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TWO-STEP EXCITATION OF MOLECULAR HYDROGEN

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Résumé. — Des molécules d'hydrogène excitées par impact électronique dans l'état métastable $c\ ^3\Pi_u$ sont portées par résonance optique dans un état $^3\Sigma_g$, $^3\Pi_g$ ou $^3\Delta_g$. La sélectivité de l'excitation électronique est mise en évidence. La lumière du laser utilisée pour la seconde excitation est polarisée linéairement ; l'alignement ainsi produit a permis d'observer l'effet Hanle du niveau $3\ d\delta\ ^3\Delta_g$, $v = 2$, $K = 2$, $I = 1$.

Abstract. — A stepwise excitation of H_2 is performed. Electron impact populates the metastable $c\ ^3\Pi_u$ state then an optical resonance allows the excitation of $^3\Sigma_g$, $^3\Pi_g$ or $^3\Delta_g$ states. We observed the selectivity of the electron impact excitation. The laser light used for the optical resonance was linearly polarized, the alignment produced allowed the observation of the Hanle effect on the $3\ d\delta\ ^3\Delta_g$, $v = 2$, $K = 2$, $I = 1$ level.

1. Introduction. — Two-step excitation of atoms is now a quite common technique : the atomic metastable level is populated by a weak discharge then, by photoabsorption higher excited levels can be reached [1]. This can also be done on a molecule. The main difference is that an atom has usually one or very few metastable levels ; a molecule has as many levels as vibrational and rotational quantum numbers. The first step excitation populates all of them, and only one among these levels can be used as a step for the second excitation. Therefore, the filling of that intermediate state is much less efficient for a molecule than for an atom. On the other hand, such an excitation is very selective and is of particular interest for molecules which have very dense emission spectra. In particular large alignment in the excited state may be created in this way.

2. Experimental set-up. — In the present experiment, the usual weak discharge [1] is replaced by slow electron impact. Thus an electron gun is placed in a tube filled with a few millitorrs of H_2 .

The resonant light is provided by a jet dye laser pumped by an Ar^+ laser. A rotating Lyot filter is used to select and tune the dye laser wavelength. The spectral width is 0.3 Å. The dye laser power used was about 200 mW. The laser light crosses the electron beam at right angle, the fluorescence is detected through a monochromator by a photomultiplier at right angles to the laser and the electron beams. A resolution of about 100 000 is needed to resolve most

of the lines of the H_2 spectrum emitted after electron impact. The laser excitation is very selective, thus the resolution needed for the detection of the two-step excitation depends only on the efficiency of this process compared to the electron impact in the same conditions. A resolution of about 2 000 was used.

The optical resonance is observed by tuning and detuning the laser wavelength.

3. The observed transitions. — The metastable state has the configuration $(1s\sigma, 2p\pi)\ ^3\Pi_u$, therefore only the $^3\Sigma_g$, $^3\Pi_g$ and $^3\Delta_g$ states can be reached through an optical resonance.

Because of the Λ -doubling, the Π and Δ levels are split into two components and therefore the $\Pi \rightarrow \Pi$ and $\Pi \rightarrow \Delta$ transitions are also split into two lines which are well observed in the spectrum produced by electron impact. In the case of the observed transitions $(1s\sigma, 2p\pi)\ ^3\Pi_u\ v = 0, 1, 2 \rightarrow (1s\sigma, 3d\delta)\ ^3\Delta_g\ v = 0, 1, 2$, their separation is between 5 and 0.6 Å, and the spectral width of the laser is narrow enough to separate them. We observed only one resonance. This fact is direct evidence for predissociation of the $2p\pi\ ^3\Pi_u^+$ component by the repulsive $2p\sigma\ ^3\Sigma_u^+$ state, through rotational interaction [2], its lifetime is then not long enough to allow absorption of photons, in our experimental conditions.

4. The excitation characteristics. — The excitation of the $3d\delta\ ^3\Delta_g\ v = 2, K = 2$ ortho component from the intermediate $2p\pi\ ^3\Pi_u^- \ v = 2, K = 1$ level by absorp-

tion of the $R(1)$ line ($5\,785\text{ \AA}$) is used as a test, the detection being on the $Q(2)$ line

$$^3\Pi_u v = 2 K = 2 \leftarrow ^3\Delta_g v = 2 K = 2 (5\,822\text{ \AA}).$$

In figure 1, one can see the change of the intensity of the $Q(2)$ line as the laser is tuned through the resonance. The amplitude of this resonance depends on the population rate by electron impact of the intermediate $2p\pi\ ^3\Pi_u v = 2$ level (located at 12.35 eV) while the off-resonance intensity is due to the impact excitation of the upper level : $3d\delta\ ^3\Delta_g v = 2$ (located at 14.50 eV). Their separation in energy is about 2 eV . Figure 2 shows the dependence of these intensities on the incident electron energy. Therefore electron impact is selective enough to populate the lower level without populating the upper one. In these conditions the cascade effects can be cancelled out and because all the H_2 levels radiating in the visible are lying between 14.4 and 15.4 eV , it seems possible to observe two-step excitation of H_2 without any selectivity at the detection.

The laser light is linearly polarized, therefore the excited level may be aligned. Indeed we observed 23% polarization on the $Q(2)$ line induced by absorption of the $R(1)$ line.

Electron impact is also known to induce some

FIG. 1. — Observation of the optical resonance. The intensity of the $^3\Pi \leftarrow ^3\Delta$ $Q(2)$ line is detected as the laser wavelength is swept through the $R(1)$ line value. The background is due to the excitation of the $^3\Delta$ level by electron impact.

FIG. 2. — The relative intensities of the $Q(2)$ line *a*) by electron impact, *b*) by stepwise excitation, as the electron energy is varied.

alignment in the excited states but in the case of molecules it is observed to be small [3]. We indeed fail to observe any on the $^3\Pi \rightarrow ^3\Delta$ transitions after electron impact.

5. Hanle effect. — All the conditions needed for the observation of Hanle effect [1] were thus fulfilled. The electron gun was placed inside Helmholtz coils, with the electron beam parallel to the magnetic field. The polarization of the laser was now parallel to the direction of observation. With these conditions we observed the Hanle effect with a typical width of 16.0 gauss .

Molecular hydrogen is known to correspond to Hund's coupling case *b* : the orbital momentum L is coupled to the internuclear axis to give Λ, Λ and the angular momentum R of nuclear rotation form the resultant N . The spins are not coupled to the internuclear axis but to N . From this model it is possible to calculate the Landé-factor of the $^3\Delta_g$ excited levels. The fine and hyperfine structures are not resolved ; the mean value of g in this particular case is about 1, therefore the deduced value for the lifetime is about 10^{-7} s . It has the same order of magnitude than the lifetime value of the corresponding configuration of the He atom.

This observation constitutes only a preliminary experiment to show that two-step excitation may align many excited molecular levels and thus allowing such experiments as : Hanle effect, level crossings, quantum beats and so on.

References

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