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Structural Health Monitoring relies on the ability of sensors to provide early warning about potential structural degradation. Sensors based on linear ultrasonics have been found to be effective in identification of flaws such as fatigue cracks, but they are reliable only when the flaw size is large enough that there is little remaining lifetime before they become critical. It is in this context that ultrasonic nonlinearity has been suggested as an effective tool for the early detection of fatigue damage in metallic structures. However, the correlation between ultrasonic nonlinearity and material damage induced by fatigue or plastic deformation has been observed to be non-monotonic by several researchers including by our group. To clarify the correlation between microstructure and ultrasonic nonlinearity, we investigate nonlinear ultrasonic behavior in model materials at multiple stages of loading: during monotonic loading; while under tension, at the loaded peak, while unloading, and after loading. The microstructures corresponding to changes in nonlinearity are examined using hardness testing and transmission electron microscopy. In the systems studied, ultrasonic nonlinearity does not monotonically increase with increased strain and dislocation density. Models that explain drops in nonlinearity are suggested, and destructive testing is used to support findings.