Structure Modulation Driven by Cyclic Deformation in Nanocrystalline NiFe

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Theoretical modeling suggests that the grain size remains unchanged during fatigue crack growth in nanocrystalline metals. Here we demonstrate that a modulated structure is generated in a nanocrystalline Ni-Fe alloy under cyclic deformation. Substantial grain coarsening and loss of growth twins are observed in the path of fatigue cracks, while the grains away from the cracks remain largely unaffected. Statistical analyses suggest that the grain coarsening is realized through the grain lattice rotation and coalescence and the loss of growth twins may be related to the detwinning process near crack tip.

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In contrast to coarse-grained material, the grain structure of nanograined metals sometimes becomes unstable under an applied stress, as suggested by the grain growth recently reported for nanocrystalline (NC) Cu deformed under indenter at liquid-nitrogen temperature and the room-temperature grain growth in NC Al [1–3]. Analytical modeling suggests that the mobility of grain boundaries (GBs) is related to dislocation activities in GBs [4]. In the case of cyclic deformation, the stability of the grain structure is of particular interest, as it is known that the adaptability of grains to the stress field in front of a fatigue crack tip governs the crack growth rate [5]. Molecular dynamics (MD) simulations suggest that a fatigue crack propagates intergranularly in NC Ni with nanovoids formed and twins nucleated in the grains in front of the crack tip, and that the grain structure remains intact in terms of size and geometry [6,7]. However, because MD simulations are computation intensive, the present simulation results only capture the early stage of the crack propagation (<100 cycles) [7]. In practical applications, fatigue samples usually undergo millions of cycles. How the nanograined structure evolves, particularly in the late stage of cyclic deformation when extensive damage occurs, is presently unknown. A careful investigation of structure evolution under high cycle fatigue conditions is therefore of both scientific and technological interest. In this Letter, we present experimental evidence in NC Ni-Fe alloys that paints a rather different picture, in that a modulated and coarsened grain structure is generated and growth twins are removed.

NC Ni-20% (w.t.) Fe alloy was synthesized by pulse electrodeposition at Integran (Canada). Transmission electron microscopy (TEM) analysis reveals slightly columnar grains with a high density of growth twins and sharp large-angle boundaries in the view perpendicular to the deposition growth direction. Selected area diffraction indicates the grain orientations are approximately random. Statistical analyses indicate that the grain size ranges from ~5 to 60 nm with an average value of ~19 nm, while most of the grains fall in the range between 10 and 20 nm. High resolution TEM (HRTEM) shows that these nanograins contain very few dislocations ($\sim 1.0 \times 10^{16}$ m^{-2} on average) in the as-received state, and that the growth twins show straight and parallel twin boundaries (TBs) with few random dislocations detected (see supplemental material [8]). Cyclic deformation was performed under a compressive-compressive mode ($R = \sigma_{\min}/\sigma_{\max} = 0.1$). Rectangular samples of $3 \times 3 \times 6$ mm^3 are used with the loading direction normal to the deposition growth direction [8]. Owing to the compressive stress (type II loading mode), a high fatigue life of $>10^6$ cycles can be achieved with $\sigma_{\max} = 2.2$ GPa, compared to very poor fatigue deformation behavior exhibited in NC materials with a tensile stress component [9].

After the cyclic deformation, the initially random structure is replaced by a modulated structure near the sample surface, i.e., alternating NC and ultrafine-grained (UFG) regions. Figure 1(a) representatively shows an integrated TEM image, where the alternation from the NC to UFG and to NC region is illustrated and transition zones are established in between the NC and UFG regions. TEM observation over wide areas indicates that the widths of the UFG regions are 10–15 μm, and the average wavelength of the fluctuation is approximately ~15 μm [Fig. 1(e)]. Careful inspection revealed not only substantial grain coarsening in these UFG regions, but also significant reduction in the growth-twin density. Most of the grains in the UFG regions are sized from 100 to 200 nm (with some as large as 500 nm) and mostly free of the growth twins [Fig. 1(d)], while in the NC region they nearly maintained their grain size and twin density similar to that prior to deformation [Fig. 1(b)]. In the transition regions, the TBs are found distorted and the original nanograins reoriented, leading to close orientation [Fig. 1(c)]. Inspection of the surface area of the fatigued sample reveals a number of

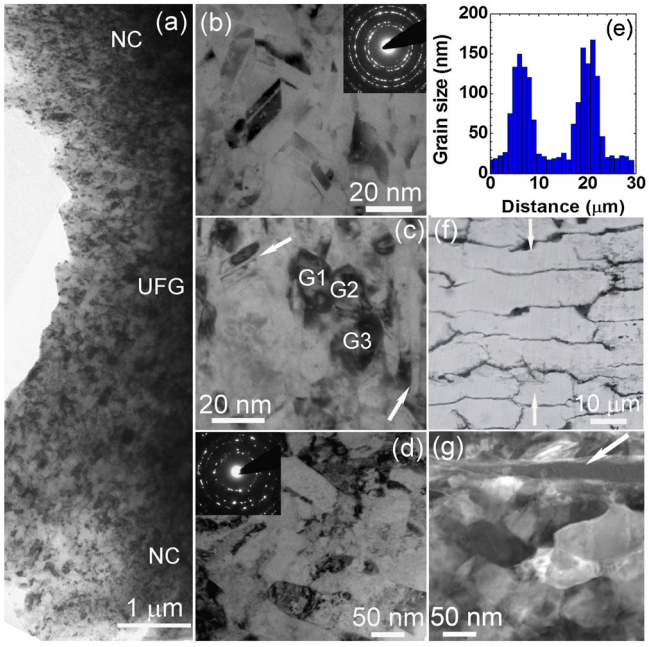

FIG. 1 (color online). (a) Integrated TEM image showing a modulated structure. (b–d) Magnified TEM images in the NC (b), transition (c), and UFG (d) regions. (e) Grain size distribution in the NC and UFG regions. (f) Surface fatigue cracks after 1.08×10^6 cycles, relative to the loading direction. (g) Scanning TEM image near fatigue crack.

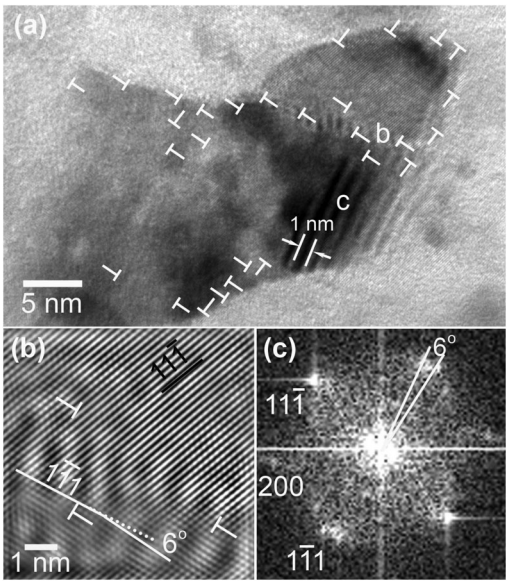

FIG. 2. (a) HRTEM image of two small grains with small-angle misorientation in the transition region. (b) Inverse Fourier transformation of area "b" in (a) showing a small-angle misorientation. (c) Electron diffraction pattern of area in (a) showing a 6° misorientation.

parallel cracks, as shown in Fig. 1(f). The surface cracks are $50 \sim 80$ μm deep. It is interesting to note that the spacing between the cracks approximates the modulation wavelength, suggesting that the grain coarsening and loss of growth twins may be driven by cracks penetrating into the sample. This hypothesis was further corroborated by scanning TEM observation of microstructures near a crack tip. With a TEM specimen prepared near a crack tip using the focused ion beam technique, we discovered the similar grain coarsening and twin density reduction associated with the crack path [Fig. 1(g)]. In addition, transgranular crack growth is identified.

HRTEM studies were performed on the NC, UFG, and transition regions. First, following fatigue deformation, the grains in NC regions still had a similar dislocation density and random distribution to those in the as-received sample. However, for small grains with $d \leq 20$ nm located in transition regions, dislocation accumulation was noted near the GBs. Figure 2(a) shows two typical grains with $d < 20$ nm, where a number of dislocations are found near the GBs, but very few are evident within the grains. Moreover, these two grains exhibit small-angle misorientation constructed with dislocations, in contrast to the large-angle GBs in the as-received samples. Inverse Fourier transformation of area "b" in Fig. 2(a) revealed a misorientation of 6° between the two grains, as shown in Fig. 2(b). This is further confirmed by an electron diffraction pattern of the area in Fig. 2(a). The diffraction arcs spread up to ~6° [Fig. 2(c)]. Moiré fringes caused by the small-angle misorientation of neighboring grains along the TEM electron beam direction are seen in area "c" in Fig. 2(a). The Moiré fringe domain size is about 10 nm, which is consistent with the original grain sizes in the as-received state. The spacing of the Moiré fringe is about 1 nm and corresponds to an 11° misorientation between the top and bottom grains.

In the case of larger grains, except for the presence of dislocations accumulated at the GBs and TBs, stacking faults were also observed near the GBs and TBs. Figure 3 shows a representative grain of ~60 nm. The HRTEM image at the site "b" of the GB displays a number of dislocations and a 15° misorientation [see Fig. 3(b)]. A higher dislocation density was observed when approaching the triple junction. Moreover, a number of stacking faults were found in the vicinity of the GB (pointed at by white arrows), which were emitted from the right side of the GB and terminated in the grain interiors, as marked by a line near the left-lower corner in Fig. 3(b). However, the initial growth twins experienced substantial topological changes during deformation. The two separate TBs marked by "a" and "c" in Fig. 3(a) are located in a straight line, suggesting they might be evolved from an initial singular TB. Detailed analysis reveals that the TB at "c" is tangled with a number of dislocations [Fig. 3(c)], and this TB was shifted due to partial dislocation emission along the TB from the GB. Dislocations can also be observed in the grain center, as shown by the inverse Fourier transformation image in Fig. 3(d), although the probability of finding dislocations in the grain center is much smaller than that near GBs.

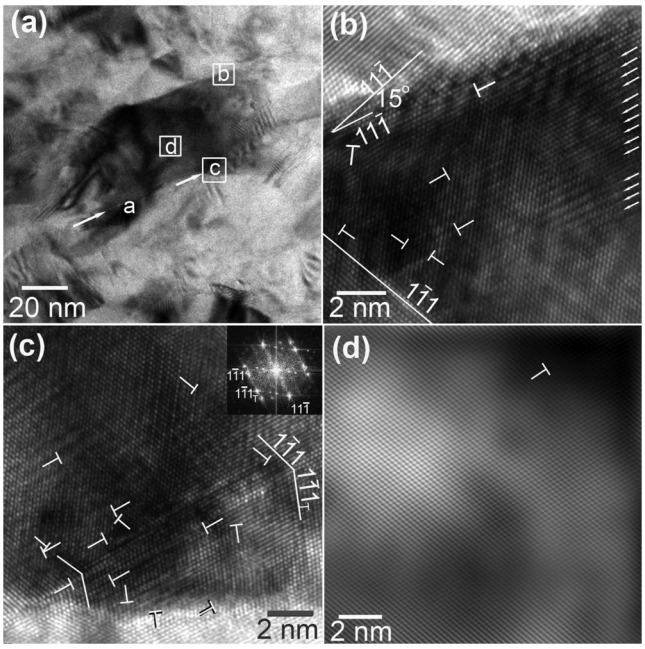

FIG. 3. (a) A large grain in the transition region. (b–d) The areas marked in "b", "c," and "d" in (a). The inset in (c) shows a twin relationship.

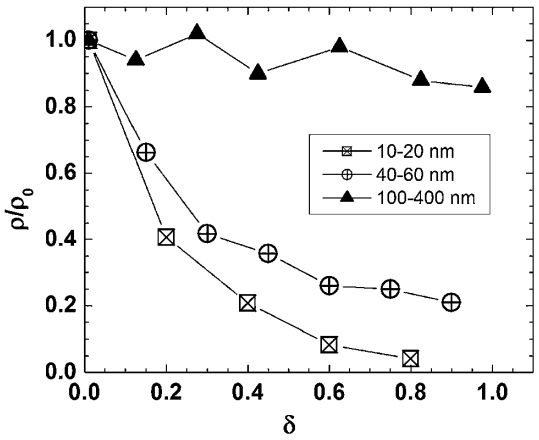

FIG. 4. Dislocation distribution $\delta = 2 \cdot L/d$, where L is the distance from the GB, and d is the grain diameter. ρ_0 near GBs are 1.2, 0.8, and 0.5×10^{17} m^{-2} for grains with diameter ranges of 10–20, 40–60, and 100–400 nm, respectively, with a measurement error of $\pm 0.1 \times 10^{17}$ m^{-2}.

In the coarsened grains (>100 nm), high dislocation density can be identified in both GBs and grain interiors, and the dislocations tend to distribute evenly across the grains [8]. Moreover, almost all the initial growth twins disappeared while deformation twins were sometimes observed. As discussed in supplemental material [8], a large twin is presumably generated by the fatigue deformation, as it is very unlikely the initial growth twins in the NC grains could align into such a large twin.

The evolving process exemplified in Figs. 2 and 3 suggests that the grain coarsening is related to dislocation accumulation at the GBs. Statistical analysis indicates that dislocation distribution in grains is grain size dependent. As shown in Fig. 4, the dislocations are predominantly located near GBs ($\delta \sim 0$) for grains smaller than 20 nm with a density of 1 order of magnitude larger than that in the as-received state, while a higher ratio of dislocations can be found in the center of larger grains (e.g., $d \sim 50$ nm). The dislocation accumulation in nanograins agrees with results of cold-rolled NC Ni [10], but seems to be in contradiction to some of the predictions by computer simulations that in NC grains discrete dislocations be emitted from GBs, and absorbed by the opposite boundary upon deformation, leaving no debris behind [11]. When the grain size is increased to the submicron scale, the dislocations are nearly evenly distributed in the grains ($\delta = 1$, corresponding to grain center) with a density of 5 times of that in the as-received state. Recent *in situ* TEM studies demonstrated that grain growth can be realized through grain rotation and agglomeration, and often occurs near crack tips [12,13]. Our observation indicates that the stress field built by the accumulation of dislocations at GBs should promote the rotation process. As suggested by Bobylev, Mukherjee, and Ovid'ko [14], the "dead-ended" dislocations are forced to climb along the GB with the assistance of a stress field, inducing global lattice rotation. With the lattice gradually aligned up, these large-angle GBs wither into sub-GBs, and the nanograins settle into subgrains [12].

Besides the dislocation activities, MD simulations suggest that deformation twinning may be an important mechanism for the crack growth during cyclic loading [6,7]. Our experimental results showed that, to the contrary, the formation of deformation twins is more likely to take place in coarsened rather than nanosized grains. Qualitatively, it is in agreement with the trend of grain-size-dependent twinning in Ni; i.e., the propensity of twinning may be enhanced with increased grain size [15,16]. Because of the growth twins, the twinning activities are more complicated in our sample. While cyclic deformation induces deformation twins (or stacking faults), a concurrent event is the removal of growth twins leading to coarsening. Despite the presence of deformation twins (or stacking faults), it is our observation that the removal of prevalent growth twins is the dominant event in the grains as they coarsen during the cyclic deformation. Stepwise detwinning may be realized by twinning partials emitting from crack tips, leading to ledged twin morphology [17]. Indeed, the stepwise twins in our sample [Fig. 3(c)] assume the characteristic detwinning ledges as in high purity Al and Cu [17,18]. With atomic simulation, detwinning of growth twins has been demonstrated through continuously shooting partials along TBs from GBs [19]. This seems to echo our experimental observations [Fig. 3(c)].

MD simulation has suggested intergranular crack propagation in NC Ni, which is assisted with nanovoid formation

in GBs. Since void nucleation and coalescence were not supported by our TEM results, apparently that crack growth mechanism does not apply to our current experimental conditions. Instead, the transgranular crack propagation observed in our crack path indicates that the crack propagation should be mediated by a dislocation slip-based mechanism [20]. Moreover, although the overall plastic strain imposed on samples is relatively small ($<2\%$ under the fatigue deformation), the deformation is under fairly high stress level (2.2 GPa, well above the yield stress of $\sim$1.8 GPa). Therefore, the plastic deformation associated with the crack tip nearby can be intense, which should drive substantial structure changes locally as the cracks propagate.

In summary, modulated grain coarsening is discovered in compressive cyclic deformation of NC Ni-Fe alloy with grain size of $\sim$19 nm. The modulation spacing is about equal to the mean spacing of cracks. This coarsening behavior was mainly driven by crack growth. Where the cracks are absent, the grain coarsening is not observed. Detailed TEM studies suggest that the grain coarsening is generated through the lattice reorientation under the high stress concentration at the crack tip, and this process is assisted with the dislocation accumulation during the cyclic deformation. The dislocations lodged in grain boundaries play an important role, causing rotations across the grain boundaries and, in the larger grains, resulting in a secondary subgrain structure. It is also revealed that twins resulting from the electrodeposition are removed by the cyclic plasticity.

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