Abstract: This dissertation is concerned with formulating the problem and developing methods for the synthesis of optimal, open-loop inputs for large numbers of identically structured dynamical systems that exhibit variation in the values of characteristic parameters across the collection, or ensemble. Our goal is to steer the family of systems from an initial state (or pattern) to a desired state (or pattern) with the same common control while compensating for the inherent dispersion caused by the inhomogeneous parameter values. We compose an optimal ensemble control problem and develop a computational method based on pseudospectral approximations to solve these complex problems. This class of ensemble systems is strongly motivated by natural complications in quantum control, especially in magnetic resonance; however, similar structures are prevalent in a variety of other applications. From another perspective, the same methodology can be used to analyze systems that have uncertainty in the values of characteristic parameters, which are ubiquitous throughout science and engineering.