New Directions in Strong Field Coherent Control. From spinning tops to ultrafast switches

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Nonadiabatic alignment is a coherent approach to control over the spatial properties of molecules, wherein a short, moderately-intense laser pulse is applied to populate a broad rotational wavepacket with fascinating properties. In the limit of small isolated molecules, nonadiabatic alignment has evolved during the past 17 years into an active field of theoretical and experimental research with a rich variety of applications.

In the present talk we extend the alignment concept to complex systems, including large polyatomic molecules, dissipative media, nonrigid systems, molecular assembly, molecular conduction junctions and dense molecular ensembles. Following a review of the essential physics underlying alignment, we consider the case of asymmetric top molecules, where alignment overcomes the mechanisms that render the rotations unstable in the classical limit. Next we focus on dissipative media, and illustrate the application of rotational wavepackets as a probe of the decohering properties of the environment. We extend alignment to control the torsional motions of polyatomic molecules, and apply torsional control to manipulate charge transfer events in solutions, suggesting a potential route to light controlled molecular switches. Turning to interfaces, we introduce a route to guided molecular assembly, wherein laser alignment is extended to induce long-range orientational order in molecular layers. Combining the nonadiabatic alignment concept with recent research on nanoplasmonics and on conductance via molecular junctions, we develop an approach to coherent control of transport in the nanoscale (Fig. 1). Finally, we explore the case of dense molecular ensembles, where alignment generalizes into a collective phenomenon that gives rise to formation of molecular assembly with long range translational and orientational order, suggesting intriguing potential applications in material design.



Fig. 1: An ultrafast, nanoscale molecular switch based on combination of the concept of nonadiabatic alignment with nanoplasmonics. Reuter, Sukharev & Seideman, Phys.Rev.Lett, 101, 208303 (2008); Nature Photonics 3, 4-5 (2009); Phys.Rev. A 86 013426 (2012).