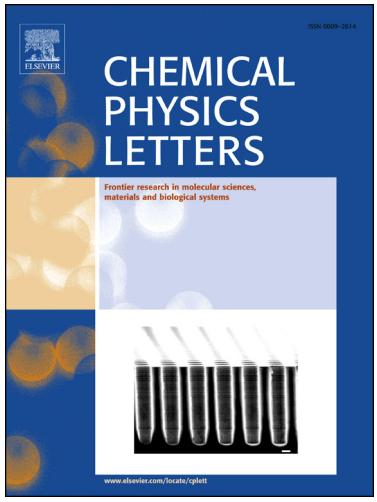

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Ab-Initio Surface Hopping and Multiphoton Ionisation Study of the Photodissociation Dynamics of CS_2

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Abstract

New *ab-initio* surface hopping simulations of the excited state dynamics of CS_2 including spin-orbit coupling are compared to new experimental measurements using a multiphoton ionisation probe in a photoelectron spectroscopy experiment. The calculations highlight the importance of the triplet states even in the very early time dynamics of the dissociation process and allow us to unravel the signatures in the experimental spectrum, linking the observed changes to both electronic and nuclear degrees of freedom within the molecule.

Keywords: Photodissociation, Photoelectron spectroscopy, Theoretical Chemistry, Non-adiabatic dynamics

1. Introduction

The dissociation dynamics of CS_2 following UV excitation have been a benchmark in chemical dynamics for many years, with numerous experimental studies in both the time and frequency domain, see for example [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. This lasting fascination with CS_2 can be traced