Feedback Quantum Control of Population Transfer

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The impossibility of exactly calculating the optimal laser pulse to control complex systems (such as polyatomic molecules in condensed phase environments) has led to the proposal that one use the sample itself to derive the optimal field [1]. In other words, a computer algorithm searches for the optimal field using feedback from the experiment to vary and improve the laser pulse. We present here an experimental implementation of this concept, applied to the maximization of electronic population transfer for the laser dye IR125 in methanol solution [2]. This is, to our knowledge, the first experimental application of feedback control to a chemical system.

Both the amplitude and phase of femtosecond laser pulse spectrum are controlled using a pulse-shaper based on the design of Warren *et al*. [3]. The shaped pulse excites fluorescence from a cell containing the dye solution, and the pulse energy and the fluorescence are detected simultaneously. The pulse shaper is controlled by a computer program which also reads the experimental measurements and uses a genetic algorithm to optimize the pulse shape. We use this algorithm to optimize two different experimental quantities: the fluorescence *efficiency* (the ratio of fluorescence power to laser power) and the fluorescence *effectiveness* (the fluorescence power only), which is related to the total population inversion.

Using experimental feedback, the algorithm converges to the optimum pulse shape within roughly 30 minutes in both cases. The most *efficient* pulse is simply a narrow bandwidth pulse tuned toward the peak of the absorption spectrum. The most *effective* pulse corresponds to a full power, full spectrum pulse with a strong positive chirp (i.e. the phase of the electric field has been modified so that low frequencies precede high frequencies in time). This has been analyzed theoretically [4], with the conclusion that such chirped, strong field control is an effective and robust way to achieve nearly 100% transfer of the electronic population. In both cases the optimization process is quite robust with respect to experimental nonidealities like noise and laser drift.

Finally, applications of shaped pulses for improving contrast in biological imaging and controlling photochemical reaction rates will be discussed.

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