effects in metal plasticity. The influence of the strain gradients is accurately captured in a number of numerical experiments, such as simple shear, torsion, void growth or nanoindentation [2, 26]. As detailed before, this is achieved by defining the flow stress as a function of a gradient-enhanced effective plastic strain $\sigma_{flow} = f\left(E^{p}\right)$, where f is an increasing function determined from a macroscopic uniaxial stress-strain test. Motivated by Taylor-based hardening, wherein increases in dislocation density underlie strain hardening, Fleck and Hutchinson [2] define E^{p} to include contributions from plastic strain gradients. Thus, as described in [23], dislocation density increases in the presence of severe plastic strain gradients: this is manifest as an increase in effective plastic strain and a subsequent increase in flow stress through the macroscopically determined strain hardening description.

2.3 Advanced Gradient Plasticity Theories

Gudmundson [3] was the first to note that the widely accepted phenomenological model by Fleck and Hutchinson [2] does not always satisfy thermodynamic requirements on plastic dissipation. With the aim of overcoming this deficiency, Gudmundson [3] and Gurtin and Anand [27] put forward a novel class of SGP theories that incorporates both energetic (or recoverable) and dissipative (or unrecoverable) higher order stresses.

2.3.1 Principle of Virtual Work and Governing Equations

Employing a small strain formulation, the total strain rate is determined from the gradients of the displacement rates: $\dot{\varepsilon}_{ij} = \left(\dot{u}_{i,j} + \dot{u}_{j,i}\right)/2$, and decomposes into an elastic part, $\dot{\varepsilon}^e_{ij}$, and a plastic part, $\dot{\varepsilon}^p_{ij}$, so that: $\dot{\varepsilon}_{ij} = \dot{\varepsilon}^e_{ij} + \dot{\varepsilon}^p_{ij}$. For a solid of volume V and external surface S, the principle of virtual work reads [3]

$$\int_V \left(\sigma_{ij}\delta\varepsilon^e_{ij} + q_{ij}\delta\varepsilon^p_{ij} + \tau_{ijk}\delta\varepsilon^p_{ij,k}\right)dV = \int_S \left(T_i\delta u_i + t_{ij}\delta\varepsilon^p_{ij}\right)dS \tag{2.36}$$

where σ_{ij} denotes the Cauchy stress, q_{ij} the so-called micro-stress tensor (work conjugate to the plastic strain, ε^p_{ij}) and τ_{ijk} the higher order stress tensor (work conjugate to the plastic strain gradients $\varepsilon^p_{ij,k}$), being the prime symbol omitted from q_{ij} and τ_{ijk} although only their deviatoric parts contribute to the principle of virtual work. The right-hand side of Eq. (2.36) includes both conventional tractions, $T_i = \sigma_{ij}n_j$, and higher order terms, $t_{ij} = \tau_{ijk}n_k$, with n_k denoting the outward normal to the surface S. The principle of virtual work can alternatively be stated as

$$\begin{aligned}\int_V \Big(&\sigma_{ij}\delta\varepsilon_{ij} + \left(q_{ij} - \sigma'_{ij}\right)\delta\varepsilon^p_{ij} \\ &+ \tau_{ijk}\delta\varepsilon^p_{ij,k}\Big)dV = \int_S \left(T_i\delta u_i + t_{ij}\delta\varepsilon^p_{ij}\right)dS\end{aligned} \tag{2.37}$$

Here, σ'_{ij} is the deviatoric part of the Cauchy stress: $\sigma'_{ij} = \sigma_{ij} - \delta_{ij}\sigma_{kk}/3$. Applying the product rule and Gauss' divergence theorem to the internal virtual work—left hand side of Eq. (2.37)—renders

$$\begin{aligned}\delta W_i = &\int_S \left(\sigma_{ij}n_j\delta u_i + \tau_{ijk}n_k\delta\varepsilon^p_{ij}\right)dS - \int_V \Big(\sigma_{ij,j}\delta u_i \\ &+ \left(\tau_{ijk,k} + s_{ij} - q_{ij}\right)\delta\varepsilon^p_{ij}\Big)dV\end{aligned} \tag{2.38}$$

Since the second integral on the right-hand side of Eq. (2.38) should vanish for arbitrary variations, two sets of equilibrium equations can be obtained

$$\sigma_{ij,j} = 0 \tag{2.39}$$

$$\tau_{ijk,k} + s_{ij} - q_{ij} = 0 \tag{2.40}$$

where the first integral on the right hand side of (2.38) may be identified as part of the external virtual work. Thus, by accounting for the right hand side of (2.36), the corresponding conventional $T_i = \sigma_{ij}n_j$ and higher order $t_{ij} = \tau_{ijk}n_k$ boundary conditions can be obtained.

2.3.2 Thermodynamically Consistent Constitutive Equations

The first two laws of thermodynamics for a given domain Ω of a continuum body within a dynamical process of specific internal energy $\mathscr{E}$ and specific entropy Λ read [28, 29]

$$\dot{\overline{\int_\Omega \mathscr{E} dV}} = W_e(\Omega) - \int_{\partial\Omega} q_i n_i dS + \int_\Omega q dV \tag{2.41}$$

$$\dot{\overline{\int_\Omega \Lambda dV}} \geqslant \int_{\partial\Omega} \frac{q_i}{\Theta} n_i dS + \int_\Omega \frac{q}{\Theta} dV \tag{2.42}$$

with $\dot{\overline{()}}$ denoting a total time derivative and where W_e is the external work, q_i is the heat flux, q is the heat absorption and Θ the absolute temperature. Within an isothermal process ($\Theta = \Theta_0$) the thermodynamic laws can be readily combined by considering the free energy $\Psi = \mathscr{E} - \Theta_0 \Lambda$, such that [30]

$$\dot{\overline{\int_\Omega \Psi dV}} \leqslant W_e(\Omega) \tag{2.43}$$

which is generally referred to as Clausius-Duhem inequality or the principle of dissipation. Given (2.36), and since $\dot{\overline{\int_\Omega \Psi dV}} = \int_\Omega \dot{\Psi} dV$ (with Ω being an arbitrary domain), the local free-energy inequality takes the form

$$\sigma_{ij}\dot{\varepsilon}^e_{ij} + q_{ij}\dot{\varepsilon}^p_{ij} + \tau_{ijk}\dot{\varepsilon}^p_{ij,k} - \dot{\Psi} \geqslant 0 \tag{2.44}$$

Within a generalized setting, the time derivative of the Helmholtz free energy is given by

$$\dot{\Psi} = \frac{\partial \Psi}{\partial \varepsilon^e_{ij}}\dot{\varepsilon}^e_{ij} + \frac{\partial \Psi}{\partial \varepsilon^p_{ij}}\dot{\varepsilon}^p_{ij} + \frac{\partial \Psi}{\partial \varepsilon^p_{ij,k}}\dot{\varepsilon}^p_{ij,k} \tag{2.45}$$

Accordingly, appropriate constitutive relations must be considered in order to fulfill the following imbalance:

$$\left(\sigma_{ij} - \frac{\partial \Psi}{\partial \varepsilon^e_{ij}}\right)\dot{\varepsilon}^e_{ij} + \left(q_{ij} - \frac{\partial \Psi}{\partial \varepsilon^p_{ij}}\right)\dot{\varepsilon}^p_{ij} + \left(\tau_{ijk} - \frac{\partial \Psi}{\partial \varepsilon^p_{ij,k}}\right)\dot{\varepsilon}^p_{ij,k} \geqslant 0 \tag{2.46}$$

It is possible to prescribe arbitrary values of $\dot{\varepsilon}^e_{ij}$ such that the inequality is violated [31]. Thus, the elastic contribution is understood to be purely energetic and the first term within brackets must vanish, leading to the constitutive equation for the Cauchy stress,

$$\sigma_{ij} = \frac{\partial \Psi}{\partial \varepsilon^e_{ij}} \tag{2.47}$$

The dissipative components of the microstress q_{ij}^D and the higher order stress τ_{ijk}^D are given by,

$$q_{ij}^D = \left(q_{ij} - \frac{\partial \Psi}{\partial \varepsilon_{ij}^p} \right) \quad \text{and} \quad \tau_{ijk}^D = \left(\tau_{ijk} - \frac{\partial \Psi}{\partial \varepsilon_{ij,k}^p} \right) \tag{2.48}$$

and the dissipation inequality (2.46) takes the form,

$$q_{ij}^D \dot{\varepsilon}_{ij}^p + \tau_{ijk}^D \dot{\varepsilon}_{ij,k}^p \geqslant 0 \tag{2.49}$$

The first term in (2.49) does not constitute a problem from the thermodynamic perspective as the effective plastic strain rate is inherently positive. Plastic deformation is generally considered to be a dissipative process and consequently the free energy is usually defined independently of the plastic strain, implying $q_{ij} = q_{ij}^D$ (this is, nevertheless, not always the case, c.f. [32]). However, the second term may be negative for strongly non-proportional histories when the stored energy associated with the plastic gradients is being released (see [30, 31]). This is the case of Fleck and Hutchinson (2001) theory [2] where the higher order stress—given by (2.28)—is purely dissipative.

One way of ensuring positive plastic work is to adopt a purely energetic formulation, implying:

$$\tau_{ijk}^E = \tau_{ijk} = \frac{\partial \Psi}{\partial \varepsilon_{ij,k}^p} \qquad \text{and} \qquad \tau_{ijk}^D = 0 \tag{2.50}$$

such that the second term in (2.49) vanishes. The earlier works of Aifantis and coworkers [20, 33, 34] fall within this approach, as the higher order stress is given by

$$\tau_i = \ell^2 H \varepsilon_{p,i} \varepsilon_{p,i} \tag{2.51}$$

such that it can be determined from a free energy,

$$\Psi = \frac{\ell^2}{2} H \varepsilon_{p,i} \varepsilon_{p,i} \tag{2.52}$$

where H is the tangent modulus, which is assumed to be a positive constant. However, as discussed by Hutchinson [35], this interpretation requires the input tensile curve to have a constant tangent modulus, which is not a realistic restriction for a general plasticity model. Moreover, from a physical standpoint it seems likely that some of the work associated with τ_{ijk} should be dissipative. The source of dissipative effects may be attributed to the movement of dislocations, a resulting resistance to dislocation motion that translates into an increase of the yield strength; while energetic effects may be associated to dislocation networks that lead to an increase in strain hardening [32, 36].

Dissipative higher order contributions can be incorporated—in a thermodynamically consistent manner—by employing higher order stress quantities that are related to increments of strain, as postulated by Gudmundson [3] and Gurtin and Anand [27]. The pivotal step in constructing this class of constitutive relations is to define an effective stress Σ work conjugate to the gradient-enhanced effective plastic strain rate $\dot{E}^p$, ensuring that the plastic work rate,

$$\Sigma \dot{E}^p = q_{ij}\dot{\varepsilon}^p_{ij} + \tau^D_{ijk}\dot{\varepsilon}^p_{ij,k} \tag{2.53}$$

is always positive. Such that, for a quadratic form of the gradient enhanced effective plastic strain rate,

$$\dot{E}^p = \sqrt{\frac{2}{3}\dot{\varepsilon}^p_{ij}\dot{\varepsilon}^p_{ij} + L^2\dot{\varepsilon}^p_{ij,k}\dot{\varepsilon}^p_{ij,k}} \tag{2.54}$$

where L is the dissipative length scale; a work conjugate effective stress can be defined

$$\Sigma = \sqrt{\frac{2}{3}q^D_{ij}q^D_{ij} + L^{-2}\tau^D_{ijk}\tau^D_{ijk}} \tag{2.55}$$

and the corresponding dissipative stress quantities (in terms of *increments* of strain) can be readily obtained:

$$q^D_{ij} = \frac{2}{3}\frac{\Sigma}{\dot{E}^p}\dot{\varepsilon}^p_{ij} \quad \text{and} \quad \tau^D_{ijk} = \frac{\Sigma}{\dot{E}^p}L^2\dot{\varepsilon}^p_{ij,k} \tag{2.56}$$

Following [3], the energetic contributions are obtained from the free energy, with a quadratic form being assumed,

$$\Psi = \frac{1}{2}\left(\varepsilon_{ij} - \varepsilon^p_{ij}\right)C_{ijkl}\left(\varepsilon_{kl} - \varepsilon^p_{kl}\right) + \frac{1}{2}\mu\ell^2\varepsilon^p_{ij,k}\varepsilon^p_{ij,k} \tag{2.57}$$

Here, C_{ijkl} is the isotropic elastic stiffness tensor, μ the shear modulus and ℓ the energetic constitutive length parameter. The conventional stresses are given through the elastic relationship:

$$\sigma_{ij} = \frac{\partial\Psi}{\partial\varepsilon^e_{ij}} = C_{ijkl}\left(\varepsilon_{kl} - \varepsilon^p_{kl}\right) \tag{2.58}$$

And the energetic higher order stresses are derived as:

$$\tau^E_{ijk} = \frac{\partial\Psi}{\partial\varepsilon^p_{ij,k}} = \mu\ell^2\varepsilon^p_{ij,k} \tag{2.59}$$

This novel class of advanced SGP theories [3, 27] is therefore able to incorporate both energetic and dissipative higher order contributions within a consistent

thermodynamic framework. The seminal theory by Fleck and Hutchinson [2] has been recently reformulated by Hutchinson [35] to meet the aforementioned thermodynamic requirements. Thus, the works by Gudmundson [3] and Gurtin and Anand [27] established a milestone in the development of gradient plasticity models. However, a number of open research issues remain to be addressed. Very recently Fleck, Hutchinson and Willis [35, 37, 38] noted that *non-incremental* dissipative higher order terms may lead to a delay in plastic flow under certain non-proportional loading conditions. This has been numerically corroborated by Bardella and Panteghini [39] and by Martínez-Pañeda et al. [40]. This physically uncertain response may favor the *incremental* modeling approach suggested by Hutchinson [35] where increments of all stress magnitudes are expressed in terms of increments of strain. However, a thermodynamically-consistent formulation with incremental constitutive relations for dissipative gradient contributions has not been proposed yet.

2.4 Distortion Gradient Plasticity

In recent years, increased attention has been focused on the need to account for the plastic spin, as proposed by Gurtin [4], to properly describe the plastic flow incompatibility and associated dislocation densities. It has been consistently shown (see, e.g., [39, 41, 42]) that the use of phenomenological higher order models that involve the whole plastic distortion (here referred to as Distortion Gradient Plasticity, DGP) leads to superior modeling capabilities. Hence, a distinct feature of Gurtin 2004 DGP [4] is the constitutive inclusion of the plastic rotation through the prescription of a free energy dependent on Nye's dislocation density tensor [43].

The equations in this section refer to the mechanical response of a body occupying a volume V with external surface S of outward normal n_i. More details about the higher order theory of distortion gradient plasticity can be found in [4].

2.4.1 Variational Principles and Balance Equations

Within a small strain formulation, the displacement gradient $u_{i,j}$ can be decomposed into its elastic and plastic parts:

$$u_{i,j} = \gamma_{ij}^e + \gamma_{ij}^p \tag{2.60}$$

where γ_{ij}^p, the plastic distortion, which characterizes the evolution of dislocations and other defects through its structure, may in turn be decomposed into its symmetric and skew parts:

$$\gamma_{ij}^p = \varepsilon_{ij}^p + \vartheta_{ij}^p \tag{2.61}$$

Unlike the plastic strain field ε_{ij}^{p}, the plastic rotation ϑ_{ij}^{p} is essentially irrelevant in the conventional theory. However, as pointed out by Gurtin [4], phenomenological models of isotropic SGP involving Nye's dislocation density tensor α_{ij} as primal higher order kinematic variable,

$$\alpha_{ij} = \epsilon_{jkl}\gamma_{il,k}^{p} \quad \left(\boldsymbol{\alpha} = \operatorname{curl} \boldsymbol{\gamma}^{p}\right) \tag{2.62}$$

must account for the plastic spin since the macroscopic characterization of the Burgers vector involves both the symmetric and skew parts of the plastic distortion

$$\epsilon_{jkl}\gamma_{il,k}^{p} = \epsilon_{jkl}\varepsilon_{il,k}^{p} + \epsilon_{jkl}\vartheta_{il,k}^{p} \quad \left(\operatorname{curl} \boldsymbol{\gamma}^{p} = \operatorname{curl} \boldsymbol{\varepsilon}^{p} + \operatorname{curl} \boldsymbol{\vartheta}^{p}\right) \tag{2.63}$$

with ϵ_{jkl} denoting the alternating symbol. Thus, in the absence of higher order surface tractions, the principle of virtual work can be expressed as:

$$\int_{V} \left(\sigma_{ij}\delta\varepsilon_{ij}^{e} + \zeta_{ij}\delta\alpha_{ij} + S_{ij}\delta\gamma_{ij}^{p} + \tau_{ijk}\delta\varepsilon_{ij,k}^{p}\right)dV = \int_{S} T_i\delta u_i dS \tag{2.64}$$

where the Cauchy stress is denoted by σ_{ij}. In addition to conventional stresses, the principle of virtual work incorporates the so-called micro-stress tensor, S_{ij} (work conjugate to the plastic distortion, γ_{ij}^{p}), the defect stress ζ_{ij} (work conjugate to Nye's tensor α_{ij}, the curl of the plastic distortion) and the higher order stress tensor, τ_{ijk} (work conjugate to the plastic strain gradients $\varepsilon_{ij,k}^{p}$). By taking into account that the micro-stress tensor can be decomposed into its symmetric and skew parts: $S_{ij} = q_{ij} + \omega_{ij}$, the virtual work statement can be expressed as:

$$\begin{aligned}\int_{V} &\left(\sigma_{ij}\delta\varepsilon_{ij} + \zeta_{ij}\delta\alpha_{ij} + \left(q_{ij} - \sigma'_{ij}\right)\delta\varepsilon_{ij}^{p} + \omega_{ij}\delta\vartheta_{ij}^{p}\right.\\ &\left. + \tau_{ijk}\delta\varepsilon_{ij,k}^{p}\right)dV = \int_{S} T_i\delta u_i dS\end{aligned} \tag{2.65}$$

with σ'_{ij} being the deviatoric part of the Cauchy stress. Applying the product rule and Gauss' divergence theorem to Eq. (2.65) renders:

$$\begin{aligned}&\int_{V}\left(-\sigma_{ij,j}\delta u_i + \left(q_{ij} - \sigma'_{ij} - \tau_{ijk,k} + \eta'_{ij}\right)\delta\varepsilon_{ij}^{p} + \left(\omega_{ij} + \varphi_{ij}\right)\delta\vartheta_{ij}^{p}\right)dV\\ &= \int_{S}\left(\left(T_i - \sigma_{ij}n_j\right)\delta u_i - \left(\Upsilon'_{ij} + \tau_{ijk}n_k\right)\delta\varepsilon_{ij}^{p} - \left(\Delta_{ij}\right)\delta\vartheta_{ij}^{p}\right)dS\end{aligned} \tag{2.66}$$

where η_{ij} and φ_{ij} are, respectively, the symmetric and skew-symmetric parts of the curl of the defect stress $\xi_{ij} = \epsilon_{jkl}\zeta_{il,k} = \eta_{ij} + \varphi_{ij}$; and equivalently, Υ_{ij} and Δ_{ij} respectively denote the symmetric and skew-symmetric parts of the cross product of the defect stress and the outward normal $\Gamma_{ij} = \epsilon_{jkl}\zeta_{il}n_k = \Upsilon_{ij} + \Delta_{ij}$, with the

prime symbol $'$ denoting deviatoric quantities. By means of the fundamental lemma of calculus of variations, the governing equations

$$\sigma_{ij,j} = 0 \tag{2.67}$$
$$q_{ij} - \sigma'_{ij} - \tau_{ijk,k} + \eta'_{ij} = 0 \tag{2.68}$$
$$\omega_{ij} + \varphi_{ij} = 0 \tag{2.69}$$

and the natural boundary conditions

$$T_i = \sigma_{ij} n_j \tag{2.70}$$
$$\Upsilon'_{ij} + \tau_{ijk} n_k = 0 \tag{2.71}$$
$$\Delta_{ij} = 0 \tag{2.72}$$

readily follow.

2.4.2 Energetic Contributions

In order to account for the influence of GNDs, the free energy is chosen to depend on both the elastic strain ε^e_{ij} and Nye's tensor α_{ij}:

$$\Psi = \frac{1}{2} C_{ijkl} \varepsilon^e_{ij} \varepsilon^e_{kl} + \Phi\left(\alpha_{ij}\right) \tag{2.73}$$

with C_{ijkl} being the elastic stiffness and $\Phi\left(\alpha_{ij}\right)$ the defect energy that accounts for the recoverable mechanisms associated with the development of GNDs. The widely used quadratic form of the defect energy is adopted

$$\Phi\left(\alpha_{ij}\right) = \frac{1}{2} \mu \ell^2 \alpha_{ij} \alpha_{ij} \tag{2.74}$$

but one should note that exploring other options may lead to further modeling capabilities (see [39, 41]), with recoverable gradient effects contributing to both hardening and strengthening. Accordingly, the defect stress equals:

$$\zeta_{ij} = \frac{\partial \Phi\left(\alpha_{ij}\right)}{\partial \alpha_{ij}} = \mu \ell^2 \alpha_{ij} \tag{2.75}$$

with μ being the shear modulus and ℓ an energetic length scale.

2.4.3 Dissipative Contributions

A gradient-enhanced phenomenological effective plastic flow rate is defined,

$$\dot{E}^p = \sqrt{\frac{2}{3}\dot{\varepsilon}^p_{ij}\dot{\varepsilon}^p_{ij} + \chi\dot{\vartheta}^p_{ij}\dot{\vartheta}^p_{ij} + \frac{2}{3}L^2\dot{\varepsilon}^p_{ij,k}\dot{\varepsilon}^p_{ij,k}} \tag{2.76}$$

where L is a dissipative length parameter and χ is the parameter governing the dissipation due to the plastic spin. Bardella [44] has analytically identified the value of χ that captures the mechanical response of a crystal subjected to multslip under simple shear:

$$\chi = \left[\frac{3}{2} + \frac{\sigma_0}{\mu\varepsilon_0}\left(\frac{L}{\ell}\right)^2\right]^{-1} \tag{2.77}$$

being σ_0 and ε_0 non-negative material parameters, which implies a value for χ bounded between 0 and 2/3. The flow resistance Σ, work conjugate to $\dot{E}^p$, is given by

$$\Sigma = \sqrt{\frac{3}{2}q_{ij}q_{ij} + \frac{1}{\chi}\omega_{ij}\omega_{ij} + \frac{3}{2L^2}\tau_{ijk}\tau_{ijk}} \tag{2.78}$$

Such that the unrecoverable stresses equal

$$q_{ij} = \frac{2}{3}\frac{\Sigma}{\dot{E}^p}\dot{\varepsilon}^p_{ij} \tag{2.79}$$

$$\omega_{ij} = \chi\frac{\Sigma}{\dot{E}^p}\dot{\vartheta}^p_{ij} \tag{2.80}$$

$$\tau_{ijk} = \frac{2}{3}L^2\frac{\Sigma}{\dot{E}^p}\dot{\varepsilon}^p_{ij,k} \tag{2.81}$$

And consequently the second law of thermodynamics is fulfilled by relating finite stress measures with rates of plastic deformation in what is referred to as a *non-incremental* form:

$$q_{ij}\dot{\varepsilon}^p_{ij} + \omega_{ij}\dot{\vartheta}^p_{ij} + \tau_{ijk}\dot{\varepsilon}_{ij,k} \equiv \Sigma\dot{E}^p > 0 \tag{2.82}$$

While higher order energetic and dissipative contributions are a common feature among the majority of the most advanced phenomenological SGP theories (see, e.g., [3, 27, 30, 45]), the need to constitutively account for the plastic spin, as proposed about ten years ago by Gurtin [4], has been mostly neglected in favor of simpler models. However, the use of phenomenological higher-order formulations that involve the whole plastic distortion has attracted increasing attention in recent years due to its superior modeling capabilities. The studies of Bardella and Giacomini [46] and Bardella [41, 44] have shown that, even for small strains, the contribution of the plastic spin plays a fundamental role in order to provide a description closer

to the mechanical response predicted by strain gradient *crystal* plasticity. This has been further assessed by Poh and Peerlings [42], who, by comparing to a reference crystal plasticity solution given by [47], showed that the plastic rotation must be incorporated to capture the essential features of crystal plasticity. Moreover, Poh and Peerlings [42] numerically elucidated that the localization phenomenon that takes place in Bittencourt et al. [48] composite unit cell benchmark problem can only be reproduced by DGP. Gurtin [4] theory has also been employed by Poh and co-workers [49, 50] through a novel homogenization formulation to describe the behavior of each grain in a polycrystal where grain boundaries are modeled to describe effects of dislocation blockage or transmittal.

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http://www.springer.com/978-3-319-63383-1

Strain Gradient Plasticity-Based Modeling of Damage and Fracture

Martínez Pañeda, E.

2018, XVII, 159 p. 66 illus., 47 illus. in color., Hardcover
ISBN: 978-3-319-63383-1