

Lyman series emission of valence and inner-shell excited gaseous H₂O

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Synopsis We present emission cross section functions for Lyman series fluorescence emission after photoexcitation of water molecules with monochromatized synchrotron radiation as function of the exciting-photon energy. Both the valence and inner-shell excitation energy range of the water molecule are covered. The results are compared to literature and contribute to complete the model of dissociative relaxation dynamics of excited small molecules.

Its unique role in nature made the water molecule always subject to intense investigations of theoretical and experimental research. Of particular interest, e.g. in atmosphere physics and chemistry, radiation biology and fundamental photochemistry, is the interaction with radiation and subsequent decay processes such as photoexcitation, -ionization, and -dissociation.

The tracking of neutral fragments is challenging, because they cannot be guided by electric or magnetic fields as it is the case for ionic particles. However, if they are excited, fluorescence spectrometry is suitable for their investigation. The Balmer emission series as well as Ly_α after photoexcitation of water molecules have been measured as functions of the exciting-photon energy in previous experiments and absolute emission cross sections are available for some exciting-photon energy ranges [1,2,3].

However, for the completion of models of dissociation dynamics, measurements of the emission of higher Lyman transitions ($\geq \text{Ly}_\beta$) are essential, as the Lyman decay is the dominant channel for all excited hydrogen fragments.

Here, we present experimentally determined cross sections for Lyman series emission as function of the exciting-photon energy. The experiments were performed with tunable synchrotron radiation in the range of valence excitations of the water molecule (15 – 34 eV) as well as for excitation in the oxygen inner-shell (1s) electron range (533 – 542 eV). In the valence energy range, thermodynamical limits of production of excited hydrogen fragments are measured and agree with previous results. Above those limits, absolute emission cross sections are determined by calibrating to literature values (see Fig. 1). Oscillator strengths at emission maxima are calculated and branching ratios between Lyman and Balmer transitions are determined and compared to theoretical values.

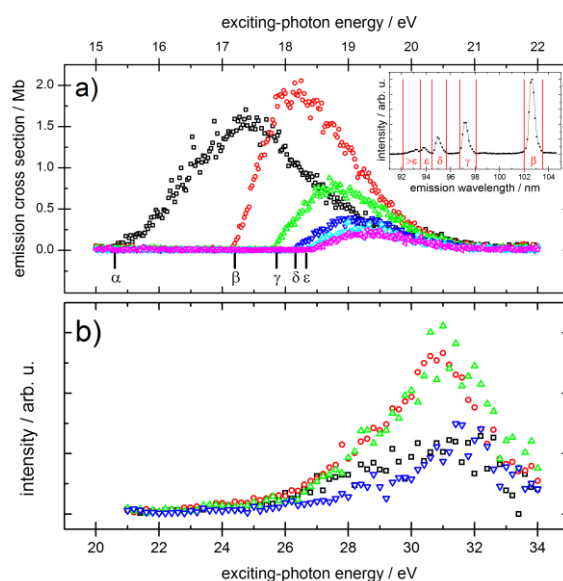


Figure 1. a) Exciting-photon energy dependent fluorescence emission cross sections: a) Absolute Lyman series emission cross sections after excitation of $1b_2^{-1}$ states of the water molecule. Black squares: Ly_α, red circles: Ly_β, green triangles: Ly_γ, blue triangles: Ly_δ, cyan diamonds: Ly_ε, magenta triangles Ly_ζ. b) Relative Lyman series emission cross sections after excitation into doubly excited and $2a_1^{-1}$ states.

In the inner-shell excitation range, the emission function is found to be strongly dependent on the character of the excitation, as observed before for the Balmer series [3].

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References

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