Atomic-scale modeling of plastic deformation of nanocrystalline copper

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Abstract

Atomic-scale simulations of nanocrystalline copper with grain sizes from 5 to 50 nm have been performed. The simulations show a clear maximum in the flow stress when the grains are 10–15 nm in diameter. At this grain size, there is a shift in deformation mechanism, from dislocation-mediated plasticity at larger grain sizes to grain boundary sliding at smaller. Above the maximum in hardness, the grain size dependence of the hardness is consistent with the Hall-Petch relation, conventionally explained by the creation of dislocation pile-ups in the grains. It has not been clear if this explanation of the Hall-Petch effect is valid for sub-micrometre grains, but the simulations presented here clearly show the existence of pile-ups in the simulation with average grain size of 50 nm. The dislocation dynamics in the grains is dominated by the grain boundaries, as almost all dislocation nucleation occurs at the grain boundaries, which also act as efficient dislocation sinks. During the plastic deformation, a large number of stacking faults and a much lower number of twin boundaries are created. These do not contribute significantly to the flow stress, as no work hardening is seen whereas the number of stacking faults increase with strain.

Key words: Computer simulations, Molecular Dynamics, Nanocrystalline metals, Flow stress, Hall-Petch relation, Deformation mechanism
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1 Introduction

Atomic-scale computer simulations have shown new deformation mechanisms in nanocrystalline metals with sufficiently small grain size[1–4]. As grain bound-

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aries act as barriers to dislocation motion[5,6], dislocations cease to play a significant role in the plastic deformation process once the grain size is below a critical value. Instead the grain boundaries become the carriers of plastic deformation through various kinds of grain boundary sliding, leading to a softening of the material as the grain size is further reduced. At larger grain sizes, where dislocation-mediated plasticity dominates, the opposite is seen. Reducing the grain size results in harder materials; this is known as the Hall-Petch effect [7,8].

Realistic atomic-scale simulations are limited in system size (up to millions of atoms) and time scale (nanoseconds), limiting simulations to the smallest grain sizes and to very high strain rates. Only recently have large-scale simulations made it possible to observe the transition in deformation mechanism, and the resulting maximum in hardness [9,10]. This also makes it possible to observe the dislocation processes leading to the Hall-Petch effect in polycrystalline metals, although with the caveat that the details of the deformation mechanism may change between the grain sizes studied in these simulations (50 nm and below), and typical grain sizes in coarse-grained polycrystalline metals (micro- to millimetres).

The purpose of this paper is to report some of these dislocation processes observed in large-scale molecular dynamics simulations of nanocrystalline copper.

2 Methods

The simulations were performed with molecular dynamics using the combination of a Nosé-Hoover thermostat and a Parrinello-Rahman barostat, as proposed by Melchionna *et al.* [11,12]. During the simulation, Newton's second law was integrated numerically with a timestep of 5×10^{-15}s. This is sufficiently small compared to the atomic vibration frequencies to ensure the stability of the dynamics. The forces between the atoms were described by the Effective Medium Theory (EMT) [13,14], which gives a realistic description of the metallic binding in the late transition metals. The EMT parameters used in this work were fitted to the elastic constants, the cohesive and vacancy formation energies and the intrinsic stacking fault energy [15].

The systems have periodic boundary conditions in all three directions, and the size and angles of the computational box were allowed to vary to keep the corresponding components of the stress to zero. The length of the computational box along the z-direction was excluded from the dynamics, and instead a constant strain rate was applied. In this way the simulation emulated a small part of an infinite (or very large) sample, which was pulled uniaxially while allowed to contract in the transverse directions.

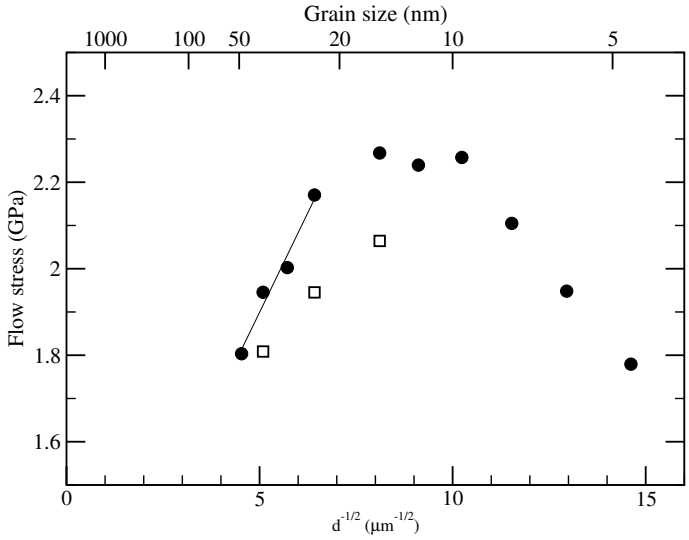

Fig. 1. Hall-petch plot of the flow stress. Filled circles are the simulations reported in Ref. [10]. Open squares are simulations with a ten times lower strain rate, the trends are the same but the flow stresses are reduced by approximately 150 – 200 MPa.

The initial configurations were created using a random Voronoi construction, as previously published [1,2]. The configurations consist of 20 grains, and contain from 91000 to 102 million atoms, corresponding to average grain diameters from 4.7 to 48.6 nm. The grains were initially defect-free, but during the simulations dislocations and other defects appear. To visualize these, Common Neighbor Analysis (CNA) [16] is used to calculate the local crystal structure. CNA identifies if the atoms are in local fcc or hcp crystal structure. Stacking faults are seen as two {111} planes of hcp atoms, twin boundaries as a single such plane. In dislocation cores and grain boundaries the atoms are neither in hcp nor fcc crystal structure.

The software used for this work is freely available under an Open Source license [17].

3 Observation of a transition in deformation mechanism

In a previous publication [10], we report molecular dynamics simulations of nanocrystalline copper, showing a clear maximum in hardness for grain sizes around 15–20 nm. This maximum is also seen in the Hall-Petch plot of Fig. 1. The line indicates that data for the largest grain sizes is consistent with a Hall-Petch relation: $\sigma_f = \sigma_{f,\infty} + kd^{-1/2}$. The Hall-Petch relation was originally proposed for the yield stress, but has been found to be valid for flow stresses too [6].

The maximum in flow stress corresponds to where the simulations show a shift in deformation mechanism between dislocation-mediated plasticity and grain boundary sliding. Yamakov *et al.* [9] report a similar shift in deformation mechanism. Their simulations are done with a constant applied stress, and the strain rate is measured. They observed a minimum in the strain rate at a grain size of approximately 18 nm, and interpret it as a maximum in hardness. Although the simulated material is different (Al instead of Cu), they are both face-centered cubic metals with similar deformation mechanisms, and indeed the maximum is found at similar grain sizes.

4 The onset of plastic deformation

As the timescale for molecular dynamics simulations is set by the vibrational frequencies of the atoms, only times up to a few nanoseconds can be simulated, and even less for the large system sizes presented here. A very high strain rate is thus required to obtain the required strain for plastic deformation. In the simulations reported here, a strain rate of $\dot{\varepsilon} = 5 \times 10^8 \mathrm{s}^{-1}$ was used.

As the starting configuration consists of dislocation-free grains, plastic deformation is initially difficult, in particularly at high strain rates where there is little time to nucleate dislocations. This results in a "bump" in the stress-strain curves, as seen in Fig. 2c. At lower strain rates, this bump diminishes, and presumably it will disappear when the strain rate becomes sufficiently low. Fig. 2 shows the effect of reducing the strain rate from $5 \times 10^8 \mathrm{s}^{-1}$ to $5 \times 10^7 \mathrm{s}^{-1}$. A factor 10 reduction of the strain rate thus results in a 15% reduction in the flow stress.

The "overshoot" observed at the high strain rate only occurs for the large grain sizes. For smaller grain sizes, dislocations are not a main contributor to the plastic deformation, and the difficulties of nucleating them are therefore irrelevant. The overshoot hides the Hall-Petch dependence of the yield stress, but at sufficiently slow strain rates it should still be present.

5 Dislocation processes during flow

During the deformation of the grains, little strain hardening is observed, indicating that dislocation tangles and other mechanisms for immobilizing dislocations either do not operate in these very small grains, or saturate so quickly that the resulting work hardening only occurs during the very first stages of plastic deformation. This lack of work hardening is also observed experimentally, and is believed to be one of the main reasons for the low tensile ductility

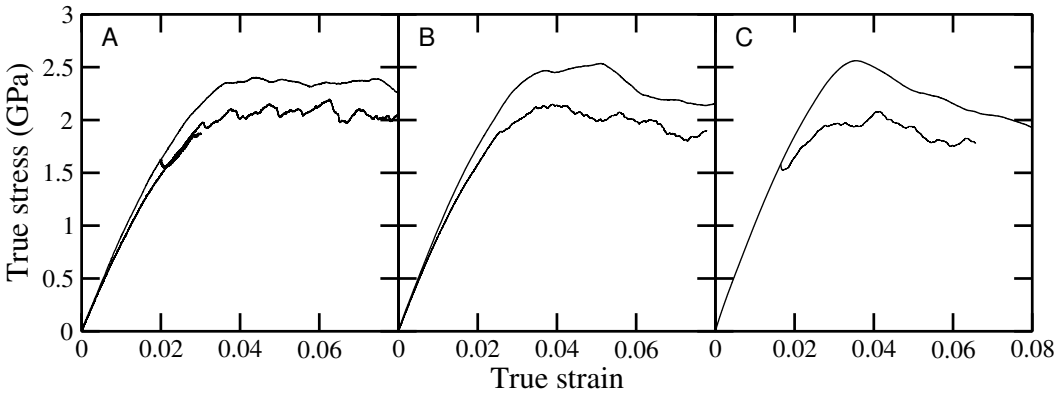

Fig. 2. Stress-strain curves at different grain sizes and different strain rates. The three panels show stress-strain curves for samples with average grain diameter of 15.2 nm (A), 24.2 nm (B) and 38.6 nm (C). Two strain rates are used; the upper curve is at $\dot{\varepsilon} = 5 \times 10^8 \mathrm{s}^{-1}$, the lower at $\dot{\varepsilon} = 5 \times 10^7 \mathrm{s}^{-1}$. In panel A a thick line shows that it is not necessary to repeat the initial (elastic) part of the simulation. This was used to save computer time for the largest system (C). Adapted from the supplementary online material of Ref. [10].

of nanocrystalline metals [18], since it leads to rapid failure through shear band formation.

Observing the grain interiors in the simulations reveals a plethora of dislocation processes. Dislocations are constantly created at grain boundary sources, propagate through the grains and vanish at the opposite grain boundaries. Very few, if any, are created at in-grain sources such as Frank-Read sources, presumably again due to the small grain size. As has previously been observed, many single Shockley partials are nucleated. This seems to be characteristic of computer simulations with small grain sizes and high strain rate. It is caused by the high stresses present during the nucleation process: the force (and energy) associated with the creation of a stacking fault is small compared to the difference in the forces on the leading and trailing Shockley partials. As the individual partials leave behind a stacking fault, which is only sometimes removed later, the grains quickly develop a rather large density of stacking faults. These are seen to act as weak barriers to the dislocation motion, delaying the dislocations rather than permanently stopping them. The effect on the flow stress is small, which can be seen by the fact that the number of stacking faults increase during the simulation, whereas the flow stress does not. It should be noted that although multiple slip systems are active in the grains, one slip system tends to dominate. The majority of the dislocations therefore move on slip planes parallel to the majority of the stacking faults.

Initially, dislocations arrive at grain boundaries and are absorbed by them, but when a number of dislocations have been absorbed, the back-stress prevents subsequent dislocations from entering the grain boundary, and a pile-up is formed. One such pile-up is seen in Fig. 3. It has not been possible to de-

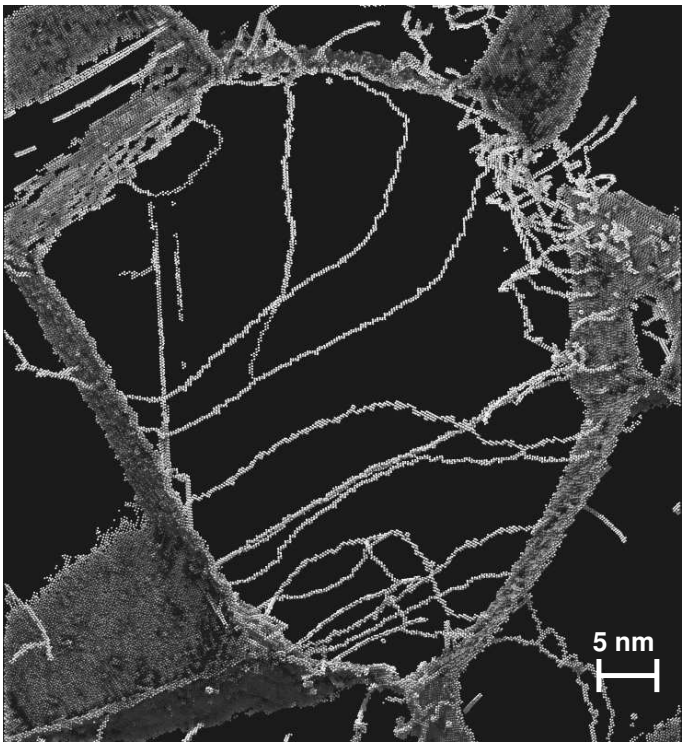

Fig. 3. A pile-up formed in a grain. The dislocations move from the upper-left to the lower-right corner. For clarity, only a thin slice through the grain is shown, and all atoms in a perfect crystalline neighbourhood have been removed. A few dislocations on nearby slip planes, but not part of the actual pile-up, are also seen. Their stress fields perturb the pile-up.

termine if there is any correlation between dislocations in the pile-up entering the grain boundary, and dislocations emitted from the grain boundary into adjacent grains. The grain boundary is a quite active dislocation source in one of the adjacent grains, but the high dislocation activity makes it impossible to correlate individual events.

The dislocation activity is qualitatively different from more coarse grained materials in one obvious way: almost all dislocations are created at grain boundary sources, as the grains are too small to contain a significant number of Frank-Read sources. This may influence the way the hardness depends on the grain size. Dislocations are seen emerging from the grain boundaries in semi-circular configurations, in agreement with assumptions that the grain boundary sources are Frank-Read-like [19]. If it is further assumed that the size of the grain boundary sources scales linearly with the grain size, then the yield stress will scale with inverse grain size ($\sigma_y \sim d^{-1}$) instead of with the usual inverse square root ($\sigma_y = \sigma_{y,\infty} + kd^{-1/2}$) [19,20]. Here it should be noted that Cheng *et al.* [19] propose a slightly more complicated functional form, which includes a term logarithmic in the grain size arising from the usual logarithmic cutoff term in the dislocation line tension. However, this cutoff is the limit of the range of the stress field of the dislocation, and there is a

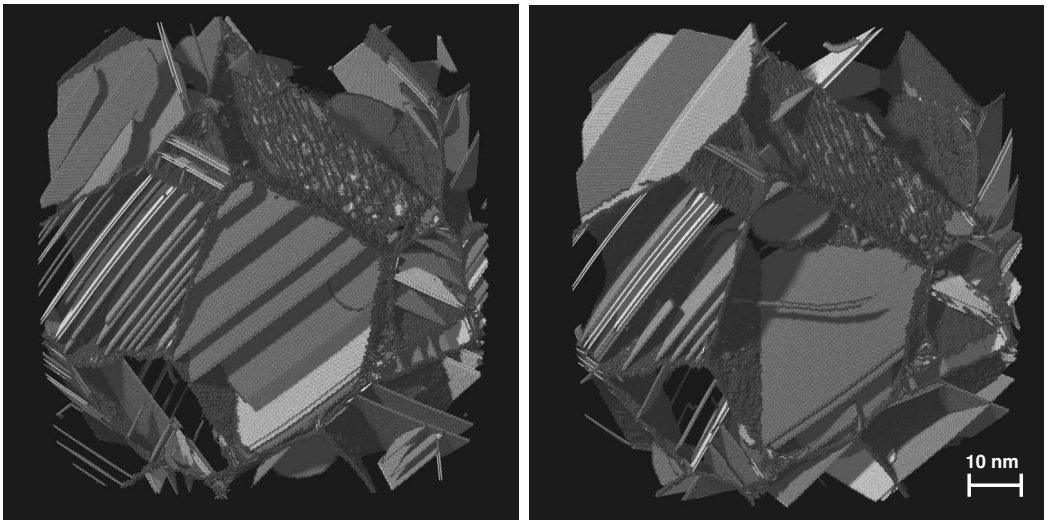

Fig. 4. Planar faults formed in nanocrystalline copper with an average grain size of 39 nm, deformed at strain rates of $\dot{\varepsilon} = 5 \times 10^8 \mathrm{s}^{-1}$ (left) and $\dot{\varepsilon} = 5 \times 10^7 \mathrm{s}^{-1}$ (right). The total strain is 6.45 %. All atoms in the perfect face-centered cubic crystal structure have been removed in the plot to expose the grain interiors, where numerous planar faults are seen. Stacking faults are dark; twin boundaries are light. In the right panel a dislocation, split into two partials separated by a narrow stacking fault ribbon, is moving through the large central grain.

priori no reason to assume that the stress field of a dislocation is restricted to a single grain.

The simulations are unfortunately not able to distinguish between the two grain size dependences, as data is only available for quite a narrow range of grain sizes. It is, however, clear that both pile-ups and grain boundary sources are important parts of the dislocation dynamics.

6 Deformation twinning

As a significant fraction of the dislocation activity is single Shockley partials, a large number of stacking faults are produced. This also leads to the possibility of deformation twinning through consecutive motion of partial dislocations on adjacent glide planes. Figure 4 shows these planar faults. It is clearly seen that although some twin boundaries are present, they are relatively rare, and the twins are only a few atomic layers wide. On the other hand there are many stacking faults, in particular at the high strain rate, indicating that the production of stacking faults is at least indirectly due to the strain rate.

The most likely cause for the strain rate dependence of the stacking fault density is the higher flow stress at the higher strain rate. At higher stresses, the cost of creating a stacking fault becomes less important compared to the

difference in resolved shear stress on the leading and trailing partials, and it is therefore more likely that there will be slip systems where production of leading partials is most likely. The opposite possibility, that the high number of stacking faults is causing the high flow stress, is less likely as the flow stress does not increase with plastic flow whereas the density of stacking faults does. The difference in flow stress is present even when the deformation begins, as is seen on Fig. 2. It cannot, however, be ruled out that there is some hardening effect of the stacking faults.

The lack of strain hardening caused by the production of stacking faults is surprising in the light of recent results by Yamakov *et al.* [20]. They compare simulations of the deformation of a material with a high stacking-fault energy (Al) with one with a low stacking-fault energy (Pd using a potential resulting in a very low stacking fault energy), and find deformation hardening in the latter case, as seen by a decrease in the strain rate at constant stress. It is unclear if this disagreement is caused by the difference in loading conditions; the strain rate at constant stress may be more sensitive to stacking faults than the flow stress at constant strain rate. In the simulations reported here, dislocations are seen to be delayed when they cross stacking faults, but in most cases they break free rather rapidly. Such delays will clearly show up in the strain rate in a stress-controlled simulation, but may only have a small effect on the overall flow stress.

Deformation twinning is expected to have a larger effect on the flow stress, as the slip systems are located differently in the twin. However, only a few twin boundaries are produced, as seen in Fig. 4, and these mainly in grains with a high stacking fault density. These are grains where a single slip system dominates, and dislocations are therefore mainly moving parallel to the twin boundaries. Twinning has previously been reported in two-dimensional simulations of nanocrystalline Al [21,22], and has also been observed experimentally in nanocrystalline Al [23]. It is, however, neither clear how important the dimensionality is (2D versus 3D simulations) [24], nor how strong an effect the twinning has on the flow stress. The experimentally found lamella-like structure of the deformation twins [23] is very similar to the structure seen in Fig. 4.

7 Conclusions

Large-scale molecular dynamics simulations of nanocrystalline copper have revealed a transition in the deformation mechanism at grain sizes around 10–15 nm, below which dislocations no longer play an important role in the plastic deformation. The flow stress, and thus the strength of the material, displays a clear maximum at the grain sizes corresponding to the transition. In systems

with larger grain sizes, intense dislocation activity occurs in the grains. The dislocations are created in grain boundary sources, move through the grains, and disappear into the opposite grain boundaries. Often dislocation pile-ups are formed in the grains. The flow stress varies with grain size in a way which is consistent with the Hall-Petch relation, although the exact exponent of the grain size dependence cannot be determined.

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