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Ultrafast ignition of a uni-directional molecular motor

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Abstract. Light-driven molecular motors convert light into mechanical energy via excited state reactions. In this work we follow sub-picosecond primary events in the cycle of a two-stroke unidirectional motor by fluorescence up-conversion and transient absorption.

1 Introduction

Light driven molecular machines are designed to convert photon energy into mechanical energy, which can often be achieved through an excited state isomerisation reaction. For the operation of molecular motors it is also clearly essential to exercise control over the direction of motion. Unidirectional operation of light-driven molecular motors was first reported by Feringa and co-workers in their studies of chiral overcrowded alkenes (Fig. 1)[1]

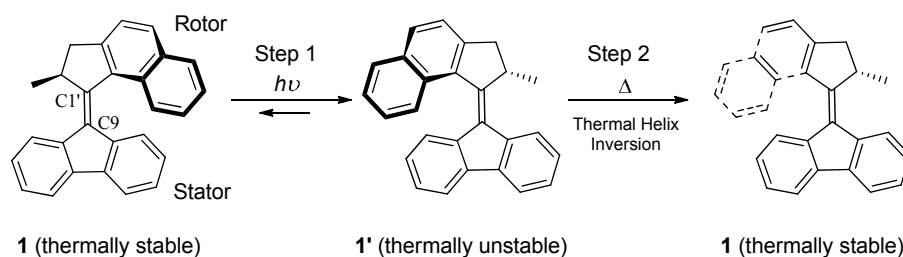


Figure 1 Structure of the molecular motor studied, indicating the rotor and stator of the motor linked by the double bond axle. The initial photochemical step is followed by the thermal stroke

In these molecules absorption gives rise to an isomerisation step which is followed by a thermal inversion in which the configuration at the stereogenic centre leads to rotation with an overwhelming preference for one direction. Subsequent sequential photo- and thermal steps result in one complete cycle. Key questions to be addressed include the rate of the isomerisation, its sensitivity to solvent friction, the extent to which the reaction occurs on the excited state surface and the possibility of controlling the rate of isomerisation through the use of shaped pulses.

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2 Results and Discussion

Time resolved fluorescence was measured at wavelengths spanning the emission spectrum. The decay on blue edge is ultrafast, revealing components on the order of 100 fs. As the observation wavelength increases the decay time lengthens, but no risetime is registered on the red edge of the spectrum (Fig. 2). Superimposed on the fast fluorescence decay is an oscillatory component which can be analysed in terms of one highly damped (90 fs) low frequency mode (113 cm^{-1}) and one less damped (220 fs) higher frequency mode (180 cm^{-1}); these are illustrated in the Fourier transform (inset). Such oscillations in the fluorescence are characteristic of vibrational modes in the excited state which are displaced with respect to the ground state. Ultrafast fluorescence is particularly well suited to the detection of such excited state modes

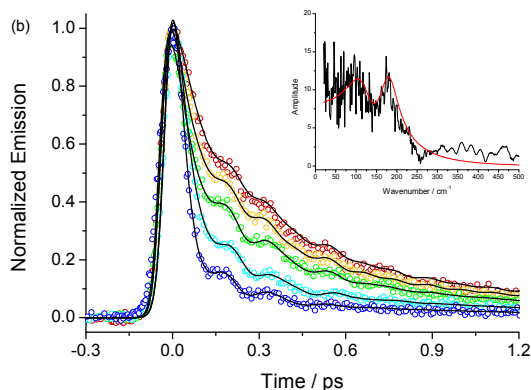


Figure 2. Time resolved fluorescence observed for the motor molecule of Fig. 1. Wavelengths span the spectrum from 455 nm (dark blue) to 556 nm (red). The inset shows a Fourier transform of the oscillatory part of the fluorescence.

The wavelength resolved time resolved fluorescence can be converted into time dependent emission spectra, which reveal that the spectral mean frequency shifts to the red as the integrated emission intensity collapses on a time scale of a few hundred femtoseconds. Subsequent to these fast events the fluorescence decays with a time constant of 1.4 ps. The oscillatory features evident in the wavelength resolved data of Fig. 2 translate to an oscillation in both the intensity and the mean frequency of the time resolved spectrum[2]. This shows that the low frequency modes excited are coupled to the electronic transition moment.

Transient absorption measurements reveal two spectral features (Fig. 3) with distinct kinetics. The lowest energy one is formed rapidly (within the 300 fs time resolution of the apparatus) and

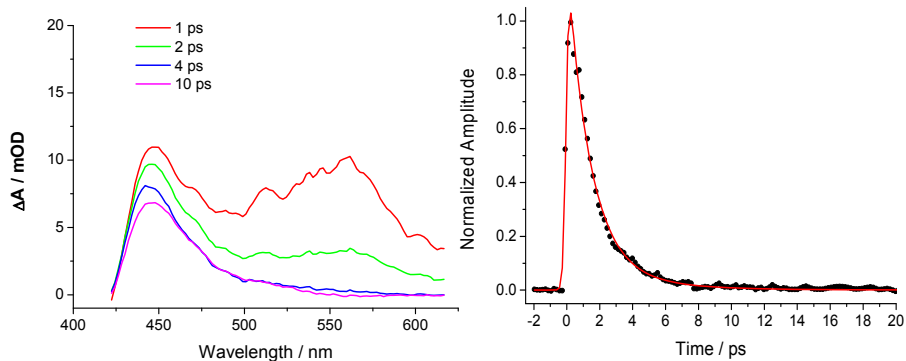


Figure 3. Transient absorption data for the motor in the visible region

