Many soft applications such as machine learning and probabilistic computational modeling can benefit from approximate but high-performance implementations. In this thesis, we study how Binary decision diagrams (BDDs) can be used to synthesize approximate high-performance implementations from high-level specifications such as program kernels written in a C-like language. We demonstrate the potential of our approach by designing nanoscale crossbars from such approximate Boolean decision diagrams. Our work may be useful in designing massively-parallel approximate crossbar computing systems for application-specific domains such as probabilistic computational modeling.