The magnetostriction of Fe-x at. % Ga (x=15, 20, and 27.5) was measured, for alloys processed under different directional solidification conditions, and the effect of partial substitution of Ga with Al on the magnetostriction of the alloys was examined. Magnetostriction measurements were carried out at different prestress levels varying from 0 to 55 MPa. Ga additions in the range of 15-27.5 at. % Ga in Fe were found to improve the magnetostriction of the disordered bcc phase of Fe by as much as 1 order of magnitude. The applied fields for saturation magnetostriction and the hysteresis observed were small. Magnetostriction values as high as $271 \times 10^{-6}$ were obtained in polycrystalline Fe-275 at. % Ga rods prepared using a directional growth (DG) process at a growth rate of 22.5 mm/h. This process, which is essentially a seedless vertical Bridgman technique, resulted in near [001] textured polycrystalline Fe-Ga alloys. The preferred [001] crystallographic orientation of the DG alloys was approximately 14° away from the rod direction. For Ga contents between 15 and 27.5 at. % in Fe, the Ga atoms increase the Fe-Fe spacing in the disordered bcc (A2) phase and reduce the magnetic moment of Fe. Substitution of Ga with Al has a significant effect on the magnetostriction of the Fe-Ga alloys. Small substitution of 5 at. % Al for Ga in the Fe-20 at. % Ga alloy increases the magnetostriction in Fe, and the value is slightly larger than that of the Fe-20 at. % Ga alloy. A higher substitution amount of Al tends to decrease the magnetostriction.