

Controlling molecular orientation and alignment: From gas phase to condensed phases

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In recent years, the non-resonant interaction of intense laser fields with molecules through their anisotropic polarizability has been successfully employed to orient and/or align molecular axes. In our theoretical work, we characterize the quantum dynamics of pendular states using a semi-analytical approach. In particular, we study pendular analogues to coherent and squeezed states and derive closed expressions for wave functions and expectation values in certain cases.

If the duration of the light pulses used to align molecules exceeds the time scale of molecular rotation, free rotor states adiabatically transform into pendular states. In the contrasting, non-adiabatic limit, shorter pulses create wave packet states exhibiting revival phenomena thus giving rise to post-pulse alignment. Recently, we have shown how suitable combinations of adiabatic and non-adiabatic scenarios can be used to achieve spectral selection for non-resonant alignment.

In other work, we consider the alignment of molecules in gaseous mixtures. We show how the timescales of dissipation and decoherence can be separately determined from the post-pulse alignment signals thereby allowing to characterize underlying collision processes.

Finally, also the orientation/alignment of molecules embedded in rare gas matrices is studied. Depending on the relative strength of molecular interaction with the internal (crystal) field and the external (laser) field, possibilities and limitations for molecular alignment in host crystals are discussed.

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