Direct Observation of Molecular Alignment in Intense Laser Fields by Pulsed Gas Electron Diffraction

Kennosuke Hoshina, Keiko Kato, Tomoya Okino, Kaoru Yamanouchi

Department of Chemistry, School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, JAPAN

Abstract. The anisotropic two-dimensional (2D) electron diffraction pattern of jet-cooled CS$_2$ in an intense nanosecond Nd:YAG laser field (1064 nm, ~0.64 TW/cm$^2$, 10 ns) was measured by a short-pulsed 25 keV electron beam packet (~7 ns) generated by irradiating a tantalum photocathode with the 4th harmonics of another nanosecond pulsed YAG laser light. It was confirmed from the numerical simulation of the observed anisotropic diffraction image that the alignment of the S-C-S molecular axis proceeds in the intense laser field along the laser polarization direction.

INTRODUCTION

Recent studies of molecules in intense laser fields (>10$^{11}$ W/cm$^2$) generated by short pulsed high-power laser radiation have revealed that molecules undergo characteristic processes such as alignment in the direction of laser polarization, structural deformation, multiple ionization, and Coulomb explosion forming singly or multiply charged atomic cations [1]. In the previous studies on the alignment [2] and geometric deformation [3] of molecules in intense laser fields, momentum vector distributions of fragment ions were measured. In most cases geometric structures of molecules just before the Coulomb explosion were estimated by solving the classical equation of motion of charged atomic fragments under the assumption that the force imposed among them was Coulombic. Therefore, a method for direct observation of the geometric structure of molecules during the laser-molecule interaction period has been awaited. We demonstrate that the alignment process of CS$_2$ molecules along the polarization direction of the intense nanosecond Nd:YAG laser field (~1 TW/cm$^2$) can be recorded as an anisotropic electron diffraction pattern [4,5].

EXPERIMENTAL

A pulsed electron beam was generated by the photoelectric effect [6-8] by irradiation of a tantalum photocathode surface of a Pierce type electron gun with the 4th harmonics (266 nm, ~50 μJ/pulse, 7 ns) of a Nd:YAG laser (GCR130 Spectra-Physics). The accelerated electron beam packets (25 keV) passed through an aperture (0.1 mm φ) and collided with a jet cooled CS$_2$ molecules in intense laser fields (~
produced by focusing a fundamental output (1064 nm, ~500 mJ/pulse, 10 ns) of another Nd:YAG laser. The temporal width of the electron beam packets was FWHM ~7 ns.

The 2D pulsed electron diffraction was projected onto a phosphor (P-31) screen with a diameter of 96 mm located \( L = 31.6 \) cm downstream from the electron-sample crossing point. The diffraction image appearing as an illumination pattern on the screen was projected through a camera lens onto an ICCD detector.

### RESULTS AND DISCUSSIONS

In the diffraction image obtained when \( \text{CS}_2 \) molecules were irradiated with a linearly polarized nanosecond YAG laser with an average intensity of \( F \sim 0.64 \) TW/cm\(^2\), a small but significant anisotropy was identified; the central region \( (s = 1.5 \sim 2 \) Å\(^{-1}\)) of the diffraction image is extended along the direction perpendicular to the laser polarization direction, where \( s = (4\pi/\lambda)\sin(\theta/2) \) with a de Broglie wavelength of electrons \( \lambda \) and a scattering angle \( \theta \). When the 2D diffraction image of \( \text{CS}_2 \) was measured without the laser field, the recorded halo pattern was concentric as expected for free molecules whose molecular axes are randomly oriented in space. Therefore, the anisotropic pattern observed in the intense laser fields indicates that the anisotropy was induced in the spatial distribution of the S-C-S molecular axis by the linearly polarized laser field.

In order to represent the aligned spatial distribution of the molecular axis, a distribution function, \( P(\chi) = 1 + \beta P_2(\cos \chi) \), was adopted, where \( \chi \) is an angle between the molecular S-C-S axis and the laser polarization direction. Assuming \( r(\text{S} \cdots \text{S}) = 2r(\text{C-S}) \), two parameters, \( r(\text{C-S}) \) and \( \beta \), were treated as independent parameters in the trial-and-error simulation of the observed anisotropic diffraction pattern, and the optimized parameters, \( r(\text{C-S}) = 1.56(3) \) Å and \( \beta = 0.17(7) \), were obtained.

The determined \( r(\text{C-S}) \) value agreed well with that of neutral \( \text{CS}_2 \) in the ground state, \( r_g = 1.5592(22) \) Å, indicating that the geometric structure of \( \text{CS}_2 \) does not change largely in an intense laser field (\( \sim 1 \) TW/cm\(^2\)). The \( <\cos^2 \chi> = 0.36(1) \) value converted from the alignment parameter \( \beta = 0.17(7) \) was well reproduced when the temporal and spatial variations of both the laser pulses and the electron beam packets were taken into account.

### REFERENCES