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Optimal control of high-dimensional Hilbert spaces.

The precise engineering of quantum states is a basic prerequisite for technologies such as quantum-enhanced sensing or quantum computing, a task which becomes more challenging with increasing dimension of the system Hilbert space. Here, we use quantum optimal control theory to derive shaped radio-frequency pulses to experimentally navigate the Stark manifold of a Rydberg atom. We demonstrate that optimal control, beyond improving the fidelity of an existing protocol, also enables us to accurately generate a nonclassical superposition state that cannot be prepared with reasonable fidelity using standard techniques.