PHYSICAL REVIEW A **82**, 040101(R) (2010)

Effect of entanglement on the decay dynamics of a pair of H($2p$) atoms due to spontaneous emission

Takehiko Tanabe,[1] Takeshi Odagiri,[1,*] Motoyoshi Nakano,[1] Yoshiaki Kumagai,[1] Isao H. Suzuki,[2,3] Masashi Kitajima,[1] and Noriyuki Kouchi[1]

[1]*Department of Chemistry, Tokyo Institute of Technology, Tokyo 152-8551, Japan*

[2]*National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8568, Japan*

[3]*Photon Factory, Institute of Materials Structure Science, KEK, Tsukuba 305-0801, Japan*

(Received 11 May 2010; published 20 October 2010)

We have measured the coincidence time spectra of two Lyman-α photons emitted by a pair of H($2p$) atoms in the photodissociation of H_2 at the incident photon energy of 33.66 eV and at the hydrogen gas pressures of 0.40 and 0.02 Pa. The decay time constant at 0.02 Pa is approximately half the lifetime of a single H($2p$) atom, 1.60 ns, while the decay time constant at 0.40 Pa is in agreement with the lifetime of a single H($2p$) atom. It turns out that the decay faster than the lifetime of a single H($2p$) atom originates from the entanglement in the pair of H($2p$) atoms. We have demonstrated an effect of entanglement on atomic decay.

DOI: 10.1103/PhysRevA.82.040101 PACS number(s): 03.65.Ud, 33.80.−b, 42.50.−p

A large number of studies have been directed toward generating and manipulating entanglement in various qubits, which not only is at the heart of quantum theory but also plays an essential role in quantum information technologies [1–4]. A deeper understanding of entanglement has become increasingly important to overcome obstacles in the realization of quantum information technologies. One of them is that the entanglement in massive quantum particles is in general fragile against interaction with environments. From the perspective of this serious issue, theoretical studies were conducted on the dynamics of entanglement in two atoms located inside spatially separated cavities and a unique feature drastically different from single-particle dynamics was predicted, i.e., in some cases the entanglement disappears in a finite time due to spontaneous emission [5,6]. It was then experimentally demonstrated for an entangled photon pair, not for an entangled pair of massive quantum particles [7]. In this Rapid Communication, we reveal that entangled H($2p$) atoms look as if they decay faster than a single H($2p$) atom. This kind of investigation promotes the understanding of the transient properties of the entanglement emerging in a pair of massive quantum particles.

Our group found an atom-pair formation process (1) followed by the emission of two photons and measured cross sections for the emission of two Lyman-α photons as a function of incident photon energy [8],

$$\begin{aligned} H_2(X\,^1\Sigma_g^+) + h\nu &\rightarrow H_2^{**}\ [\text{the } Q_2\,^1\Pi_u(1) \text{ state}] \\ &\rightarrow H(2p) + H(2p) \\ &\rightarrow H(1s) + H(1s) + 2 \text{ Lyman-}\alpha \text{ photons}. \end{aligned} \tag{1}$$

Recently, our group published two papers on the theoretical and experimental studies of the angular distribution of two Lyman-α photons in process (1). In the first paper, Miyagi *et al*. [9] pointed out that a pair of H($2p_0$) and H($2p_{\pm1}$) atoms produced from an H_2 molecule in the $^1\Pi_u$ state, where the lower subscripts of 0 and ±1 denote the magnetic quantum numbers m with respect to the internuclear axis, is entangled as follows:

$$\begin{aligned} |^1\Pi_u^+;\, r = +\infty\rangle = \frac{1}{2\sqrt{2}}\big[&\big|2p_1^a(1)2p_0^b(2)\big\rangle + \big|2p_1^a(2)2p_0^b(1)\big\rangle \\ &- \big|2p_0^a(1)2p_1^b(2)\big\rangle - \big|2p_0^a(2)2p_1^b(1)\big\rangle \\ &- \big|2p_{-1}^a(1)2p_0^b(2)\big\rangle - \big|2p_{-1}^a(2)2p_0^b(1)\big\rangle \\ &+ \big|2p_0^a(1)2p_{-1}^b(2)\big\rangle + \big|2p_0^a(2)2p_{-1}^b(1)\big\rangle\big], \end{aligned} \tag{2}$$

where two protons are labeled a and b and the two electrons are labeled 1 and 2. The internuclear distance r is infinite in the entangled state of two H($2p$) atoms in Eq. (2). In fact, the value of r is 93 μm when the H($2p$) atoms emit the Lyman-α photons, as calculated from the incident photon energy (33.66 eV as mentioned later), the dissociation limit of H($2p$) + H($2p$), and the lifetime of the H($2p$) atom, i.e., 1.60 ns [10]. The dissociation is in general much faster than the spontaneous emission. The system of interest is not a molecule but an entangled pair of atoms. The entangled pair of H($2p$) atoms in Eq. (2) emits an entangled pair of Lyman-α photons, i.e., the entanglement in the pair of H($2p$) atoms is copied to the pair of Lyman-α photons, resulting in strong anisotropy in the angular distribution of two Lyman-α photons as calculated by Miyagi *et al*. [9] (solid line in Fig. 2). More recently Jänkälä *et al*. [11] formulated a general theoretical model to calculate the angular distribution of two fluorescence photons from entangled atomic fragments in the photodissociation of a diatomic molecule. They calculated the angular distribution of two Lyman-α photons in process (1), which shows a shape similar to that by Miyagi *et al*. [9] but weaker anisotropy.

In a paper subsequent to that of Miyagi *et al*. [9], Tanabe *et al*. [12] reported the experimental angular distribution of two Lyman-α photons in process (1) measured at the incident photon energy of 33.66 eV and at the hydrogen gas pressures of 0.40 and 0.13 Pa. The results were compared with the theoretical prediction by Miyagi *et al*. [9]. Tanabe *et al*. [12] found that (i) the experimental angular distributions seem to approach the theoretical one with decreasing pressure, which indicates the generation of an entangled pair of H($2p$) atoms expressed by Eq. (2), and (ii) the dependence of the

*odagiri.t.aa@m.titech.ac.jp

angular distribution on the pressure cannot be explained by the reactions of a H($2p$) or H($2s$) atom with an H_2 molecule, but by the reaction of an entangled pair of H($2p$) atoms with an H_2 molecule. It turned out that the reaction of an entangled pair of H($2p$) atoms is two orders of magnitude faster than that of a single H($2p$) atom.

In the present investigation we have obtained an unexpected result: the decay time constant in the coincidence time spectrum of two Lyman-α photons in process (1) is approximately half the lifetime of a single H($2p$) atom, 1.60 ns [10], at the hydrogen gas pressure of 0.02 Pa and at 33.66 eV incident photon energy, while it is expected to be 1.60 ns when each H($2p$) atom in the entangled pair emits a Lyman-α photon independently with the lifetime of a single H($2p$) atom [9]. The decay time constant at 0.40 Pa, on the other hand, is in agreement with the lifetime of a single H($2p$) atom. We demonstrate an effect of entanglement, i.e., each H($2p$) atom in the entangled pair [Eq. (2)] apparently decays with a lifetime shorter than that of a single H($2p$) atom.

The experiments were carried out at the beam line BL20A of the Photon Factory, KEK. The experimental apparatus was the same as that used in our previous measurements of the angular distribution of two Lyman-α photons [12] and thus is described in brief. Linearly polarized synchrotron radiation was introduced into a gas cell. Two photon detectors were placed on a line perpendicular to the incident light beam with the same distance from the crossing point of the line and the incident light beam, and were opposite to each other. The solid angle subtended by each photon detector from the crossing point was 0.64 sr, covering 5% of 4π sr. Each vacuum ultraviolet photon detector is composed of a microchannel plate and a MgF_2 window. Two pulses from two detectors were fed into a standard delayed-coincidence system, and then coincidence time spectra of two Lyman-α photons were obtained. The time interval per channel in the coincidence time spectra was 0.204 ns/channel, which was measured using an Ortec 462 time calibrator. The linear dependence of the Lyman-α photon count rate on the hydrogen gas pressure was confirmed up to 0.80 Pa.

The incident light at the Photon Factory consists of a train of pulses with a duration of several hundred picoseconds and a pulse-to-pulse interval of 2 ns. It takes 624 ns for a bunch of charged particles to come full circle in the storage ring. The accidental coincidences should have the same time structure as the incident light: a peak-to-peak interval of 2 ns and a period of 624 ns. In fact, however, a time structure of the accidental coincidences was not seen and they just fluctuated randomly, mainly due to the much weaker accidental coincidences. The ratios of the true coincidence at the peak to the accidental ones were approximately 300 at 0.40 Pa and better than that at 0.02 Pa. The average of the accidental coincidences over the channels was hence subtracted. The results in Figs. 1 and 2 are not influenced by the accidental coincidences. The coincidence time spectra were measured at several angles between the unit polarization vector of the linearly polarized incident light and the line joining two detectors. They were summed after the subtraction of the accidental coincidences to improve the statistics and to make the statistics approximately the same at 0.40 and 0.02 Pa. In the procedure of summation the small

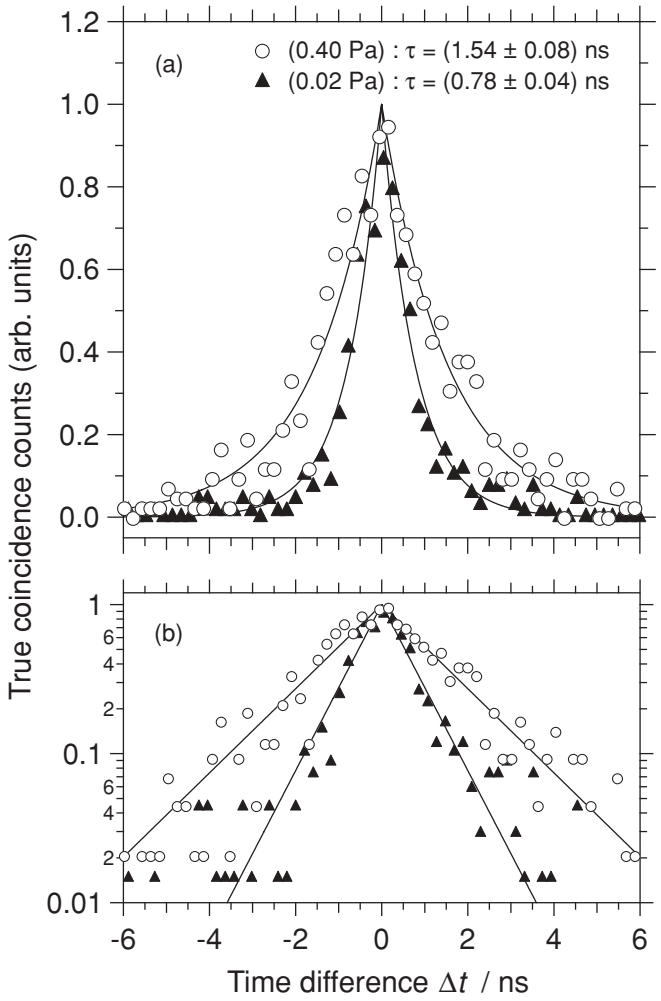

FIG. 1. Coincidence time spectra of two Lyman-α photons emitted by a pair of H($2p$) atoms in the photodissociation of H_2 measured at the hydrogen gas pressures of 0.40 Pa (◯) and 0.02 Pa (▲) with a linear scale of the vertical axis in (a) and a logarithmic scale in (b). The incident photon energy was 33.66 eV. The accidental coincidences were subtracted and the results were summed over the angles (see the text). The solid lines show the results of the fits of Eq. (6).

difference in the peak channels of the true coincidence peaks was taken into account.

The coincidence time spectra of two Lyman-α photons after the subtraction of the accidental coincidences and summation over the angles are shown in Fig. 1, which were measured at the hydrogen gas pressures of 0.40 and 0.02 Pa and at 33.66 eV incident photon energy. They are normalized to unity at time difference $\Delta t = 0$. The time difference Δt is defined as

$$\Delta t = t_{\mathrm{stop}} - t_{\mathrm{start}}, \tag{3}$$

where t_{stop} is the time when the photon is detected by the *stop* detector and t_{start} is the time when the photon is detected by the *start* detector. The coincidence time spectra in Fig. 1 were obtained within the same beam time and thus the time resolution seems to have been the same. The cascade contribution from H($n \geqslant 3$) is not involved in Fig. 1 since the lifetime of H($3s$) is 160 ns and that of H($3d$) is 15.6 ns [13]. There is also no

contribution of the reactions of a H($2p$) or H($2s$) atom with an H_2 molecule, as discussed in detail in [12]. The coincidence time spectra in Fig. 1 are hence attributed to process (1).

Let us obtain the decay time constants in the coincidence time spectra in Fig. 1. We first derive the fitting function to do so. In the theoretical treatment of our group [9] the single-photon state of the Lyman-α radiation is written using the Weisskopf-Wigner theory of spontaneous emission [14]. The entangled two-photon state $|\psi\rangle$ emitted by the entangled pair of H($2p$) atoms [Eq. (2)] is then obtained, considering that each H($2p$) atom in the entangled pair emits a Lyman-α photon independently with the lifetime of a single H($2p$) atom, denoted by $\tau_{2p} = 1.60$ ns [the lifetimes of H($2p_{0,\pm1}$) atoms are 1.60 ns] [10]. The two-photon correlation function $G^{(2)}(\boldsymbol{r}_c,t_c,\boldsymbol{r}_d,t_d)$, proportional to the coincidence detection probability of two Lyman-α photons at position $\boldsymbol{r}_c$ and the time t_c and at position $\boldsymbol{r}_d$ and the time t_d, is calculated as follows [9]:

$$\begin{aligned} G^{(2)}(\boldsymbol{r}_c,t_c,\boldsymbol{r}_d,t_d) &= \langle\psi|\boldsymbol{E}^{(-)}(\boldsymbol{r}_c,t_c)\boldsymbol{E}^{(-)}(\boldsymbol{r}_d,t_d)\boldsymbol{E}^{(+)}(\boldsymbol{r}_d,t_d)\boldsymbol{E}^{(+)}(\boldsymbol{r}_c,t_c)|\psi\rangle \\ &= A(\boldsymbol{r}_c,\boldsymbol{r}_d)\exp\left[-\tau_{2\mathrm{p}}^{-1}\left(t_c-\frac{R}{c}\right)\right]\exp\left[-\tau_{2\mathrm{p}}^{-1}\left(t_d-\frac{R}{c}\right)\right]\Theta\left(t_c-\frac{R}{c}\right)\Theta\left(t_d-\frac{R}{c}\right), \end{aligned} \tag{4}$$

where $\boldsymbol{E}^{(+)}$ and $\boldsymbol{E}^{(-)}$ are the positive and negative frequency parts of the electric field operators, respectively, R the distance between each photon detector and the incident light beam, c the light velocity, and $\Theta(\cdot)$ the step function. The origin of time is taken at the time when the entangled pair of H($2p$) atoms is produced. The coincidence time spectrum of two Lyman-α photons, $F(\Delta t)$, is given by integrating $G^{(2)}(\boldsymbol{r}_c,t_c,\boldsymbol{r}_d,t_d)$ along the straight line of $t_d = t_c + \Delta t$, where t_c and t_d correspond to t_{start} and t_{stop} in Eq. (3), respectively. The result is shown below [9]:

$$\begin{aligned} F(\Delta t) &= K\int_{-\infty}^{+\infty} G^{(2)}(\boldsymbol{r}_c,t_c,\boldsymbol{r}_d,t_c+\Delta t)dt_c \\ &= \frac{K\tau_{2p}A(\boldsymbol{r}_c,\boldsymbol{r}_d)}{2}\exp\left(-\tau_{2p}^{-1}|\Delta t|\right), \end{aligned} \tag{5}$$

where K is the apparatus constant. Equation (5) shows that the coincidence time spectrum of two Lyman-α photons is proportional to $\exp(-\tau_{2p}^{-1}|\Delta t|)$ when each H($2p$) atom in the entangled pair emits a Lyman-α photon independently with the lifetime of a single H($2p$) atom τ_{2p}. We have fitted $F'(\Delta t)$ to obtain the experimental decay time constants,

$$F'(\Delta t) = A'\exp(-\tau^{-1}|\Delta t|), \tag{6}$$

where A' and the decay time constant τ, the apparent lifetime of each H($2p$) atom in the pair, are fitting parameters. As shown by the solid lines in Fig. 1, good fits have been obtained without considering the time resolution of the present apparatus. The result of the fits is as follows: $\tau = (0.78 \pm 0.04)$ ns at 0.02 Pa and $\tau = (1.54 \pm 0.08)$ ns at 0.40 Pa. The apparent lifetime of each H($2p$) atom in the pair at 0.02 Pa is approximately half the lifetime of a single H($2p$) atom and that at 0.40 Pa is in agreement with the lifetime of a single H($2p$) atom.

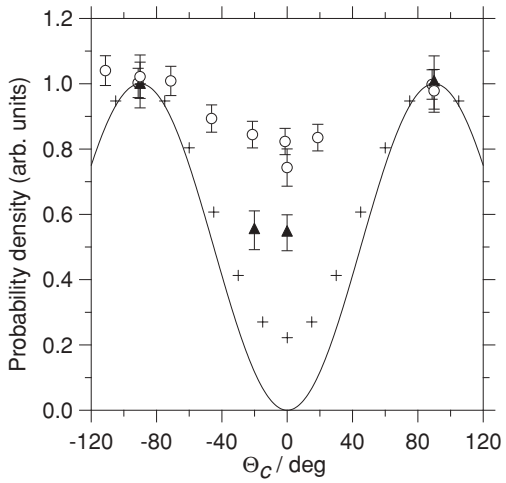

FIG. 2. Angular distributions of two Lyman-α photons measured at the same hydrogen gas pressures and incident photon energy as in Fig. 1. ◯, 0.40 Pa; ▲, 0.02 Pa; solid line, theoretical angular distribution of two Lyman-α photons emitted by the entangled pair of H($2p$) atoms [Eq. (2)] [9]; +, convoluted result of the solid line with the angular resolution [12].

Let us show that the entanglement in the pair of H($2p$) atoms expressed by Eq. (2) makes the apparent lifetime shorter than that of a single H($2p$) atom. More entangled pairs survive at 0.02 Pa than at 0.40 Pa as shown below. The angular distributions of two Lyman-α photons have been obtained from the coincidence measurements [12] and are displayed against angle Θ_c in Fig. 2; the angle specifies the direction of the detector c with respect to the unit polarization vector of the linearly polarized incident light. The detectors c and d were on the line perpendicular to the incident light beam as mentioned. The present angular distribution at 0.40 Pa does not coincide well with the previous one by Tanabe *et al.* [12], probably because the alignment was not so good and the scatter of coincidence counts was large in the previous experiment. In the present experiment the coincidence measurements were carried out at $\Theta_c = \pm 90^\circ$ to check the alignment more precisely, although Tanabe *et al.* [12] measured only at $\Theta_c = -90^\circ$. The coincidence time spectrum at 0.40 Pa in Fig. 1 was obtained from the measurements at $\Theta_c = \pm 90^\circ$ and 0° and that at 0.02 Pa from the measurements at $\Theta_c = \pm 90^\circ, -20^\circ$, and 0°. The theoretical angular distribution by Miyagi *et al.* [9] is also shown (solid line). It was convoluted with the angular resolution (+) [12]. As pointed out by Tanabe *et al.* [12], the experimental angular distributions approach the theoretical prediction based on the entanglement

in Eq. (2) (+) with decreasing pressure. There still exists a discrepancy between the experimental distribution at 0.02 Pa and theoretical prediction (+), which is probably because the pressure was still too high, the incident light was not completely linearly polarized, or both. The angular distribution of two Lyman-α photons will be discussed in detail in the following paper. We conclude from Fig. 2 that more entangled pairs of H($2p$) atoms expressed by Eq. (2) survive at 0.02 Pa than at 0.40 Pa.

It thus follows from Figs. 1 and 2 that entangled H($2p$) atoms apparently decay with a lifetime shorter than that of a single H($2p$) atom. The decay dynamics of each H($2p$) atom in the entangled pair due to the spontaneous emission is fundamentally different from that of a single H($2p$) atom. We note that the present finding of $\tau \approx (1/2)\tau_{2p}$ at 0.02 Pa is different from the fact that the coincidence detection probability $G^{(2)}(\boldsymbol{r}_c,t,\boldsymbol{r}_d,t)$ is proportional to $\exp(-2\tau_{2p}^{-1}t)$ for $t > R/c$.

In conclusion, we have demonstrated an effect of entanglement: each H($2p$) atom in the entangled pair [Eq. (2)] looks as if it decays faster than a single H($2p$) atom. The decay dynamics of each H($2p$) atom in the entangled pair due to the spontaneous emission is fundamentally different from that of a single H($2p$) atom. Our group recently demonstrated that the reaction cross section of an entangled pair of H($2p$) atoms with an H_2 molecule is roughly two orders of magnitude larger than the reaction cross section of a single H($2p$) atom [12], which means that the reaction dynamics of the entangled pair of H($2p$) atoms is much different from that of a single H($2p$) atom. The entanglement has a large influence on the decay and reaction dynamics of atoms, and possibly in general of massive quantum particles.

The experiment was carried out under the approval of Photon Factory Program Advisory Committee for Proposal No. 2008G107. This work was partially supported by Grants-in-Aid for Scientific Research (C) (No. 19550011 and No. 22550008) from the Japan Society for the Promotion of Science. T.T. wishes to acknowledge the financial support by a Sasakawa Scientific Research Grant from the Japan Science Society, T.O. that of the Matsuo Foundation and Reimei Research Promotion Project of the Japan Atomic Energy Agency, and N.K. that of Research Foundation for Opto-Science and Technology. The authors are grateful to Dr. Kouichi Hosaka of the Department of Chemistry, Tokyo Institute of Technology, Dr. Atsushi Ichimura of the Institute of Space and Astronautical Science, JAXA, and Dr. James Harries of JAEA/SPring-8 for their fruitful discussions.

[1] K. Edamatsu, Jpn. J. Appl. Phys. **46**, 7175 (2007).
[2] V. Vedral, Nature (London) **453**, 1004 (2008).
[3] R. Blatt and D. Wineland, Nature (London) **453**, 1008 (2008).
[4] I. Bloch, Nature (London) **453**, 1016 (2008).
[5] T. Yu and J. H. Eberly, Science **323**, 598 (2009).
[6] T. Yu and J. H. Eberly, Phys. Rev. Lett. **93**, 140404 (2004).
[7] M. P. Almeida, F. de Melo, M. Hor-Meyll, A. Salles, S. P. Walborn, P. H. S. Ribeiro, and L. Davidovich, Science **316**, 579 (2007).
[8] T. Odagiri, M. Murata, M. Kato, and N. Kouchi, J. Phys. B **37**, 3909 (2004).
[9] H. Miyagi, A. Ichimura, and N. Kouchi, J. Phys. B **40**, 617 (2007).
[10] B. H. Bransden and C. J. Joachain, *Physics of Atoms and Molecules*, 2nd ed. (Pearson Education, Harlow, 2003), pp. 200–201.
[11] K. Jänkälä, P. V. Demekhin, S. Heinäsmäki, I. Haar, R. Hentges, and A. Ehreshmann, J. Phys. B **43**, 065104 (2010).
[12] T. Tanabe, T. Odagiri, M. Nakano, I. H. Suzuki, and N. Kouchi, Phys. Rev. Lett. **103**, 173002 (2009).
[13] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Plenum, New York, 1977), p. 266.
[14] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, 1997), pp. 206–210.