THE OPTICAL BICHROMATIC FORCE IN MOLECULAR SYSTEMS[a]

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The bichromatic optical force (BCF), which can greatly exceed radiative forces, seems ideal for laser slowing and cooling of molecules because it minimizes the effects of radiative decay. However, it relies on sustained coherences between optically coupled states, and molecules, with their many sublevels and decay pathways, present new challenges in maintaining these coherences compared with simple atoms. We have conducted extensive numerical simulations of BCFs in model molecular systems based on the $B \leftrightarrow X$ transition in CaF, and have begun experimental tests in a molecular beam.

In our modeling, the effects of fine and hyperfine structure are examined using a simplified level scheme that is still sufficiently complete to include the major pathways leading to loss or decoherence. To circumvent optical pumping into coherent dark states we explore two possible schemes: (1) a skewed dc magnetic field, and (2) rapid optical polarization switching. The effects of repumping to compensate for out-of-system radiative decay are also examined. Our results verify that the BCF is a promising method for creating large forces in molecular beams while minimizing out-of-system radiative losses, and provide detailed guidance for experimental designs. Compared to a two-level atom, the peak force is reduced by about an order of magnitude, but there is little reduction in the velocity range over which the force is effective. Our experiments on deflection and slowing using the CaF $B \leftrightarrow X$, (0-0) transition, still at an early stage, include studies of both the $P_{11}(1.5)/^{P}Q_{12}(0.5)$ branch, a quasi-cycling configuration with extensive hfs, and the $R_{11}(0.5)/^{R}Q_{21}(0.5)$ branch, which has a much simpler hfs but requires rotational repumping.

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