Experimental Evidence of Twin Fast Metastable $H(2^2S)$ Atoms from Dissociation of Cold H_2 Induced by Electrons

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We report the direct detection of two metastable $H(2^2S)$ atoms coming from the dissociation of a single cold H_2 molecule, in coincidence measurements. The molecular dissociation was induced by electron impact in order to avoid limitations by the selection rules governing radiative transitions. Two detectors, placed close to the collision center, measure the neutral metastable $H(2^2S)$ through a localized quenching process, which mixes the $H(2^2S)$ state with the $H(2^2P)$, leading to a Lyman-α detection. Our data show the accomplishment of a coincidence measurement which proves for the first time the existence of the $H(2^2S)$-$H(2^2S)$ dissociation channel.

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In the 1960s, Leventhal, Robiscoe, and Lea [1] performed the first measurements of the TOF and angular distribution of $H(2^2S)$ atoms produced by H_2 dissociation. Since then, the structure of H_2 and the dynamics of its fragmentation channels have been subjects of renewed experimental studies, by synchrotron radiation or laser techniques or in ion beam experiment [2–4] via the study of dissociative recombination [5,6], as well as theoretical ones, by *ab initio* calculations or by the method of the molecular multichannel quantum defect [7–13]. Despite the wealth of information on fragmentation of H_2, according to our knowledge, the production of two metastable atoms $H(2^2S)$ arriving from the dissociation of the same molecule has not yet been reported.

In this Letter we report the accomplishment of successful measurements in detecting two metastable atoms $H(2^2S)$ arriving from the dissociation of the same molecule. This result is of general interest. For instance, it may stimulate theoretical computations regarding the production of $H(2^2S)$-$H(2^2S)$ coming from the doubly excited states, by electron impact, of the molecular hydrogen. In addition, an emerging pair of atoms with mean lifetime of the order of $1/10$ s and with polarized angular momentum (polarized spin or polarized total angular momentum) may provide a new manner to obtain insight into the complex field of the molecular interactions, from the short-distance to the long-distance domain of interactions between moving atoms [14]. Moreover, the potential of this system as a source of entangled particles is clear.

In regard to this last subject, it is interesting to follow the original ideas of Bohm [15] when he proposed a variant of the Einstein-Podolsky-Rosen system [16], involving a Stern-Gerlach interferometer. In fact, a twin-atom experiment should be closer to the experimental suggestions of Englert, Schwinger, and Scully, who followed Bohm's ideas (see, for instance, [17]). It is worth mentioning that other proposals of experimental realizations of Bohm's spin-$1/2$ particle experiment, in its original form by making use only of atomic fragments and not photons, were already suggested [18]. However, according to our knowledge, the production of twin atoms with the possibility of direct manipulation of them, according to Bohm's ideas, has not yet been achieved. On the other hand, recently another group has performed measurements involving the photodissociation of H_2 and has analyzed the entanglement of the H atoms generated in this process by detecting the two Lyman-α photons emitted by the pair of short-life $H(2^2P)$ atoms [19]. In this case, no manipulation of the atoms themselves is possible due to the short lifetime ($\approx 10^{-9}$ s) of the handled atoms.

The fact that until now no research group has reported the evidence of detection of twin $H(2^2S)$-$H(2^2S)$ atoms is due to the small cross section of this dissociation channel. Consequently, the corresponding coincidence rate will be small and care has to be taken and specific experimental conditions have to be fulfilled to observe it.

Our setup adapts a high-precision time-of-flight spectroscopy experiment, with which we have studied the production of fast and slow metastable $H(2^2S)$ atoms coming from the dissociation of cold H_2 molecules [20,21]. This setup placed the detection system according to our collision kinematics analysis, positioning the detection active volume with very high precision to sample the direction where we believed that the twin atoms should be found. The active volume detected was defined by the very small spot where the quenching occurred due to the applied electric field by two needles. In addition, the flight distance of this detection

system was large. Both aspects provided an extremely narrow solid angle associated with the region, from which we detected the $H(2^2S)$ atoms. For details we refer the reader to our previous works mentioned earlier. The joint effect of that small solid angle together with the small cross section of the $H(2^2S)$-$H(2^2S)$ dissociation channel resulted in the ability to observe only the random coincidence rate of $H(2^2S)$ atoms from the $H(2^2S)$-$H(2^2P)$ dissociation channel. According to that, we mounted another experiment, designed for our specific purpose. In this second setup the detectors themselves were directed to the collision region and brought close to it. This caused a loss of precision in the $H(2^2S)$ point of origin, but yielded a very much larger solid angle, which increased the coincidence rate enormously. Under these conditions, we began to see a coincidence rate that could be assigned to the $H(2^2P)$-$H(2^2P)$ dissociation channel and another one which we recognized as being the twin $H(2^2S)$-$H(2^2S)$. Then we returned to the first setup, adjusting it according to the conditions of the second setup in order to obtain the results we display in this Letter.

Thus, in the present Letter we discuss our experimental setup and the analysis of the data that provide evidence of the coincidence measurement of two $H(2^2S)$ atoms arriving from the same H_2 molecule. The experimental setup is in general the same as that which was discussed in recent papers of our group [20,21], except for the detection system. We next briefly present the experimental details and then we discuss our data.

Our apparatus consists of a supersonic jet source of molecular hydrogen, whose stream crosses an electron beam coming from a pulsed electron gun. In all results appearing in this Letter we have used a 100 ns electron pulse width and 10 kHz repetition rate. The detection of the metastable atoms is performed by two detectors directly facing the collision zone, placed in opposite sides of the plane defined by the electron and the H_2 beams. We have placed the detectors at different distances from the collision region, in order to avoid the coincident counts overlapping with any electric noise which could be picked up simultaneously by both detectors. In Refs. [20,21] we were concerned with the precision of the time-of-flight spectra, which led us to use a specially designed detection system, which ensured we read only $H(2^2S)$ atoms, and placed them as far as possible from the collision region. In the present case we have been concerned with enhancing the coincidence signals. Thus, we brought the detectors closer, placing them at 58 and 67 mm from the collision region and facing it directly. The effective area of the detectors is 78.5 mm^2, leading to a solid angle of 0.017 and 0.023 sr., respectively. Each detector was contained in a small aluminium box with a grounded grid shielding the entrance aperture. Contrary to the experimental setup of our previous works, no MgF_2 window was employed, so the channeltron cone was biased negatively in order to avoid being saturated with scattered electrons. Since there is direct line of sight between the active area of the detectors and the collision region, we can detect, in addition to excited atoms, both UV radiation and ions, although we believe ions have very low probability of detection since there is little acceleration for them to gain energy and efficiently produce secondary electrons at the front surface of the detectors. The time-of-flight system enables us to separate the Lyman-α radiation emitted by $H(2^2S)$ from all the other contributions, since the Lyman-α coming from the metastable $H(2^2S)$ is produced in the detector's neighborhood where the quenching occurs, at larger times, whereas photons from $H(2^2P)$ will appear practically at the origin of the TOF spectra. The signals from the detectors are separately preamplified and amplified by standard Nuclear Instrument Module electronics and a commercial time analyzer card (FastComTec 7888) processes and records the data. Throughout the experiment we have set the time analyzer card with a time resolution of 16 ns per channel (bin).

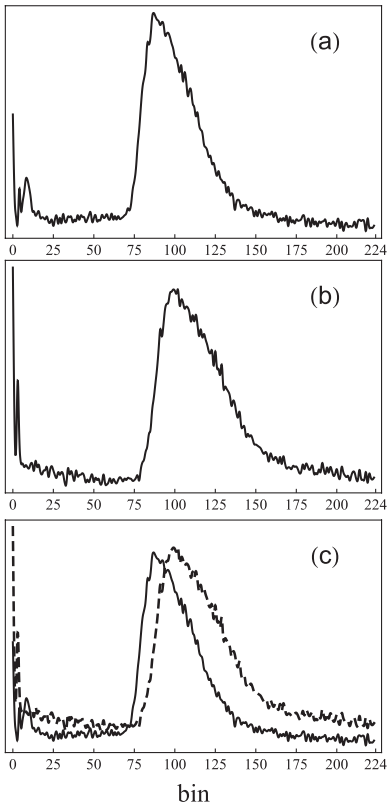

FIG. 1. Time-of-flight spectrum obtained with detectors A and B placed opposite of each other with respect to the collision plane and at distance of (a) 58 mm and (b) 67 mm from the H_2 dissociation spot. The H_2 excitation is accomplished by electron impact. The energy of the electrons is 200 eV and the electron pulse width is 100 ns. The horizontal axis displays time in units (bins) of 16 ns. The vertical axis corresponds to counting rate in arbitrary units. (c) Spectra A (full line) and B (dashed line) together displaying the shift associated with the distance difference.

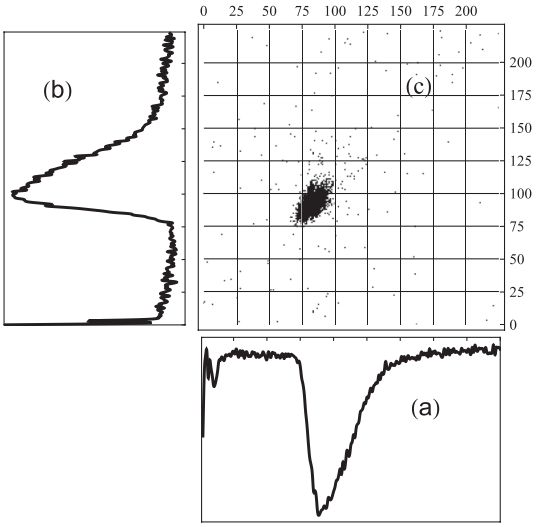

FIG. 2. (a) Same as Fig. 1(a). (b) Same as Fig. 1(b). (c) Coincidence counting spectrum, for the same experimental conditions of Fig. 1, containing a total of 3164 coincidence counts.

In Figs. 1(a) and 1(b) we show the TOF spectrum registered by both detectors, A and B, respectively. On the very left of both graphs we can observe a vertical line which corresponds to the H(2^2P) atoms, whose lifetime is of the order of 1 ns, and to the photons produced by the excitation of the radiative states of the hydrogen molecule. We observe that its width is of the order of 100 ns (the electron pulse width) as expected. In Fig. 1(a) we also still observe the presence of the scattered electrons, although not so clearly. Finally, in both figures we see the (large) peak corresponding to the *fast* H(2^2S) atoms. The peak of the *slow* atoms produced in the H_2 dissociation does not appear there and it is further to the right. In Fig. 1(c) we display both spectra in the same diagram to make clear the shift in time between them due to the different position of the detectors.

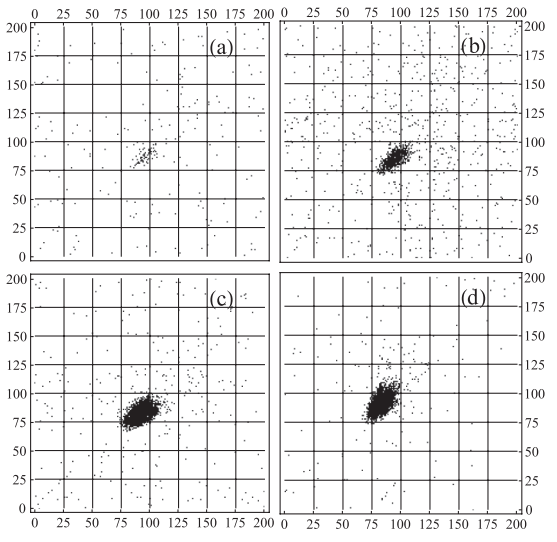

FIG. 3. Top view of the coincidence counting rate spectrum for different electron energies (a) 60 eV, (b) 80 eV, (c) 200 eV, and (d) 200 eV. (a)–(c) $L_A = 67$ mm and $L_B = 58$ mm, (d) $L_A = 58$ mm and $L_B = 67$ mm.

Now, let us discuss our coincidence measurement. The *start* of each sweep is established at the beginning of the electron beam pulse, while the signal coming from each detector feeds a different *stop* input into our time-of-flight acquisition card. Discrimination against noise is performed within the acquisition card by setting a ''lower threshold'' via software. Given a specific start signal, the arrival times provided by the stop's inputs of valid pulses are recorded with 16 ns resolution as integers corresponding to their bin number, where each detector furnishes a different column in the file. We label the bins of one detector as β_A and the other β_B. The analyzer card records all events where at least one stop for a given start was valid, and in this case a new line is added to the data file.

In Fig. 2 we display the top view of the coincidence counting spectrum [Fig. 2(c)] with its corresponding TOF spectra placed accordingly to the axis associated with each one; on the bottom panel [Fig. 2(a)] we have the former Fig. 1(a) and, on the left panel, Fig. 1(b). The coincidence spectrum reveals random coincidences occurring over the whole domain in the $\beta_A \times \beta_B$ plane with a very low count. It also shows a concentration of coincidences in a region which corresponds to the first third part of the fast H(2^2S) peak on both detectors. In fact, the coincidences occurring in this region have a counting rate much higher than the average, as we can see in Fig. 4, forming a peak inside the region expected to find the pair H(2^2S)-H(2^2S). This result was obtained during a round of the experiment which lasted 2 h and 8 min and yielded a total of 3164 coincidence counts.

In Fig. 3 we display the top view of the coincidence counting rate spectrum for different electron energies.

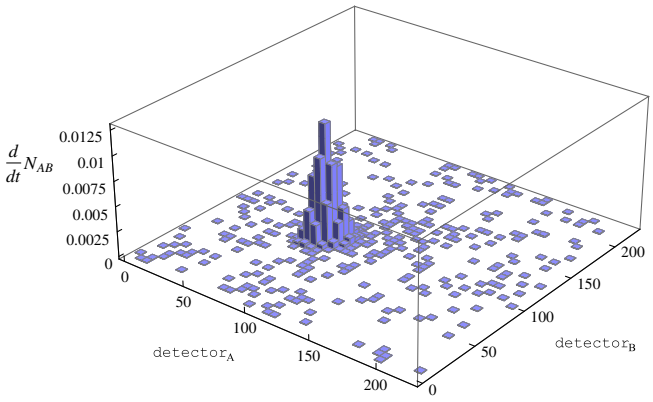

FIG. 4 (color online). Coincidence counting rate spectrum as function of the bins for the electron energy 200 eV and $L_A = 67$ mm and $L_B = 58$ mm.

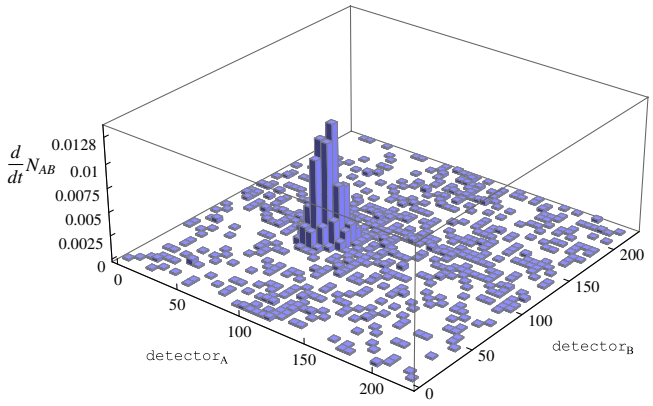

FIG. 5 (color online). Coincidence counting rate spectrum as function of the bins for the electron energy of 80 eV and $L_A = 67$ mm and $L_B = 58$ mm.

Additionally, regarding Figs. 1 and 2, the distance of the detectors were interchanged. Figure 3(a) shows the result obtained when the electron's energy was 60 eV, with a data acquisition period of 5 h and 51 min and resulted in a total of 286 coincidence counts. Figure 3(b) shows the result taken during 1 h and 56 min, for electrons with energy of 80 eV, involving a total counting number of 1424. Observe that, in spite of the lower acquisition time, the total counting number increased considerably in the whole $\beta_A \times \beta_B$ space. In addition, the small concentration, which barely appeared in Fig. 3(a), became stronger, revealing now the coincidence peak. In Fig. 3(c) the result for electrons with energy of 200 eV electrons and a period of 9 h and 17 min of data acquisition is shown. The total counting number in this case is 3362. As a result, when normalized with respect to the electron flux the relative cross section for pair production is in arbitrary units 1 for 60 eV, 12.27 for 80 eV, and 4.2 for 200 eV. This shows that the process obeys a threshold law and qualitatively shows that the probability of pair production, and its corresponding cross section, is larger for an electron energy of 80 eV than for 60 and 200 eV. However, these numbers are only an indication, since our electron gun cannot be guaranteed to remain stable over such long periods of time. Further work will be done in order to better estimate the cross sections.

In Figs. 4 and 5, we display a three-dimensional representation of the coincidence counting rate spectrum for different electron energies corresponding to Fig. 3(c) (200 eV) and Fig. 3(b) (80 eV). Comparison between the two 3D spectra show clearly that the random count rates with respect to the true ones are lower for the higher energy.

In conclusion, we have shown for the first time unambiguous experimental evidence of the measurement of fast metastable twin atoms. From this work several questions arise [for example, the cross section for the production of metastable H(2^2S) pairs as a function of the electron impact energy, the existence of other possible production channels, etc.]. These studies will require better precision in the detector positioning as well as much larger measuring times. Finally, from the theoretical point of view, a long path still remains to be followed in order to properly describe the superexcited state dynamics, not to mention the study of the entanglement of the two atoms.

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