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Journal of Photochemistry and Photobiology A: Chemistry 145 (2001) 17–21

Journal of Photochemistry and Photobiology A:Chemistry

www.elsevier.com/locate/jphotochem

Development of attosecond optical-phase manipulation for the wave-packet engineering

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Received 10 April 2001; received in revised form 21 May 2001; accepted 4 July 2001

Abstract

We have developed the "attosecond phase modulator" (APM) for the manipulation of optical phases with an accuracy of attoseconds. The interference of two 300 fs laser pulses at 252 nm has been taken by using this APM; the time delay between those two pulses is tuned with an accuracy of attoseconds by APM to induce constructive and destructive interferences. The interferogram measured by scanning the time delay shows a high-frequency oscillation with a period of 840 as. Quantum mechanical calculations have been performed to demonstrate that APM is useful in manipulating the relative phase of two nuclear wave packets created in a single molecule and controlling their interference with an accuracy of attoseconds. Applications of such an ultraprecise phase-manipulation technique are discussed to propose a possible new field of science and technology which we call the "attosecond wave-packet engineering".

Keywords: Wave-packet engineering; Attosecond phase modulator; Optical interference

1. Introduction

Suppose you shine a femtosecond optical pulse on a diatomic molecule with its bandwidth broad enough to cover several vibrational levels in the excited state. Then you can create a nuclear wave packet which is spatially localized and is no longer stationary. The wave packet thus created is the linear superposition of several eigenstates and follows classical motions such as stretching and shrinking that are readily understandable with our ordinary feeling. Now, a question arises: what is the relation between the wave packet and the optical pulse? The optical pulse is a periodic function oscillating with a period from attoseconds to a few femtoseconds. And its phase is transported into the molecule and stored as the quantum phase of the nuclear wave packet. If you can manipulate the phase of the optical pulse with an accuracy of attoseconds, you can precisely manipulate the quantum phase of the wave packet with the same accuracy. This precise quantum-phase manipulation serves as the heart of the "attosecond wave-packet engineering", we are proposing in this paper, and we believe its application could contribute to the drastic development of various fields such as material, biological, and information sciences and technologies in this new century.

The quantum-phase manipulation should allow you, for example, to control the interference of two nuclear wave packets created in a single molecule to induce constructive and destructive interferences for enhancement and disappearance of the probability of finding the molecule. In this interferometric control, the sequence of two identical ultrashort laser pulses generates two identical wave packets in the molecule. The relative phase of these two wave packets are controlled by tuning the relative phase of the laser pulses with an accuracy better than the optical period of the laser field. Scanning the relative phase of the laser pulses is thus expected to give an interferogram which shows a high frequency oscillation corresponding to the temporal interference of wave packets modulated by a slow envelope arising from the wave-packet motion in the excited state. This high-frequency component could be directly related to the matter-wave interference or atomic interferometry in the atom optics [1–3]. Scherer et al. have employed a beautiful phase-lock technique to eliminate rapidly oscillating interfernce and extracted slow periodic envelope resulting from the wave-packet motion in the excited state of I_2 [4]. Later on, Blanchet et al. have succeeded in scanning the time delay between two femtosecond laser pulses with an accuracy better than the period of the interference and have resolved the rapid oscillation arising from the intereference of two nuclear wave packets in the B state of Cs_2 [5]. The period of the observed oscillation is about 2.7 fs, which is

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1010-6030/01/$ – see front matter
PII: S1010-6030(01)00560-3

in agreement with the optical period of the laser field at 769 nm they have used. This is indeed the only one example where the temporal wave-packet interferogram has ever been taken for the nuclear motion.

The wave-packet interferogram has been so far taken in the near-IR and IR wavelength regions, so that the period of the observed oscillation has been no shorter than 2 or 3 fs. If one can manipulate optical phases with an accuracy of attoseconds, much more precise control of wave-packet interference becomes possible even in the UV and X-ray regions. The application of such a technique greatly enhances the future possibilities of the wave-packet engineering including material, biological, and information sciences and technologies. From these scientific and technological points of view, we have developed the "attosecond phase modulator" (APM) for the manipulation of optical phases with an accuracy of attoseconds. The optical interference of two temporally-overlapped 300 fs laser pulses at 252 nm has been taken by using this APM; the relative phase between those two pulses is tuned with an accuracy of attoseconds by APM to induce constructive and destructive interferences. The interferogram measured by scanning the relative phase shows a high-frequency oscillation with a period of 840 as. Moreover, we have carried out a quantum mechanical simulation where the sequence of two identical 300 fs laser pulses generated by APM creates two identical nuclear wave packets in the $A^3 0^+$ state of the single Hg–Ar van der Waals (vdW) complex. In this simulation, it is found that APM is useful in manipulating the relative phase of those two nuclear wave packets and controlling their interference with an accuracy of attoseconds. Applications of such an ultraprecise phase-manipulation technique are discussed to propose a possible new field of science and technology which we call the "attosecond wave-packet engineering".

2. Experiment

Fig. 1 shows a schematic of the present experimental setup. The output of a mode-locked Ti:Sapphire laser pumped by Ar^+ laser was amplified by a double-stage regenerative amplifier pumped by two 10 Hz Nd:YAG lasers to give the final output around 760 nm. It was frequency tripled to generate a 300 fs pulse around 253 nm. It was introduced into APM which we have developed, and a pair of 300 fs pulses is generated. Fig. 2 shows the inside of APM. The input pulse was devided by a half mirror and then recombined on another half mirror to generate the pulse pair. The delay Δt between two of those output pulses was coarsely tuned by sliding the mechanical stage placed on one of their optical paths. The minimum step of this coarse tuning was about 3 fs. An Ar cell was placed on the other path, and its pressure change was used for the fine tuning of Δt with an accuracy better than the optical period of the

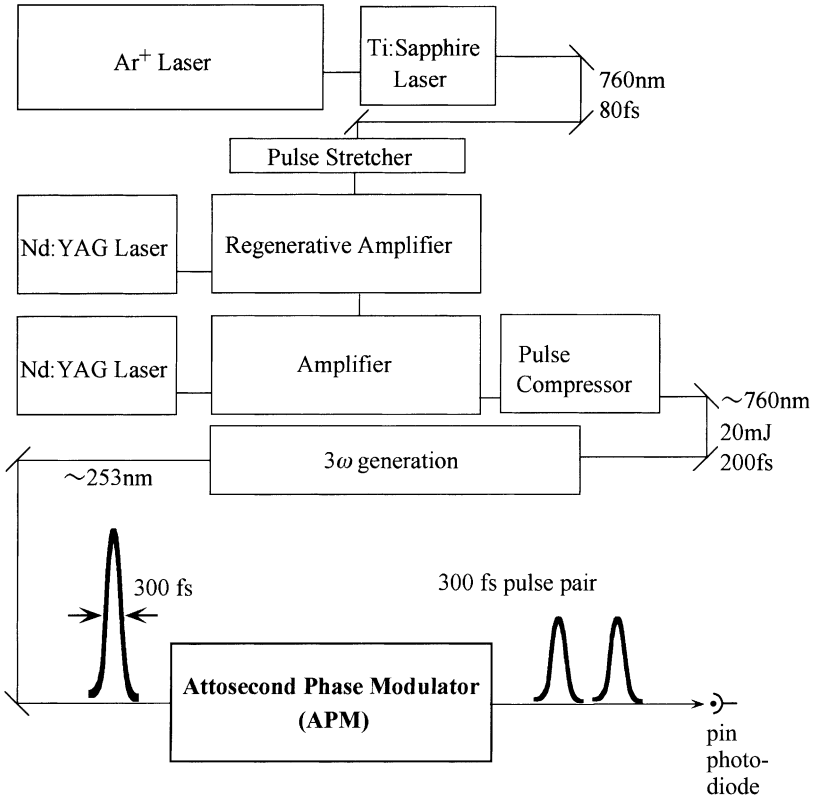

Fig. 1. Schematic of the experimental setup.

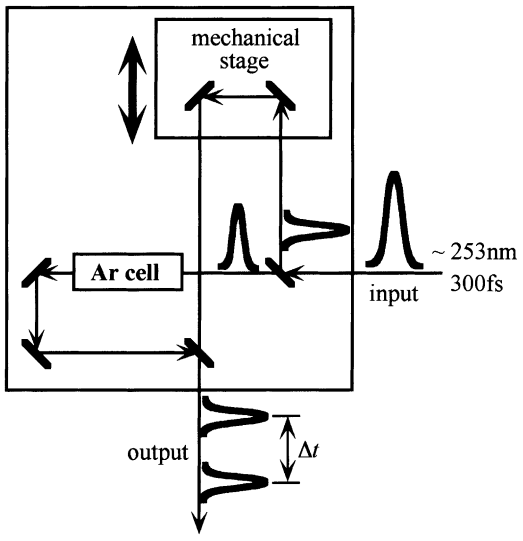

Fig. 2. Attosecond phase modulator (APM).

pulses, so that the relative phase of the output pulses was precisely tuned. For the present measurement of the optical interference, Δt was set around 0.0 ps by the mechanical stage so that the two output pulses were temporally overlapped. The intensity of the superposed pulse was measured by a pin photodiode.

3. Results

We have measured the optical interference of the two 300 fs pulses at 252 nm by scanning the Ar pressure with Δt set around 0.0 ps by the mechanical stage. The measured interferogram is shown in Fig. 3 as a function of the pressure of the Ar cell. The period of the oscillation corresponds to 840 as, which is the optical period of the present laser fields. It is seen from this figure that our APM can manipulate the optical phase with an accuracy of attoseconds. The intensity ratio of the two femtosecond pulses is 2:3 for this measurement, and this inequality is the major source of the background in Fig. 3.

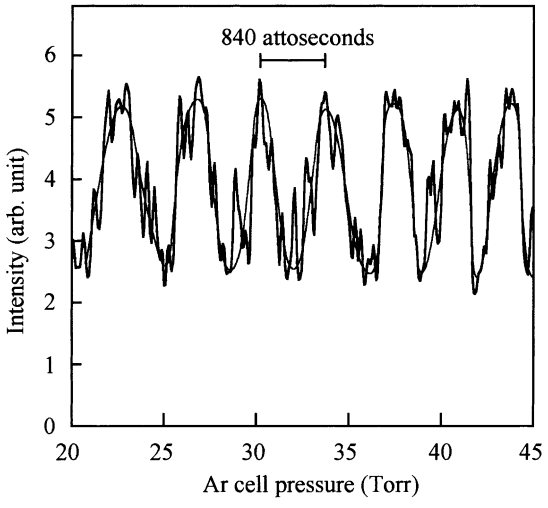

Fig. 3. Optical interference of two 300 fs laser pulses tuned to 252 nm. The period of the interferogram is 840 as.

4. Discussion

In our previous experiment, we have succeeded in creating a spatially localized vibrational wave-packet on the excited state potential of the Hg–CO vdW complex generated in a supersonic beam using the present femtosecond laser system [6]. We have also theoretically shown that the interference of those wave packets can be controlled by the present phase-locked pulse pair [7]. Here, we design an experiment where a similar interference control is performed in the Hg–Ar vdW complex. Hg–Ar is an ideal candidate for the target of the experimental trial since it is a simple one-dimentional system in the nuclear coordinate, and the vibrational-level spacing of its A^30^+ state matches our 300 fs laser pulse for the creation of a stable vibrational wave packet. Moreover, its potential curves have been fully investigated in a number of spectroscopic studies [8].

Fig. 4 shows a schematic of the designed experiment. The Hg–Ar vdW complex is irradiated by the sequence of two identical 300 fs pulses generated by APM. The wavelength of the pulses is tuned to 254.3 nm so that the $v = 3$, 4, and 5 levels of the A^30^+ state are coherently superposed, and two identical nuclear wave packets are created sequentially

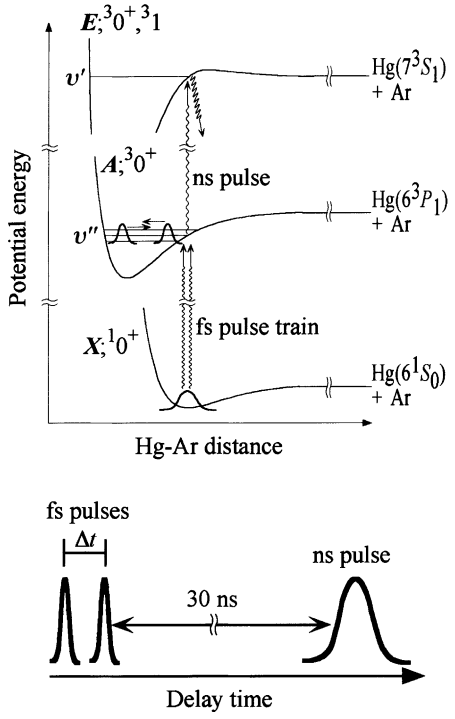

Fig. 4. Schematic of the wave-packet interference experiment.

near the outer turning point of the relevant potential curve. Δt between the two pulses is coarsely set around 2.2 ps by the mechanical stage of APS so that the two pulses are well temporally separated. This Δt corresponds to twice of the wave-packet vibration period, so that the second packet is created when the first packet recurs to the outer turning point for the second time. They interfere constructively or destructively depending on their relative phase, which is controlled by tuning Δt with an attosecond accuracy with the Ar cell of APM. Another nanosecond laser pulse is delayed by 30 ns from the femtosecond pulses and is used for the laser induced fluorescence (LIF) detection of the quantum amplitude of one of the vibrational eigenstates initially superposed by the femtosecond pulses. The ratio of the quantum amplitudes of the eigenstates involved is estimated to be 2:7:2 for the $v = 3$, 4, and 5 levels under the present experimental conditions, so that the LIF signal is expected to be most intense with the nanosecond pulse frequency is tuned to the $v' \leftarrow 4$ band of the E $\leftarrow$ A transition. By scanning Δt with a resolution of attoseconds, the fluorescence signal is expected to show an oscillation corresponding to the wave packet interference. Fig. 5 shows theoretical simulations of the wave packet motion on the A-state potential under the present experimental conditions. In these simulations, we solve the one-dimentional time-dependent Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi(R,t) = [H(R) - \mu\varepsilon(t)]\Psi(R,t). \quad (1)$$

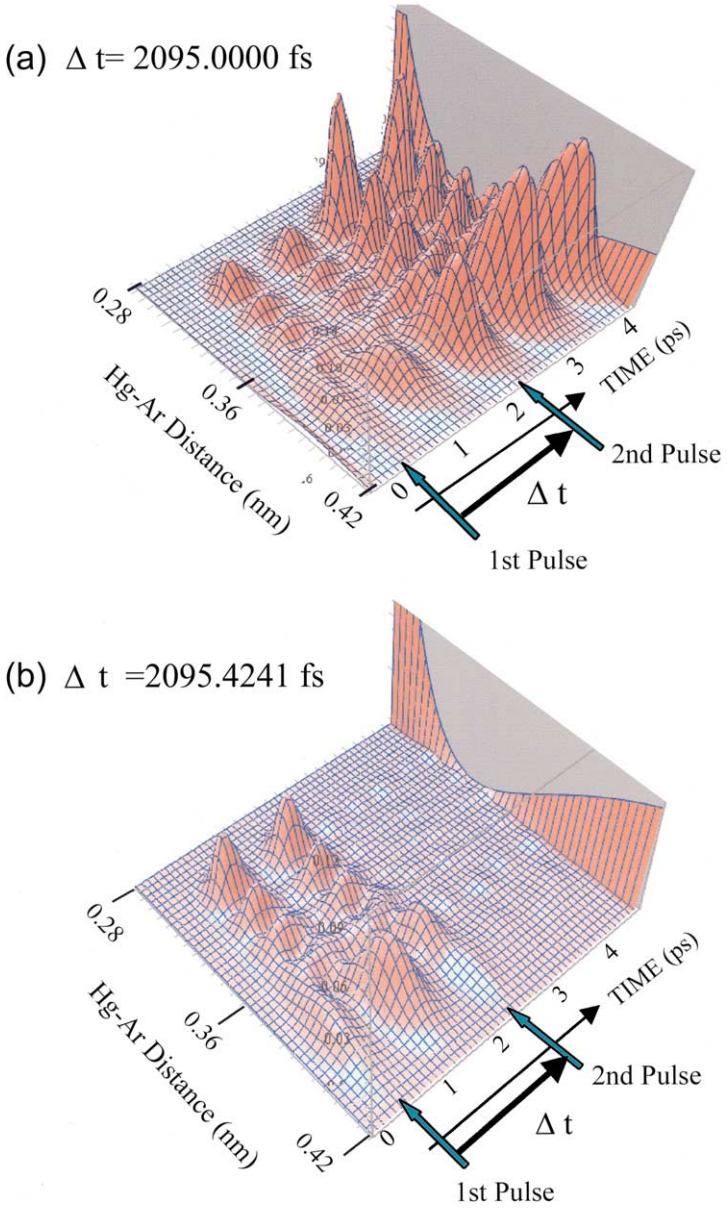

Fig. 5. Theoretical simulation of the wave-packet interference. The ordinate represents $\Psi^2(R,t)$.

For the sequence of two femtosecond laser pulses:

$$\varepsilon(t) = \varepsilon_{\max} \exp\left(-\frac{t^2}{2\delta^2}\right) \cos\omega t + \varepsilon_{\max} \exp\left[-\frac{(t-\Delta t)^2}{2\delta^2}\right] \cos\omega(t-\Delta t), \quad (2)$$

where δ is the temporal width of each pulse, ω the center frequency, and the second pulse is delayed from the first one by Δt. The radiative and non-adiabatic decay of the wave packet to the $X^1 0^+$ and $A^3 0^-$ states is negligible [9–14], and it is not included in the present simulations. It is seen in Fig. 5 that the interference changes drastically from constructive to destructive in changing Δt by only 424 as, which corresponds to π in the relative phase of the two wave packets. This quantum interference experiment is actually in progress in our group at this moment.

The present attosecond phase-manipulation technique leads to a variety of useful applications which we call the wave-packet engineering. One possibility lies in the coherent control of elementary chemical reactions. This type of coherent reaction control is expected, for example, to greatly improve the quality of lithography, where the UV light is used to minimize the diffraction effect for the fabrication of the large scale circuit such as LSI and VLSI. The attosecond accuracy realizes the coherent control in the UV region and should be highly useful in the lithography for those large scale circuits. Furthermore, it is being revealed recently that the quantum coherence plays a key role also in the complicated biological processes such as electron transfer in the reaction center of photosynthesis and the *cis–trans* molecular isomerization in vision [15,16]. Those kinds of complicated systems may also be the targets of the present attosecond interference control. Ahn et al. [17] have recently suggested the possibility of a single atom quantum computer where a specific information is encoded in the form of Rydberg electronic wave packet and is retrieved through the interference with another wave packet. In this paper, we have demonstrated the feasibility of a similar quantum manipulation of nuclear wave packets in a single molecule even with an accuracy in attosecond time-scale. The attosecond interference control will also be useful in the development of such a quantum information science.

Acknowledgements

K. Ohmori thanks Prof. Elmar Schreiber (Princeton University) for his friendship and a helpful guidance to the fantastic femtoworld. K. Ohmori also thanks Prof. Masaki Yamamoto (Tohoku University) for stimulating discussion about the applications of the attosecond optical-phase manipulation. This research was partly supported by the Grant-in-Aid for Scientific Research on Priority Areas "Molecular Physical Chemistry" and by the Grant-in-Aid for Science (No. 11640383) from the Ministry of Education, Science, Sports, and Culture.

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