H^- shape-resonance studies in an arc plasma

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Calculations by Macek have shown that the H⁻ photoabsorption cross section should be affected by a shape resonance at 1129.5 Å. This experiment is an attempt to observe the resonance in emission according to the reaction $e + H \rightarrow [H^-]^* \rightarrow H^- + h\nu$. The plasma is in a condition of local thermodynamic equilibrium and is generated by a stationary wall-stabilized hydrogen arc. With an axis temperature of 14000 K and 1 atm pressure, the H⁻ free-bound continuum contributes only about 2% of the total radiation which is dominated in the 1130-Å region by the Ly- α wing. However, the peak of the shape resonance, according to Macek, should have a cross section about 25 times greater than the continuous free-bound cross section; therefore, it should appear as a very noticeable 50% structure superimposed on the Ly- α wing. Except for some small features which are attributed to weak molecular emission, there is no obvious indication of the shape resonance in either deuterium or hydrogen spectra between 1105 and 1135 Å. It is estimated that the minimum feature which could have been detected at 1129 Å was about 2% of the total signal.

I. INTRODUCTION

There have been several theoretical and experimental investigations in recent years concerning resonances associated with the H⁻ negative ion.¹⁻⁶ Since this atomic system is one of the simplest and most basic, it is of fundamental importance that these resonances be well understood. Closecoupling calculations of low-energy electron-hydrogen-atom collision cross sections by Burke and collaborators^{4,6,7} indicated a series of Feshbach resonances associated with the formation of virtual H⁻ doubly excited states which are intermediate in the production of excited hydrogen atoms. They also predicted,⁴ for the first time, an unusually narrow shape resonance just above the n = 2 excitation threshold. The shape resonance differs from the Feshbach resonance in that it lies energetically above rather than below the channel or channels to which it is most strongly coupled. Transition to these states is energetically allowed and quite rapid, generally resulting in a relatively wide feature.

One can consider that the resonating electron is captured in a weak potential well from which it escapes by tunneling. Figure 1 shows schematically how this might look when the outgoing channel is an H atom in the n = 2 state. There is a dominant long-range $l(l+1)r^{-2}$ repulsive barrier and a small bump or well which is produced by an ad-

ditional short-range attraction. The actual potential situation is more complicated, as coupling to both the 2²S and 2²P excited states and the 1²S ground state must be considered. Taylor and Burke calculated the energy of the shape resonance to be 18 meV above the n = 2 level of the neutral atom with a full width at half-maximum (FWHM) of about 15 meV, which means that the natural lifetime is about 10⁻¹⁴ sec.

Figure 2 is an energy-level diagram of the H⁻ system. A well-known H⁻ photoabsorption continuum⁸ is produced for $h\nu \ge 0.754$ eV with the maxi-



FIG. 1. Potential-energy diagram illustrating schematically the effect of a shape resonance on the electronhydrogen-atom system.

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mum cross section occurring at about 1.5 eV. As $h\nu$ increases, additional channels involving H(n)are opened. The H⁻ shape resonance is illustrated as an energy level just above the level representing the e + H(n = 2) channel. The configuration of the shape resonance is ${}^{1}P$ and consequently is dipole connected to the ¹S H⁻ ground state. The effect of the resonance on the H⁻ photoabsorption cross section has been calculated by Macek.⁵ He finds an absorption resonance at $\lambda = 1129.54$ Å with a FWHM of 1.5 Å and oscillator strength f = 0.044. The total cross section at line center was calculated to be 1.5×10^{-16} cm², which is about 25 times the strength of the bound-free background H⁻ continuum at that wavelength and about four times the maximum absorption at 1.5 eV. Hyman $et al.^2$ have done a similar close-coupling calculation and are in complete agreement with these values. They also calculate that the relative strengths of the outgoing channels near the resonance peak are approximately 2p: 2s: 1s: :4:1:3.

A significant astrophysical application involving the predicted shape resonance has been described by Snow⁹ and serves to show why these properties of the resonance need verification. There has been some controversy over the last few years concerning the possibility that the infrared excess in extreme Be stars is produced by H⁻ free-bound and free-free emission from a hot plasma surrounding the stars. From the measured infrared excess, Snow has calculated the column density of H⁻, which would follow from this hypothesis. An independent determination of the column density was then made by measuring the strength of the autoionization absorption feature in the corresponding vacuum-uv stellar spectrum near 1130 Å. However, no resonance structure was observed above

the background, leading to a discrepancy of at least a factor of ten in the column density determinations. At the present time, it is not clear whether this is evidence that the above interpretation of the ir excess is incorrect or that the theoretical calculations describing the H⁻ resonance absorption are inadequate.

Evidence at least for the existence of the ^{1}P shape resonance has already been demonstrated in electron-hydrogen-atom inelastic scattering experiments,^{1,3} but these experiments have relatively low energy resolution, about 70 meV, as compared with the 15-meV resonance width. Conventional spectroscopy, on the other hand, can often be done with better than 0.1 Å resolution, which corresponds to about 1 meV at 1129 Å. Such a capability would allow a measurement of both oscillator strength and line shape if the resonance could be seen. The experiment described in Sec. II is an attempt to observe the shape resonance in the emission spectrum of a laboratory hydrogen plasma which is in a condition of local thermodynamic equilibrium (LTE).

II. EXPERIMENTAL DETAILS

A. Physical properties of source

A stationary plasma is generated in a wall-stabilized arc operating at atmospheric pressure. Figure 3 illustrates some of the physical details of this plasma source. The dc arc discharge is struck between a set of tungsten electrodes located slightly off axis at each end of the figure. It is defined and stabilized by a 2-mm-diam arc channel formed by a stack of 20 water-cooled, electrically insulated, center-bored copper plates. The total anode-to-cathode length of the cylindrically symmetric discharge is 5.3 cm. A LiF window seals the light source at each end and allows vacuum-ultraviolet (vuv) observations along the arc axis. The gas ports are arranged so that the cen-



FIG. 2. Energy-level diagram of the hydrogen negative-ion system.



FIG. 3. Schematic of the wall-stabilized arc source.

ter section of the arc can be operated in pure hydrogen while the ends are operated in pure argon. The use of such a high-temperature transparent argon buffer plasma ia essential, since it allows the monochromator to see an essentially homogeneous length of high-temperature hydrogen plasma free of any cool boundary layers where molecular hydrogen emission and/or absorption (Lyman and Werner bands) would dominate the vuv spectrum. Flow rates are adjusted so that the two gases are mixed only in short 3-mm zones. The voltage drop across the hydrogen plasma is about four times that across the same length of argon, so that contamination either by argon in the center portion or by hydrogen in the ends can easily be detected on voltmeters and corrected by readjustment of the flow rates. The arc is stable in such a configuration to within 2% over the period of the experiment.

At a current of 60 A, an axis temperature of about 16000 K and a radial temperature distribution as shown in Fig. 4 are obtained.¹⁰ A simple vuv optical system¹¹ defines the effective light source to be a collimated cone of light (f/200) emitted from along the arc axis. The dashed lines in the figure indicate the limited area viewed by the spectrometer both at focus (0.3-mm diameter) and at the extreme ends of the arc. The temperature is nearly homogeneous and varies only about 7% over this region. Also plotted in Fig. 4 is the corresponding electron density appropriate for an LTE plasma.¹² At such high temperatures, the equilibrium concentration of molecular hydrogen in the pure hydrogen section is on the order of 1



ppm, and molecular absorption and emission are negligible. However, there is strong molecular emission from the cooler off-axis regions which must be kept out of the cone of observation. This is illustrated in Fig. 5, which is a scan across the arc diameter at a wavelength corresponding to one of the strongest H₂ emission lines. The on-axis radiation is pure hydrogen continuum but is one or two orders of magnitude less intense than the off-axis molecular line radiation. This clearly illustrates the need for limiting the field of view to the near-axis region and for carefully aligning the arc along the optical axis. Further details concerning the arc apparatus and plasma diagnostics are described elsewhere.^{11,13,14}

B. Equilibrium properties of source

The hydrogen wall-stabilized arc is a high-density collision-dominated plasma with electron density $N_e \gtrsim 10^{17}$ cm⁻³ and electron-atom gas-kinetic collision frequencies on the order of 10^{10} sec⁻¹. Extensive studies of such arcs have shown that the condition of LTE is well fulfilled under typical



FIG. 4. Radial dependence of the electron density and temperature for an LTE arc plasma having a 2-mm-diam channel and operating at 1 atm, 60 A. The dashed lines define the dimensions of the plasma that are observed by the vacuum-uv spectrometer.

FIG. 5. Radial dependence of the spectral radiance at 1606 Å for a hydrogen arc with axis temperature of about 19500 K. The intense off-axis radiation is due to Lyman molecular-band emission. The nearly uniform radiation in the vicinity of the axis contains negligible molecular contributions and is due primarily to the electron-proton recombination continuum.

operating conditions. Rather than review the various experiments and criteria which can be used to establish a case for LTE,¹⁵⁻¹⁷ it is perhaps better to consider those cases where small deviations from LTE are known to exist. In an ideal collisiondominated stationary plasma, the system is essentially closed, and no net energy escapes either in the form of energetic particles or photons. The plasma thermalizes, the various constituents come to a Saha equilibrium, and the population densities are described according to a Maxwell-Boltzmann distribution. Deviations from this ideal system can occur when (a) the plasma properties deviate significantly before a particle has undergone sufficient thermalizing collisions (loss of particles), and (b) the plasma is optically thin, and radiative emission processes are not exactly balanced by radiative absorption (loss of radiation).

The first of these effects has been encountered and observed in hydrogen arcs with axis electron temperatures less than about 15000 K, where the radial electron density gradient is rather steep and ground-state atoms may not make a sufficient number of equilibrating collisions before they diffuse into significantly different conditions. This results in a slight overpopulation of ground-state atoms along the axis of such a low-current hydrogen arc compared to what one might expect from a plasma in LTE at the electron temperature. Because the temperature characterizing the groundstate atoms is lower than the electron temperature (typically 10%), radiation from the excited states which are to some degree coupled with the ground state is less than what would be calculated assuming LTE.^{11,18} In the present experiments, data were taken at currents of 45, 60, and 75 A, corresponding to axis electron temperatures of about 14000, 16000, and 18000 K.¹⁰ Therefore, LTE should be a good assumption in all but the 45-A data, where the above-described conditions can be expected. However, even in this case, where one should assume at most a state of partial LTE among the electrons and excited atoms, ¹⁸ the data are useful since the significant observation, as described in Sec. IIC, is the ratio of the shape resonance to essentially the Lyman- α wing intensity. Since both the 2s and 2p excited states of atomic hydrogen and the ${}^{1}P$ H⁻ resonance state are energetically very close and are coupled strongly with each other, radiation from the excited atoms and those channels associated with the 2s and 2p partial cross sections² in the reaction

$$e^{-} + H(1s, 2s, 2p) \rightarrow H^{-}(^{1}P) \rightarrow H^{-}(^{1}S) + h\nu$$
 (1)

will be similarly affected by any small deviations from LTE. Any overpopulation of the 1s ground state tends to increase the effect of the resonance relative to the Lyman- α wing, since the 1s partial cross section also exhibits the resonance structure.² This means that the ratio of the resonance to the Lyman- α wing intensity obtained by assuming LTE and detailed balance for the reaction given in Eq. (1) is, if anything, an underestimate. In any case, the measured ratio at 45 A is not expected to be significantly different from that which is calculated assuming LTE.

The significance of the second effect, namely, radiative losses, can be determined by applying to the equilibrium H⁻ resonant-state population density N_{eq} (H⁻)* the relation¹⁹

$$N(H^{-})^{*} = [A_{a}/(A_{a}+A_{r})]N_{eq}(H^{-})^{*},$$
 (2)

where A_a is the autoionization rate or the probability that the state decays by ejection of an electron, and A_r is the radiative transition probability. Using Macek's calculated f value for the transition $H^{-}({}^{1}P) - H^{-}({}^{1}S)$, we obtain $A_r \sim 10^8 \text{ sec}^{-1}$. This is entirely negligible compared to the autoionization rate of 10^{14} sec^{-1} , and no deviations from equilibrium that are due to radiative losses will occur. The H⁻ resonant-state population ought to be found in LTE, since it is governed almost entirely by collisional processes. Physically, one can imagine that there are about 10^6 shuffles in and out of the ${}^{1}P$ H⁻ level before a single radiative transition takes place. The number of autoionizing transitions into the kth channel is given by

$$\left(\frac{dN(\mathrm{H}^{-})^{*}}{dt}\right)_{\mathrm{out}} = N(\mathrm{H}^{-})^{*}A_{ak}.$$
 (3)

This is balanced by the number of transitions into the ${}^{1}P$ level,

$$\left(\frac{dN}{dt}\right)_{\rm in} = N_k({\rm H})A_{ck}\,,\tag{4}$$

where N_k (H) is the number density of hydrogen atoms in either the ground state or the n = 2 state and A_{ck} is the appropriate electron collision rate which can be given in terms of the velocity v of the interacting electron, the number of electrons having a velocity in the interval between v and v+dv, $(dn_e/dv)_v \Delta v$, and the collision cross section Q_k :

$$A_{ck} = \left(\frac{dN_e}{dv}\right)_v \Delta v N_k(\mathbf{H}) v Q_k(v).$$
⁽⁵⁾

Typically, A_{ck} is several orders of magnitude less than A_{ak} , but this is compensated by the fact that $N_k(H)$ is the same amount larger than $N(H^-)^*$ in equilibrium.

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Since the plasma can be considered an LTE source, calculation of its spectrum is straightforward.^{12,20} For example, at atmospheric pressure, a hydrogen arc plasma at a typical temperature of 15 000 K is composed almost entirely of neutral hydrogen, protons, and electrons. By combining the Saha equation with the condition of charge neutrality and the equation of state, the following densities are obtained¹²:

$$N(H) = 1.8 \times 10^{17} \text{ cm}^{-3},$$

 $N_p = N_e = 1.5 \times 10^{17} \text{ cm}^{-3},$
 $N(H^-) = 2.9 \times 10^{12} \text{ cm}^{-3}.$

The emission coefficient for the H^- resonance produced in the reaction given in Eq. (1) is then calculated according to the expression

$$\epsilon_{1129}(\mathrm{H}^{-})^{*} = N(\mathrm{H}^{-})\sigma(\mathrm{H}^{-})^{*}B_{\lambda}(\mathrm{T}), \qquad (6)$$

using Macek's resonance absorption cross section $\sigma(H^-)^*$ and the Planck blackbody function $B_\lambda(T)$. The dominant emission from the plasma near 1129 Å comes from the Stark-broadened Lyman- α line, centered at $\lambda_0 = 1216$ Å. Its emission is given by Eq. (7) in terms of the transition probability A_{21} , the n = 2 hydrogen-atom density $N_2(H)$, and the line-



FIG. 6. Temperature dependence of the various processes contributing to the emission coefficient of a 1-atm-pressure hydrogen plasma in LTE at 1130 Å.

shape function $\phi(\lambda)$

$$\epsilon_{\lambda}(Ly-\alpha) = (hc/4\pi\lambda_0)A_{21}N_2(H)\phi(\lambda).$$
(7)

From the Boltzmann equilibrium equation, one obtains $N_2(H) = 2.7 \times 10^{14} \text{ cm}^{-3}$. For the line-shape function it is sufficient to use just the asymptotic $\lambda^{-5/2}$ Holtsmark wing,¹⁶ since 1129 Å is far enough into the wings that quasistatic Stark-broadening theory is accurate. In this way, Roberts and Voigt²⁰ have evaluated Eq. (7) for the first four lines of the Lyman series and have listed the line sum as a function of wavelength and temperature. However, the line shape function $\phi(\lambda)$ must be modified to account for the various trivial sources of asymmetry discussed, for example, by Griem.¹⁶ Thus a Boltzmann factor $\exp[-\hbar(\omega - \omega_0)/\kappa T]$ and a factor $(\omega/\omega_0)^6$ combine to make the line sum at 1129 Å about 11% lower than the value listed by Roberts and Voigt.

Figure 6 shows the contributions to the emission at 1130 Å from the Lyman line wings, the H⁻ freebound continuum (not the resonance), and several other relevant processes.²⁰ The total emission coefficient is the top curve, and the H free-bound continuum is second from bottom. The dashed line is the ratio of this H⁻ continuum to the total emission coefficient. Clearly, it would be difficult to observe the H⁻ continuum in competition with the Lyman line wings, but the peak of the resonance cross section is predicted to be about 25 times the H⁻ free-bound continuum. Thus, the resonance should be seen as a feature whose peak intensity is the following percentage of the total background signal at 1129.54 Å: 46% at 14000 K, 30% at 16000 K, and 18% at 18 000 K.



FIG. 7. Strip-chart recordings of the hydrogen and deuterium spectra in the vicinity of the predicted shape resonance at an arc current of 45 Å.

III. DATA

Figure 7 shows some typical unsmoothed spectra taken on a strip-chart recorder, both for a pure hydrogen plasma and a pure deuterium plasma. Background signal due to scattered light and detector current was determined for each scan by measuring the signal with the monochromator set at a wavelength below the LiF-window cutoff. As presented in Fig. 7, the spectra have been corrected for the background and are superimposed on each other (with the zero offset indicated) to facilitate comparison and discussion. The location of the predicted resonance is indicated either with an arrow or with dashed lines representing the expected structure. The monochromator resolution is 0.3 Å FWHM (about 3 meV), which would degrade the effect of the resonance only slightly. The current of 45 A gives a 14000 K axis temperature. The three NI lines near 1135 Å are atomic resonance lines from a nitrogen impurity in the gas and serve as convenient wavelength calibration points. The hydrogen was obviously more pure than the deuterium, as is seen in the strength of the NI lines. About 5% of the intensity shown here is due to the argon buffer gas. The decrease of the signal towards lower wavelengths is due primarily to a decreasing system efficiency. As can be seen, there is neither an enhancement by a factor of 1.46 in the intensity at 1129.54 Å nor any structure with a FWHM of 1.5 Å as anticipated.

The small undulations shown have widths that are about the same as the instrumental width and are the result of weak molecular emission from the Lyman and Werner bands of hydrogen (deuterium). The source of this radiation is most likely from the rich off-axis emission which is somehow scattered, for example, by optical imperfections



FIG. 8. Strip-chart recordings of the hydrogen and deuterium spectra in the vicinity of the predicted shape resonance at an arc current of 60 Å.

into the collimated optical beam. Support for this stems from the fact that the undulations show none of the current dependence which would be expected if the origin were from on-axis molecular or H⁻ resonance radiation. There is some structure in the hydrogen scan near 1129 Å and even a slight peak near 1132 Å. Although the position and width of this peak did not correspond to that of the anticipated resonance, the deuterium scan was made in an effort to establish whether this was indeed an atomic or molecular line. Atomic lines should have isotope shifts only on the order of 0.3 Å, which is the Ly- α shift, while the molecular rotational structure is generally shifted much more and can be expected to be considerably scrambled since the isotope shift is strongly dependent on the quantum numbers. The bump at 1132 Å is absent in the deuterium spectrum, and the two superimposed spectra have no obvious features in common other than the impurity lines. Even with the undulations, the spectra are smooth to about the 15% level.

We also considered the possibility that the arc has some unexpected LTE problem at 45 A and took some scans at higher currents, where LTE is more assured and the effect of the resonance should be reduced only slightly. Figures 8 and 9 show the spectra at 60 A (16000 K) and 75 A (18000 K). Again, the argon plasma contributes less than 10% of the signal, and there is no evidence for the shape resonance.

Finally, successive scans at the same current were averaged with the use of an analog-to-digital data recorder and then smoothed with a 1-Å line-width. This essentially removes the narrow mole-cular lines, but there is still nothing at 1129.5 Å, at least down to the 2% level.



FIG. 9. Strip-chart recordings of the hydrogen and deuterium spectra in the vicinity of the predicted shape resonance at an arc current of 75 Å.

IV. CONCLUSIONS

So why is the resonance not seen? Figures 7-9 are high-resolution spectra from a good LTE source, and yet they show at most only very weak molecular structure superimposed on the background. Let us first consider the resonance position, width, and oscillator strength calculation. The position and width are no problem. The n = 2 threshold is near 1132 Å; certainly, the resonance must appear on the short-wavelength side of that. In addition, scattering experiments like the one by Williams and Willis have shown the resonance to be at the predicted energy, at least to within their resolution, which for that experiment was 70 meV, or about 7.5 Å in wavelength units. The spectra we have taken scan all the way to 1105 Å. The same scattering experiment indicates that the width is probably not larger than predicted. A width narrower than expected would increase the effect of an emission line.

However, there may be some question about the oscillator strength. Macek's f-value calculation⁵ is based on three-state close-coupling theory, but the position and width of the line come from a different calculation. This calculation is by Taylor and Burke⁴ and is also a three-state close-coupling calculation, but with some 20 correlation terms included. The added correlation terms have the effect of moving the resonance somewhat closer to the n = 2 threshold and reducing the linewidth by a factor of 5.6. Macek has suggested that his oscillator strength from the simpler calculation should be accurate and that it can be applied to the position and width found by Taylor and Burke. Macek's prediction that the peak resonant cross section is 25 times that of the H⁻ background is based on the narrower linewidth, and his peak cross section would be reduced by a factor of 5.6 without this scaling. However, even then we would have expected an emission increase of about 10% in the 45-A scans, which certainly would have been observable.

We must also consider plasma effects. As explained in Sec. II B, deviations from LTE will, if anything, enhance the resonance relative to the Lyman wing background. Impact broadening will be insignificant since the resonance natural decay rate is 10^{14} sec^{-1} , and the collision rate with electrons is only on the order of 10^{10} sec^{-1} . Static Stark broadening, associated with shifts of energy levels, should also be insignificant. Energy-level perturbations due to local electric fields at the 10^{17} electron densities are on the order of no more than a few meV, while the resonance already has a natural width of 15 meV. However, these fields may be strong enough to alter the very



FIG. 10. Potential-energy diagram illustrating the effect of an electric field perturbation on the H^- shape resonance.

nature of the resonance. The local instantaneous electric field can be estimated from the Holtsmark field distribution to be about 1.4 mV/Å. If we consider that the resonating electron is confined to a space of a few Å, then the potential is modified by a *-Eez* term, which may be several meV. This is shown schematically in Fig. 10. The decay rate by tunneling might be severly changed, since it is extremely sensitive to barrier size; perhaps the small barrier no longer even supports a bound state.

The actual parameters of the potential barrier are difficult to determine, since the interaction is simultaneously coupled to three open channels, not to mention that it is not obvious which electron is resonating. Lin²¹ has recently obtained an estimate for these potential curves by use of a technique in which the Schrödinger equation is written in hyperspherical coordinates.²² He finds a barrier height of about 65 meV; however, his curves predict the resonance to be about 36 meV above the n = 2 atomic level with a width of 27 meV (in contrast to Macek's⁵ values of 18 and 15 meV, respectively). The mean electric field in this experiment may depress the barrier on the order of 14 meV at the peak. Simple WKB estimates of the barrier penetration show that such fields could possibly explain the disappearance of the resonance, but only if the resonance position were taken at 36 meV or more, the lower value being consistent with Lin's potential curves. However, because the position of the resonance is very sensitive to correlations, ranging from 65 meV in three-state close coupling without correlations to 18 meV with correlations, the latter being considered the best value, $4 \cdot 5 \cdot 21$ such an explanation must be regarded as showing only plausibility. We therefore conclude that further theoretical work is necessary. The close-coupling calculations (including correlations) should be repeated, first to check Macek's value of the oscil-

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lator strength, and second to obtain a quantitative estimate of the barrier penetration in the presence of a static electric field.

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