Large Pressure Effect on the Angular Distribution of Two Lyman-α Photons Emitted by an Entangled Pair of H($2p$) Atoms in the Photodissociation of H_2

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The angular distribution of two Lyman-α photons, i.e., the probability density that two Lyman-α photons are emitted in given directions, in the photodissociation of a hydrogen molecule have been measured at the hydrogen gas pressures of 0.40 and 0.13 Pa. We have found that the experimental angular distributions seem to approach the theoretical one by our group [J. Phys. B **40**, 617 (2007)] with decreasing pressure, which indicates the generation of the entangled pair of H($2p$) atoms shown in the theory and the role of the reaction of the entangled pair of H($2p$) atoms with an H_2 molecule that efficiently changes the entanglement.

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Entanglement plays an important role in quantum information and communication and thus the generation of entangled pairs of photons and massive quantum particles, e.g., atoms, atomic ions, and so on, has been widely investigated [1–3]. The advantage of massive quantum particles, in comparison to massless photons, is that they can remain stationary while the disadvantage is that their entanglement is readily changed by interaction with the environment. Hence, we need to understand the mechanism of the change of the entanglement in massive quantum particles by the interaction with the environments. We have found in the present study that the entanglement in a pair of H($2p$) atoms, generated in the photodissociation of an H_2 molecule, is efficiently changed by the reaction of the entangled pair of H($2p$) atoms with an H_2 molecule. The change of the entanglement in massive quantum particles by the interaction with the environments may in general be considered as reactions, and the fruitful knowledge on reactions and atomic collisions accumulated so far will probably promote the investigations on the change of the entanglement.

Our group recently found process (1), i.e., the emission of two photons in the photodissociation of a molecule, and measured doubly differential cross sections for the emission of two Lyman-α photons as a function of incident photon energy [4]

$$\begin{aligned} H_2(X^1\Sigma_g^+) + h\nu &\rightarrow H_2^{**} \text{ in a doubly excited state} \\ &\rightarrow H(2p) + H(2p) \\ &\rightarrow H(1s) + H(1s) \\ &\quad + 2 \text{ Lyman-}\alpha \text{ photons.} \end{aligned} \tag{1}$$

They concluded that H_2^{**} in process (1) lies in the doubly excited $Q_2{}^1\Pi_u(1)$ state.

Miyagi *et al.* [5] in our group calculated the angular distribution of two Lyman-α photons generated by process (1), i.e., the probability density that two Lyman-α photons are emitted in the direction (Θ_c, Θ_d), where H_2^{**} is in the doubly excited $Q_2{}^1\Pi_u(1)$ state. The point detectors c and d are placed on the plane including the center of mass of the two protons and perpendicular to the incident light beam. The directions of the point detectors c and d are specified by angles Θ_c and Θ_d, respectively, measured from the unit polarization vector of the linearly polarized incident light. The positive direction of Θ_c and Θ_d is defined as the counterclockwise direction when facing into the propagation direction of the incident light beam. The angular distribution calculated for $H_2(X^1\Sigma_g^+)$ molecules randomly oriented in space is shown in Fig. 1. In this Letter we report the first result of the experimental angular distribution of two Lyman-α photons in process (1) at the hydrogen gas pressures of 0.40 and 0.13 Pa, which is compared with the theoretical prediction in Fig. 1. We have found that (1) the

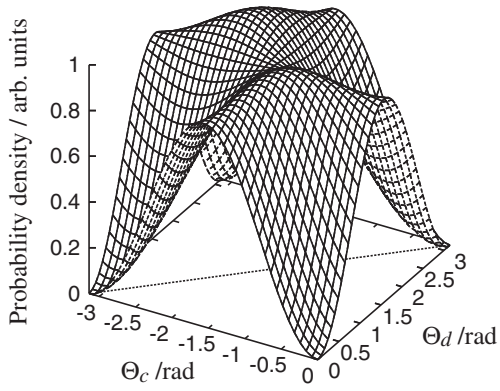

FIG. 1. The calculated angular distribution of two Lyman-α photons in the photodissociation of H_2 [5].

experimental angular distributions seem to approach the theoretical one with decreasing the pressure, which indicates the generation of an entangled pair of $H(2p)$ atoms expressed by Eq. (2) below [5], and (2) there is a large pressure effect originating from the reaction of an entangled pair of $H(2p)$ atoms with an H_2 molecule to change the entanglement.

Let us describe the calculation of the angular distribution in brief [5]. A pair of $H(2p_0)$ and $H(2p_{\pm 1})$ atoms is produced from an H_2 molecule in the $^1\Pi_u$ state, where the lower subscripts in $H(2p_0)$ and $H(2p_{\pm 1})$ show the magnetic quantum numbers m with respect to the internuclear axis. Miyagi *et al.* [5] pointed out that the pair of $H(2p)$ atoms is entangled as expressed by

$$|^1\Pi_u^+\rangle = \frac{1}{2\sqrt{2}}(|2p_1^a(1)2p_0^b(2)\rangle + |2p_1^a(2)2p_0^b(1)\rangle - |2p_0^a(1)2p_1^b(2)\rangle - |2p_0^a(2)2p_1^b(1)\rangle - |2p_{-1}^a(1)2p_0^b(2)\rangle - |2p_{-1}^a(2)2p_0^b(1)\rangle + |2p_0^a(1)2p_{-1}^b(2)\rangle + |2p_0^a(2)2p_{-1}^b(1)\rangle), \quad (2)$$

where two protons are labeled a and b and two electrons are labeled 1 and 2. The entangled pair of $H(2p)$ atoms expressed by Eq. (2) emits an entangled pair of the Lyman-α photons expressed as

$$|\psi\rangle = \tfrac{1}{2}[(|\gamma_a\phi_b\rangle - |\phi_a\gamma_b\rangle) - (|\rho_a\phi_b\rangle - |\phi_a\rho_b\rangle)], \quad (3)$$

where the ket-vectors $|\gamma\rangle$, $|\phi\rangle$, and $|\rho\rangle$ are single-photon states of the Lyman-α radiation generated through the $2p \rightarrow 1s$ transition in an H atom with $\Delta m = -1$, 0, and 1, respectively. The calculation of the second-order correlation function in quantum optics for the photon-pair state $|\psi\rangle$ in Eq. (3) gives the angular distribution of two Lyman-α photons generated from $H_2(X^1\Sigma_g^+)$ molecules randomly oriented in space as seen in Fig. 1. The entanglement in the pair of $H(2p)$ atoms [Eq. (2)] is copied to the pair of the Lyman-α photons [Eq. (3)] to yield the strong anisotropy seen in Fig. 1. The measurement of the angular distribution of a photon pair is equivalent to probing the entanglement in the photon pair and atom pair.

The experiments were carried out at the BL20A of the Photon Factory, KEK. Linearly polarized synchrotron radiation monochromatized by a 3 m normal incidence monochromator was introduced into a gas cell filled with molecular hydrogen. The hydrogen gas pressure was constant from the interaction volume viewed by the two photon detectors mentioned below up to the wall of the cell, on which the photon detectors were mounted. The angular distribution was measured at 33.66 eV incident photon energy, which gives the maximum value of the doubly differential cross sections for the emission of two Lyman-α photons [4]. Each vacuum ultraviolet photon detector is composed of a microchannel plate and an MgF_2 window that provides a filter range of approximately 115–150 nm. Only Lyman-α radiation, 121.6 nm wavelength, is detected at 33.66 eV incident photon energy. The solid angle subtended by each Lyman-α photon detector from the origin, i.e., the crossing point of the incident light beam and the axes of the two detectors, was 0.64 sr. Let us also label the real detectors c and d as is the same for the point detectors for convenience and thus keep using Θ_c and Θ_d to specify the directions of the real detectors c and d from the unit polarization vector of the linearly polarized incident light, respectively. The real detectors c and d were mounted on the wall of the gas cell in such a way that they were on the line perpendicular to the incident light beam with the same distance from the origin and opposite to each other. The detectors were rotatable around the direction of the incident light beam thus changing the angle Θ_c with keeping the relation of $\Theta_d = \Theta_c + \pi$. This means measuring the angular distribution along the dotted line on the $\Theta_c\Theta_d$ plane in Fig. 1. The pair of the Lyman-α photons was counted with a standard delayed-coincidence system. The true coincidence peaks in the photon-photon coincidence time spectra seem to have two decay components with the time constant of 1.6 ns, the lifetime of $H(2p)$ atom [6], on both sides of the peaks, which indicates that the cascade contribution from $H(n \geq 3)$ is small [4]: the lifetime of $H(3s)$ is 160 ns and that of $H(3d)$ is 15.6 ns [6,7]. The coincidence rate measured at each value of Θ_c was normalized for the flux of the incident photons and the pressure in the gas cell, and the normalized rates were plotted against Θ_c to give the angular distribution. It was shown from the ray tracing of Lyman-α photons that the integral of the product of the solid angles subtended by the detectors c and d over the interaction volume is constant irrespective of Θ_c when the detectors are aligned well. The alignment was examined by measuring the angular distribution of photoelectrons from He at the incident photon energy of 33.66 eV since the asymmetry parameter has been well known to be 2 [8]. Such measurements gave also the direction of the unit polarization vector of the linearly polarized incident light. The experimental angular distributions of photoelectrons from He at 33.66 eV incident photon energy showed that the rotation axis of the electron energy analyzer and the photon detectors, both of which were mounted on the same gas cell, was in good agreement with the axis of the incident light beam.

Figure 2 shows the angular distribution of two Lyman-α photons in the photodissociation of an H_2 molecule measured at the hydrogen gas pressures in the gas cell of 0.40 and 0.13 Pa. The error bar indicates the statistical uncertainty. The linear dependence of the Lyman-α photon count rate on the hydrogen gas pressure was confirmed at least up to 0.80 Pa. There exists the repulsive $b^3\Sigma_u^+(2p\sigma_u)$ state of H_2 dissociating into two $H(1s)$ atoms around 10.2 eV, the energy of the Lyman-α photon, within the Franck-Condon region [9]. However, a spin flip during the absorption of the Lyman-α photon would be unlikely and

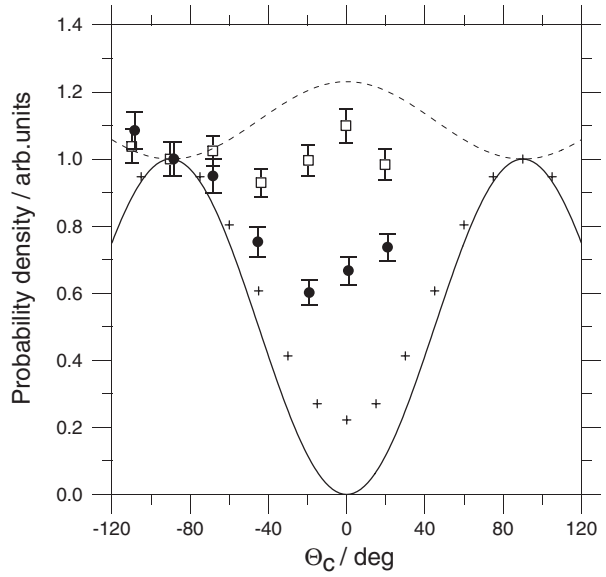

FIG. 2. Angular distribution of two Lyman-α photons in the photodissociation of a hydrogen molecule under the relation of $\Theta_d = \Theta_c + \pi$. □: experimental results at 33.66 eV incident photon energy and 0.40 Pa hydrogen gas pressure; ●: those at 0.13 Pa, solid line: theoretical angular distribution for the photon-pair state $|\psi\rangle$ in Eq. (3) [5]; dashed line: that for the photon-pair state $|\gamma_a \phi_b\rangle$ [5]; +: convoluted result of the solid line with the angular resolution. All the results are normalized to unity at $\Theta_c = -90°$.

thus the Lyman-α photon will not be absorbed. The radiation trapping of Lyman-α photons need hardly be taken into account. The theoretical prediction, the diagonal cross section of Fig. 1 [5], is also shown (solid line). It was convoluted considering that the solid angle subtended by each detector was 0.64 sr and process (1) did not occur at a point but occurred along the incident light beam. The result is shown by the symbol of +. The experimental angular distributions seem to approach the theoretical prediction (+) with decreasing the pressure. As mentioned before, the strong anisotropy predicted theoretically (solid line) originates from the entangled pair of $H(2p)$ atoms expressed by Eq. (2). Figure 2 seems to indicate the generation of the entangled atom pair. It is remarkable that a large difference is seen between the angular distributions at 0.40 and 0.13 Pa. The large pressure effect seems to be attributed to the reaction of a pair of $H(2p)$ atoms or an $H(2p)$ atom with an H_2 molecule.

Let us show that the large pressure effect is not able to be explained by the reactions of an $H(2p)$ and $H(2s)$ atom with an H_2 molecule that may influence the angular distribution. They are listed below [the partner $H(2p)$ fragment is not shown],

$$H(2p_m) + H_2 \xrightarrow{\sigma_{pp}} H(2p_{m'}) + H_2, \tag{4}$$

$$H(2s) + H_2 \xrightarrow{\sigma_{sp}} H(2p) + H_2, \tag{5}$$

where σ_{pp} and σ_{sp} are the cross sections for reactions (4) and (5), respectively. Reaction (4) includes the m-changing reaction as well as the transfer to the non-m eigenstate. The magnetic quantum number m is defined with respect to the internuclear axis of a pair of $H(2p)$ atoms as mentioned before. We also consider reaction (6),

$$H(2p) + H_2 \xrightarrow{\sigma_{ps}} H(2s) + H_2, \tag{6}$$

where σ_{ps} is the cross section, because it does not influence the angular distribution but the successive reactions (6) and (5) may influence. Let us calculate mean free times of reactions (4)–(6), which are compared with the lifetime of $H(2p)$ atom, 1.6 ns [6], as mentioned later. The mean free time τ of the reaction of which the cross section is σ is given by

$$\tau = 1/(\sigma v n), \tag{7}$$

where v is the relative velocity between an $H(2p)$ or $H(2s)$ atom and H_2 molecule and n the number density of H_2 molecules. The value of v is approximately calculated to be 2.9×10^6 cm s^{-1} from the incident photon energy of 33.66 eV and the dissociation limit of $H(2p) + H(2p)$ with respect to the zero point energy of the $H_2(X^1\Sigma_g^+)$ molecule, i.e., 24.875 eV [9], since the velocity of an $H(2p)$ atom against the center of mass of the two protons is much faster than the thermal velocity of an H_2 molecule against the laboratory frame at room temperature. Terazawa *et al.* [10] measured cross sections for reactions of an $H(2s)$ and $H(2p)$ atom with an H_2 molecule in the range of the relative velocity 4.6×10^5–12×10^5 cm s^{-1} by means of the time-dependent intensity of the Lyman-α radiation in the photodissociation of H_2. Let us use their values of σ_{sp} and σ_{ps} extrapolated to 2.9×10^6 cm s^{-1}, i.e., $\sigma_{sp} = (0.4 \pm 0.2) \times 10^{-14}$ cm^2 and $\sigma_{ps} = (2 \pm 1) \times 10^{-14}$ cm^2. The errors show those in extrapolation. Flemming *et al.* [11] estimated the value of σ_{pp} to be $(1.0 \pm 0.2) \times 10^{-14}$ cm^2 from the pressure dependence of the polarization degree of the Lyman-α radiation in the photodissociation of H_2 in almost the same range of the relative velocity as Terazawa *et al.* [10] and found that σ_{pp} is independent of the relative velocity. Let us thus use their value of σ_{pp} as it is, i.e., $\sigma_{pp} = (1.0 \pm 0.2) \times 10^{-14}$ cm^2. The mean free times of reactions (4)–(6), τ_{pp}, τ_{sp}, and τ_{ps}, respectively, were calculated at the hydrogen gas pressure of 0.40 Pa to be

$$\tau_{pp} = 3.5 \times 10^{-7} \text{ s}, \quad \tau_{sp} = 9 \times 10^{-7} \text{ s}, \quad \text{and} \quad \tau_{ps} = 2 \times 10^{-7} \text{ s}. \tag{8}$$

Those at 0.13 Pa are approximately 3 times. Most of the $H(2p)$ atoms do not undergo reactions (4) and (6) since the lifetime of $H(2p)$ atom, 1.6 ns [6], is much shorter than the mean free times of reactions (4) and (6) in Eq. (8). The $H(2s)$ atoms, on the other hand, undergo reaction (5) since they are metastable. The component due to reaction (5) in

the true coincidence peak of the coincidence time spectrum may have the width of $\approx 2\tau_{sp}$. In fact the shape of the true coincidence peak is dominated by the lifetime of H($2p$) atom as mentioned before and thus the contribution of reaction (5) to the true coincidence peak seems small. Reactions (4)–(6) are hence not likely to be able to explain the large pressure effect seen in Fig. 2, even if we take account of the uncertainty in the cross sections mentioned above. The observed linear dependence of the photon count rate on the hydrogen gas pressure up to 0.80 Pa mentioned before is consistent with the results in Eq. (8). In conclusion, it is likely that the origin of the large pressure effect is the reaction of the entangled pair of H($2p$) atoms expressed by Eq. (2) with an H_2 molecule to transfer to another entangled pair of H($2p$) atoms. We refer to this reaction as the entangled atom-pair reaction. Let us show the angular distribution calculated by Miyagi *et al.* [5] for the photon-pair state $|\gamma_a \phi_b\rangle$, i.e., the first term on the right-hand side of Eq. (3), to see the effect of the atom-pair state (dashed line in Fig. 2). The photon pair in the $|\gamma_a \phi_b\rangle$ state is emitted by the entangled atom-pair in the $\frac{1}{\sqrt{2}}(|2p_1^a(1)2p_0^b(2)\rangle + |2p_1^a(2)2p_0^b(1)\rangle)$ singlet Π state, i.e., the first two terms on the right-hand side of Eq. (2).

Let us roughly estimate the cross section of the entangled atom-pair reaction σ_{ap}. The mean free time of the entangled atom-pair reaction seems to be of the order of 1 ns at the hydrogen gas pressures of 0.40 and 0.13 Pa considering the large pressure effect in this pressure range and the lifetime of H($2p$) atom, 1.6 ns [6]. This shows that σ_{ap} is roughly two orders of magnitude larger than σ_{pp}, σ_{sp}, and σ_{ps}, i.e., $\sigma_{ap} \approx 10^{-13}$–$10^{-12}$ cm^2. We note that σ_{ap} is the cross section of the reaction of the entangled pair of H($2p$) atoms expressed by Eq. (2) with an H_2 molecule to transfer to another entangled pair of H($2p$) atoms while σ_{pp}, σ_{sp}, and σ_{ps} are those of reactions of an H($2p$) and H($2s$) atom with an H_2 molecule. We have found a new kind of reaction, the entangled atom-pair reaction, which is not a reaction of an atom or molecule but a reaction of a pair of atoms from an entangled pair state to another entangled pair state.

In conclusion, we have for the first time measured the angular distribution of two Lyman-α photons in the photodissociation of a hydrogen molecule at the hydrogen gas pressures of 0.13 and 0.40 Pa, and found that (1) the experimental results seem to approach the theoretical prediction by our group [5] with decreasing the pressure and (2) there is a large pressure effect. The former seems to show the generation of the entangled pair of H($2p$) atoms expressed by Eq. (2). The latter is explained by a new kind of reaction referred to as the entangled atom-pair reaction, i.e., the reaction of the entangled pair of H($2p$) atoms expressed by Eq. (2) with an H_2 molecule to transfer to another entangled pair of H($2p$) atoms, in other words to change the entanglement. The cross section of the entangled atom-pair reaction σ_{ap} has been estimated to be roughly two orders of magnitude larger than those of the reactions of an H($2p$) and H($2s$) atom with an H_2 molecule, i.e., $\sigma_{ap} \approx 10^{-13}$–$10^{-12}$ cm^2. The change of the entanglement in massive quantum particles by the interaction with the environments may in general be considered as reactions. More investigations are strongly required to substantiate the generation of the entangled pair of H($2p$) atoms and entangled atom-pair reaction in much more detail. In particular, the much larger cross section of the entangled atom-pair reaction attracts much interest.

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[1] K. Edamatsu, Jpn. J. Appl. Phys. **46**, 7175 (2007).
[2] C. A. Sackett, D. Kielpinski, B. E. King, C. Langer, V. Meyer, C. J. Myatt, M. Rowe, Q. A. Turchette, W. M. Itano, D. J. Wineland, and C. Monroe, Nature (London) **404**, 256 (2000).
[3] E. Hagley, X. Maître, G. Nogues, C. Wunderlich, M. Brune, J. M. Raimond, and S. Haroche, Phys. Rev. Lett. **79**, 1 (1997).
[4] T. Odagiri, M. Murata, M. Kato, and N. Kouchi, J. Phys. B **37**, 3909 (2004).
[5] H. Miyagi, A. Ichimura, and N. Kouchi, J. Phys. B **40**, 617 (2007).
[6] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Plenum, New York, 1977), p. 266.
[7] S. Lauer, H. Liebel, F. Vollweiler, O. Wilhelmi, R. Kneip, E. Flemming, H. Schmoranzer, and M. Glass-Maujean, J. Phys. B **31**, 3049 (1998).
[8] V. Schmidt, *Electron Spectrometry of Atoms Using Synchrotron Radiation* (Cambridge University Press, Cambridge, 1997), pp. 43–45, 273–277.
[9] T. E. Sharp, Atomic Data **2**, 119 (1971).
[10] N. Terazawa, M. Ukai, N. Kouchi, K. Kameta, Y. Hatano, and K. Tanaka, J. Chem. Phys. **99**, 1637 (1993).
[11] E. Flemming, O. Wilhelmi, H. Schmoranzer, and M. Glass-Maujean, J. Chem. Phys. **103**, 4090 (1995).