Autler-Townes Effect in Atoms and Molecules with Hyperfine Structure

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The Autler-Townes (AT) effect [1] is the high-frequency analogue of the splitting of two degenerate levels by a dc Stark field. It has potential for development of new applications to molecular spectroscopy, like obtaining the transition dipole moment matrix element [2, 3] and as well as lifetime and branching ratios of highly excited molecular states using cw laser fields [3]. The possibility to achieve quantum state control using the AT effect has been studied by using the Nonresonant Dynamic Stark Effect (NRDSE) [4] and the Selective Population of Dressed States (SPODS) [5] techniques. An all-optical method for aligning non-polar molecules for chemical reactions was demonstrated [2b] based on AT effect and its dependence on the molecular magnetic quantum number (M_J). The latter enables resolution of individual M_J peaks, leading to additional state selectivity and better control of the molecular properties.

In our present work we extend the quantum control schemes to more complex energy level systems. In particular, the effect of coupling between nuclear and electronic angular momentum, which leads to occurrence of hyperfine structure, is studied in atomic and molecular level systems. Each hyperfine level is 2F+1-fold degenerate over the projection of F (i.e., M_F). Since the coupling of hyperfine components by the laser field depends on the hyperfine and magnetic quantum numbers F and M_F, this will lead to a different AT splitting for each involved hyperfine level. Simulations based on solving the density matrix equations of motion show that application of a strong coupling field leads to lifting of the M_F degeneracy, thus giving the opportunity to selectively address hyperfine components of excited atomic and molecular states. The latter is only true when hyperfine structure components can be resolved by the usual spectroscopic means. Further work is necessary to investigate the above effects in the case of unresolved hyperfine structure.

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