Photodetachment of H^-

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In a recent experiment [Andersen *et al.*, Phys. Rev. Lett. **79**, 4770 (1997)], the H^- photodetachment cross section (PDCS) and resonances are measured to high precision. Detailed resonance profiles are obtained. To make a critical comparison with experiment, the H^- PDCS below the $n=2$ and $n=3$ thresholds is calculated with a saddle-point complex-rotation method. The theoretical PDCS are convoluted with a Gaussian profile using experimental resolution for full width at half maximum. The theoretical results are calculated with length, velocity, and acceleration gauges. They are compared with the precision data in the literature. Improved widths are obtained for some of the narrow resonances below the $n=3$ threshold. [S1050-2947(99)07207-8]

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I. INTRODUCTION

The study of photodetachment of H^- has a long history, both theoretically and experimentally. This process is known to be an important cause for stellar opacity [1]. Early theoretical calculation has been carried out by Chandrasekhar [2] to determine its cross section. He also showed the equivalence of the length, velocity, and acceleration formulas when exact wave functions for the initial and final state wave functions are used. Early experimental measurement has been carried out for photon energies near the ionization threshold [3,4]. The lowest Feshbach resonance below the $n=2$ threshold and many of the H^- resonances above the $n=2$ threshold have been experimentally investigated by Bryant and collaborators [5–11]. The second Feshbach resonance below the $n=2$ threshold has not been seen in these experiments due to the extremely narrow width. Recently, Andersen *et al.* [12] used Doppler-tuned collinear laser spectroscopy technique with a resolution of 0.180 meV; they were able to observe this second Feshbach resonance. They have also obtained detailed line profiles for the two Feshbach resonances below the $n=2$ threshold and the shape resonance above this threshold.

In the early theoretical calculations, photodetachment cross sections (PDCS) from length and velocity gauges are computed with a discrepancy of 20% for some energies near the detachment threshold [13–15]. This discrepancy is improved slightly to about 8% by Ajmera and Chung [16], who use a highly accurate ground-state wave function and the Feshbach projection operator formalism for the final state. More recently, the theoretical accuracy has been much improved and many new methods have been developed for the H^- PDCS, for example, J matrix [17], R matrix [18,19], nonvariational CI [20], L^2 basis [21], hyperspherical coordinate close coupling [22,23], B spline [24–26], etc. PDCS are calculated near the Feshbach resonances in both length and velocity gauges [21] or in length and acceleration gauges [23]. Close agreement is obtained.

Our interest on the H^- PDCS is stimulated by the recent experimental success in the ultrahigh resolution spectra [12]. We hope to make a detailed calculation on the H^- resonance profile and to compare the theoretical prediction with experiment. PDCS from the detachment threshold to the $n=3$ threshold will be calculated using length, velocity, and acceleration gauges. The resonance parameters obtained in this work will be compared with the accurate results in the literature.

II. THEORY

The PDCS of H^- in the length gauge is given by

$$\sigma(\omega)=\frac{4\pi^2}{3c}\omega\sum_{E_f=E_0+\omega}|\langle\Psi_0|\mathbf{D}|\Psi_f\rangle|^2, \tag{1}$$

where ω is the photon energy, $\mathbf{D}$ is the dipole operator, Ψ_0 is the initial ground-state wave function, and Ψ_f is the final state wave function.

In this work, the cross sections are also calculated with velocity and acceleration gauges. The equality of the three oscillator strength expression

$$f_L=\frac{2}{3}(E_f-E_0)|\langle\Psi_0|\mathbf{D}|\Psi_f\rangle|^2, \tag{2}$$

$$f_V=\frac{2}{3}(E_f-E_0)^{-1)}|\langle\Psi_0|\nabla|\Psi_f\rangle|^2, \tag{3}$$

and

$$f_a=\frac{2}{3}(E_f-E_0)^{-3}\left|\left\langle\Psi_0\left|Z\sum_i \mathbf{r_i}/r_i^3\right|\Psi_f\right\rangle\right|^2 \tag{4}$$

is well known. By replacing (E_f-E_0) with ω, PDCS for the velocity and acceleration gauge can immediately be obtained from Eq. (1).

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In this work, the cross sections are calculated using a complex-rotation technique suggested by Rescigno and Mckoy [27]. The formulation of this method has been presented in Chung [28]. These details will not be repeated here. To describe the method briefly, it is pointed out in Ref. [27] that the cross-section expression of Eq. (1) is proportional to the imaginary part of the frequency-dependent polarizability. Since this polarizability can be calculated with the standard variation-perturbation technique using the square integrable wave functions and a complex-rotation method, one can thus obtain the cross section with the square integrable wave function only. Rescigno, McCurdy, and Mckoy have also applied this method to H^- PDCS with an approximate wave function [29]. Recently, Chung has applied this method to the photoionization of lithium both from $1s\,^22s$ and $1s\,^22p$ states; high-precision results are obtained [28,30,31].

The basis wave functions used in this work are multiconfiguration (CI) wave functions using Slater-type orbitals. For the $1s1s$ ground state the spatial part of the wave function is given by

$$\Psi_0(\mathbf{r_1},\mathbf{r_2})=\sum_{j,l} C_{jl}(r_1^{k_j}r_2^{n_j}e^{(-\alpha_l r_1-\beta_l r_2)}Y_{ll}^{00}+1\leftrightarrow 2), \quad (5)$$

where C is the linear parameter and α and β are nonlinear parameters. The angular part is given by

$$Y_{ll}^{00}=\sum_m \langle 00|lml-m\rangle Y_{lm}(\Omega_1)Y_{l,-m}(\Omega_2). \quad (6)$$

In this work, we have chosen a 384 term Ψ_0 which include l from 0 to 9. The nonrelativistic energy of this wave function is $-0.527\,737\,15$ a.u. This energy is not as accurate as the ''exact'' energy of Pekeris [32]. However, this energy happens to be the exact energy plus relativistic and mass polarization corrections for H^-.

For the wave function in the continua, a saddle-point method is used. The same type of CI basis functions as that of ground state is adopted. For the two-electron system, the saddle-point method [33] is very similar to the Feshbach projection operator formulism [34]. The minimum principle in these methods allows us to obtain an optimized closed-channel wave function $\Psi_{\rm cl}$ with basis functions of reasonable size. For example, for the region below the $n=2$ threshold, an 175 term $\Psi_{\rm cl}$ will give us very accurate resonance energies for both $^1P^o$ resonances. For the region between $n=2$ to $n=3$ thresholds, a 293 term $\Psi_{\rm cl}$ is adopted which gives accurate energies for all six Feshbach resonances in this region.

The resonance width is obtained by using the saddle-point complex-rotation method [35]. The wave function in the continua is given by

$$\Psi(\mathbf{r_1},\mathbf{r_2})=\Psi_{\rm cl}(\mathbf{r_1},\mathbf{r_2})+\sum_{nl}(\psi_{nl}(\mathbf{r_1})\phi_{l\pm1}(\mathbf{r_2})+1\leftrightarrow 2), \quad (7)$$

where ψ_{nl} are the hydrogenic open channel target states, and the radial part of $\phi_{l\pm1}$ is given by

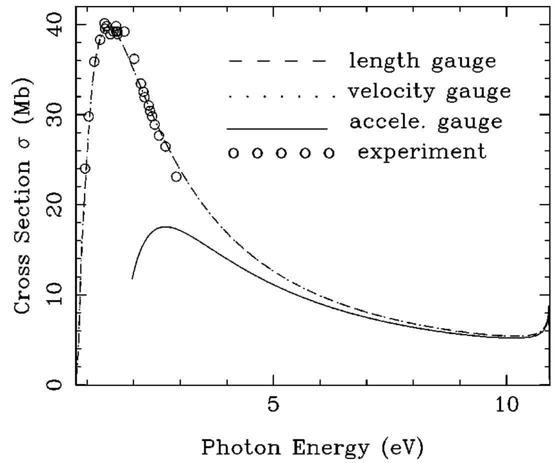

FIG. 1. Photodetachment cross section of H^- below 10.8 eV. Experimental data are from Smith and Burch [3], accele. denotes acceleration.

$$\phi_k=\sum_{j=k}^{K} C_j r^j e^{-\alpha r}. \quad (8)$$

Proper angular coupling is implicitly assumed in Eq. (7).

In the complex-scaling processes, the Hamiltonian, $\Psi_{\rm cl}$, and ψ_{nl} are unchanged. Only the r in $\phi_{l\pm1}$ is scaled to $re^{-i\theta}$. The same wave function for the resonance is used for that of the continuum. A similar complex-scaling procedure is adopted in the PDCS calculation.

III. RESULT AND DISCUSSION

Immediately above the detachment threshold, the PDCS profile exhibits a shape resonance. The profile of this resonance is measured by Smith and Burch [3]. These experimental data are presented in Fig. 1. Since the experimental result is a relative intensity measurement, we have multiplied the experimental data by a constant. The theoretical results are calculated using length, velocity, and acceleration gauges. It is clear from this figure that the agreement between the length and velocity gauge is excellent but the result from the acceleration gauge is very poor for photon energy below 5 eV. We have tried to improve the final state wave function to make it more complete but no significant improvement in the PDCS result is observed. The comparison between theory and experiment appears to be excellent in Fig. 1. The experimental uncertainty is about 3% [3] and it appears that almost every experimental data point agrees with theory. However, one needs to remember that this is a relative intensity measurement. It will be more conclusive if an absolute cross section can be made. In Table I, the calculated PDCS for a few photon energies below 10.5 eV is tabulated.

From photon energy above 7 eV to the $n=2$ threshold, the results from the three gauges agree closely. Near 10.9 eV there are two Feshbach resonances. The first one has been measured in MacArthur *et al.* [7]. The second resonance was only observed recently [12]. Both resonances are extremely narrow. These resonances have been investigated with various theories. In Table II, we present a comparison on some of the results. For the lowest resonance, the results of Ho

TABLE I. H^- photodetachment cross section (in cm^2) below the $n=2$ threshold. The number in square brackets represents the power of 10 by which the preceding number is to be multiplied.

ω (eV)	σ_l	σ_v
0.805	3.493[−18]	3.654[−18]
1.055	2.987[−17]	2.987[−17]
1.455	3.981[−17]	3.963[−17]
1.855	3.667[−17]	3.668[−17]
2.255	3.174[−17]	3.177[−17]
3.055	2.337[−17]	2.333[−17]
3.555	1.956[−17]	1.951[−17]
5.055	1.245[−17]	1.243[−17]
7.555	7.314[−18]	7.297[−18]
9.555	5.599[−18]	5.576[−18]

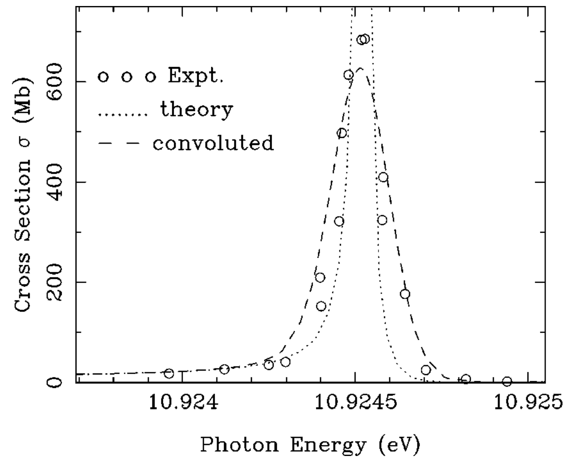

FIG. 2. H^- photodetachment cross section near the lowest $^1P^o$ Feshbach resonance at 10.924 52 eV. The theoretical data (dotted line) are convoluted with a Gaussian profile using 0.18 meV as FWHM. Experimental data are from Andersen *et al.* [12].

[36], Lindroth *et al.* [25], and Chen [26] agree extremely well with this work. We note that all these works use the complex-rotation method in one form or another but their wave functions are quite different. The results of Cortés and Martín [21] also agree closely with these theories but the width of Tang *et al.* [23] for the lowest Feshbach resonance is almost twice as much as that of others. This is very significant because it is claimed in Tang *et al.* [23] that their length and acceleration gauge agree to three digits ‘‘in the whole energy range.’’ This should indicate remarkable accuracy in the wave function. Hence, the discrepancy of their result with rest of the theories is especially significant.

In our calculation, it is quite challenging to determine the width for the second Feshbach resonance accurately. Part of the reason could be that it is extremely small. Different results are obtained when different choices of outgoing wave functions are used. The values range from 0.0016 to 0.0020 meV. Hence a value of 0.0018(2) eV is adopted. This result agrees with the theoretical data in the literature, which range from 0.0017 to 0.002 meV. The correct nonrelativistic energy for this resonance is probably around −0.125 035 a.u. The result given in Tang *et al.* [23] is −0.125 03 a.u. They have not given any width for this resonance. To compare with the experimental energy positions, we need to include relativistic and mass polarization corrections. The total correction for the lowest resonance is −1.04 μau and it is −1.48 μau for the second resonance. With these corrections, the predicted positions become 10.924 52 and 10.952 08 eV. These results are very close to the uncertainty quoted in Andersen *et al.* [12] but the lowest resonance is outside of the uncertainty quoted in MacArthur *et al.* [7].

The energy resolution in Andersen *et al.* [12] is 0.180 meV. The theoretical width for the lowest Feshbach resonance is about one-fifth of this value. The width of the second Feshbach resonance is narrower than the first by a factor of 20. This means that a direct comparison between the theoretical and experimental PDCS will not be meaningful for the resonances. To make a proper comparison, we convolute our PDCS with a Gaussian profile using the experimental resolution for the full width at half maximum (FWHM). These results are presented in Figs. 2 and 3.

It should be pointed out that the profile data presented in Andersen *et al.* [12] are for D^-. We converted their D^- data into data for H^- with corrections for specific mass shift and mass polarization difference. Figure 2 is the comparison for the lowest Feshbach resonance. It appears that the experimental data show a broader spectrum than that of the calculated data. This is expected. However, the convoluted data seem to be slightly broader than the experimental profile. A

TABLE II. H^- $^1P^o$ Feshbach resonances below the $n=2$ threshold ($E_{res}-E_{1s1s}$ in eV).

	$^1P^o(1)$			$^1P^o(2)$		
Authors	E_{nonrel} (a.u.)	$E_{res}-E_{1s1s}$	Γ (meV)	E_{nonrel} (a.u.)	$E_{res}-E_{1s1s}$	Γ (meV)
Ho [36]	−0.126 049		0.034			
Tang *et al.* [23]	−0.126 06		0.065	−0.125 03		
Sadeghpour *et al.* [19]	−0.126 014		0.0288			
Lindroth *et al.* [25]	−0.126 049 9	10.924 5	0.0372	−0.125 035 1	10.952 1	0.002
Chen [26]	−0.126 049 9		0.0356	−0.125 034 9		0.0019
Cortés and Martín [21]	−0.126 049		0.0324	−0.125 035		0.0017
This work	−0.126 048 9	10.924 52	0.0369	−0.125 035 3	10.952 08	0.0018(2)
Expt. [7]		10.926 4(6)				
Expt. [12]		10.924 3(2)			10.951 9(2)	

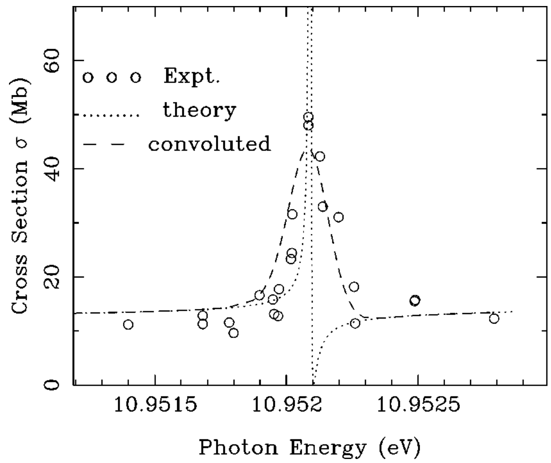

FIG. 3. H$^-$ photodetachment cross section near the second lowest $^1P^o$ Feshbach resonance at 10.952 08 eV. The theoretical data (dotted line) are convoluted with a Gaussian profile using 0.18 meV as FWHM. Experimental data are from Andersen *et al.* [12].

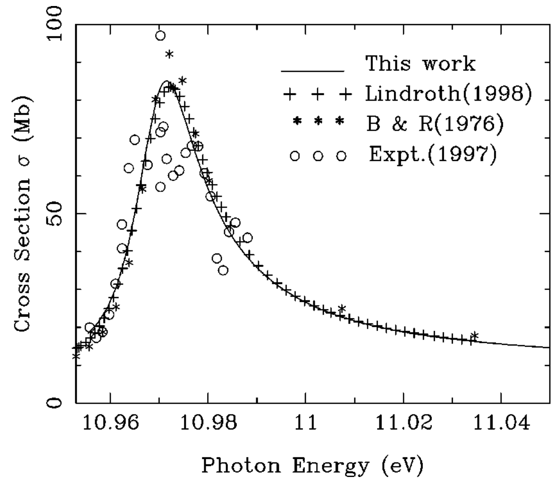

FIG. 4. H$^-$ photodetachment cross section near $^1P^o$ shape resonance above the $n=2$ threshold. Other theories are Lindroth [25] and Broad and Reinhardt [17]. Experimental data are from Andersen *et al.* [12].

similar comparison for the second lowest resonance is given in Fig. 3.

It is interesting to note that although the quoted positions for the H$^-$ Feshbach resonances in Andersen *et al.* [12] are slightly lower than our prediction, the energy position converted from their D$^-$ data agrees with our prediction almost exactly (see Figs. 2 and 3).

Above the $n=2$ threshold, the PDCS of H$^-$ rises due to the presence of a shape resonance. This resonance has been investigated by many theorists. It is also studied experimentally in Bryant *et al.* [6] and Andersen *et al.* [12]. In Fig. 4, the result of our calculation is compared with the recent data of Andersen *et al.* and with Broad and Reinhardt [17] and Lindroth [25]. It appears that we agree with Lindroth extremely well.

For the Feshbach resonances below $n=3$, the result of

TABLE III. H$^-$ $^1P^o$ Feshbach resonances below the $n=3$ threshold. Experimental energy is converted into a.u. using $E_{1s1s}=-0.527\,737\,15$ a.u. and 1 a.u.=27.196 58 eV. The number in parentheses is the uncertainty in the last digit quoted.

Authors	$^1P^o(1)$	$^1P^o(2)$	$^1P^o(3)$	$^1P^o(4)$	$^1P^o(5)$	$^1P^o(6)$
			Energy (a.u.)			
Sadeghpour *et al.* [19]	−0.062 695	−0.058 866		−0.055 832		
Tang *et al.* [23]	−0.062 72	−0.058 59	−0.056 14	−0.055 91		
Cortés and Martín [21]	−0.062 646 8	−0.058 569 7	−0.056 075 9	−0.055 836 7	−0.055 629 0	
Lindroth [24]	−0.062 73	−0.058 57	−0.056 12	−0.055 90	−0.055 66	−0.055 58
Ho [37]	−0.062 716 75	−0.058 571 8	−0.056 116 7	−0.055 907		
This work (E_{nonrel})	−0.062 716 68	−0.058 570 8	−0.056 114 4	−0.055 898 1	−0.055 662 8	−0.055 574 5
This work (E_{rel})	−0.062 715 70	−0.058 570 3	−0.056 114 7	−0.055 898 3	−0.055 663 2	−0.055 574 8
Expt.						
Hamm *et al.* [5] [a]	−0.062 61(15)			−0.055 73(15)		
Cohen *et al.* [8]	−0.062 605(37)					
Halka *et al.* [10]	−0.062 53(11)					
			Width (meV)			
Sadeghpour *et al.*	33.4	0.402		1.16		
Tang *et al.* [23]	32.6	0.261		1.55		
Cortés and Martín [21]	27.55	0.1935	0.06466	1.790	0.00807	
Lindroth [24]	32.6	0.24	0.06	1.93	0.01	0.1
Ho [37]	32.40	0.2444	0.057	1.9		
This work	32.36	0.2442	0.0547(1)	1.916	0.0113(1)	0.1342
Expt.						
Hamm *et al.* [5]	27.5(8)			1.6(3)		
Cohen *et al.* [8]	39(2)					
Halka *et al.* [10]	30(3)					

[a]Position of the dip rather than the true resonance energy.

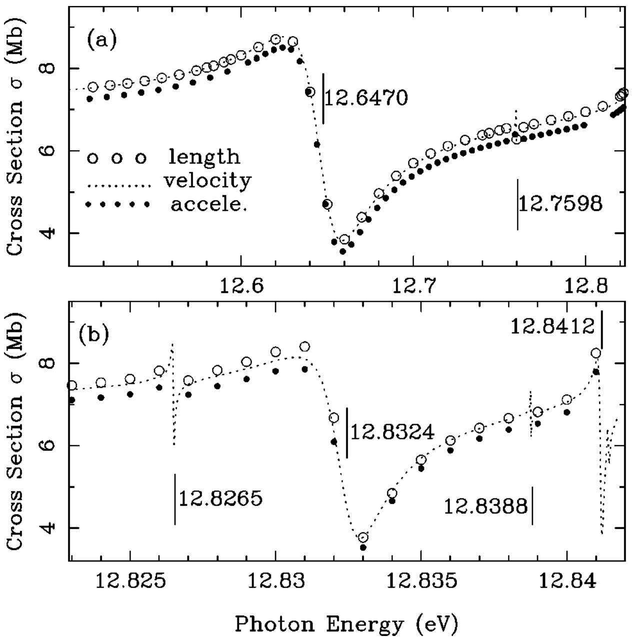

FIG. 5. H^- photodetachment cross section below the $n=3$ threshold. The vertical bars give the location of the resonances, accele. denotes for acceleration gauge.

this calculation is given in Table III. Both the nonrelativistic energy and the energy with relativistic and mass polarization corrections are listed. The relativistic correction includes P^4, the Darwin term, the electron-electron contact term, and orbit-orbit interaction. The last two types of corrections are extremely small for H^-. It is interesting to note that the total correction actually raises the energy for the lowest and second resonances. This is because the mass polarization corrections are positive for the two states and they are larger than the corresponding relativistic corrections. Our energy and width results agree well with Ho [37] and Lindroth [24] but deviate somewhat with those of Cortés and Martín [21]. Out of the five resonance energies calculated in this reference, only one is close to our result. This is significant because for the width of the lowest resonance, that of Cortés and Martín is the only one which agrees with experiment whereas all other theories agree with each other. For higher resonances, more discrepancies appear among the theories. However, our results consistently agree with those of Ho [37] and Lindroth [24]. The widths for the fifth and sixth resonances are small. Lindroth has quoted only one significant figure for these states. These widths converge well in our calculation. Hence, we are able to give more digits in this table.

In Fig. 5, we show the PDCS near the Feshbach resonances. This is a four open channel calculation. The results with length, velocity, and acceleration formulas are compared in this figure. It is clear from this figure that except in a very small energy region near 12.831 eV, the length and velocity results agree closely (to within 1%). The result from the acceleration is only slightly poorer. In this figure, the resonance positions are marked with vertical bars. As can be seen from this figure, the true resonance position of the broad states is substantially different from their dip. The position for the lowest Feshbach resonance is quoted to be 12.650(4) eV in Hamm *et al.* [5]. In Cohen *et al.* [8], this position is corrected to be 12.646(4) eV. It agrees well with the 12.6470 eV shown in this figure. The dip for the second broad resonance occurs at 12.833 eV. This is also within the uncertainty 12.837(4) eV quoted in Hamm *et al.* [5]. Other resonances have not been reported in the experiments thus far. With the high resolution achieved in Andersen [12], we hope they can be measured in the near future.

IV. CONCLUSION

In this work, we have studied the H^- PDCS below the $n=2$ and $n=3$ thresholds using a saddle-point complex-rotation method. Two Feshbach resonances below the $n=2$ threshold and six Feshbach resonances below the $n=3$ threshold are obtained. In addition, the PDCS for the two shape resonances in these energy regions are also obtained.

Our work is stimulated by the recent high-resolution experiment of Andersen *et al.* [12]. For the resonances reported in the experiment, we have made a detailed comparison between the theoretical results and measured data. Many of the narrow resonances below the $n=3$ have yet to be measured in the experiment. They are actually broader than the two resonances reported in Andersen *et al.* We hope a detailed theory-experiment comparison can soon be made on these resonances in the near future.

The PDCS in this work are calculated using length, velocity, and acceleration gauges. Above 6 eV (photon energy), the results from the three formulas agree quite well. However, below 6 eV, the results from the acceleration gauge are very poor. The reason for this is not clear; it remains a challenge to overcome.

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