ENANTIOMER-SELECTIVE POPULATION TRANSFER IN THE GAS PHASE USING PHASE-CONTROLLED RESONANT MICROWAVE FIELDS

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Chiral molecules are present ubiquitously in nature. They have two enantiomeric forms, which are mirror images of each other and cannot be superimposed onto one another through rotation. Due to the mirror-imaged mass distribution, the two enantiomers have the same moments of inertia (I_A, I_B, and I_C) in the principal axis system and thus have almost identical rotational signatures and cannot be distinguished with conventional microwave spectroscopy. In order to achieve chiral analysis for the chiral molecules in the gas phase, microwave three-wave mixing (M3WM) technique has been developed,[a] which exploits three-dimensional light-matter interactions in the dipole approximation. Beyond chiral analysis, this technique is further extended to achieve enantiomer-selective chiral control in the rotational state of interest, which is capable of inducing a state-specific enantiomeric excess "on the fly" when starting from a racemic mixture.

Previously, it has been reported that enantiomeric excess of about 0.5% and 6% were successfully generated using this approach with 1,2-propanediol[b] and carvone,[c] respectively. The enantiomer in excess can be induced selectively by tuning the phase of the microwave pulses. Here we present our recent investigation aimed at further improving the efficiency of this technique with solely microwave fields. We performed the population transfer experiment with a racemic sample of 2-trifluoromethyl oxirane and show that an enantiomeric excess of about 13% was induced by employing a population transfer scheme starting from the ground rotational state $|0_{00}\rangle$, which diminishes the spatial degeneracy of the rotational states. Furthermore, by depleting the initial thermal population with a resonant π-pulse or a microwave chirp in the rapid adiabatic passage regime, the obtained enantiomeric excess can be significantly improved to over 40%. These effects will be discussed in detail along with the theoretical simulations.

[a] D. Patterson, M. Schnell, J. M. Doyle, *Nature* **497**, 475–477 (2013).

[b] S. Eibenberger, J. Doyle, D. Patterson, *Phys. Rev. Lett.* **118**, 123002 (2017).

[c] C. Pérez, A. L. Steber, S. R. Domingos, A. Krin, D. Schmitz, M. Schnell, *Angew. Chem. Int. Ed.* **56**, 12512–12517 (2019).