Abstract Submitted for the MAR07 Meeting of The American Physical Society

Femtosecond pump – shaped-dump – probe quantum control PATRICK NUERNBERGER, Physikalisches Institut, PHILIPP MARQUETAND, Institut fuer Physikalische Chemie, GERHARD VOGT, TOBIAS BRIXNER, Physikalisches Institut, VOLKER ENGEL, Institut fuer Physikalische Chemie, GUSTAV GERBER, Physikalisches Institut, Universitaet Wuerzburg, Am Hubland, 97074 Wuerzburg, Germany — We present a three pulse pump-shaped-dump-probe scheme for femtosecond spectroscopy. The objective is a reversion of regular control schemes for optimal excitation in which the pump pulse is shaped. Instead, we seek optimal de-excitation with a shaped dump pulse. Besides variation of the time delay between pump and dump pulses, the versatility of a femtosecond pulse shaper furthermore allows to record systematic fitness landscapes as a function of selected pulse parameters, providing additional information on wave-packet evolution and the potential energy surfaces of the system under study. Since the dump pulse is independent from the pump pulse, the pump- shaped-dump-probe scheme facilitates control of molecular systems away from the initial Franck-Condon window in regions of the potential-energy landscape where the decisive reaction step occurs, e.g. near conical intersections. Experimental results on the retinal photoisomerization reaction in bacteriorhodopsin and exemplary model calculations demonstrate the potential of this new scheme.

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Date submitted: 21 Nov 2006

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