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学位論文題目	Using high-precision coherent control to investigate molecular wave-functions distorted by intense femtosecond laser pulses; Toward a model study of decoherence
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論文内容の要旨

Introduction

Coherent control is a technique that manipulates the interference of wave functions by tuning their amplitudes and phases using coherent light. It is a basic scheme of controlling a variety of quantum objects ranging from simple atoms to nano structures with possible applications to novel quantum technologies such as bond-selective chemistry and quantum computation. Besides those technological applications, coherent control is heavily indebted to the wave nature of matter so that it should be a promising approach to explore quantum world-view. Although almost 90 years have passed since quantum mechanics was founded in 1920's, it is not yet well understood how the microscopic quantum world, where matter waves are clearly seen, is smoothly connected to the macroscopic classical world without wave-nature of matter. One possible mechanism of this quantum-classical boundary is decoherence. Decoherence is the process in which a quantum system loses its wave nature as a result of the interaction with its surrounding environment. In this study the authors have employed their high-precision coherent-control with attosecond-precision, which has been developed in their research group, to better understand the mechanism of decoherence.

One possible straightforward approach to the test of decoherence is measuring the fringe-contrast of matter-wave interference as a function of the size and complexity of environment. Another approach will be the use of synthesized electric fields to mimic the interactions with environment. The authors have employed the latter approach in the present study. They consider collision-induced decoherence of a diatomic molecule. When a molecule is collided by another particle, the temporal oscillation of its wave function is phase-shifted and its population is redistributed. These collision-induced dephasing and population redistribution are the elementary processes of decoherence. In an ensemble of molecules, collisions occur randomly to be incoherently superposed in real experiments, even with the maximally coherent ensemble such as BEC. Fundamental physics that dominates atomic and molecular collisions are electric interactions between two particles each of which is surrounded by an electron cloud. The impulsive electric interaction during a collision scrambles these electron clouds in a very short time scale which typically ranges from femtoseconds (fs) to picoseconds (ps), depending on the temperature of the ensemble. Because of this ultrafast timescale and randomness, each individual collision is impossible to be observed selectively. However, one can use an external electric field, ideally designed to mimic such an ultrafast electric interaction during the collision. The only possibility to prepare such an ultrafast designed electric field is to use a laser pulse. An ensemble of molecules can be irradiated with a strong fs laser pulse so that it interacts with that laser field to serve as a simulator of the ultrafast electric interaction during a single collision, in which the electronic cloud is scrambled on the same ultrafast timescale.

The authors have applied this idea to a jet-cooled ensemble of the iodine molecules. The iodine molecule is irradiated with a fs laser pulse to create a vibrational wave packet (WP), which is a coherent superposition of multiple vibrational eigenstates. The molecule is then irradiated with an intense fs near infrared (NIR) laser pulse to induce population redistribution and phase-shift of each individual eigenstate.

Experiments

The iodine molecules were prepared in the ground electronic state (X) by an expansion of heated I_2/Ar mixture into a vacuum chamber through a nozzle. The authors conducted two series of experiments with this iodine sample; (i) the population redistribution measurement and (ii) the combination of the phase measurement and the fs probe measurement. In (i) the population redistribution measurement, the initial vibrational WP was created on the excited-state (B) with a 540 nm fs pump pulse and then irradiated with a 1400 nm fs pulse to mimic the impulsive electric interaction during a single collision (hereafter referred to as "NIR" pulse). This WP was interrogated with a narrowband nanosecond (ns) probe pulse so that the population of each eigenstate is observed state-selectively, by the laser-induced fluorescence (LIF) technique with the E state being the fluorescent state. In (ii) the combination of the phase measurement and the fs probe measurement, on the other hand, they employed a 535 nm fs pump pulse and an 800 nm fs NIR pulse to mimic the impulsive electric interaction. For the phase measurement, this pump pulse was input to an attosecond phase modulator (APM), which is their homemade Michelson-type interferometer highly stabilized in another vacuum chamber to generate a pair of laser pulses whose delay was stabilized on the attosecond time scale. With this APM, they conducted ultrahigh-precision WP interferometry, and observed phase shift of each eigenstate due to the NIR pulse radiation, employing a ns probe pulse for the state-selective measurement. As to the fs probe measurement, a 430 nm fs probe pulse was employed for the real-time observation of the WP temporal evolution.

Result and discussion

In the population redistribution measurement, the authors observed that the population of each eigenstate was modulated and showed oscillation as a function of the delay between the pump and NIR pulses with the period almost equal to the classical vibrational period of the WP. This oscillatory modulation of population is elucidated in terms of the population redistribution among the eigenstates within the WP, due to the NIR pulse radiation. As to the phase measurement, the phase shift on the each eigenstate was observed and the amount of the phase-shift depended on the timing of the NIR pulse so that it can be controlled. Furthermore this phase-shift control has allowed us to decelerate and accelerate the spreading of the WP, which arises from dephasing among the multiple eigenstates within the WP induced by potential anharmonicity. Comparison with theoretical simulation shows that these findings can be elucidated by the distortion of the relevant potential curve induced by the NIR pulse. The population redistribution and phase-shift of each individual eigenstate observed in the series of experiments demonstrate that the present experiment serves as a good simulator of an ultrafast electronic interaction during a single binary collision.

博士論文の審査結果の要旨

本論文は、４章から構成されている。

第１章『Introduction』では、研究の背景、目的、意義、概要について述べている。デコヒーレンスが波動-粒子二重性や量子-古典境界といった量子論的な世界観の謎を解明する上で重要な現象と考えられる事、そのデコヒーレンスの素過程として他の粒子との衝突による分子波動関数の振幅と位相の変動に着目する事、しかしながら個別の衝突を分離して観測する事は不可能なので近赤外領域のフェムト秒レーザーパルス（以下、NIRパルス）によって衝突の際の撃力的な電気的相互作用をシミュレートする事、等が述べられている。

第2章『Modulation on the population of each eigenstate – Laser Induced Interference』では、ヨウ素分子の電子励起状態に発生させた振動核波束にNIRパルスを照射した際に起こる各振動固有状態のポピュレーション変動について述べている。NIRパルスの照射のタイミングを掃引すると、単一の固有状態の観測にも関わらず、複数の固有状態を重ね合わせて初めて観測されるはずのビートが観測され、このビートがNIRパルスの照射によって誘起されるコヒーレントなポピュレーション移動によって説明される事を明らかにしている。NIRパルスの照射によって、単一の衝突過程で起こるポピュレーション移動をシミュレートできることがわかった。

第3章『Modulation on the phase and the wave-packet evolution』では、第２章と同じくヨウ素分子の電子励起状態に発生させた振動核波束にNIRパルスを照射した際に起こる各振動固有状態の位相変動について述べている。位相変動は波束干渉法を応用した量子状態ホログラフィーと呼ばれる手法によって測定している。これはNIRによって擾乱を受けた波束に既知の参照波束を重ね合わせて干渉させ、この干渉信号から擾乱を受けた位相情報を再構築するという手法である。その結果、NIRパルスの照射によって各固有状態の位相がシフトする様子が観測された。第2章のポピュレーション移動の場合と同様に、NIRパルスの照射によって複数の振動固有状態がコヒーレントに混ざり合うことで位相がシフトするものと理解される。NIRパルスの照射によって、単一の衝突過程で起こる位相シフトをシミュレートできることがわかった。また、位相シフトの量は、NIRパルス照射のタイミングと振動準位に依存して変化した。このことを利用して、NIRパルスの照射のタイミングを調節する事によって、ポテンシャルの非調和性によって誘起される振動固有状態間のdephasingを遅らせたり早めたりすることに成功している。

第4章『Conclusion』では、本研究で得られた結果をまとめている。

以上、本研究はデコヒーレンスの素過程を超短パルスレーザー電場によってシミュレートするという新たな実験手法を提唱し、その有効性を実証しただけでなく、デコヒーレンスの素過程の一つである波動関数の位相のずれを修復するための基盤技術を開発したものであり、理学上貢献するところが大である。よって、本論文は博士(理学)の学位論文として十分に価値のあるものと認める。