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Quantum Control of Radical-Pair Dynamics beyond Time-Local Optimization

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We realize arbitrary-wave-form-based control of spin-selective recombination reactions of radical pairs in the low-magnetic-field regime. To this end, we extend the gradient-ascent pulse engineering (GRAPE) paradigm to allow for optimizing reaction yields. This overcomes drawbacks of previously suggested time-local optimization approaches for the reaction control of radical pairs, which were limited to high biasing fields. We demonstrate how efficient time-global optimization of the recombination yields can be realized by gradient-based methods augmented by time blocking, sparse sampling of the yield, and evaluation of the central single time-step propagators and their Fréchet derivatives using iterated Trotter-Suzuki splittings. Results are shown for both a toy model, previously used to demonstrate coherent control of radical-pair reactions in the simpler high-field scenario and, furthermore, for a realistic exciplex-forming donor-acceptor system comprising 16 nuclear spins. This raises prospects for the spin control of actual radical-pair systems in ambient magnetic fields, by suppressing or boosting radical reaction yields using purpose-specific radio-frequency wave forms, paving the way for reaction-yield-dependent quantum magnetometry and potentially applications of quantum control to biochemical radical-pair reactions. We demonstrate the latter aspect for two radical pairs implicated in quantum biology.
(https://journals.aps.org/prxquantum/abstract/10.1103/PRXQuantum.5.020303)

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