A2.3 Precision experiments for parity violation in chiral molecules: the role of STIRAP

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Roadmap on STIRAP applications by

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Status. Precision experiments measuring the extremely small energy difference $\Delta_{pv}E$ between the enantiomers of chiral molecules, predicted to be of the order of 100 aeV to 1 feV (depending on the molecule), are among the greatest challenges in physical-chemical stereochemistry relating also to the standard model of particle physics (SMPP) [31–37]. So far, no successful experiments have been reported, and it turns out that STIRAP [38] may contribute importantly to enabling such precision experiments.

Following the discovery of parity violation in nuclear and particle physics in 1956/57 it has been surmised qualitatively that the ground-state energies of the enantiomers of chiral molecules are slightly different, as are also their absorption frequencies in the infrared or other spectral ranges. Thus, parity violation is of fundamental importance for our understanding of the structure and dynamics of chiral molecules with potential implications for the evolution of biomolecular homochirality, which has been an enigma of stereochemistry for more than a century (for in depth reviews see [31–34]). A scheme of how to measure the parity violating energy difference $\Delta_{pv}E$ between the ground state of the enantiomers of chiral molecules (and also $\Delta_{pv}E^*$ between corresponding excited rovibronic states) was proposed in 1986 [35]. At that time, however, the spectroscopic ground work of high-resolution analyses of rovibronic spectra of chiral molecules was not available and appeared very difficult. Also, theories available at that time were incorrect, predicting values too low by a factor of 100 for typical prototype molecules (see review [33]), and the proposed experiment on $\Delta_{pv}E$ appeared correspondingly almost impossible.

The situation has changed importantly over the last decades. Theoretical approaches developed by many groups over the last decades following our discovery in 1995 of the new orders of magnitude agree to converge today to the larger values. At the same time, there has been substantial progress in the high-resolution spectroscopy of chiral molecules, as well as in laser technology, enabling efficient and selective population transfer, therefore making the observation of $\Delta_{pv}E$ a realistic goal for our current experiments (see [33, 36]).

The basic experimental scheme is shown in figure 6. In brief, a first-step laser excitation of a chiral molecule (either enantiopure or simply from a racemate) leads to the preparation of an excited rovibronic state of well-defined parity, which is therefore achiral. Such a state can either arise from an excited electronic state with an achiral (for example planar) equilibrium structure, or it can arise from rovibrational-tunneling sublevels in the electronic ground state,

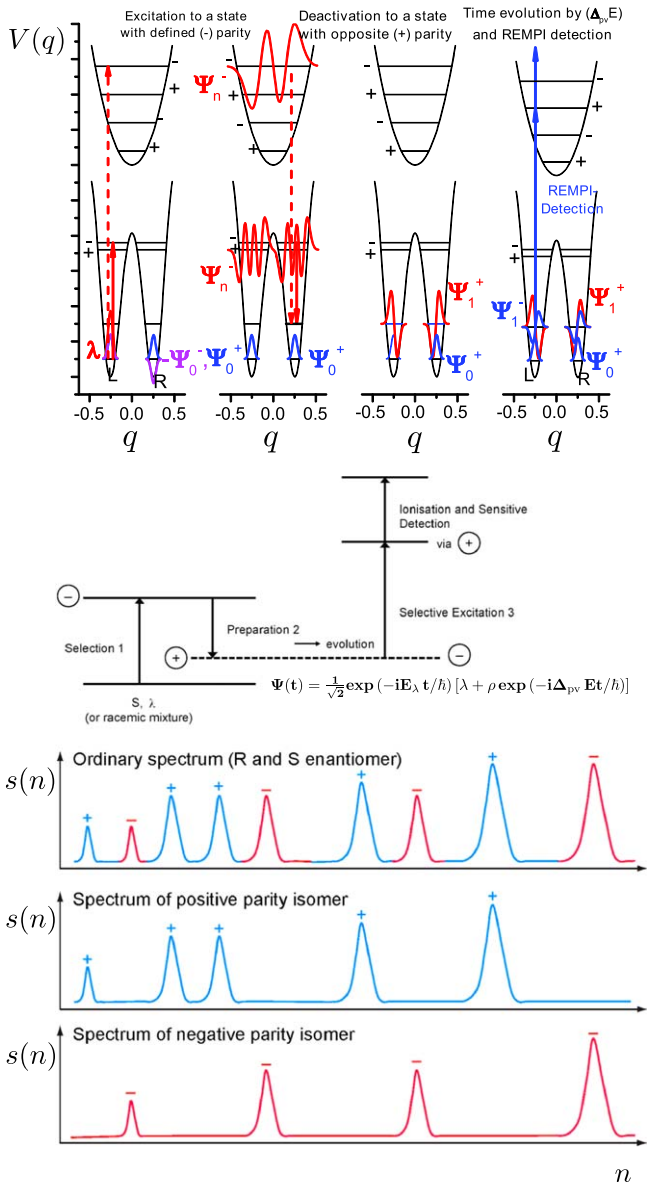

Figure 6. A scheme of the preparation and detection steps for the time resolved experiment to measure $\Delta_{pv}E$. Top: the transitions to the intermediate states are indicated together with the corresponding wave functions for an excited state with well-defined parity close to the barrier of a double minimum potential (full line) or an achiral electronically excited state (dashed line) as an intermediate. The right-hand part shows the sensitive detection step with REMPI. Middle: a summary scheme for the three steps. Bottom: the spectra of the normal enantiomers (top) and of the selected positive (blue) and negative (red) parity isomers (modified after [34–36]). Here, n is a reduced frequency difference $(\nu - \nu_0)/\nu_0$, where the frequency spacings between lines are of the order of MHz in order to separate lines connecting states of different parity (+ or −) in the rovibronic resolved spectrum. The high resolution (Hz to subHz) needed for $\Delta_{pv}E$ is obtained by measuring the time evolution of the spectrum in the middle towards the spectrum at the bottom at very high sensitivity in the ms timescale.

which are near or above the potential barrier for interconversion between the enantiomers. Such tunneling sublevels can therefore satisfy the condition that the tunneling splitting $\Delta E^*_{\pm}$ between sublevels of well-defined parity in that excited state

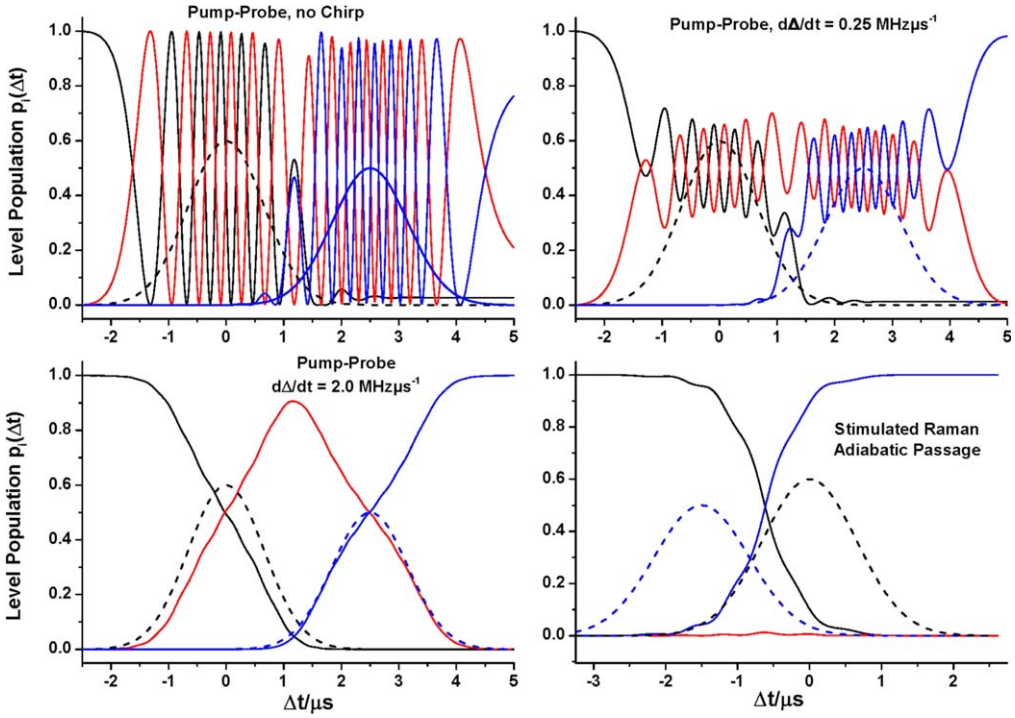

Figure 7. Time evolution of a three-level system exposed to two laser pulses nearly resonant with $|1\rangle \rightarrow |2\rangle$, and with $|2\rangle \rightarrow |3\rangle$ transition for different pulse conditions: Pump—Dump (Stokes), no frequency chirp (upper left), Pump—Dump (Stokes), small frequency chirp (0.25 $MHzs^{-1}$, upper right), Pump—Dump (Stokes), larger frequency chirp (2.0 $MHzs^{-1}$, lower left), STIRAP Dump (Stokes)—Pump (lower right). Time-dependent level populations: $|1\rangle$: black, $|2\rangle$: red, $|3\rangle$: blue. The laser pulses are indicated by the dashed line (Pump: black, Dump (Stokes): blue). Experimental conditions: vibrational transition moments: $\mu_{12} = \mu_{23} = 0.0262$ D, laser power: Pump: 0.6 W, Stokes: 0.5 W, pulse duration: $\tau = 1.31$ s. Reprinted from [36], with the permission of AIP Publishing.

is much larger than $\Delta_{\rm pv}E^*$,

$$\boldsymbol{\Delta E}^*_{\pm} \gg \boldsymbol{\Delta}_{\rm pv}\boldsymbol{E}^*. \tag{8}$$

This then allows for a spectroscopic selection of states of well-defined parity. In a second step in the scheme of figure 6 one prepares a state of well-defined parity in the ground state (or some other low energy state), which satisfies the condition

$$\boldsymbol{\Delta}_{\rm pv}\boldsymbol{E} \gg \boldsymbol{\Delta E}_{\pm}. \tag{9}$$

The parity selection arises from the electric dipole selection rule connecting levels of different parity. Thus, if in the first step one has selected a state of some given parity, in the second step one prepares a state of the opposite parity. Such a state is a superposition of the energy eigenstates of the two enantiomers separated by $\Delta_{\rm pv}E$, and therefore shows a periodic time evolution with a period

$$\tau_{\rm pv} = \frac{h}{\Delta_{\rm pv}E}. \tag{10}$$

In such a state parity evolves in time due to parity violation, and parity is not a constant of the motion. The probability of finding a given parity (p^+ for positive parity and p^- for negative parity) is given by equation (11)

$$p^-(t) = 1 - p^+(t) = \sin^2\left(\frac{\pi\boldsymbol{\Delta}_{\rm pv}Et}{h}\right). \tag{11}$$

In the third step the initially 'forbidden' population of negative parity $p^-(t)$ is probed very sensitively, for example, by resonantly enhanced multiphoton ionization (REMPI). This is possible, because the line spectra of positive and negative parity isomers are different, with lines that are well separated at high resolution (figure 6 and [37]). In this fashion it is not necessary to wait for a whole period, but it is sufficient to probe the initial time evolution at very early times. The sensitivity in the probe step determines in essence how small a value of $\Delta_{\rm pv}E$ can be measured. In a recent test experiment with a current experimental setup in our laboratory, on the achiral molecule ammonia, NH_3, it was estimated that an energy difference as small as 100 aeV should be measurable with the existing current experiment.

The original proposal of 1986 [35] preceded the invention of STIRAP [38], and therefore assumed population transfer using pulse shaping or chirp by rapid adiabatic passage (RAP). It is clear, however, that STIRAP is an ideal technique for generating population transfer in a well-controlled fashion.

Current and future challenges. Figure 7 shows simulations of population transfer using various methods, including STIRAP (see [36]). In the experiment using RAP we could demonstrate a population transfer efficiency for the combined two-step procedure of about 60%. Because of the better and more flexible control of experimental parameters in the STIRAP process [38], it should be possible to achieve a transfer efficiency near to 100%. The modifications needed to implement the STIRAP process in the current experimental setup are relatively straightforward, although not trivial. The

major current and future challenges are related to the much greater complexity of the rovibrational-tunneling spectra of chiral molecules compared to the test molecule NH_3 with the well-known spectra. However, the first spectroscopic investigations on two candidate molecules proved promising (1, 2-dithiine, $C_4H_4S_2$ [39] and trisulfane HSSSH [40]).

Advances in science and technology to meet challenges. The current CW-OPO laser systems (coupled to a frequency comb) only cover spectral ranges above about $2500\,cm^{-1}$ in the infrared. This limits the choice of molecules. Further development in laser technology, e.g. of quantum cascade lasers with power and resolution meeting our needs in the future, might make other molecules accessible, e.g. the simpler molecule ClOOCl, for which complete theoretical simulations of the experiment have been achieved already [37].

Concluding remarks. While the experiment to measure $\Delta_{pv}E$ might have appeared impossible, when it was first proposed in 1986 [35] the current outlook for a successful experiment is excellent. Indeed, provided that adequate funding for the continuation of the current project is guaranteed and required spectroscopic analyses can be completed, most significant results can be expected for any of two possible outcomes:

1. Either one finds experimentally the theoretically predicted values for $\Delta_{pv}E$, then one can analyze the results of the precision experiments in terms of the SMPP in a range not yet tested by previous experiments.
2. Or else one finds values for $\Delta_{pv}E$ different from the theoretical predictions. This will then lead to a fundamental revision of current theories for $\Delta_{pv}E$ also with the potential for new physics.

In addition, the experimental results will have implications for our understanding of the evolution of biomolecular homochirality.

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