Theoretical model of dynamic Stark induced degenerate vibronic state using two non-resonant shift up lasers

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Abstract: Recently we demonstrated by that the unidirectional π-electron rotation can be generated even in low symmetry aromatic ring molecules using the two linearly polarized lasers with a relative phase [1]. The key point of the mechanism of the coherent π-electron rotation originates from the degenerate electronic state of two quasi-degenerate electronic excited states by applying two non-resonant lasers. Here, vibrational degrees of freedom have not been taken into account. In our recent work [2, 3] we have taken into account the nuclear vibrational effects on the unidirectional π-electron rotation in the adiabatic approximation, where a weak coupling model of two electronic states with a few vibrational states is adopted [2, 3], where the two lowest vibronic states in two electronic excited states were set to be degenerate by two lasers (Dynamic Stark induced-Degenerate vibronic state DSIDVS). Here vibrational states in the electronic excited state 1 are shifted up by the laser *a*, and the lowest vibrational state in the electronic excited state 2 is shifted down by the laser *b*, on the other hand the other vibrational states in the electronic excited state 2 are shifted up by the laser *b*. Such opposite behaviours in the level shift of the vibronic states in the electronic excited state 2 make it complicate to describe the behaviours of dynamic Stark-induced vibronic states. In this work we propose theoretical scenario for the generation of coherent π-electron angular momentum in a low-symmetry aromatic molecule using two nonresonant lasers to avoid the complication mentioned above.

REFERENCES

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