TABLE I. The values of lifetimes and g factors of $^1P_1$ Ba and Ca.

	$\tau(^1P_1)$ (nsec)	$g_J(^1P_1)$
Barium	8.3(0.5)[a]	1.00(0.01)[a]
	8.37(0.20)[b]	1.0039(0.0008)[b]
	8.36(0.25)[c]	
Calcium	...[d]	1.00(0.01)[a]
	4.48(0.15)[e]	
	4.67(0.11)[c]	
	4.62(0.15)[f]	

[a]Present work.

[b]M. Swagel and A. Lurio, Phys. Rev. 169, 114 (1968).

[c]E. Hulpke, E. Paul, and W. Paul, Z. Phys. 177, 257 (1964).

[d]The ≃ 3-nsec width of the pulse response of the 1P21 precludes measurement of such a short time. The effect on the Ba lifetime, however, is small.

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ies of molecular excited states where one could measure the lifetime and g factor of several rotational levels of the same electronic state. In addition, tunable dye lasers with pulse lengths short compared to excited-state lifetimes would be useful tools for the study of radiation trapping in optically dense vapors. Finally, it may be possible to distinguish experimentally between the currently accepted theory of electromagnetic radiation and the recently proposed "neoclassical" theory.[15]

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Experimental Evidence for New Dissociation Channels in Electron-Impact Ionization of H_2

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Proton energy spectra have been obtained from the dissociative ionization of H_2 by electrons which reveal the existence of many new states lying between the $1s\sigma_g$ ground state of H_2^+ and the $2p\sigma_u$ repulsive state.

Considerable interest is currently being focused on the various possible ionization and dissociation channels in the simplest molecule H_2 under electron impact. Considerable evidence is available for the existence of high-lying H_2^- or excited H_2 states above the threshold for direct ionization of H_2 at 15.4 eV though the relative importance of autoionization and negative-ion formation in the ionization process has not yet been clearly established.[1-3] Autoionization is very prominent in the photoionization of H_2.[4]

Dissociative ionization by electron impact has been studied by Dunn and Kieffer[5,6] and by Van Brunt and Kieffer.[7] They demonstrated the existence of fast protons as a result of the dissociation process, and interpreted their results in terms of excitation to the repulsive $2p\sigma_u$ state of H_2^+, Fig. 1. To explain a discrepancy between the observed and calculated kinetic-energy distributions of H^+ from H_2, they suggested that some of the H^+ could be formed by autoionization from repulsive high-lying Rydberg states of H_2. They were not, however, able to obtain any direct evidence for the existence of any such states.

Recently, Misakian and Zorn[8,9] monitored the kinetic-energy spectra of fast metastable $2s$ H atoms produced by electron impact on H_2, and confirmed an earlier suggestion[10] that these were due to the dissociation of a doubly excited state of H_2. This state has an asymptotic energy of 24.9 eV and crosses the Franck-Condon excitation region around 33 eV. Very recently, Czuchlewski and Ryan[11] have been able to construct part of the potential-energy curve involved, and have found that the curve lies above the $2p\sigma_u$ curve.

To date there has been no direct evidence for

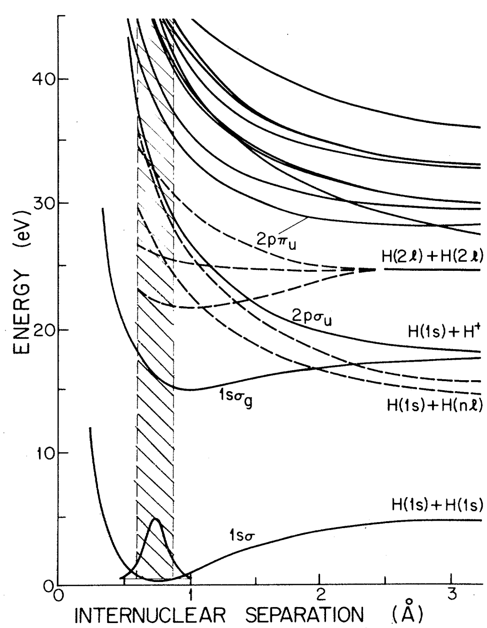

FIG. 1. Simplified potential-energy diagram of H_2 and H_2^+. Dashed curves, positions of some of the possible states seen in this work. The very approximate nature of these curves should be stressed.

the existence of any molecular states between the $1s\sigma_g$ ground state of H_2^+ and the $2p\sigma_u$ repulsive state, though very recently in experiments studying the dissociative ionization of H_2 by photon[12] and electron impact,[13] protons have been observed with appearance potentials in the neighborhood of 25–26 eV, an energy at which the $2p\sigma_u$ state should not be accessible. To date there does not seem to have been any detailed calculations published of potential energy curves which might cross the Franck-Condon region in this energy interval, 18–30 eV, with the exception of the preliminary work on diabatic states.[14]

The present work, which presents detailed ion energy spectra arising from the dissociative ionization of H_2 by electron impact, is the first direct evidence for the existence of a number of other states in this energy region.

The apparatus used is a modified version of that described previously in detail.[13,15] Basically, it consists of a modified Pierce-type electron gun producing a beam of electrons which is fired through a static gas target. Ions produced in a field-free region are allowed to drift through two apertures, defining the angular resolution of the system, before entering the ion energy analyzer. This is a cylindrical capacitor electrostatic analyzer[16] which is preceded and followed by ion lenses carefully designed[13] to handle low-energy ions. Energy-selected ions are accelerated and focused into a quadrupole mass spectrometer, and ions of a particular mass are detected using a Channeltron electron multiplier. Output pulses from the Channeltron are routed directly to a multichannel scaler or PDP-8/E laboratory computer. A ramp voltage generated by the multiscaler is used to automatically sweep the deflecting voltage on the energy analyzer over the region of interest. In this way ion energy spectra could be obtained directly on the display of the multiscaler. The electron gun is rotatable relative to the detection system so that angular distributions of fragment ions can be studied. Also, by looking at forward and backward angles relative to the electron beam direction, momentum-transfer effects from the incident electron to the parent molecule can be investigated.

The energy resolution ($\Delta E/E$) of the ion energy analyzer was typically 2–3%, where ΔE is the full width at half-maximum. Although the transmission of the system as a function of ion energy has not yet been accurately determined, the potentials on the ion lenses were adjusted so that the transmission stayed approximately constant.

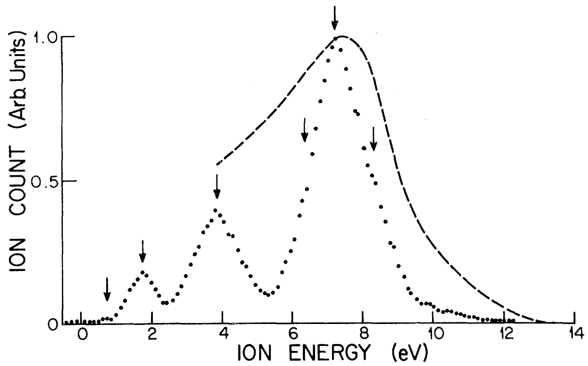

FIG. 2. Proton energy distribution obtained at an angle of 27° to the incident electron beam at an incident energy of 50 eV. Some of the features are indicated by the vertical arrows. See text for further details. The energy scale has been corrected for a contact potential of 0.57 eV. Dashed line, normalized data of Van Brunt and Kieffer (Ref. 7) taken at the same electron energy and at an angle of 23°.

The normal precautions were taken to ensure that the detected ion signal was proportional to electron beam current and target gas pressure. Base pressures in the system were $\sim 2\times10^{-8}$ Torr.

Figure 2 shows the proton energy spectrum obtained for an incident electron energy of 50 eV together with the data from Ref. 7 for the sake of comparison. Both sets of data have been normalized to unity at their maxima, and although both Kieffer and Dunn[6] and Van Brunt and Kieffer[7] found some evidence of structure at about 5 eV, a a wealth of hitherto unsuspected structure is apparent in the present data. In addition to the expected peak in the region of 8 eV (from the $2p\sigma_u$ state) and a contribution at close to zero energy (from the $1s\sigma_g$ ground state), there are pronounced peaks in the neighborhood of 1, 2, and 4 eV, with some evidence of a high-energy tail above 10 eV. The 8-eV peak is distinctly narrower than that given by the data of Ref. 7 and is, in fact, closer in shape to the theoretical curve predicted by Misakian, Pearl, and Mumma.[17] It is not clear why Van Brunt and Kieffer failed to see some of the structure shown in Fig. 2 if their resolution was 10% as they claimed.

Closer examination of the curves reveals the presence of additional fine detail; for example, that indicated by the arrows on both sides of the 8-eV peak. The 4-eV peak also exhibits a triple structure when examined in greater detail.

The variation of the ion-energy spectra with

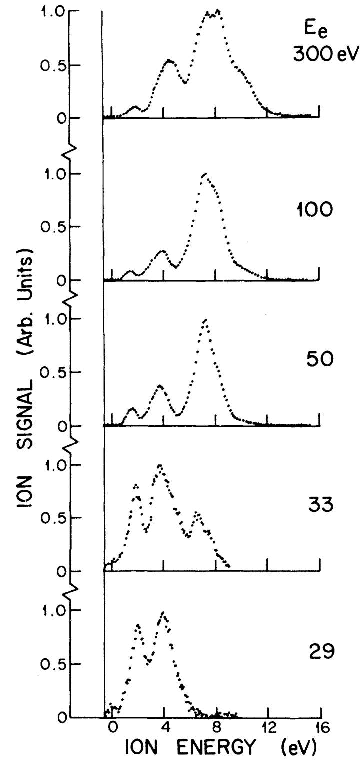

FIG. 3. Proton energy spectra obtained at the incident electron energies indicated. Data were taken at an angle of 27° to the electron beam direction. The curves have been normalized so that the maximum signal in each spectrum is unity.

incident electron energy is illustrated in Fig. 3. A number of facts are evident. First, and perhaps most significant, is the fact that, at 29 eV, where the $2p\sigma_u$ state is not accessible, a rich proton spectrum is still obtained indicating the existence of some hitherto unsuspected channels for H^+ production. As the electron energy is increased, the 8-eV peak appears as expected, but the structure mentioned above introduces considerable distortion to the shape of this peak especially at the highest energies. At 300 eV the high-energy (10 eV) feature is particularly prominent.

The fact that the 8-eV peak is composite in nature suggests that analyses such as that of Van Brunt and Kieffer,[7] who measured momentum transfer to the parent molecule by observing the energy shift of the 8-eV peak between observations made in the forward and backward directions, need to be interpreted with some caution. Even very close to threshold the 8-eV feature is composite in nature. This is evident from observations which we have made in the forward and backward directions at an incident electron energy of 33 eV. In the forward direction the peak is considerably broadened with respect to what is seen at backward angles. This could be caused by one component being more affected by momentum transfer from the incident electron. This direct evidence of the composite nature of the 8-eV peak confirms the suggestion[6] that the $2p\sigma_u$ state of H_2^+ is not the only parent state for protons of this energy. This also probably explains why the angular distribution of 8-eV protons near threshold for their production does not go to zero at 90° to the incident electron beam as would be expected from symmetry arguments[18] for a Σ_g^+-Σ_u^+ transition. If, as seems likely, some of the 8-eV protons are coming from autoionization of high-lying states of H_2 with potential-energy curves very close to the $2p\sigma_u$ curve in the Franck-Condon region, then an isotropic component would be expected. Two possible autoionization channels exist. First a high-lying Rydberg state, for example, one of the $l\sigma_u n\sigma_g$ or $l\sigma_u n\pi_g$ series which converge on the σ_u state, may autoionize into the ground-state H_2^+ continuum near the internuclear separation where the curve crossing occurs (see Fig. 1). Hazi[19] has calculated that the lifetime of these states against autoionization is very short in this region. Alternatively, a curve crossing may occur between one of the doubly excited states of H_2 and the repulsive $2p\sigma_u$ state, both states crossing the Franck-Condon region rather close to one another.

Whether or not the subsidiary peaks seen at higher impact energies are due to autoionizing states in this region, or to some of the output channels which open up at these higher impact energies, where more of the high-lying repulsive states cross the Franck-Condon region, must await more detailed appearance potential measurements. If other channels are significant, then pronounced breaks in a graph of ion intensity against electron energy should occur for a particular ion energy.

Appearance potential measurements on the 4-eV ions point to a definite threshold for their production of 26 eV. They may be due to excitation, first of a doubly excited state of H_2 [the asymptotic energy of $H(2n)+H(2n)$ is 24.9 eV], followed by autoionization to the repulsive $2p\sigma_u$ state at or near the region where the curves cross (see Fig. 1). Because some of the ions in this group have energies considerably less than 4 eV (as low as 2.5 eV), it is necessary to postulate that the parent doubly excited state is bound.

The 2-eV peak is even more difficult to explain as appearance potential measurements suggest that it is made up of at least two components, one with an onset at about 23 eV and the other with an onset some 4 eV higher at 26.5–27 eV. (Because of the low count rates the uncertainty in these appearance potentials was of the order of ±0.5 eV.) Again, high-lying autoionizing states of the neutral molecule may be involved. However, if this occurs, one would have to postulate that the ejected electron must carry off considerable kinetic energy in order that total energy may be conserved.

Measurements near threshold indicate that the angular distributions of both the 4- and 2-eV ions possess some degree of anisotropy in the same sense, but not as pronounced, as for the 8-eV ions (i.e., more are observed in the forward and backward directions than at right angles to the electron beam). This suggests Σ_u^+ symmetry characteristics for the parent states.[18,20]

Because of the very low signal levels involved, it has not yet been possible to obtain appearance-potential data for the 1-eV peak, and so no suggestions can yet be made regarding its possible origin.

Other possible channels for proton production which should be considered are the formation of excited states of H_2^- as intermediate states or excitation to the repulsive region of an H^+-H^- curve. Depending on where such a curve crossed the Franck-Condon region above the H_2^+ dissociation limit, then protons of the proper energy might be produced. Unfortunately, current calculations[21] of the H^+-H^- potential curves are rather inaccurate at small internuclear distances.

In order to identify further some of the processes which are occurring, additional measurements of appearance potentials and angular distributions are currently underway, in addition to similar measurements on D_2 and parallel investigations of scattered electron and H^- production.

It is interesting to note that we have also made a series of measurements of dissociative-ion energy spectra for electron impact on N_2. Highly structured curves have been found for both N^+ and N^{++} production. Most of these structures were not seen in earlier work.[22] Clearly, measurements of this type open up whole new areas of interesting physics.

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Phase Variation in Coherent-Optical-Pulse Propagation

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It is shown that the stipulation of lossless propagation provides sufficient asymptotic information on the pulse-propagation process to permit determination of both amplitude and phase of a class of coherent optical pulses ($2n\pi$ pulses, $n=0,1,2,\ldots$) by the inverse method. Results for $n=1$ and 2 are summarized.

The purpose of this note is to show that information on the phase variation of coherent optical pulses propagating in an attenuating medium consisting of inhomogeneously broadened, nondegenerate, two-level atoms can be obtained by employing the inverse method. The inverse method has been used previously for solving the Korteweg–de Vries equation,[1-4] the modified Korteweg-de Vries equation,[5,6] the equations governing self-modulation and self-focusing of light waves in nonlinear media,[7] and the equations of coherent-optical-pulse propagation when phase variation is ignored.[8]

In the present instance, one finds that the equations solved by the inverse method, namely the Bloch equations with phase terms included, are equivalent to the equations constructed by Zakharov and Shabat[7] in their application of the method of Lax[9] to the equations governing self-focusing and self-modulation!

After the time dependence of the solution is determined by solving the Bloch equations by the inverse method, the spatial dependence is obtained by requiring that the solution also satisfy an appropriate wave equation. Nonresonant loss, Kerr-type nonlinearities,[10,11] etc., are not considered.

For the 2π pulse one merely finds that there is no frequency sweep and, for an asymmetric spectrum of inhomogeneous broadening, the phase term contains only spatial dependence. This agrees with early assumptions[12] and recent experimental results[11] on self-induced transparency. No restriction to form-preserving solutions is introduced in obtaining this result. For a 4π pulse it is found that an initially chirped pulse can decompose into a pair of 2π pulses that have different carrier frequencies.

For the plane polarized electric field vector

$$E(x,t)=\mathcal{E}(x,t)\cos[k_0x-\omega_0t+\phi(x,t)], \tag{1}$$

the slowly varying envelope and phase approxima-