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Interpretation of the Stark effect of H^- resonances

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Resonances in the photodetachment cross sections of H^- in a strong dc electric field are studied theoretically in hyperspherical coordinates in the adiabatic approximation. In terms of the variation of one-dimensional effective potentials with electric field for different channels, the experimental results of Gram *et al.* [Phys. Rev. Lett. 40, 107 (1978)] are interpreted. In particular, the linear Stark splitting of the Feshbach resonance at low fields and the stability of the shape resonance against electric fields up to a few thousand kV/cm are understood from their associated effective potential curves. A new mechanism of field ionization for the "blue-shifted" Stark states is also discussed.

The photodetachment cross sections of H^- ions in the neighborhood of $h\nu \sim 11$ eV were first systematically studied by Bryant *et al.*[1] with use of the 800-MeV H^- beams at the LAMPF accelerator in collision with an intense laser beam. Two resonances were observed near the excitation to the $H(n=2)$ threshold ($h\nu = 10.95$ eV): (1) a narrow Feshbach resonance at $h\nu \sim 10.925$ eV and (2) a broad shape resonance at $h\nu \sim 10.98$ eV. This shape resonance was estimated to have full width at half maximum of 23 ± 6 meV. Subsequently, Gram *et al.*[2] studied the effects of electric fields on these resonances. The Feshbach resonance was found to split into three components where the two outer components exhibit linear Stark shifts and the middle one exhibits quadratic Stark shift. Later experiments[3] by the same group with use of polarized laser light confirmed that the two outer components belong to states which have a magnetic quantum number $M=0$ and the middle one has $|M|=1$. The lowest Stark component has been observed to quench at $\mathscr{E} \sim 130$ kV/cm while the other two components still exist for $\mathscr{E} \sim 300$ kV/cm. On the other hand, the shape resonance was found to be very stable against the electric field except for small gradual broadening; it was still clearly observed for $\mathscr{E} \sim 1200$ kV/cm.

Early theoretical interpretations of these experimental results have been limited to small electric fields. Callaway and Rau[4] studied the Stark effect of the Feshbach resonance and Wendoloski and Reinhardt[5] studied the broadening of the shape resonance in the electric field. In this Rapid Communication these experimental results are interpreted in terms of effective one-dimensional potentials $U_\mu(R)$, in hyper-radius R, where $R=(r_1^2+r_2^2)^{1/2}$ and r_1 and r_2 are the electron radii. The potential curves for different channels μ at different electric fields are obtained by solving the Schrödinger equation in hyperspherical coordinates for H^- in an electric field.

The field-free Feshbach and shape resonances of $H^-\,{}^1P^o$ states near the $H(n=2)$ threshold were studied previously by Lin[6,7] in terms of effective channel potentials in hyperspherical coordinates. In Fig. 1, the + and − ${}^1P^o$ curves, which support the shape and Feshbach resonances, respectively, are reproduced. Also shown is the attractive ${}^1S^e$ curve which converges to the $H(n=2)$ limits. It turns out that the *second* ${}^1S^e$ resonance is nearly degenerate with the ${}^1P^o$ Feshbach resonance; therefore, a *weak* electric field easily mixes the $M=0$ components of ${}^1S^e$ and ${}^1P^o$ to produce two linearly split Stark terms while the $|M|=1$ component follows the general quadratic shift. This explains the Stark effect of the Feshbach resonance at low-$\mathscr{E}$ field but the Stark quenching is not incorporated in this picture.

In this paper, I report the modification of the effective potentials for the three channels shown in Fig. 1 in an electric field. The details of the theoretical method and the results for other channels will be described in a later publication. Basically, the Schrödinger equation in hyperspherical coordinates for H^- in an electric field is solved in the adiabatic approximation. The potential curves at each R are obtained by diagonalizing the Hamiltonian using analytical channel functions.[8] In actual calculations, two-electron basis functions for all singlet channels up to $L=12$ are included in the diagonalization.

The resulting potential curves for the two $M=0$ components of the split Feshbach resonance are shown in Figs. 2(a) and 2(b). The $|M|=1$ component has weaker dependence with electric field (quadratic shift at low fields) and will not be discussed here. Horizontal arrows in the figures indicate the position of the field-free resonances and the vertical arrow indicates where the potential has the minimum.

The potential curves shown in Fig. 2(a) can be easily understood. Since the static electric potential is proportional to R, modification of effective potentials first occurs at large

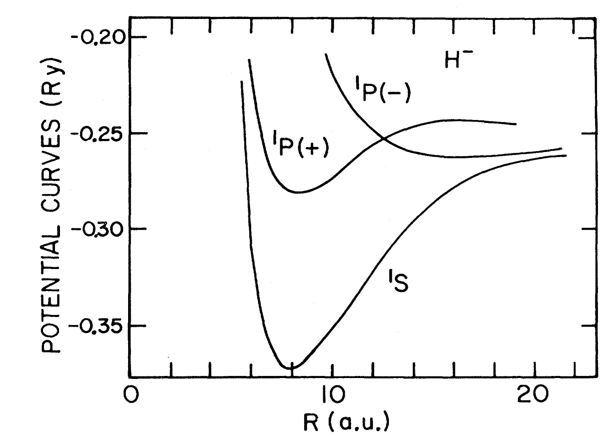

FIG. 1. Field-free potential curves of H^- converging to the $H(n=2)$ limits. The + and − ${}^1P^o$ curves support the shape and Feshbach resonances, respectively. The *second* resonance of ${}^1S^e$ is nearly degenerate with the Feshbach resonance of the ${}^1P^o$ (−) channel.

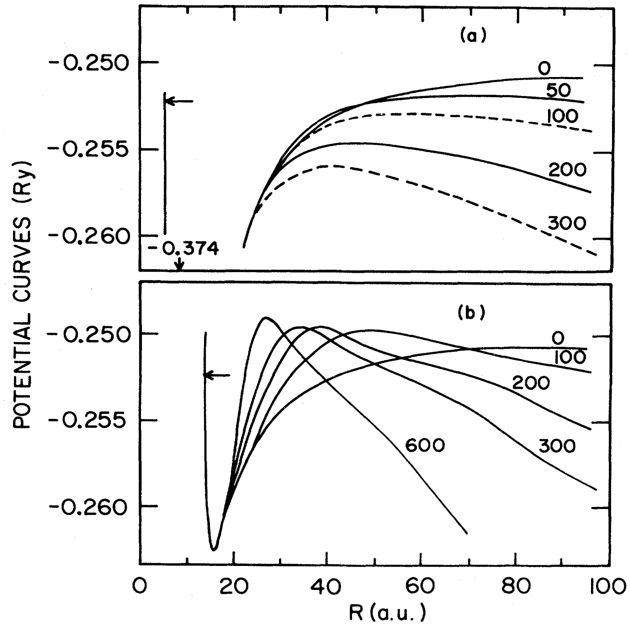

FIG. 2. (a) The variation of the zero-field $^1S^e$ potential curve with electric fields. The electric fields are given in units of kV/cm. (b) Similar to (a) but for the zero-field $^1P^o$ (−) curve. The two horizontal arrows indicate the position of field-free resonances.

R. In Fig. 2(a) we notice that, at the field strengths shown, the inner region of the potential is hardly changed while the outer region is modified from an attractive dipole form for $\mathcal{E}=0$ to a linearly decreasing potential. Thus, the maximum of the potential in the outer region decreases and shifts to smaller values of R with increasing field strengths. Since the field-free Feshbach resonance (corresponding to the second $^1S^e$ resonance) is supported primarily by the weak dipole potential, the downward shift of the effective potential results in a downward shift in energy (or red shift in spectral line) and gradual Stark quenching through quantum tunneling over the barrier. A simple estimate based upon first-order perturbation theory indicates that the energy shift is linear with the strength of the electric field and classical field ionization occurs at $\mathcal{E} \sim 100$ kV/cm. This latter value is consistent with the experimental value of $\mathcal{E} \sim 130$ kV/cm.

In Fig. 2(b) the potential curves for the upper $M=0$ Stark component of the Feshbach resonance are shown for several values of the electric field. These curves exhibit common unusual structure such that the inner portion of each curve is shifted upward, while the outer portion of each curve is shifted downward with increasing electric fields. Thus while the potential at large R becomes more negative as $\mathcal{E}$ increases, the outer barrier height actually increases with $\mathcal{E}$ while shifting to smaller R. In Fig. 2(b) we notice that the barrier height for each $\mathcal{E} \neq 0$ case is above the field-free threshold at -0.25 Ry.

The behavior of potential curves in Fig. 2(b) clearly indicates an upward Stark shift of its eigenvalue (blue-shift in spectral line) with $\mathcal{E}$ field. At small field strength, calculations from first-order perturbation theory indicate that the shift in eigenvalue is linear with $\mathcal{E}$ At higher $\mathcal{E}$, the width of the attractive potential well becomes *narrower* while the height of the outer barrier becomes *higher*. These results clearly point out a new mechanism of field ionization (or detachment for H^-). In this mechanism, the detachment proceeds through the leakage of the resonance over an *ever-increasing* potential barrier *forced by the narrowing* of the potential well by the electric field. In other words, while the potential barrier becomes higher with $\mathcal{E}$, because of the narrowing of the potential well, the resonance is "squeezed" out of the well. This is to be compared with the "normal" Stark field detachment [such as in Fig. 2(a)], where the resonance is quenched by quantum tunneling through an *ever-decreasing* potential barrier, while at the same time, the width of the potential well remains basically constant. The normal Stark field detachment is characterized by a downward shift of its eigenvalue while the new mechanism is characterized by an upshift of its eigenvalue and by a higher field strength for detachment.[9] A simple estimate based upon the Wentzel-Kramers-Brillouin method indicates that the inner potential well in Fig. 2(b) is no longer attractive enough to support a bound state for $\mathcal{E} \sim 350$ kV/cm. This is consistent with the experimental result in Ref. 2 where the upper resonance is still clearly observable for $\mathcal{E} \sim 300$ kV/cm.

The fact that the potential curves in Fig. 2(b) become more repulsive at small R can be understood in terms of the general behavior of spectral repulsion. Since the strength of perturbation in an electric field is proportional to R, the perturbation makes little modification of the field-free potential curves at small R. At large R, when the perturbation is small or comparable to the separation between the field-free $^1S^e$ curve and the $^1P^o$ (−) curve, the general characteristic of spectral repulsion implies that the $^1P^o$ (−) curve will be pushed upward and the $^1S^e$ curve downward. At larger R or stronger fields, the perturbation due to the electric field is so large as to mix many field-free curves (those due to higher L) to result in the downward shift of potential curves shown in Fig. 2(b). This behavior of the potential curves with $\mathcal{E}$ field is expected to be true also for other higher potential curves.

In Fig. 3, the potential curves associated with the shape

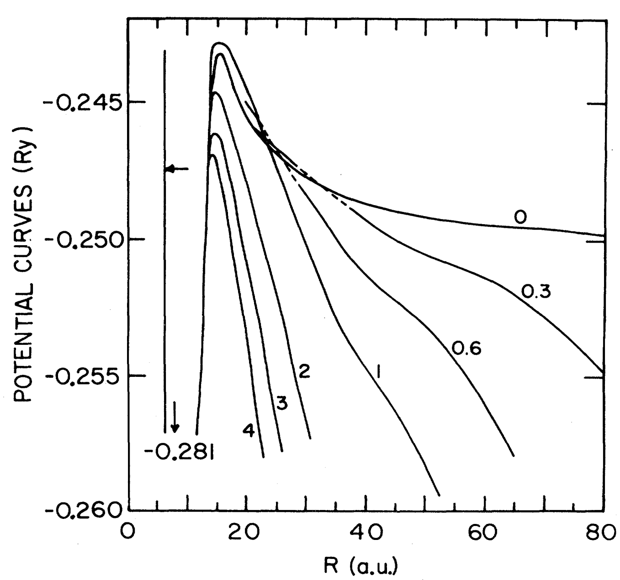

FIG. 3. Similar to Fig. 2 but for the field-free $^1P^o$ (+) curve. The electric fields are given in units of MV/cm.

resonance are shown in the presence of electric fields. For $\mathscr{E}=0$, the potential curve already exhibits a barrier. For $\mathscr{E}\leq 600$ kV/cm., the barrier height and the potential well are hardly modified except that the outer region of each potential curve becomes more negative. In terms of the simple one-dimensional effective potential model, we would thus expect that the energy and width of the shape resonance remain fairly constant until $\mathscr{E}\sim 1000$ kV/cm. This appears to be consistent with the early data of Gram *et al.*[2] but not with the newer data (see Fig. 16 of Ref. 3), where the broadening of the resonance is quite significant for $\mathscr{E}\geq 600$ kV/cm. From the potential curves estimated at higher $\mathscr{E}$, we expect that the detachment of the shape resonance by tunneling through the potential barrier and the shift of resonance position will not be important until $\mathscr{E}\geq 1000$ kV/cm. If the new data[3] are to be confirmed by future experiments, then it is possible that the decay of the shape resonance through autoionization to lower channels becomes more efficient in an electric field.

In summary, the observed Stark effects of the Feshbach and shape resonances of H^- are interpreted in terms of the associated one-dimensional effective potential curves for each channel in hyperspherical coordinates. Two different mechanisms were identified for the Stark field detachment of the Feshbach resonance, one for the red-shifted component and another for the blue-shifted component. The shape resonance is shown to be very stable against the electric field because the effective potential barrier is not significantly modified until $\mathscr{E}>1000$ kV/cm.

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