学位論文（要約）

Development of an apparatus for femtosecond laser-assisted elastic electron scattering with high-sensitivity and the observation of high-order multiphoton processes

（高感度フェムト秒レーザーアシステッド弾性電子散乱観測装置の開発と高次多光子過程の観測）

平成 28 年 12 月博士（理学）申請

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Abstract

When an electron is scattered by an atom or a molecule in a laser field, the energy of the scattered electron can be shifted by multiples of photon energy. This process is called laser-assisted elastic electron scattering (LAES). This thesis consists of two studies related to the LAES processes.

In the first study, multiphoton free-free transitions were observed in the LAES processes by Xe atoms in a femtosecond near-infrared intense laser field. The distinct peak structures at the energy shifts of *n*-photons (n = +1, +2, +3, +4, +5, and +6) were identified in the observed energy spectrum, and the energy and angular distributions of the LAES signals were in good agreement with those obtained by numerical simulations based on the Kroll-Watson theory. The LAES signal intensities at the scattering angles at 9.1 deg. and 11.8 deg. exhibited a clear plateau structure as a function of the harmonic order *n*, and the mechanism of these non-perturbative LAES processes was interpreted by a classical mechanical description.

In the second study, a second-generation apparatus equipped with an angular-resolved time-of-flight electron analyzer was developed for observing the LAES processes in femtosecond intense laser fields. The energy and the scattering angle of the scattered electrons by Xe atoms in the absence of the laser fields were measured. From the energy spectra of the scattered electrons, the energy resolution of the apparatus was found to be sufficiently high to detect the femtosecond-LAES signals. The detection efficiency was improved by ~50 times as high as that achieved in the first-generation apparatus used in the first study.

Contents

1. Introduction

Changes in the geometrical structure of molecules occur on a femtosecond timescale, and in the case of electrons within the molecules or atoms, the motion occurs on a sub-femtosecond or few-femtosecond timescale. The motivation of my research is to investigate these dynamics, and for this aim, laser-assisted elastic electron scattering process was utilized.

As shown FIG. 1(b), laser-assisted elastic electron scattering (LAES) is a process in which electrons change their energy by $n\hbar\omega$ (n: integer, $\hbar\omega$: photon energy) through elastic scattering by atoms or molecules in a laser field [1]. The LAES process occurs only when three beams, i.e., an atomic beam, an electron beam, and a laser beam, collide with each other, and consequently, three different kinds of interactions among them, i.e., the electron-atom interaction, the laser-electron interaction, and the laser-atom interaction, are involved in the LAES process, attracting the interest of researchers in collision physics as well as in laser physics and chemistry [2-5].

Kroll and Watson developed the theoretical framework of the LAES processes [6] and categorized the LAES processes into three regimes, i.e., (i) perturbative regime, (ii) intermediate regime, and (iii) nonperturbative regime. When the LAES process is

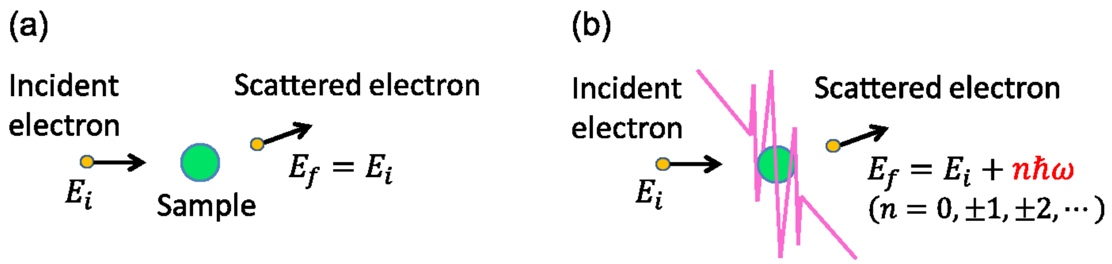

FIG. 1. Schematic of (a) the ordinary elastic electron scattering and (b) the LAES processes. When the electron with the energy of E_i is elastically scattered by the sample in the absence of the laser field, the energy of the scattered electron, E_f, is equal to that of the incident electron and is expressed as $E_f = E_i$. When the electron is scattered by the sample in the laser field, the energy of the scattered electron can be shifted by multiples of photon energy and is expressed as $E_f = E_i + n\hbar\omega$.

in the nonperturbative regime, the process can be treated as the dynamics of the classical electron-particle, and therefore, the time when the electron-atom collision occurs can be estimated from the observable momentum of the scattered electrons by tracing the trajectory of the electron-particle. Consequently, the LAES process can be utilized to the investigation of ultrafast dynamics in target atoms and molecules. In the present study, high-order multiphoton LAES processes in the nonperturbative regime by Xe atoms in a mode-locked femtosecond intense laser field (λ = 800 nm, Δt = 100 fs, I = 8.8×10^{12} W/cm^2) were observed by using the apparatus [7] used in our previous studies, and I showed that we can interpret the ultrafast electron collision in the LAES processes by classical mechanics and that slight differences in the electron-atom collision times of the order of 10 attoseconds can be discriminated. This classical mechanical interpretation of the LAES processes in terms of the collision time will be of use for the investigation of ultrafast electron collisions by target atoms and molecules by the measurements of the LAES signals.

Furthermore, from the femtosecond-LAES measurements, we are able to investigate ultrafast responses of the electrons within an atom in an intense laser field [8] and determine temporal variation of geometrical structure of molecules in the gas phase in the course of chemical reactions with femtosecond temporal resolution [9]. In 2014, we demonstrated that the instantaneous geometrical structure of static CCl_4 molecules during the femtosecond laser pulse duration can be determined by laser-assisted electron diffraction (LAED) [10]. However, the femtosecond-LAES or LAED signals were in general extremely weak, and significant improvements in signal-to-noise ratios of LAES signals are necessary for time-resolved measurements. In the present study, I have developed a second-generation femtosecond-LAES apparatus with a high collection efficiency of scattered electrons in order to achieve high signal-to-noise ratios of LAES signals.

Chapter 2 gives the study of the high-order multiphoton LAES processes, Chapter 3 gives the details of the second-generation apparatus, and Chapter 4 gives a summary of this thesis.

第 2 章

　本章については、5 年以内に雑誌等で刊行予定のため、非公開。

第 3 章

本章については、5 年以内に雑誌等で刊行予定のため、非公開。

4. Summary

Two studies which relate to the femtosecond-LAES processes were described in this thesis.

In the first study, I performed the measurement of high-order multiphoton LAES processes in a femtosecond near-infrared intense laser field (Δt = 100 fs, λ = 800 nm, I = 8.8×10^{12} W/cm^2), and succeeded in observing the signals of the scattered electrons with n-photon (n = +1, +2, +3, +4, +5, and +6) energy shift. In the energy spectra of the scattering angles at 9.1° and 11.8°, clear plateau structures were identified. I interpreted the mechanism of the LAES signals in the plateau region in terms of classical trajectories of the electron in the well-defined laser electric field, and slight differences in the electron-atom collision time of the order of 10 attosecond were discriminated for each n-photon LAES signals. The result of this measurement will be of use to measure the electron dynamics within an atoms or molecules in intense laser field.

In the second study, I have developed the second-generation apparatus for femtosecond-LAES measurements equipped with the angular-resolved time-of-flight electron energy analyzer. The detection efficiency became around 50 times as high as that achieved in the first-generation apparatus. By performing measurements with this apparatus, the time-resolved pump-probe LAED measurement, i.e., the determination of the geometrical structure change of the isolated molecules in the course of chemical reactions with sub-10 fs temporal resolution, can be realized. Furthermore, the measurement of the high-order multiphoton LAES signals around the small angle region, from which the electron dynamics within an atom in intense laser fields can be investigated, will also be achieved by using the new apparatus.

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