Non-adiabatic effects in laser orientation and alignment of molecules

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We present a theoretical study of the laser-alignment and mixed-field-orientation of polar molecules. In these experiments, pendular states were created by means of linearly polarized strong laser pulses combined with tilted weak electric fields. The time dependent Schrödinger equation is solved into account the time profile of the experimental laser pulse and the theoretical results compared to the experimental observations. For perpendicular fields one obtains pure alignment, we demonstrate and analyze a strongly driven quantum pendulum in the angular motion of state-selected and laser-aligned OCS molecules. For tilted fields, we show that a fully adiabatic description of the process does not reproduce the experimentally observed orientation, and that it is mandatory to perform a time-dependent study taking into account the time profile of the laser pulse. Our results show that the adiabaticity of the mixed-field orientation depends at weak ac fields on the energy splitting of the states in a J-manifold, as well as on the formation of the quasi-degenerate doublets at stronger ac fields. These pendular doublets result in the transfer of population from a single occupied field-free rotational state into two strongly oriented and anti-oriented pendular states, reducing the overall orientation. Hence, we demonstrate that under ns laser pulses the weak dc field orientation is, in general, not adiabatic, so that a time-dependent description of the orientation process becomes mandatory. A revised condition for achieving adiabatic mixed-field orientation is provided.