Physical foundations of crystal structure refinement under severe plastic deformation

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Abstract. The problem of crystal structure refinement under severe plastic deformation has been examined in the framework of the multiscale approach used in strength physics and nonequilibrium thermodynamics. This approach assumes that fragmentation of a solid in the entire hierarchy of the structural-scale levels (nano-, micro-, meso- and macroscale levels) plays a crucial role in the plastic deformation and fracture of materials. Formation of submicrocrystalline or nanocrystalline structure in solids subjected to severe plastic deformation is associated with fragmentation of the strain-induced band structures in the bending internal stress field. Flowing defect boundaries at the lower scale accommodate fragmentation of any curvature in a highly nonequilibrium material under SPD.

1. Introduction
Translation invariance is known to assure the thermodynamic stability of crystals. Violation of the translation invariance in a defect crystal causes its internal energy to increase. Because of this, the crystal undergoes continuous fragmentation, and defects are displaced toward fragment boundaries. Processes of this kind obey laws of nonequilibrium thermodynamics. The nonequilibrium thermodynamics of a deformed solid as a multiscale system and the corpuscular-wave dualism of plastic shear were discussed elsewhere [1]. In this work, the thermodynamical approach is applied to a description of fragmentation of solids under severe plastic deformation (SPD).

2. The nonequilibrium thermodynamics of fragmentation of solids in external force fields
Metastable mesoscopic substructures formed in a crystal fragmented at different scale levels are responsible for the local minima seen in the plot of the Gibbs thermodynamic potential against the molar volume, $F(v)$, shown in figure 1. To describe the local minima in terms of the thermodynamics of nonequilibrium states, we will consider the effective potentials $U(v, \alpha)$ in local defect-structure fields $\alpha$ [2], because this parameter accounts for the local fall in the magnitude of $F(v, \alpha)$ due to the entropy production during defect phase formation.

The relation for the Gibbs thermodynamic potential is of the form:

$$F(v) = U - TS + \sum_{i=1}^{n} \mu_i c_i,$$

(1)
Figure 1. Dependence of the Gibbs thermodynamic potential $F$ on the molar volume $v$ with allowance made for local hydrostatic tension zones of different scale levels.

where $U$ is the internal energy, $T$ is the temperature, $S$ is the entropy, $p$ is the pressure, $v$ is the molar volume and $\mu_i$ is the chemical potential of the $i$th element with concentration $c_i$.

Since defect phases occur in local hydrostatic tension zones [1], $v$ is used as an independent variable for the Gibbs thermodynamic potential. In the equilibrium state of a crystal for $v = v_0$, $F(v_0)$ has a minimum value. For any change in $v$, the value of $F(v)$ for the examined crystal is increased. By virtue of (1) the local minima in the ascending branch of the $F(v, \alpha)$ curve are attributable to the entropy production and redistribution of alloying elements between the basic crystal and the defect phases because of the difference in the chemical potentials of each of the $i$th components.

Following [3], the entropy production $\sigma_s$ is written as

$$\sigma_s = \chi \left( \frac{\nabla T}{T^2} \right)^2 + \rho \frac{\nabla \sigma}{T} - \rho \frac{\nabla v}{T^2} \left( [\overline{\nabla v}] \nabla T \right),$$

where $\chi$ is the thermal conductivity, $\rho$ is the density of the material, $\overline{\nabla}$ is proportional to the density of the defect flux and $\overline{\sigma}$ is the hydrostatic tension. The augend in (2) is the entropy production due to heat release. The addend defines the work done by the stress field on the defect flux to provide the motion of defects. The third term is related to the energy flux of the mechanical field of the crystal through the surface bounding the local structural transformation zone during the formation of the defect phase. The first two terms always have positive values and specify the entropy production. The third term has a negative value and reduces the entropy production. This term accounts for the relaxation of the disturbance of the mechanical field of the strained crystal, as the local structural transformation takes place in the local hydrostatic tension zone.

The fact that easy local structural transformation may take place during generation of dislocation nuclei near the ground state of a solid (the average molar volume $v = v_0$) determines a high value of the negative term in (2) for $\sigma_s$. In a thermodynamically stable crystal, the splitting of dislocations is limited by the energy flux of the mechanical field. As a consequence the entropy production is small during generation of dislocations and shallow minima appear in the $F(v)$ curve for $v = v_f$. For this reason, dislocations propagate along the slip plane as solitons without leaving any traces. The generation of dislocation nuclei is classified as local structural transformations in the hydrostatic tension zones.

As the degree of nonequilibrium of a solid is increased, it moves away from the ground state when $F(v_0)$ is at a minimum. For a certain range of values of $v$, local structural-phase transformations take place in the local hydrostatic tension zones, as seen in the ascending branch of the $F(v) = f(v)$ curve to the right of figure 1. The transformations are not suppressed by weak energy fluxes of the mechanical field of a nonequilibrium crystal. In this case, the negative term plays a secondary role in the entropy production, whereas the positive terms are growing in importance. The depth of the local minima in
the \( F(v) = f(v) \) curve increases. Irreversible defect structures (twins, bands of different scales and strain-induced martensite) are formed in the strained material.

As evident from figure 1, the condition \( F(v) < 0 \) is met in the range \( v_0 - v_{cr} \). This means that all crystal fragmentation stages under SPD in this range can be described in the framework of existing theories of deformation defects and their self-organization.

Notably, a nonequilibrium solid under SPD demonstrates a new peculiar quality where the Gibbs thermodynamic potential for the strained material approaches zero, and a two-phase pre-transition state is formed in the material (region \( C \) in figure 1). In this state, some of the nanocrystals with molar volume \( v_4 \) are surrounded by quasi-amorphous interlayers with molar volume \( v_5 \). We emphasize that the Gibbs thermodynamic potential for the nanocrystals is negative, even though their molar volume is dramatically increased (which is why the material is in a highly nonequilibrium state), whereas the quasi-amorphous interlayers have \( F(v) > 0 \). The shear instability of the quasi-amorphous interlayers accounts for the anomalous dynamic behavior of the Hall-Petch relation for nonequilibrium two-phase nanocrystals-amorphous phase systems. The pre-transition two-phase state is referred to as the nanostructural state of a solid for a grain-size range \( d < 30 \) nm. In addition to that, a solid may undergo fragmentation of nanosized structures with the proviso that \( F(v) < 0 \) (region \( B_1 \) in figure 1). Mechanical behavior of solids of this kind can be described using Hall-Petch relation. Finally, local zones formed for \( v > v_5 \), wherein \( F(v) > 0 \), are evidence of microporous structure developing in these regions of the material [1]. To prevent the porosity formation, SPD must be accomplished under hydrostatic compression.

3. The influence of solid nonequilibrium degree on mechanisms of its refinement at SPD

Two kinds of SPD were used to refine structure of specimens under study and, by doing so, to change their nonequilibrium degree: cold rolling and high cycle fatigue testing.

![Figure 2](image)

**Figure 2.** Formation of macroscopic localized-deformation bands at different stages of cold rolling of high-nitrogen austenitic steel Cr20NMn20V: \( T = 293 \) K and \( \varepsilon > 95 \% \): (a) macrobands as a system of contacting subbands of crystal and defect phases, \( \times 18160 \); (b) macroband fragmentation under superplastic flow, \( \times 16000 \); (c) cellular substructure, \( \varepsilon = 99\% \), \( \times 30000 \).

As an example, figure 2 shows the evolution of the mesoband structure in high-nitrogen austenitic steel Cr20NMn20V subjected to very high degrees of cold rolling. In the initial stage of cold rolling, a typical mesoscopic structure is formed in the material. However, very high degrees of cold rolling give rise to macroscopic bands evident as a defect phase (figure 2(a)). Notably, the macrobands experience severe bending. The macroband length increases dramatically, and continuous and discrete misorientations as high as several tens of degrees are accumulated at the macroband boundaries. The electron diffraction patterns from the regions within the bands exhibit a system of reflections arranged in pairs, which is evidence of two-phase macroband structure. One type of plastic-shear subbands has the same crystal structure as the initial material with a certain density of dislocations characterized by chaotic distribution. Another type of subbands is a defect phase in its own right. On further increase in the degree of cold rolling the bent crystal subbands are fragmented. The space between the fragments is filled with the defect phase (figure 2(b)). Increasing of \( \varepsilon \) up to 99% is accompanied by formation of...
cellular substructure with the characteristic cell size of 100 nm (figure 2(c)). Cold-working dynamic recrystallization develops in some zones. In cold-rolled materials, structures of this kind are formed in the superplasticity regime.

An ingenious method of SPD was offered in [4] making it possible to obtain high severe plastic deformation. Thin film of high-purity Al glued to a commercial Al substrate is subjected to fatigue testing. It allows to realize very high degree of SPD within Al-film under alternating bending with the number of cycles N up to $18 \times 10^6$. This method was also used in this investigation.

Figure 3(a) shows the 3D micrograph to characterize extrusion of material from surface layer of Al-film at $N=2.9 \times 10^6$. It is easily seen that quasiperiodical extruded mesobands of localized plastic deformation appear. Each mesoband consists of a set of lamellae being displaced along each other. Very high curvature of lamellae indicates about high nonequilibrium state of the extruded material. As it is seen in figure 3(b), plastic flow of extruded lamella is realized by propagation of flowing spiral - wise defect boundaries, whose trajectory and curvature depend upon internal stress field. The results shown in figure 3 testify that the refinement of a very nonequilibrium solid develops as multiscale process where at the lower scale the flowing defect boundaries accommodate fragmentation of any curvature. Sophisticated description and discussion of plastic flow mechanisms of highly nonequilibrium solid will be published soon in a separate paper.

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References
[1] Panin V E and Egorushkin V E 2008 Phys. Mesomech. 11 105
[2] Leontovich M A 1938 Zh. Exp. Teor. Fiz. No. 7 844
[3] Egorushkin V E 1990 Russ. Phys. J. 33 135
[4] Panin V E, Elsukova T.F, Egorushkin V E, Vaulina O Y, Pochivalov Y I 2009 Deformation and Fracture of Materials No. 6 2 (in Russian)