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Frequency Domain Control by the Autler-Townes Effect ERGIN AHMED, XINHUA PAN, MARJATTA LYYRA, Temple University — In recent years Quantum Optics has been expanded from atomic to molecular systems despite much weaker oscillator strengths and complex relaxation pathways that have presented serious challenges in the past. The richness of molecular excitation pathways and the variety of molecular interactions combined with the Autler-Townes (AC Stark) effect has made it possible to develop novel applications in this field,¹ such as control of molecular angular momentum $alignment^2$ and the electronic transition dipole moment function.³ In addition, the electric field amplitude of the control laser Rabi frequency can be used as a "tuning" mechanism for the mixing coefficients of energy levels that are weakly perturbed by the spin-orbit interaction,⁴ i.e. to control the valence electron spin polarization and the spin multiplicity of molecular quantum states. Experimentally we demonstrate that the enhancement of the spin-orbit coupling between a pair of weakly interacting singlet-triplet rovibrational levels depends on the Rabi frequency (laser power) of the control laser. We will discuss how this mechanism can be used to optimize collisional population flow between singlet and triplet electronic states, through such control of the spin-orbit perturbation based gateway levels.

¹E. H. Ahmed et al., Adv. At., Mol., Opt. Phys., Vol. 61, 467 - 514, (2012).
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³J. Qi, et al., PRL 88, 173003 (2002).
⁴E. H. Ahmed, et al., PRL 107, 163601 (2011).

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