17a-C4-9

Vibrational wavepacket reconstruction with frequency-resolved optical gating technique

Yasuo Nabekawa[1*], Yusuke Furukawa[1], Tomoya Okino[1], A. Amani Eilanlou[1], Eiji J. Takahashi[1], Kaoru Yamanouchi[2], and Katsumi Midorikawa[1]

[1] RIKEN Center for Advanced Photonics, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan
[2]Department of Chemistry, the University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
e-mail address: nabekawa@riken.jp

Wave nature of matter attracts our great interests not only in a point of view of fundamental physics but also in the technology for the information processing. We now direct our attention to a quantum system of vibrational wavepacket of a diatomic molecule both in the point of views of the both fields. A vibrational wave packet at a time t, $\varphi(R,t)$, is generally expressed as $\varphi(R,t)=\sum_v a_v \chi_v(R)\mathrm{e}^{-i\omega_v t}$, where $\chi_v(R)$ is the v th vibrational wave function with respect to the internuclear distance of R, and $\hbar\omega_v$ is the eigen energy of v th vibrational state. In order to identify the vibrational packet, we have to determine the complex amplitude, a_v, of each vibrational state.

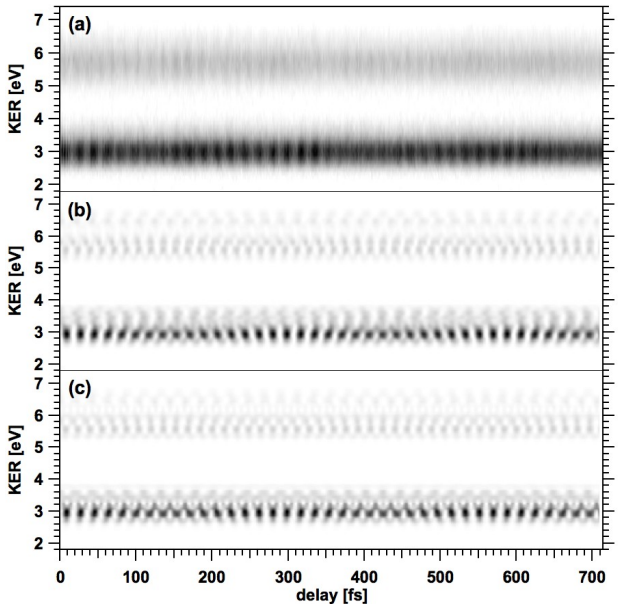

Fig. 1. (a) Measured delay-KER spectrogram of H^+ fragments. (b) Target delay-KER spectrogram obtained by applying a bandpass filter to the spectrogram in Fig.1(a). (c) Retrieved delay-KER spectrogram.

In this paper, we report on the novel method to retrieve the vibrational wavepacket of H_2^+ molecule generated with one-photon ionization of H_2 molecule irradiated by a sub-10-fs XUV pulse. The frequency resolved optical gating (FROG) [1], which is now conventionally used for reconstructing the magnitude and phase of a ultrashort optical pulse, is a key technique.

The experimental scheme and preliminary results for the observation of the real time evolution of the vibrational wavepacket of D_2^+ ionic molecule was already reported in ref. [2]. In brief, high-order harmonic fields of Ti:sapphire laser pulse with a duration of 14 fs reflected from a pair of silicon beam splitter (SiBS) mirror, which acts as a correlator, is focused in a H_2 molecular gas jet, and the angle-resolved momentum of generated fragment ion of H^+ is measured with an ion spectrometer recording velocity map image (VMI). We can obtain kinetic energy release (KER) spectrum of the H^+ ion by angularly integrating the measured VMI. By scanning the delay between the two replicas of all the harmonic synthesized field with the SiBS correlator, we obtained a delay-KER spectrogram shown in Fig. 1(a). We can clearly see the intensity modulations with a period of 16 fs in the KER region A and with a period of 18 fs in region B as the evidence of real time evolution of vibrational wavepacket of H_2^+ on the bound ground electronic state ($1s\sigma_g$). The assignment of the electronic state is ensured by the agreement of beat frequencies appearing on the Fourier transform of the spectrogram with the known frequency differences of vibrational states.

We know that the fragments of H^+ ion in the KER region A (B) originates from the electronic transition from the $1s\sigma_g$ bound state to the $2p\sigma_u$ repulsive state with one-photon absorption of the 5th (3rd) harmonic field. The transition amplitude of this one-photon process at probe-delay τ can be expressed as $T(\omega^u;\tau) = \sum_v \mathcal{M}(\omega^u;\omega_v)\widetilde{G}(\omega^u-\omega_v)\, a_v \mathrm{e}^{-i\omega_v\tau}$, where $\hbar\omega^u$ is KER, $\mathcal{M}(\omega^u;\omega_v)$ is a dipole transition matrix element from the $1s\sigma_g$ to $2p\sigma_u$ states, and $\widetilde{G}(\omega)$ is the Fourier amplitude of the optical field with a synthesis of the 3rd and 5th order harmonic fields. We can find the $\widetilde{G}(\omega)$ acts as a gate field to resolve a_v by recognizing the equivalence of this equation to the FROG amplitude with a gate and an optical field to be retrieved, $\widetilde{\epsilon}(\omega)$, in frequency domain, that is $T_{\mathrm{frog}}(\Omega;\tau) = \int d\omega \widetilde{G}(\Omega-\omega)\widetilde{\epsilon}(\omega)\mathrm{e}^{-i\omega\tau}$.

Although the spectrogram in Fig.1(a) contains the sufficient information for a_v and $\widetilde{G}(\omega)$, the signal-to-noise ratio and KER resolution is not sufficient for applying the FROG algorithm. Thus, we have applied a bandpass filter to the measured spectrogram, resulting in the spectrogram in Fig.1(b), then implemented FROG algorithm with the calculated $\mathcal{M}(\omega^u;\omega_v)$ from known literatures and with the filtered spectrogram in Fig.1(b) as a target. The retrieved spectrogram is shown in Fig. 1(c), which is in good agreement with the target spectrogram. The retrieved wavepacket amplitude, a_v, (not shown) exhibits a non-trivial group delay difference, which may be related to a settling time needed for wavepacket formation immediately after the ionization from H_2 to H_2^+. We are now studying for solving this issue. This work was a part of APSA project commissioned by MEXT. Y. N., T. O., E. J. T., and K. M. acknowledge the support from KAKENHI, No. 26247068, 40431895, 26600122, and 26220606.

References

1. Rick Trebino, et al., Rev. Sci. Instrum. **68**, 3277–3295 (1997).
2. Yusuke Furukawa, et al., Opt. Lett. **37**, 2922–2924 (2012).