Spinning Tops: A Tale for Chanuca

Benjamin Ashwell, Sai Ramakrishna and Tamar Seideman

Northwestern University, Evanston, IL 60208-3113

t-seideman@northwestern.edu

Strong field, nonadiabatic alignment, a subfield of strong field coherent control, is an old problem but one that was the subject of much discussion and some controversy at our summer meeting in British Columbia, and was one of Moshe's favorite topics, although it was not part of his own research. It 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 decade into an active field of experimental and theoretical research with a rich variety of applications.

Given that most of the audience is familiar with this problem, and that our focus in the present meeting is on current and future trends, I will skip the introductory material as well as the theory and experimental methods and will focus on what I personally regard some of the interesting future directions. In particular, I will 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 brief review of the essential physics underlying laser alignment, we will consider the case of asymmetric top molecules, where alignment can win over the mechanisms that make 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

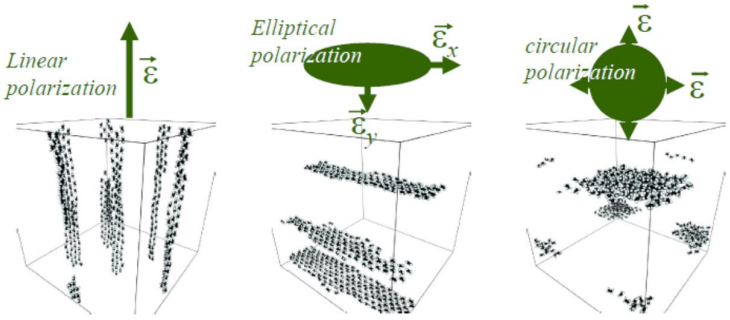

Fig. 1. Strong field-driven molecular assembly. Both the short-range order of the molecules within an assembly and the long range order of the assembly with respect to one another are controlled by the laser polarization.

M. Artamonov & T. Seideman, Phys. Rev. Lett. 109, 168302 (2012)

an approach to coherent control of transport in the nanoscale. 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.