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Spinning Tops in External Fields. From High Harmonic Generation to Control of Transport in the Nanoscale[1]

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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 in recent years into an active field of theoretical and experimental research with a rich variety of applications. Following a brief review of the essential physics underlying nonadiabatic alignment, we discuss one of these applications, namely the use of high harmonics generated from aligned molecules as a probe of the underlying electronic dynamics and rotational coherences. Next, we extend the alignment concept to dissipative media, including dense gases, solutions, and interfaces. We illustrate the application of rotational wavepackets as a probe of the dissipative properties of dense media and propose a means of disentangling population relaxation from decoherence effects via strong laser alignment. We extend alignment to control the torsional motions of polyatomic molecules, and apply torsional control in solutions to manipulate charge transfer events, 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. Finally, we combine the nonadiabatic alignment concept with recent research on nanoplasmonics and on conductance via molecular junctions to develop an approach to optical control of transport in the nanoscale.

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