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High Resolution Spectroscopy on Balmer-α Line Excited by Controlled Electron Impact

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An instrument with a high resolution interferometer was constructed for the determination of the line shape of the Balmer-α line of H* (n=3). The finesse of the etalons was 140, and its resolution was 0.020 Å. The high resolution spectra of the Balmer line produced in e-H_2, H_2O and CH_4 collisions were measured and their translational energy distributions were obtained. The fine structure splitting was observed for the slow component of H* (D*) from H_2 (D_2). This instrument is very useful for investigations of the mechanism of the dissociative excitation of hydride molecules by controlled electron impact.

1. Introduction

A state-to-state treatment of the reaction dynamics can be most easily carried out for the unimolecular decomposition of an excited diatomic molecules, since freedoms of the products are only the excitation energy and the translational energy of fragment atoms. The emission spectrum of the fragment atom gives these information[1)2)].

The translational energy distributions of ions and metastables were measured before by mass spectroscopic methods. Those of excited short-lived atoms, however, have become obtainable only recently; the derivative of their line shapes taken in a high resolution has been correlated with the translational energy distribution of the emitting atom[1)2)]. The spectral line shape has to be measured with a high resolution and accuracy for the successful application of this method. The translational energy distributions thus obtained are useful for the analysis of the mechanism of the dissociative excitation of hydride molecules upon controlled electron impact. The investigations on H_2[1)3)], D_2[4)], HCl[2)], H_2O[5)] and CH_4[6)] have been carried out.

In the present paper, the experimental details for obtaining a high resolution spectrum of the Balmer-α line of H*(n=3) and D*(n=3) are shown together with some applications.

2. Instrumentation

The experimental apparatus consists of a collision chamber, a high-resolution spectrometer, a photon counting system, and a vacuum line for sample preparation. The sample is excited in the collision chamber by a collision with an electron. The photoemissions produced are observed and analyzed. The schematic diagram of the apparatus is shown in **Fig. 1**.

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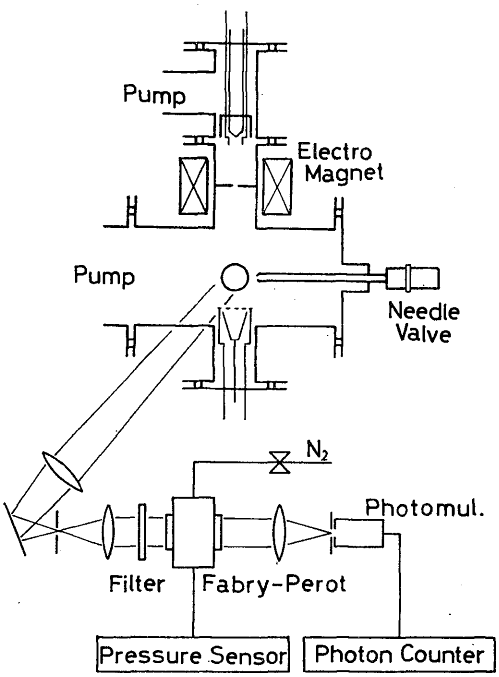

Fig. 1. Schematic diagram of the apparatus.

Collision chamber is made of stainless steel. Two oil diffusion pumps of 650 ls^{-1} and 250 ls^{-1} continuously evacuate it; the base pressure is of the order of 10^{-7} Torr and the operating pressure of 10^{-4} Torr as measured by an ULVAC GI-TL2 ionization gauge. The electrons from a heated tungsten filament are accelerated, collimated and introduced into the collision region. An electromagnet of about 30 G is used for the collimation. The sample gas, which may be degassed by repeated freeze-pump-thaw cycles, is jetted into the collision region through a needle valve and a multichannel nozzle. The collision chamber has two quartz windows and the photoemission can be measured at angles of 90° and 55° with respect to the electron beam; 55° is the magic angle where the effect of anisotropy can be ignored in the Born approximation.

The photoemission is collected with a spherical mirror and made a parallel beam through a pinhole (1 mm) and a lens. The Balmer-α line is separated with an IF-B interference filter (Vacuum Opt. Jpn.). A Fabry-Pérot interferometer (Mizoziri Optics) is used for recording the Balmer-α line in a high resolution; the wavelength scanning of which was carried out by varing the pressure of nitrogen gas between two etalon plates. The etalon plates are made of quartz, polished for a surface flatness of better than 1/250 λ and coated with dielectric material at about 98 % reflectance for the 630–660 nm region.

The photons are detected with an HTV R649 photomultiplier with a C659 thermoelectric cooler, and they are counted with an NF-PC545A or an HTV-C1230 photon counter. The data are accumulated and analyzed with a SORD M170 microcomputer.

3. Instrument performance

The performance of the interferometer for the Balmer-β line was shown before[7]; the finesse was 100 and the resolution was 0.024 Å for a Cd line at 4799 Å.

This interferometer is found to be also very excellent for the Balmer-α line. The performance test was carried out with an NEC GLG5000 He-Ne laser at 6328 Å as the light source. A typical line shape of the laser is shown in **Fig. 2**. The finesse is 140 and the resolution is 0.020 Å; this result is better than the previous one for the Balmer-β line.

The slow component of the excited hydrogen atom produced in e-H_2(D_2) collisions gives a relatively narrow line[3].

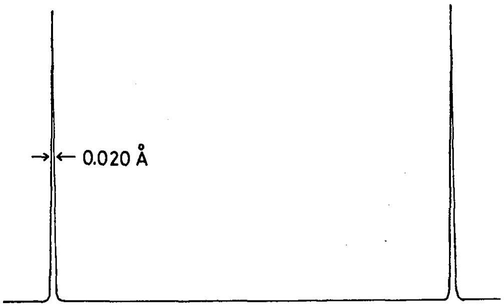

Fig. 2. A typical spectrum of a He-Ne laser (6328 Å).

This component was observed separately and the result is shown in **Fig. 3.**

The Balmer-α line consists of several fine structure components and appears as a doublet in an usual high-resolution spectrum; they are the $2P_{1/2}$-$3D_{3/2}$ and the $2P_{3/2}$-$3D_{5/2}$ transitions[8]. The line in **Fig. 3** reveals this doublet structure as a shoulder. Thus, the analysis of the line shape of this component has to be carried out with the consideration of the fine structure.

These results indicate that the interferometer is suitable for the line shape analysis at a resolution of 0.02–0.04 Å.

4. Applications

The high-resolution spectra of H*(n=3) produced in e-H_2, H_2O and CH_4 collisions have been measured as are shown in **Fig. 4** (top). The electron energy is 100 eV and the electron-beam current is 600–800 μA. The line shape is different from each other and reveals the difference of mechanisms of the formation of H* from the three molecules.

The translational energy distribution of H* was obtained by the method described before[1)2)] as is shown in **Fig. 4** (bottom). The line shapes are smoothed once by using a three point averaging and then differentiated. This method is strictly correct when the observed line shape is equal to the true line shape. The anisotropy is usually small at such a high excitation energy. Thus, in the present experiment the largest source of deformation of the line shape comes from the experimental uncertain-

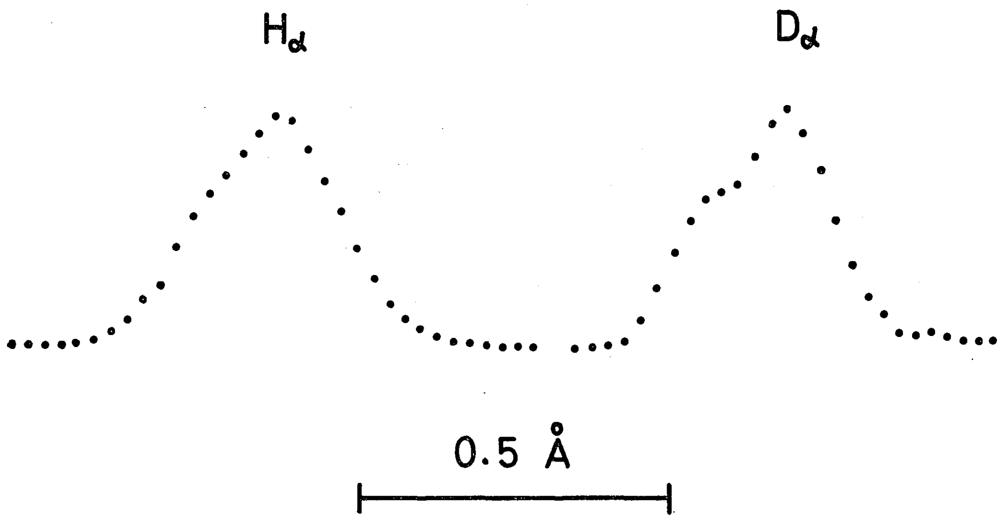

Fig. 3. A typical spectrum of the slow component of H*(n=3) and D*(n=3) obtained by controlled electron impact on H_2 and D_2 (100 eV).

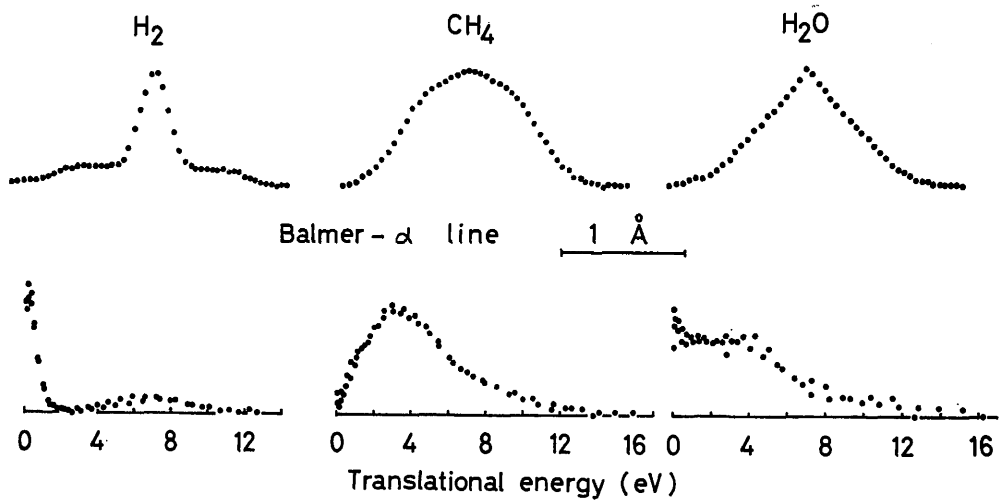

Fig. 4. Line shapes and translational energy distributions of H*(n=3) from H_2, CH_4 and H_2O (100 eV).

ties.

The translational energy distributions of H* from the three molecules differs considerably from each other. All of them, however, consists of two or more components. H* from H_2 and H_2O has a slow component, which has a peak of translational energy at about 0 eV; however, the slow component is weak for methane. The excited hydrogen atom produced through a repulsive potential curve has a large translational energy. Such fast component is one of the major one for all of the three molecules. The details of assignments has been or will be described elsewhere[1)~6)].

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