Doppler tuning vuv spectroscopy of D^- over an extended photon-energy range around the $n=2$ threshold

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The giant (or shape) resonance in the photodetachment cross section of D^- above the $n=2$ threshold has been subjected to high-resolution vacuum ultraviolet spectroscopy performed by Doppler tuning of ions stored in the ASTRID storage ring. In order to prevent changes in the overlap between laser and ion beams over the large kinetic-energy range employed in the experiment, a new ion-beam-positioning method based on a quadrupole-shunt technique was applied. The study presents an accurate measurement of parameters for the resonance, which is also denoted ${}_2\{0\}_2^+\ {}^1P^o$. The resonance has a width of 26(2) meV while the asymmetry parameter q of the Fano profile is 3.2(0.4), which is at variance with the most recent theoretical calculations.

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The photodetachment spectrum of the negative hydrogen ion is dominated by a number of series converging to the thresholds associated with excited states of the neutral hydrogen system. The entire spectrum of H^- has been investigated in the group of Bryant and co-workers (see [1] and references herein). This experimental effort has provided a wealth of information about the fundamental negative-ion system, but unfortunately the limited resolution in the investigations has hampered detailed conclusions on structural parameters. During the last ten years, a complementary study has been undertaken at the Aarhus heavy-ion storage ring in Denmark, ASTRID [2–5]. These studies have demonstrated a much improved resolution over small photon-energy regions [5]. A previous investigation of the broad ${}_2\{0\}_2^+\ {}^1P^o$ resonance above the $n=2$ threshold was, however, of limited use, since the measurement was strongly influenced by systematic changes in the overlap between the ion and laser beams during large changes in the ion kinetic energy.

In the present study, we present the results of an investigation of the ${}_2\{0\}_2^+$ resonance by means of a method for ion-beam positioning in storage rings [6] that allows for a detailed control of the overlap between the laser and ion beams over an extended storage-energy range. The measured cross-section profile exhibits a single broad resonance, which can be fitted with a Fano profile. The resonance parameters resolve a discrepancy in previous experimental investigations and allow a detailed comparison with theoretical results.

The experimental procedure has been described in detail in previous publications [4,5]. Negative deuterium ions are formed in a duo-plasmatron ion source, accelerated to 150 keV and injected into the ASTRID storage ring. In the ring, the ions are further accelerated by a radio-frequency cavity to a final kinetic energy in the few MeV range. Along one of the straight sections of the storage ring, the ion beam is overlapped almost collinearly with a 118-nm laser beam. By tuning the ion-beam velocity, the Doppler-shifted photon energy covers the region of interest around the $n=2$ threshold. The negative ions that are neutralized along the straight section due to both collisions and photodetachment are not deflected by the bending magnets of the storage ring and impinge on an electron-multiplier detector after the magnet. The light flux is measured by determining the photocurrent on a biased aluminum plate at the exit of the storage ring, which allows a normalization of the detachment signal to the flux.

A measurement of the neutral-atom production (corrected for collisionally induced neutrals) versus the photon energy provides a measurement of the photodetachment cross section, assuming that the overlap between the laser and ion beams is constant. While the geometry of the light beam is easily controlled and kept stable, the same is not true for the ion beam. The position of an ion beam stored in a heavy-ion storage ring is influenced by two factors. First, the ions follow a certain trajectory in the ring. The ideal trajectory describes a straight line between the bending magnets and going through the middle of focusing quadrupole magnets, but much freedom exists, and in general a stored beam will have an average trajectory that differs from this ideal trajectory. Second, the ions perform oscillations in the harmonic trapping potential, the so-called betatron oscillations.

In the previous photodetachment studies at ASTRID, the ion-beam positioning was performed by means of electrostatic pickups positioned at eight locations around the ring. These pickups allow a quite sensitive position measurement at the point where they are located, but the ion-beam position throughout the ring (i.e., between pickups) can be estimated only by modeling the ion trajectory in the magnetic lattice. Our previous investigations have demonstrated that this method could lead to strong variation in the laser-ion-beam overlap when the ion kinetic energy is scanned over a significant range. The most probable explanation for these fluctuations are slight differences in the ion trajectory that are caused by deviations from the ideal orbit.

The quadrupole magnets of the storage ring serve to focus the ion beam and hence avoid beam loss due to a gradual expansion of the ion beam. However, quite analogous to

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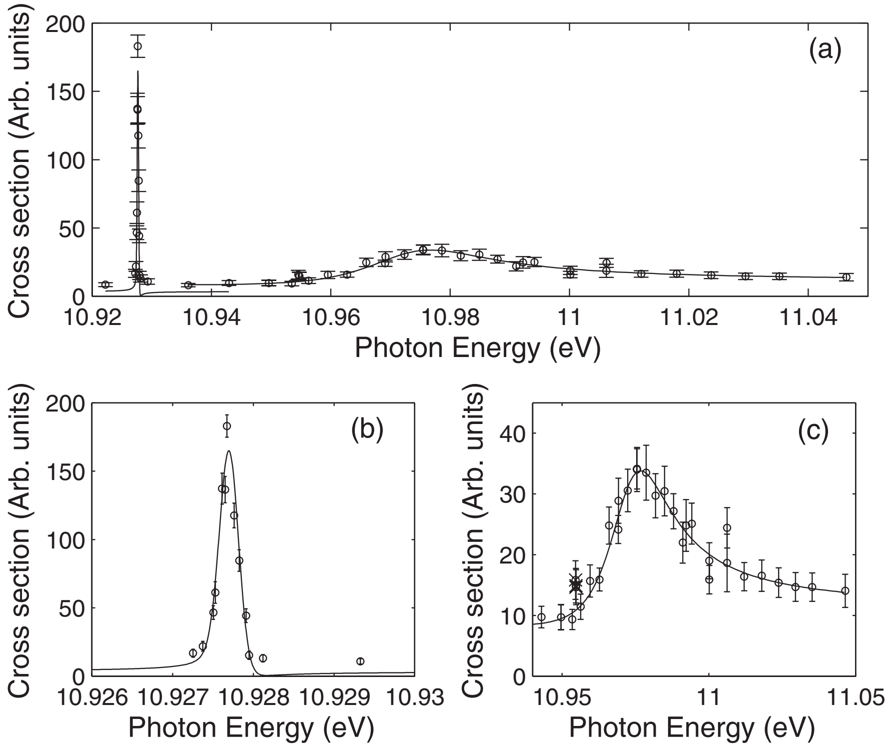

FIG. 1. Relative D^- photodetachment cross section of the region around the $n=2$ threshold. The narrow ${}_2\{0\}_3^-$ resonance is located below the threshold, while the broad ${}_2\{0\}_2^+$ (shape) resonance is located above the threshold. The weak ${}_2\{0\}_4^-$ resonance just below the $n=2$ threshold is barely visible above the noise. The error bars indicate a statistical uncertainty of one standard deviation. The solid curves represent the best fits to the two major resonances, as described in the text. (b) and (c) show expanded views of the two major resonances. In (c), the data points corresponding to the ${}_2\{0\}_4^-$ resonance are marked with crosses. These points have been left out of the fit.

classical optics, an ion beam passing through the quadrupole lens off axis will also experience a deflection. This deflection is, of course, dependent on the ion-beam energy. Since a change in the storage energy in ASTRID is associated with a scaling of all magnetic fields proportional to the momentum, this deflection should also remain constant to lowest order. However, the previous studies show that the compensation is not perfect.

In order to reduce this problem, a new method for ion-beam alignment in a storage ring has been introduced. The technique was first implemented for optimization of laser-cooling experiments and worked only at the injection energy for the storage ring, but it was subsequently extended to allow for diagnostics on an accelerated beam [6]. The technique relies on the above observation that an ion beam passing through the quadrupoles off axis is deflected. Thus, by inducing a small change in the current of one specific quadrupole (a so-called quadrupole shunt) and using the position pickups to measure the change in the position of the stored beam, it is possible to infer the position through the quadrupole. By performing this procedure on all the 16 quadrupoles of the ring, a much more accurate position of the ion beam can be obtained, and the ability to obtain storage with a trajectory closer to the ideal orbit can potentially reduce or eliminate changes in the overlap.

In practice, the quadrupole-shunt beam positioning was performed at two storage energies corresponding to a photon energy on either side of the resonance under investigation. Between these two reference points, the magnetic fields were interpolated, and the quality of this interpolation was checked by a position measurement in between the two reference points. No deviation from an ideal orbit was observed to within ~ 1 mm.

The measured neutral-atom signal versus photon energy is shown in Fig. 1. Data points have been taken in random order to reveal any possible long-term drifts, which were fortunately not observed. The cross section shows two pronounced features: A narrow resonance below the $n=2$ threshold and a broad resonance above this threshold. These have previously been observed both by Bryant and co-workers (see [1] and references therein) and in our previous investigations [2]. There is in fact a third resonance between the two large features, as previously reported [3]. The aim of the present study is to investigate the broad resonance above the threshold, and the weak third resonance can only just be seen above the noise in Fig. 1.

The two narrow resonances are the two lowest-lying members of a series of resonances. The resonances are labeled according to the ${}_n\{\nu\}_m^{A}\ {}^{2S+1}L^{\pi}$ scheme [7] as the ${}_2\{0\}_3^-\ {}^1P^o$ and the ${}_2\{0\}_4^-\ {}^1P^o$ resonances. The broad resonance is the ${}_2\{0\}_2^+\ {}^1P^o$ resonance, historically often denoted the shape resonance. In order to quantify the resonances, they are fitted to appropriate functions. All three resonances should in principle be well described by a Fano profile, but the two narrow resonances have a width that is small compared to the experimental resolution. The experimental reso-

TABLE I. A comparison of experimental and theoretical results for the $^1P^o$ resonance of D^-.

	E_R (eV)	Γ (meV)	q	$S({}_2\{0\}_2^+)/S({}_2\{0\}_3^-)$
	Experiment			
Present	10.9727(9)[a]	26(2)	3.2(4)	16(3)
Bryant *et al.* [8]	10.9735(7)[b]	21.2(1.1)	4.9(3)	18(2)
Comtet *et al.* [9]	10.9766(3)[b]	22.2(6)	5.5(2)	18(2)
Halka *et al.* [10]	10.9753(3)[b]	21(1)	5.3(2)	
Halka *et al.* [10]	10.9713(3)[b]	30(1)	4.5(1)	
	Theory			
Sadeghpour *et al.* [13][c]	10.9769	18.6		
Ho and Bhatia [14][c]	10.9737	18.8		
Cortes and Martin [15]	10.9770	22.6		
Tang *et al.* [16][c]	10.9748	16.9		
Lindroth [17]	10.9734	18.5	4.0	14
Bürgers and Lindroth. [18]	10.9728	19.1		
Bylicki and Nicolaides [19]	10.9730	19.3		

[a]Determined from the measured separation between the two resonances ${}_2\{0\}_2^+$ and ${}_2\{0\}_3^-$ of 0.0450(9) eV and using the position of the ${}_2\{0\}_3^-$ resonance from Ref. [3].
[b]Based on measurements on resonance positions in H^- corrected for a systematic error of 1.9(9) meV; see [11,12]. Converted to D^- energies using the conversion factor 27.211 383$M/(M+m)$ eV [20]. Ground-state energies of 14.356 80 eV (D^-) and 14.352 64 eV (H^-) relative to the two-electron escape threshold are derived from Ref. [21], neglecting the small mass dependence of the relativistic and QED corrections.
[c]Converted from the total energy of the system (in a.u.) to the energy with respect to the ground state of D^- using a conversion factor of 27.211 383$M/(M+m)$ eV [20]. Ground-state energies for D^- and H^- are described in footnote b above.

lution, which is limited by the momentum spread of the ions in the storage ring, must therefore be included. This is done by convolving the Fano profile with a Gaussian. In a previous study, momentum-spread reduction (electron cooling) has been applied, leading to a much improved resolution (see Ref. [5]). Since the aim here is to investigate the broad resonance, electron cooling was not applied in the present study. Consequently, for the ${}_2\{0\}_3^-\ {}^1P^o$ resonance, the intrinsic resonance parameters—the width Γ and the asymmetry parameter q—are taken from the previous investigation. The data are fitted by adjusting the width of the Gaussian representing the momentum spread and the position of the resonance. For the ${}_2\{0\}_2^+\ {}^1P^o$ resonance, the position, width, and asymmetry parameters are fitted. It should be noted that the photon-energy scale is normalized to previous measurements of the position of the ${}_2\{0\}_3^-\ {}^1P^o$ resonance, so the present investigation does not represent an independent measurement of this resonance energy, but the splitting between the two resonances is well determined in the present work.

Figures 1(b) and 1(c) show an expanded view of the two main resonances. The data in both panels represent the same experimental run and the vertical scales of the two measurements can be directly compared. This means that the relative strength of the two resonances can be measured. The solid curves in Fig. 1 represent the best fit to Fano functions (convolved with a Gaussian for the narrow ${}_2\{0\}_3^-$ resonance, as described above). The fitting range is the same as the range where the fitted curve has been plotted. The parameters from the fit are given in Table I. For the ${}_2\{0\}_2^+\ {}^1P^o$ resonance, the data points corresponding to the position of the small ${}_2\{0\}_4^-\ {}^1P^o$ resonance are excluded from the fit [marked with crosses in Fig. 1(c)]. A separate fit to this resonance (not shown) was consistent with previous position measurements [3].

The previous experimental determinations of the width of the ${}_2\{0\}_2^+\ {}^1P^o$ resonance exhibit a discrepancy. While the most recent values for the width are in the ~20 meV range [10], earlier data for unexplained reasons indicated a significantly larger width of ~30 meV [10]. The present value is right between the two earlier values. It should be noted that the resolution in previous measurements corresponded to 1/3 of the resonance width, while, in the present study, the experimental resolution is negligibly small compared to the resonance width.

Generally, the most recent theoretical calculations predict a position for the resonance that is in agreement with our measurement. The predicted width is, however, in general smaller than the present measurement. In other words, the lifetime of the ${}_2\{0\}_2^+\ {}^1P^o$ state is shorter than predicted by theory. The reason for this is not clear. Bryant has previously proposed that an additional decay mechanism should originate from "nuclear recoil" phenomena [22]. However, to the extent that these finite-nuclear-mass effects can be described perturbatively in a complex-coordinate-scaling calculation, Bürgers and Lindroth calculate a contribution to the width that is only ~30 μeV. Another issue is the validity of the Fano parametrization for such a relatively broad resonance. For instance, any variations in the background cross section will perturb the resonance shape. However, within the statistical limitations of the current experiment, we cannot observe any deviation from a perfect Fano profile. In addition, fits to cross-section profiles corrected for an energy-dependent background cross section in the analysis of previous experimental investigations yielded very similar results for the resonance parameters [10].

The relative strength of the ${}_2\{0\}_2^{+}\,{}^1P^o$ and the ${}_2\{0\}_3^{-}\,{}^1P^o$ resonances is in perfect agreement with the previous investigations by Bryant and co-workers. This is also expected since this number is to a first approximation independent of the experimental resolution. It can be taken as an indication that the measures taken to ensure a constant overlap in the present study are successful. The only available theoretical calculation predicts a slightly lower ratio of the strength, which may reflect the above-mentioned underestimation of the width of the ${}_2\{0\}_2^{+}\,{}^1P^o$ resonance.

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