

Supporting Information

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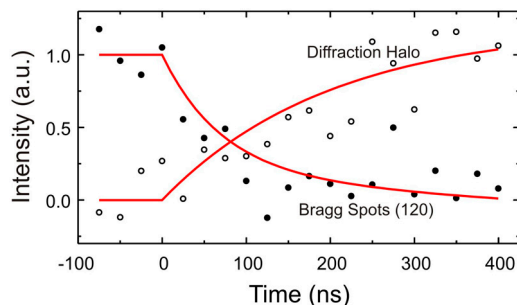


Fig. S1. Bragg spots vs. diffused ring diffraction. Following the T -jump, the loss of Bragg peak intensity (closed circles) due to ordered structure is concomitant with intensity increase of halo rings (open circles) due to the presence of disorder.

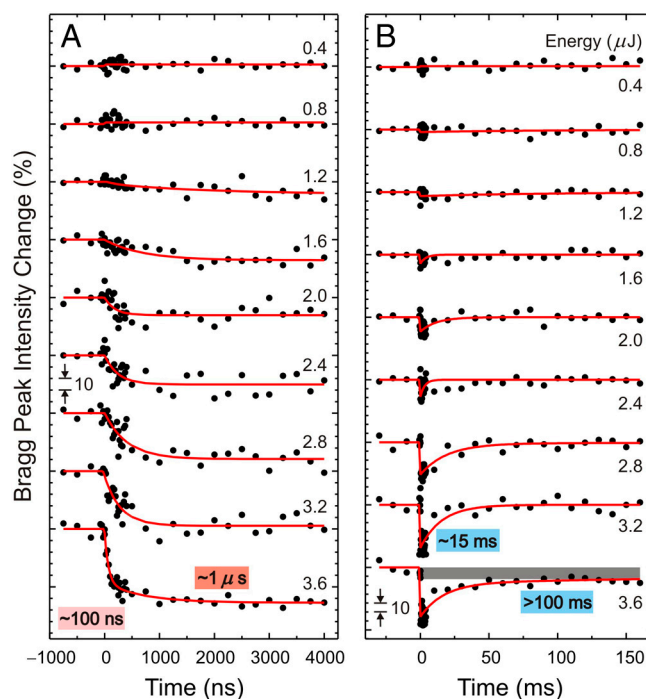


Fig. S2. Energy dependence of disordering and reordering. (A) Loss of diffraction and (B) recovery of diffraction. Shown are the temporal profiles of all Bragg (120) family of spots as a function of the pulse energy of the T -jump. The change at a given energy is normalized to that measured before t_0 . Up to approximately $1 \mu\text{J}$, the Bragg intensity does not change. From 1.2 to $3.2 \mu\text{J}$, a trend was observed; on the ns time scale, the diffraction intensity drops in nearly 100 ns and recovers in $15 \pm 8 \text{ ms}$. At the maximum energy of $3.6 \mu\text{J}$, an additional drop component (10–30%) on the time scale of approximately $1 \mu\text{s}$ appears and is also accompanied with a recovery component that decays on a longer than 100-ms scale. These results at the highest energy used are consistent with partial melting of nanoscale crystals in the imaged specimens. It is known that the melting temperature increases with the increase in the rate of heating (1) and at our rate of $>10^9 \text{ K/s}$ the melting temperature must be higher than the measured equilibrium value of 63°C (Fig. S3). Indeed, at $3.6 \mu\text{J}$ of energy we obtained a corresponding temperature of 70°C (Fig. 3C) that is a higher threshold above the equilibrium value. Due to their interfacial structure, the surface and coiled regions of the lamellae are expected to be the first to melt (2); note that if restructuring were to occur from a complete melt, the orientation of Bragg peaks must change because the crystal orientation is no longer preserved, and this was not observed here. Finally, a note regarding the loss of order in diffraction by the Debye–Waller effect follows: unlike monoatomic crystals such as gold (3), the intensities of diffraction here are sensitive to macromolecular helicity and chain conformations because of the nature of Bragg spots being probed and the fact that the effect of other helices in the unit cell at approximately 0.5 nm is remote. On the other hand, the Bragg peaks separations at such a distance reflect the change in displacement of each macromolecule from its equilibrium position (see text).

- 1 Madbouly SA, Wolf BA (2002) Equilibrium phase behavior of polyethylene oxide and of its mixtures with tetrahydronaphthalene or/and poly(ethylene oxide-*block*-dimethylsiloxane). *J Chem Phys* 117:7357–7363
- 2 Hu W, Frenkel D, Mathot VBF (2008) Free energy barrier to melting of single-chain polymer crystallite. *J Chem Phys* 118:3455–3457.
- 3 Barwick B, Park HS, Kwon O-H, Baskin JS, Zewail AH (2008) 4D imaging of transient structures and morphologies in ultrafast electron microscopy. *Science* 322:1227–1231.

