Collisional Excitation and Decay of the ${}^{1}P$ Shape Resonance of H⁻

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The ${}^{1}P$ shape resonance of H⁻ has been populated in collisions between H⁻ and Xe. We report on the decay H⁻(${}^{1}P$) \rightarrow H⁰(n=2) + e^{-} , which for the first time is measured by high-resolution electron spectroscopy. New information on the angular distribution of ejected electrons is obtained by use of a Shore parametrization to describe the cross section. The Shore parameters are related to matrix elements of the transition operator. The obtained resonance energy and width are in good agreement with recent theoretical calculations as well as other experimental results.

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Doubly excited electronic states of ions and atoms have received much attention during the last decade. In particular, electronic states of two-elecron systems such as H^- and He have been the subject of continuous interest. Such states are found to behave quite differently from what is expected on the basis of the independent-particle model.¹ From a theoretical point of view, the treatment of radial and angular correlations between the two active electrons constitutes a considerable challenge. Such correlations are expected to be very important for the negative ion H^- because of the small Coulomb field of the nucleus.

The present Letter studies the formation and subsequent decay of the 2s2p¹P shape resonance of H⁻. The state is formed in collisions between 100-keV H⁻ and Xe:

$$\mathbf{H}^{-} + \mathbf{X}\mathbf{e} \rightarrow \mathbf{H}^{-**}(^{1}P) + \mathbf{X}\mathbf{e}.$$
 (1)

The doubly excited ${}^{1}P$ state of H⁻ couples strongly to the continuum of H⁰, which results in autodetachment (see Fig. 1),

$$H^{-**(1_P)} \to H^0(n \le 2) + e^-.$$
 (2)

When the hydrogen atom is left in the exctied state n=2, the energy of the ejected electron is about 20 meV. We have measured the energy of these extremely low-energy electrons by kinematically shifting the energy. This was done by measurment of the decay from H⁻ ions moving at a velocity of 2 a.u. Because of the small laboratory scattering angle of the emitted electrons, 0° electron spectroscopy was applied.

As a result of the interference between the direct detachment process,

$$H^- \to H^0(n=2) + e^-,$$
 (3)

and that via the resonant ${}^{1}P$ state, the shape of the autodetaching line is in general asymmetric. Since we consider the energy and angular distribution of the ejected rather than absorption cross sections, we used the Shore parametrization² instead of the more com-

monly applied parametrization of Fano.³ Fits to the experimental data yield the resonance energy E_r and the characteristic width Γ of the ¹P state. These parameters are independent of the excitation mechanism. The parameters describing the shape and size of the cross section, on the other hand, depend on the excitation process. Here we report on the spectroscopic quantities E_r and Γ as well as the Shore parameters, which reflect the dynamics of the collision.

The existence of the ${}^{1}P$ shape resonance has been verified in a number of experiments such as the electron-hydrogen scattering experiment,^{4,5} hydrogenarc emission experiment,⁶ and the experiments with crossed laser and H⁻ beams.^{7,8} To the best of our knowledge, there has been no previous report on



FIG. 1. Energy-level diagram of the negative hydrogen system. (1) is collisional excitation of the ¹*P* resonance, (2) is autodetachment of the ¹*P* resonance to H⁰(n = 2), and (3) is direct detachment yielding H⁰(n = 2).

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high-resolution electron spectroscopy applied to the ¹P shape resonance although the possible appearance of the decay to $H(n=2)^9$ as well as to $H(n=1)^{10}$ has been discussed. Electron ejection from autodetaching states (¹S, ³P, and ¹D) below the n=2 level has been reported by Risley, Edwards, and Geballe.¹⁰

The H⁻ ions were produced in a duoplasmatron ion source and accelerated to the desired energy of 100 keV. From the ion source, beam intensities of ~ 20 μA were achievable. The ion beam was collimated and directed through a small gas cell of 4 mm length. Immediately before the interaction with the target, the beam was deflected 3° and thereby cleaned of undesired charge-state components (H⁰, H⁺). Electrons ejected in the forward direction underwent energy analysis by a 30° parallel-plate spectrometer and detected by a Channeltron. For the data presented here, the acceptance angle of the spectrometer θ_0 was 0.506°. The ion beam passed almost undeflected through the spectrometer. A set of electrostatic plates was available for analysis of the final charge state of the beam particles after passage through the spectrometer. This allowed coincidence measurements to be performed. Three pairs of Helmholtz coils were used to compensate for magnetic stray fields and the Earth's magnetic field in the target region. Electrons created at beam-defining apertures and slits were to a large extent removed from the beam by an electrostatic trap located in front of the target-gas cell. To produce background-free electron-energy spectra, spectra were taken without gas in the target cell and subtracted from the primary spectra. For further details about the experimental equipment see Andersen et al.¹¹

Figure 2 shows the experimentally obtained yield of electrons as a function of energy in the laboratory



FIG. 2. Electron-energy spectrum measured with $\theta_0 = 0.506^\circ$ in the forward direction for 100-keV H⁻ on Xe. The ¹P resonance is seen on both sides of the cusp. The curve represents the best fit obtained from Eq. (13).

frame. Detachment via the ¹P shape resonance clearly manifests itself as two lines on each side of the cusp corresponding to forward- and backward-ejected electrons. The presence of the ¹P resonance was also observed in collisions with He, Ne, Ar, and Kr targets. The coincidence technique verified that the two lines belonged to the $H^- \rightarrow H^0 + e^-$ channel.

Information from the experimentally obtained electron-energy spectra was obtained by writing the cross section in the projectile rest frame in the form

$$\left(\frac{d^2\sigma}{dE \ d\Omega}\right)_p = \left(\frac{d^2\sigma}{dE \ d\Omega}\right)_{\rm NR} + \frac{\alpha(\theta')\epsilon + \beta(\theta')}{1 + \epsilon^2}, \quad (4)$$

where

$$\epsilon = (E - E_r) / \frac{1}{2} \Gamma, \qquad (5)$$

and α,β are the so-called Shore parameters that describe the shape of the resonance. *E* is the energy of the ejected electron. In the following, we assume that α, β , and Γ do not vary with energy in the resonance region. Thus at fixed collision energy, α and β depend only on the emission angle θ' because of axial symmetry. $(d^2\sigma/dE d\Omega)_{\rm NR}$ is the nonresonant cross section which will be discussed later.

The Shore parameters can be expressed in rather simple terms. Consider the transition matrix element for the direct process,

$$t_{lm}^{0} = \langle \psi_{i} | T | \psi_{E,lm}^{0} \rangle, \qquad (6)$$

where T is the T operator, ψ_i is the initial wave function, and $\psi_{E,lm}^0$ is the unperturbed continuum wave function for a free electron with energy E and angular momenta *l,m*. In the general case of a resonance with angular momenta *L,M*, we find¹²

$$\alpha = 2q \operatorname{Re}(z) + 2\operatorname{Im}(z), \qquad (7)$$

$$\beta = (q^2 + 1) \left| \sum_{M} t_{LM}^0 Y_{LM} \right|^2 - 2 \operatorname{Re}(z) + 2q \operatorname{Im}(z),$$
(8)

where

$$z = \sum_{IMm} t_{LM}^0 t_{Im}^{0*} Y_{LM} Y_{Im}^*.$$
(9)

 $Y_{LM} = Y_{LM}(\theta', \psi')$ are the spherical harmonics and q is the well-known Fano shape parameter.³ The *T*-matrix element is considered to be independent of the energy *E* in the resonance region. These equations relate the Shore parameters and thereby the angular distribution of electrons to the collision dynamics expressed through the transition matrix element.

The *T*-matrix elements of Eqs. (6)–(9) are not known for the present case. Rather we treat α and β as parameters to be determined from the experiment. We shall assume α and β to be constant within the angular resolution at $\theta'=0^{\circ}$ (forward-directed electrons) and $\theta'=180^{\circ}$ (backward-directed electrons). With the present ion velocity and acceptance angle $\theta_0 = 0.506^\circ$, the angular acceptance in the H⁻ rest frame is $\sim \pm 30^\circ$ at $E = E_r$. (The acceptance angle is sufficiently small that the resolution of the spectrometer is much smaller than the width of the resonance line in the laboratory frame.)

In Eq. (4), $(d^2\sigma/dE d\Omega)_{NR}$ is the cross section one would find if there had been no resonance. We use the following expansion:

$$\left(\frac{d^2\sigma}{dE \ d\Omega}\right)_{\rm NR} = \sum_{l} a_l(v') P_l(\cos\theta'), \qquad (10)$$

where v' is the electron velocity in the rest frame of the projectile in atomic units and p_l are the Legendre polynomials. We retain the first two Legendre polynomials l=0, 1 and expand the coefficients a_l in powers of v' since we deal with electrons of very low energy in the H⁻ frame,

$$\left(\frac{d^2\sigma}{dE\,d\,\Omega}\right)_{\rm NR} = a_0^0 + a_0^1\,\upsilon' + (a_1^0 + a_1^1\,\upsilon')\cos\theta'. \quad (11)$$

This expansion has proved very useful to describe low-energy continuum electrons for other processes as well.^{11,13}

The yield of electrons can be expressed as

$$Y = \int_{\Delta E} \int_{\Delta \Omega} \left(\frac{d^2 \sigma}{dE \ d \ \Omega} \right) dE \ d \ \Omega \tag{12}$$

$$= \int_{\Delta E} \int_{\Delta \Omega} \frac{v}{v'} \left(\frac{d^2 \sigma}{dE \ d \Omega} \right)_p dE \ d \Omega , \qquad (13)$$

where the integration extends over the acceptance in energy and angle of the spectrometer (this includes an integration over θ'). The symbol v is the laboratory velocity of the ejected electron in atomic units. With Eqs. (13), (4), and (11) we have an expression that can be fitted to the experimental data; the coefficients, which describe the nonresonant process (a_l) as well as those due to the presence of the resonance $(\alpha, \beta, E_r,$ and Γ), can be determined. Note that since the cross section is finite at v'=0, the division by v' in Eq. (13) will produce the well-known cusp in the yield of electrons at v'=0.

Figure 2 shows the result of the fit to the experimental electron energy spectrum. Obviously, the Shore parametrization of the cross section describes the data very well. The parameter values obtained from the fit are listed in Table I. The values of a_l , α , and β are normalized so that $a_0^0 = 1$. Our values of E_r and Γ have been compared with various theoretical results.¹⁴⁻¹⁸ The present experimental findings are in very close agreement with the calculations of Callaway¹⁸ which yielded $E_r = 0.0165$ eV and $\Gamma = 0.020$ eV. Our results are also in reasonably good agreement with TABLE I. Fitted values for 100-keV H⁻ on Xe. Cross sections are normalized so that $a_0^0 = 1$. Estimated uncertainties are listed.

the results of the experiment with crossed laser and H⁻ beams by Bryant *et al.*,¹⁹ who found $E_r = 0.0194 \pm 0.0004$ eV and $\Gamma = 0.0212 \pm 0.0011$ eV.

When inspecting Table I one finds that $\beta(\theta'=0^\circ) \simeq \beta(\theta'=180^\circ)$ and $\alpha(\theta'=0^\circ) \neq \alpha(\theta'=180^\circ)$, which shows that the emission in the H⁻ rest frame is not isotropic. By integration over the azimuth angle and by omitting terms with $\sin\theta'$ we obtain from Eqs. (7)-(9)

$$\alpha = 2\pi \{ 2q \operatorname{Re}(z) + 2\operatorname{Im}(z) \}, \qquad (14)$$

$$\beta = 2\pi \{ (q^2 + 1) | t_{10}^0 Y_{10} |^2$$

$$-2 \operatorname{Re}(z) + 2q \operatorname{Im}(z) \},$$
 (15)

with

$$z = \sum_{l} t_{10}^{0} t_{10}^{0*} Y_{10} Y_{10}.$$
 (16)

Information as to which angular momenta l to include must await direct calculations of the transition-matrix elements of Eq. (6).

The cross section of Eq. (4), with the parameters given in Table I, is shown in Fig. 3 as a function of energy in the H⁻ rest frame. Clearly, a very pronounced and asymmetric profile of the resonant cross section $\alpha\epsilon + \beta/(\epsilon^2 + 1)$ is observed both in the forward and backward directions. It is seen that the cross-section maximum is not at the resonance energy, and that different positions of the maximum are observed for forward- and backward-directed electrons. Further, the interference with the direct process is destructive at very low energy. The insets in Fig. 3 show the nonresonant cross sections $\sigma_{\rm NR} = (d^2\sigma/dE d\Omega)_{\rm NR}$ as well as the total cross sections. The relatively large nonresonant contribution is due to detachment to



FIG. 3. Cross sections (arbitrary units) in the H⁻ rest frame as a function of electron energy in that frame. Shown are the cross sections in for forward direction ($\theta'=0^\circ$) and in the backward direction ($\theta'=180^\circ$). The total cross section σ_T is the sum of σ_R and σ_{NR} .

 $H^0(n=1)$ which is the dominant detachment channel.²⁰ The contribution from double detachment $H^- \rightarrow H^+ + 2e^-$ amounts to about 10% of the total yield and is also included in σ_{NR} . Thus the autodetachment of the ¹P state interferes only with a small fraction of σ_{NR} . In an ideal experiment, one should therefore measure the ejected electrons in coincidence with the Lyman- α photon stemming from the decay of $H^0(n=2)$.

In conclusion, we have observed the autodetaching ${}^{1}P$ shape resonance of H⁻ in the ejected-electron energy spectrum. Characteristic for the state, we find

 $E_r = 0.017 \pm 0.001$ eV and $\Gamma = 0.021 \pm 0.001$ eV. These results are in close agreement with recent theoretical results by Callaway as well as with the experimental results of Bryant *et al.* Further, we have obtained new angular information about the electron emission in the H⁻ rest frame around $\theta' = 0^\circ$ and $\theta' = 180^\circ$. The data indicate that the emission is not isotropic. Finally, we would like to emphasize that the present technique may be applied to other resonances in negative ions when the resonances are located just above an excitation threshold.

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- ¹C. D. Lin, Phys. Rev. Lett. 51, 1348 (1983).
- ²B. W. Shore, Rev. Mod. Phys. **39**, 439 (1967).
- ³U. Fano, Phys. Rev. 124, 1866 (1961).
- ⁴J. F. Williams and J. W. McGowan, Phys. Rev. Lett. 21, 719 (1968).
- ⁵J. W. McGowan, J. F. Williams, and E. K. Curley, Phys. Rev. 180, 132 (1969).
- ⁶K. Behringer and P. Thoma, Phys. Rev. A 17, 1408 (1978).
- ⁷H. C. Bryant, B. D. Dieterle, J. Donahue, H. Sharifian, H. Tootoonchi, D. M. Wolfe, P. A. M. Gram, and M. A. Yates-Williams, Phys. Rev. Lett. **38**, 228 (1977).
- ⁸D. W. MacArthur, K. B. Butterfield, D. A. Clark, J. B. Donahue, P. A. Gram, H. C. Bryant, C. J. Harvey, W. W. Smith, and G. Comtet, Phys. Rev. A **32**, 1921 (1985).
- ⁹N. Maleki and J. Macek, Phys. Rev. A 26, 3198 (1982).
- $^{10}\text{J}.$ S. Risley, A. K. Edwards, and R. Geballe, Phys. Rev. A 9, 1115 (1974).
- ¹¹L. H. Andersen, M. Frost, P. Hvelplund, and H. Knudsen, J. Phys. B 17, 4701 (1984).

¹²K. Taulbjerg, private communication.

- ¹³L. H. Andersen, K. Jensen, and H. Knudsen, J. Phys. B 19, L161–L166 (1986).
- ¹⁴J. Macek and P. G. Burke, Proc. Phys. Soc. London **92**, 351 (1967).
- ¹⁵C. D. Lin, Phys. Rev. Lett. **35**, 1150 (1975).
- ¹⁶J. T. Broad and W. P. Reinhardt, Phys. Rev. A 14, 2159 (1976).
- 17 J. J. Wendoloski and W. P. Reinhardt, Phys. Rev. A 17, 195 (1978).
- ¹⁸J. Callaway, Phys. Lett. **81A**, 495 (1981).
- ¹⁹H. C. Bryant, David A. Clark, Kenneth B. Butterfield, C. A. Frost, H. Sharifian, H. Tootoonchi, J. B. Donahue, P. A. M. Gram, M. E. Hamm, R. W. Hamm, J. C. Pratt, M. A. Yates, and W. W. Smith, Phys. Rev. A 27, 2889 (1983).
- ²⁰K. L. Bell, A. E. Kingston, and P. J. Madden, J. Phys. B 11, 3357 (1978).