PHYSICAL REVIEW A **85**, 034502 (2012)

Experimental isotope shifts in Stark recurrence spectra of Rydberg D_2 and H_2 molecules

J. Haestad, A. Murphy, J. D. Wright, and T. J. Morgan
Department of Physics, Wesleyan University, Middletown, Connecticut 06459, USA

(Received 25 January 2012; published 12 March 2012)

D_2 and H_2 Stark recurrence spectra have been measured at scaled energies of $\epsilon = -2.3$ and $\epsilon = -3.2$. An isotope shift in the scaled-action locations of recurrence peaks is found and explained by the increased density of states associated with perturbing rotational series. This reinforces the insight gained in previous work on the correspondence between the structures of a quantum spectrum and its classical periodic orbits.

DOI: 10.1103/PhysRevA.85.034502 PACS number(s): 33.15.−e, 33.80.−b, 32.60.+i, 32.80.Ee

Recently, Wright *et al.* [1] measured the Stark recurrence spectrum of H_2 and observed two primitive recurrences in the spectrum due to the presence in the photoabsorption spectrum of two Rydberg series converging to different rotational core states. Diffractive inelastic scattering recurrence peaks were also found resulting from configuration interaction between the two Rydberg series. We have carried out a new set of measurements using both H_2 and D_2 in order to study the effect of mass on these recurrences.

The experimental apparatus used is identical to that in Ref. [1] with the only change being a different experimenter and the use of D_2 as well as H_2. A brief outline of the experiment follows. A 4-keV D_2^+ ion beam is neutralized in potassium vapor and the resulting neutral molecules in the $2p\pi\ c\,^3\Pi_u$ metastable state are excited to Rydberg states in the presence of an electric field using a tunable dye-laser system. The Rydberg states are field ionized and the resulting ions detected with a channel plate. A field-ionization spectrum is recorded over the principal-quantum-number range of $n = 16$ to 26 while holding the scaled energy $\epsilon = EF^{-1/2}$ constant. The energy E is the electron binding energy relative to a Rydberg-series limit and F is the applied electric field. This maintains the classical dynamics unchanged over the entire spectrum [2]. The scaled spectrum is Fourier transformed to obtain the recurrence spectrum, i.e., signal strength versus orbit scaled action S [3].

Figure 1 shows the measured recurrence spectrum of D_2 (upper panel) and H_2 (lower panel) at $\epsilon = -3.2$ and Fig. 2 shows the spectrum at $\epsilon = -2.3$, both for scaled action up to $S = 4.0$. Both energies are beyond the Inglis-Teller limit with two (three) n manifolds overlapping for $\epsilon = -3.2$ ($\epsilon = -2.3$). We refer the reader to Ref. [1] for additional details about molecular recurrence spectra.

The peak labeled p in the figures is the primitive (smallest-action) recurrence associated with the scaled Rydberg excitation series ($R = 1$ for H_2 and $R = 2$ for D_2, where R is the rotational quantum number). This peak occurs at scaled action $S = 1$ due to scaling. Each recurrence spectrum is normalized such that this peak has unit height. The peak labeled p' in the figures is due to the presence in the photoabsorption spectrum of interloping Rydberg series ($R > 1$ in H_2 and $R > 2$ in D_2). As can be seen in the figures, p' is at lower action than p. This occurs because the interloper series in both the H_2 and D_2 spectra is at a lower principal-quantum-number range compared to the main series and has larger energy separation between Rydberg levels, thus contributing a lower-frequency component to the Fourier transform.

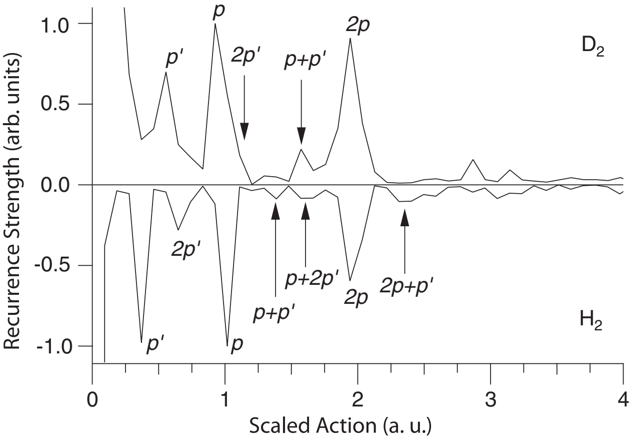

FIG. 1. Recurrence spectra at $\epsilon = -3.2$ of D_2 and H_2 obtained by Fourier transform of experimental photoabsorption spectra. For D_2 (H_2), the peaks labeled p' and p are the primitive recurrences of the $R > 2$ and $R = 2$ ($R > 1$ and $R = 1$) rotational series. The first primitive recurrence repetitions are labeled $2p'$ and $2p$. Other labeled peaks are combination diffractive recurrences from inelastic scattering between the two rotational series.

An isotope effect is observed in the spectra with the p' peak occurring at higher action in D_2 compared to H_2. The reason for this is that the interloper-series ionization limit is raised above the main-series ionization limit by less in D_2 than in H_2. The rotational constant of D_2 is

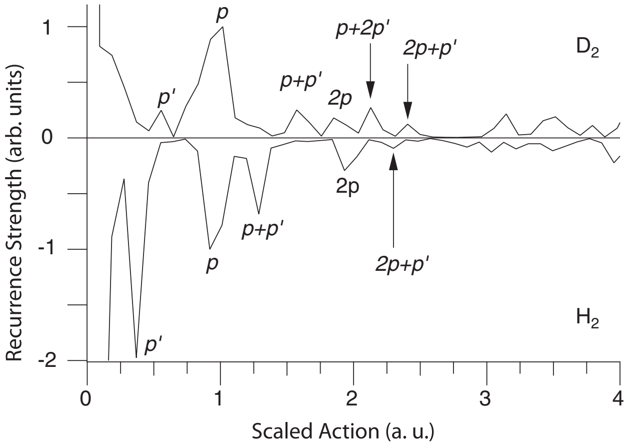

FIG. 2. As Fig. 1 but for $\epsilon = -2.3$.

approximately half that of H_2 (3.71 versus 7.35 meV), due primarily to the mass difference. This results in a higher density of lower-n interlopers in the experimental range of the photoabsorption spectrum in D_2 compared to H_2 and consequently a slightly higher frequency in the recurrence spectrum. There is also an isotope shift in the diffractive inelastic scattering peaks in the spectra as can be seen by examining the action location of the recurrence peak labeled $p + p'$ in the figures. This peak is due to the inelastic scattering between the two primitive recurrence trajectories and is the signature in the spectrum of interaction between rotational series [1,4,5].

The recurrence spectra at the two scaled energies are basically the same with a few differences in details such as the amplitude of particular peaks. The spectra characterize the multichannel semiclassical dynamics between the Rydberg electron and the molecular ion core. By using different isotopes, the Coulomb dynamics is preserved while the rotational dynamics is changed. These observations provide insight into the differences between molecular hydrogen and deuterium and emphasize the correspondence between the quantum structure and the classical limit [6,7].

We wish to acknowledge the NSF for support.

[1] J. D. Wright, J. M. DiSciacca, J. M. Lambert, and T. J. Morgan, Phys. Rev. A **81**, 063409 (2010).

[2] U. Eichmann, K. Richter, D. Wintgen, and W. Sandner, Phys. Rev. Lett. **61**, 2438 (1988).

[3] M. L. Du and J. B. Delos, Phys. Rev. A **38**, 1896 (1988).

[4] A. Matzkin and T. S. Monteiro, Phys. Rev. Lett. **87**, 143002 (2001).

[5] A. Matzkin, P. A. Dando, and T. S. Monteiro, Phys. Rev. A **66**, 013410 (2002).

[6] R. V. Jensen, H. Flores-Rueda, J. D. Wright, M. L. Keeler, and T. J. Morgan, Phys. Rev. A **62**, 053410 (2000).

[7] J. D. Wright, H. Flores-Rueda, W. Huang, and T. J. Morgan, Phys. Rev. A **79**, 052510 (2009).