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The Dissociation of OH and OD in Comets by Solar Radiation

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The radiative lifetimes of cometary OH are calculated as a function of the heliocentric velocity of the comet and the velocity distributions of the product atoms are determined. At a distance of 1 AU from the Sun, the lifetimes vary between 1.2×10^5 and 1.9×10^5 sec at solar minimum and between 1.0×10^5 and 1.4×10^5 sec at solar maximum, depending upon velocity. Continuous absorption into the repulsive $1^2\Sigma^-$ state is a major destruction path. The calculated lifetimes are generally consistent with the lifetimes inferred from observations, but suggest some elaboration of the models is necessary. Photodissociation of OH produces a low-velocity component of hydrogen atoms at 8 km sec^{-1} relative to the parent OH molecule and a high-velocity component between 17 and 27 km sec^{-1}. Photodissociation of OH leads to metastable $O(^1D)$ and $O(^1S)$ and is an additional source of the red and green line emission of atomic oxygen. The lifetime of OD is estimated to be about 4.3×10^5 sec at solar minimum and 2.6×10^5 sec at solar maximum so that the OD/OH ratio in comets is enhanced relative to the HDO/H_2O production ratio by a factor between 2 and 3. Photodissociation of OD produces only high-velocity D atoms with a mean value of 17 km sec^{-1}.

INTRODUCTION

Measurements of the spatial distribution of the hydroxyl radical in cometary atmospheres have been made by observations of ultraviolet emission at 309 nm and of radio emission and absorption at 18 cm. The distribution depends upon the velocities of the parent water molecules from which OH is produced by photodissociation and on the lifetime of OH (cf. Keller, 1976; Festou, 1981a). The lifetime of OH can be inferred from models of the distribution. The ultraviolet data have been interpreted to yield a lifetime of OH at 1 AU from the Sun for Comet Bennett (1970 LI) of $2(+1,-1)10^5$ sec (Keller and Lillie, 1974), for Comet Kobayashi–Berger–Milon (1975 IX) a lifetime of $2.3(+1.5,-1.3)10^5$ sec (Festou, 1981b), for Comet Kohoutek (1973 XII) a lifetime of $2(+2,-0.7)10^5$ sec (Blamont and Festou, 1974; Festou, 1981b), and for Comet Bradfield (1979X) a lifetime between 5×10^4 and 1.6×10^5 sec (Weaver *et al.*, 1981a). The radio data tend to suggest longer lifetimes. For five comets, including Kobayashi–Berger–Milon (1975 IX) and Bradfield (1978 VII), Despois *et al.* (1981) derived lifetimes of $4(+2,-1) \times 10^5$ sec. It may be possible to resolve the discrepancies by modifying the parameters in the models (Bockelée-Morvan *et al.*, 1981; Bockelée-Morvan and Gerard, 1984) and by taking into account variations in the solar flux (Oppenheimer and Downey, 1980). A reliable determination of the lifetime of OH would constrain the possible models and permit an accurate estimate of the cometary production rates of water.

Measurements of hydrogen Lyman alpha emission from comets indicate the presence of two populations of hydrogen atoms, one with a velocity of about 20 km sec^{-1}, the second with a velocity of about 8 km sec^{-1} (Keller, 1976; Drake *et al.*, 1976; Keller and Thomas, 1975; Opal and Carruthers, 1977; Festou *et al.*, 1979; Keller and Meier, 1980; Weaver *et al.*, 1981a,b; Festou *et al.*, 1983). It has been suggested that the high-velocity component arises from photodissociation of H_2O and the low-velocity component from photodissociation of OH (Keller, 1976; Festou, 1981b). Keller (1976) pointed

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out that OH is photodissociated by absorption into the $v' = 2$ vibrational level of the excited $A^2\Sigma^+$ state followed by predissociation, a sequence which produces hydrogen atoms with velocities of 8 km sec^{-1}. Jackson (1980) noted that the destruction rate of OH by absorption of solar radiation into the predissociating levels of the $A^2\Sigma^+$ state varies with heliocentric velocity and presented estimates of the corresponding lifetimes for a few velocities. Using more reliable molecular data, Singh *et al.* (1983) have calculated the destruction rate as a function of the radial velocity.

Other channels exist for the photodestruction of OH (van Dishoeck *et al.*, 1983; van Dishoeck and Dalgarno, 1983). The OH radical is dissociated by absorption into the repulsive $1^2\Sigma^-$ and $1^2\Delta$ states, into the repulsive branch of the $B^2\Sigma^+$ state, into the discrete levels of the $D^2\Sigma^-$ state, and into the mixed $2^2\Pi$–$3^2\Pi$ states. Hydrogen atoms with velocities ranging up to 30 km sec^{-1} are produced. The end products of absorption into the $1^2\Delta$ and $2^2\Pi$–$3^2\Pi$ states are hydrogen atoms in the ground state and oxygen atoms in the metastable O(^{1}D) state. The end products of absorption into the continuum of the $B^2\Sigma^+$ state are hydrogen atoms in the ground state and oxygen atoms in the metastable O(^{1}S) state. Thus photodissociation of OH is a source of emission of the red and green lines of atomic oxygen.

We present here calculations of the radiative lifetimes of OH as a function of the radial velocity of the comet and of the velocity distributions of the hydrogen and oxygen atoms produced by photodissociation of OH. The contribution of the photodissociation of OH to the emission of the red and green lines of atomic oxygen is assessed. An estimate is made of the lifetime of OD and of the velocity distribution of the deuterium atoms.

PHOTODESTRUCTION RATES OF OH

Figure 1 is an illustration of the potential energy curves of the low-lying doublet states of OH that are important in the destruction of OH by the absorption of solar radiation. The $X^2\Pi$ and $A^2\Sigma^+$ curves are taken from calculations by Langhoff *et al.* (1982), the $^2\Sigma^-$ curves from calculations by van Dishoeck *et al.* (1983), and the $1^2\Delta$, $2^2\Pi$, and $3^2\Pi$ curves from calculations by van Dishoeck and Dalgarno (1983).

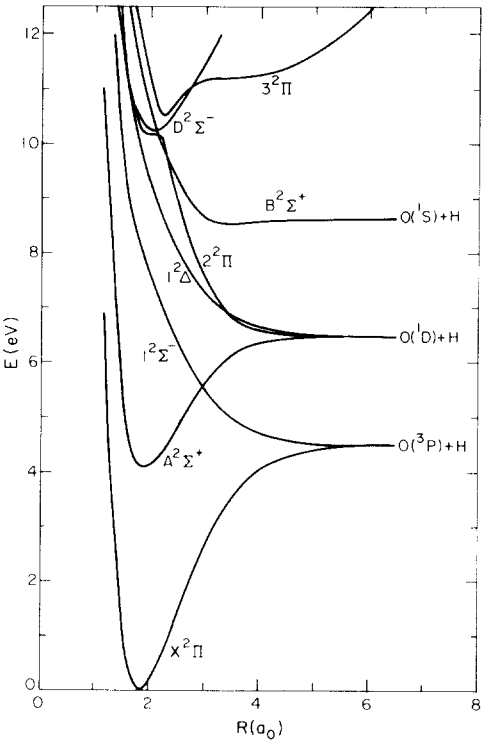

FIG. 1. Calculated potential energy curves of the doublet states of OH.

Discrete absorption into vibrational levels $v' \geq 2$ of the $A^2\Sigma^+$ state occur at wavelengths shorter than 260 nm and are followed by predissociation. The corresponding destruction rates for the $v' = 2$ level vary with the radial velocity of the comet relative to the Sun (Jackson, 1980) and take values between 6.1×10^{-6} and 3.0×10^{-6} sec^{-1} at 1 AU from the Sun (Singh *et al.*, 1983) (the signs of the velocity in this reference should be reversed). There is an additional contribution from absorption into the $v' = 3$ level. Because it is a small fraction of the total rate, we ignore its variation with velocity and take it to be con-

stant with a dissociation rate of 0.5×10^{-6} sec^{-1}. The resulting lifetimes against photodissociation are shown as the dotted line in Fig. 2 for velocities in the range ±60 km sec^{-1}. The solar radiation field intensities near 260 nm, which were obtained from the atlas of Kohl *et al.* (1978), vary by not more than 4% between solar maximum and minimum (Oppenheimer and Downey, 1980; Cook *et al.*, 1980) so that the lifetimes can be regarded as constant through the solar cycle.

At higher photon energies absorption occurs into the lowest-lying $1^2\Sigma^-$ state. The photodissociation cross section attains a maximum value of 3.0×10^{-18} cm^2 at a wavelength of 157 nm. The intensity of solar radiation between 180 and 140 nm varies from solar maximum to solar minimum by a factor of 1.5 to 2.0 (Oppenheimer and Downey, 1980; Cook *et al.*, 1980; Mount *et al.*, 1980; Rottman, 1981; Hinteregger *et al.*, 1981; Mount and Rottman, 1981, 1983; Lean *et al.*, 1982). The corresponding destruction rates at 1 AU, obtained using the solar fluxes of Mount *et al.* (1980) at solar maximum and of Mount and Rottman (1983) or Rottman (1981) at solar minimum, vary between 2.3×10^{-6} and 1.4×10^{-6} sec^{-1}. The lifetimes of OH resulting from absorptions into the $A^2\Sigma^+$ and $1^2\Sigma^-$ states for solar maximum conditions are shown in Fig. 2 as the short-dashed line.

The photodissociation of OH in comets by absorption into the $1^2\Delta$ and $B^2\Sigma^+$ states is driven by Lyman α radiation, the intensity of which varies considerably through the solar cycle (cf. Oppenheimer and Downey, 1980; Mount *et al.*, 1980; Bossy and Nicolet, 1981). The destruction rates for a solar Lyman α flux of 2.5×10^{11} photons cm^{-2} sec^{-1}, appropriate to solar minimum conditions (Bossy and Nicolet, 1981), for absorption into the $1^2\Delta$ and $B^2\Sigma^+$ states are, respectively, 3.0×10^{-7} and 5.0×10^{-8} sec^{-1}. The cross sections for these states at

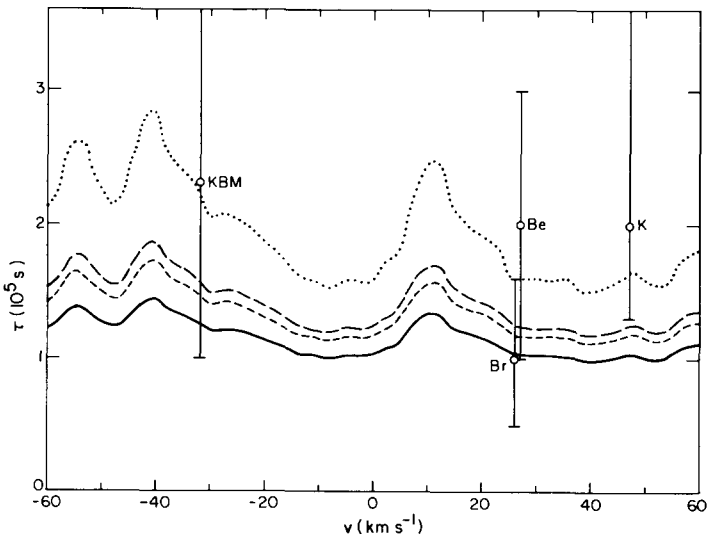

FIG. 2. The lifetime τ against photodissociation of OH as a function of the heliocentric radial velocity v of the comet. The dotted curve refers to absorptions into the $A^2\Sigma^+$ state only. The short-dashed curve refers to absorptions into the $A^2\Sigma^+$ and the $1^2\Sigma^-$ states at solar maximum. The long-dashed curve refers to absorptions into all states at solar minimum and the full curve to absorptions in all states at solar maximum. The open circles show the lifetimes inferred from UV observations with their uncertainties indicated. KBM: Comet Kobayashi–Berger–Milon (1975IX) (Festou, 1981b); Br: Comet Bradfield (1979X) (Weaver *et al.*, 1981a); Be: Comet Bennett (1970II) (Keller and Lillie, 1974); K: Comet Kohoutek (1973XII) (Festou 1981b).

the Lyman α wavelength are 1.2×10^{-18} and 0.2×10^{-18} cm^2, respectively. At solar maximum, the rates are enhanced by a factor of about 2.5.

Discrete absorptions into the lower levels of the $D^2\Sigma^-$ state, which occur at wavelengths near 120 nm, are followed by spontaneous radiative emission into the repulsive $1^2\Sigma^-$ state with an efficiency of about 0.2 (van Dishoeck *et al.*, 1983). The corresponding photodestruction rate is less than 10^{-8} sec^{-1}.

Absorption into the $2^2\Pi$–$3^2\Pi$ states has an effective threshold at 130 nm and maximizes at 104 nm. Because of nuclear radial couplings between the $^2\Pi$ states, the absorption structure is very complex (van Dishoeck *et al.*, 1984). However, the strong Lyman α line, where the cross section is 0.4×10^{-18} cm^2, is responsible for most of the dissociating events in the $^2\Pi$ channels and it gives a destruction rate of 1×10^{-7} sec^{-1} at solar minimum increasing to 2.5×10^{-7} sec^{-1} at solar maximum.

The photodissociation rates for each individual channel of OH in the solar radiation field are summarized in Table I. Figure 2 shows the lifetimes of OH resulting from photodissociation into all channels as functions of the radial velocity of the comet for solar minimum (long-dashed line) and solar maximum (full line). For velocities in the range ± 60 km sec^{-1} at a distance of 1 AU, the lifetimes vary between 1.2×10^5 and 1.9×10^5 sec at solar minimum and between 1.0×10^5 and 1.4×10^5 sec at solar maximum. The relative variation of the lifetime of OH with radial velocity is substantially diminished by the inclusion of the $1^2\Sigma^-$ channel.

The total lifetimes of OH, presented in Fig. 2, have a possible systematic error of $\pm 30\%$ because of uncertainties in the solar fluxes, oscillator strengths, and cross sections. The uncertainties in the solar fluxes are $\pm 15\%$, in the oscillator strength for the $A^2\Sigma^+$ state $\pm 10\%$, and in the cross sections for the $1^2\Sigma^-$ channel $\pm 10\%$. Because the solar flux diminishes rapidly toward shorter

TABLE I

SUMMARY OF CALCULATIONS OF THE PHOTODISSOCIATION RATES OF OH BY THE SOLAR RADIATION FIELD AT A DISTANCE OF 1 AU FROM THE SUN FOR EACH OF THE CHANNELS OF OH

State	λ_{eff}[a] (nm)	Photodissociation rate (10^{-6} sec^{-1})		Product	v_H (km sec^{-1})	v_O (km sec^{-1})
		Solar maximum	Solar minimum			
$A^2\Sigma^+$						
$v' = 2$	261	3.0–6.1[b]	3.0–6.1	$O(^3P) + H$	8.0	0.5
$v' = 3$	245	0.5[c]	0.5		11.0	0.5
$1^2\Sigma$	140–180	2.3[d]	1.4[e]	$O(^3P) + H$	22–26	1.6
$1^2\Delta$	Ly α	0.75[f]	0.30[g]	$O(^1D) + H$	26.3	1.6
$B^2\Sigma^+$	Ly α	0.13	0.05	$O(^1S) + H$	17.1	1.1
$2^2\Pi$–$3^2\Pi$	Ly α	0.25	0.10	$O(^1D) + H$	26.3	1.6
$D^2\Sigma$	120	<0.01	<0.01	$O(^3P) + H$	~22	1.4

[a] Wavelength where photodissociation by the solar radiation field is most effective.

[b] Obtained using the solar fluxes of Kohl *et al.* (1978); the rate depends on the radial velocity of the comet.

[c] Estimated average contribution; the dependence on velocity of the comet is neglected.

[d] Obtained using the solar fluxes of Mount *et al.* (1980).

[e] Obtained using the solar fluxes of Mount and Rottman (1983).

[f] Obtained by multiplying the rate at solar minimum by a factor 2.5.

[g] Obtained using an integrated Ly α flux of 2.5×10^{11} photons cm^{-2} sec^{-1} (cf. Bossy and Nicolet, 1981).

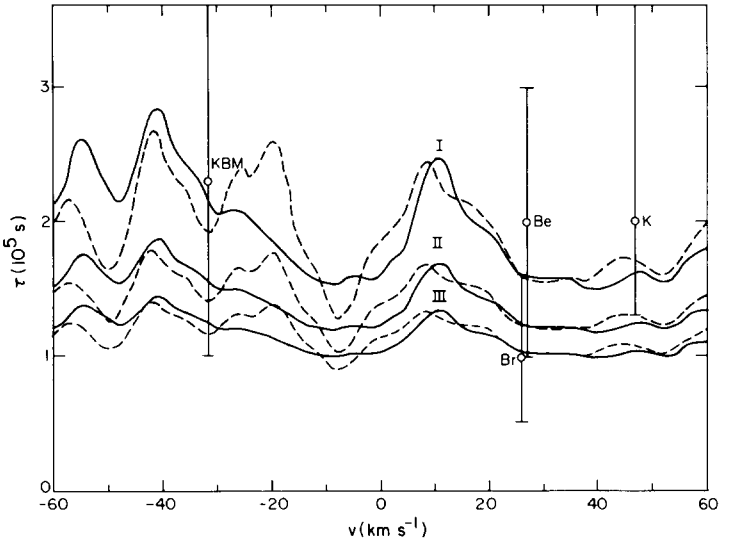

FIG. 3. Comparison between the lifetimes τ against photodissociation of OH without (full lines) and with (dashed lines) inversion in the ground-state λ doublet levels taken into account, as functions of the heliocentric radial velocity v of the comet. (I) Absorptions into the $A^2\Sigma^+$ state only; (II) absorptions into all states at solar minimum; (III) absorptions into all states at solar maximum. The open circles show the lifetimes inferred from UV observations, with their uncertainties indicated. For explanation of the symbols, see Fig. 2.

wavelengths, the rate for the $1^2\Sigma^-$ channel is very sensitive to the calculated excitation energy of the $1^2\Sigma^-$ state and the possible error of about 0.1 eV in the excitation energy gives rise to an uncertainty of ±25% in the rate. The cross sections for the $1^2\Delta$, $B^2\Sigma^+$, and $2^2\Pi$–$3^2\Pi$ channels at the wavelength of Lyman α are uncertain by ±30%, again because of uncertainties in the excitation energies of the states.

The lifetimes presented in Fig. 2 were calculated for an equal population of each of the two λ doublet components of the ground state. The population distribution is affected by solar ultraviolet pumping in the $A^2\Sigma^+$ state and it depends upon the radial velocity and on the differential motions of OH in the coma (Biraud *et al.*, 1974; Despois *et al.*, 1981). Photodissociation in the $A^2\Sigma^+$ channel will be modified at velocities where the population inversion is large (Schleicher and A'Hearn, 1983). Schleicher and A'Hearn (1982) have calculated the dependence of the population ratio on the radial velocity and the corresponding photodissociation rates for absorption into the $A^2\Sigma^+$ state are shown in Fig. 3. To obtain them, we employed the solar fluxes listed by A'Hearn *et al.* (1983) which are similar near 260 nm to those of Kohl *et al.* (1978). Figure 3 includes the total destruction rates at solar maximum and minimum. The effect of population inversion is greatest at a velocity near -20 km sec^{-1} where the photodissociation rate at solar maximum is diminished from 8.8×10^{-6} to 7.3×10^{-6} sec^{-1} and at solar minimum from 7.2×10^{-6} to 5.7×10^{-6} sec^{-1}.

The lifetimes inferred from analyses of the ultraviolet data (Keller and Lillie, 1974, 1978; Festou, 1981b; Weaver *et al.*, 1981a) on the assumption of an outflow velocity of H_2O between 0.5 and 1 km sec^{-1} are presented in Figs. 2 and 3 for several comets. Within the observational uncertainties, the empirical and theoretical values are consistent. For Comet Bradfield (1979X), Weaver *et al.* (1981a) were able to represent the data equally well by either an outflow velocity of 1 km sec^{-1} and an OH lifetime of 5

$\times 10^4$ sec or an outflow velocity of 0.5 km sec^{-1} and an OH lifetime of 1×10^5 sec. Our calculations then support the lower velocity of 0.5 km sec^{-1}.

The radio data (Despois *et al.*, 1981; Bockelée-Morvan *et al.*, 1981) yield lifetimes that are generally longer than we predict. Some elaboration of the theoretical models of the spatial distribution of cometary OH appears to be required to bring the empirical lifetimes into harmony with each other and with the theoretical lifetimes.

VELOCITY DISTRIBUTIONS

Energetic hydrogen and oxygen atoms are produced in the photodissociation of OH. In the predissociation of the $v' = 2$ level of the $A^2\Sigma^+$ state, a photon of energy 4.74 eV is absorbed, whereas the dissociation energy for the ground state of OH into $O(^3P) + H$ is 4.39 eV. The resulting hydrogen and oxygen atoms have velocities of respectively 8.0 and 0.5 km sec^{-1} relative to the parent OH molecule.

Absorption into the $1^2\Sigma^-$ state takes place between 140 and 180 nm and the dissociation products have a broad range of velocities. We have used the solar fluxes measured by Mount *et al.* (1980) and the cross sections of van Dishoeck *et al.* (1983) to derive the velocity distribution. The results for the hydrogen atoms are shown in Fig. 4. The central velocity is 24 km sec^{-1} and the half-width of the distribution is about 4 km sec^{-1}.

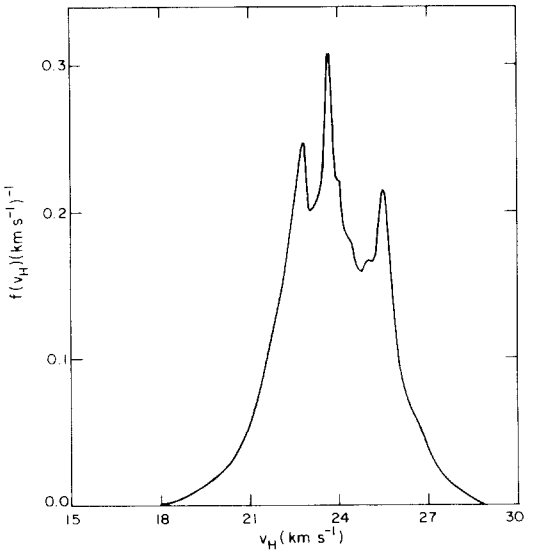

FIG. 4. The hydrogen atom velocity distribution resulting from the photodissociation of OH by absorption into the $1^2\Sigma^-$ state.

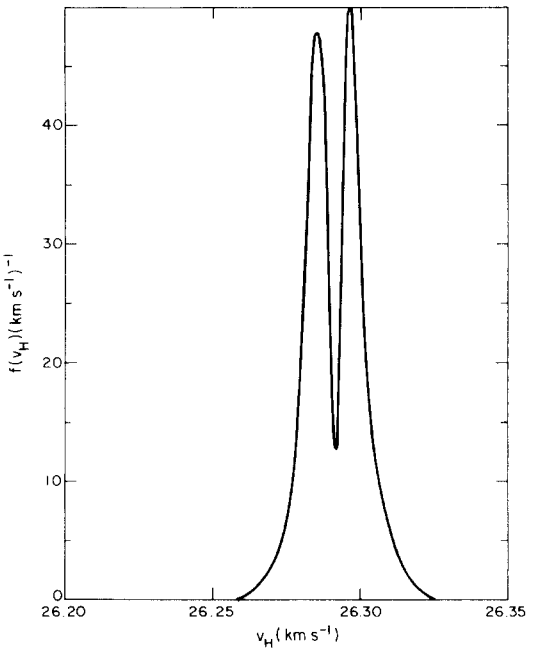

FIG. 5. The hydrogen atom velocity distribution resulting from the photodissociation of OH by absorption of Lyman alpha radiation into the $1^2\Delta$ or $2^2\Pi$–$3^2\Pi$ states.

In the solar radiation field, transitions into the $1^2\Delta$ state are dominated by Lyman α radiation. The $1^2\Delta$ state separates to ground-state hydrogen atoms and $O(^1D)$ oxygen atoms. The resulting hydrogen atom velocity distribution has been calculated from the Lyman α profile measured by Gouttebroze *et al.* (1978) and is illustrated in Fig. 5. It is sharply peaked at a velocity of 26.3 km sec^{-1}.

Lyman α radiation is also absorbed in transitions to the $2^2\Pi$–$3^2\Pi$ and $B^2\Sigma^+$ states. The $^2\Pi$ states have the same end products as the $1^2\Delta$ state and produce a similar velocity profile. Absorption into the vibrational

continuum of the $B^2\Sigma^+$ state leads to an oxygen atom in the metastable O(^{1}S) state and a ground-state hydrogen atom. The resulting hydrogen atom velocity distribution is sharply peaked at 17.1 km sec^{-1}, and has a profile similar to that of Fig. 5.

In the cometary atmosphere, the velocity distributions, whose shapes are illustrated in Figs. 4 and 5, are weighted by the associated destruction rates. Thus at solar minimum, between 65 and 80% of the hydrogen atoms have a velocity of 8 km sec^{-1} and the remainder, velocities between 17 and 27 km sec^{-1}. At solar maximum the fraction of the faster atoms is approximately doubled and the fraction of atoms at 8 km sec^{-1} is reduced to between 35 and 60%, depending on the radial velocity of the comet.

Table I contains a summary of the hydrogen atom velocities v_H and oxygen atom velocities v_O produced by the different channels. Absorption into the $v' = 2$ level of the $A^2\Sigma^+$ state is a satisfactory explanation of the observed low-velocity component of hydrogen atoms. The other channels provide rapidly moving hydrogen atoms at a rate which is about one-fifth of that arising from the photodissociation of H_2O (Oppenheimer and Downey, 1980; Festou, 1981b). The H_2O and OH sources of fast atoms have a similar variation through the solar cycle. There is some observational evidence (Festou *et al.*, 1979) that the more energetic component is produced closer to the nucleus which is consistent with H_2O as the major source.

METASTABLE OXYGEN

Emissions from the metastable O(^{1}D) and O(^{1}S) states of atomic oxygen have been observed in cometary atmospheres (cf. Festou and Feldman, 1981; Cosmovici *et al.*, 1982; Spinrad, 1982; Delsemme and Combi, 1983; Fink and Johnson, 1984). The observations indicate that the lines are produced close to the nucleus and that the metastable atoms are the daughter products of the same parent molecule. Plausible candidates for the parent are H_2O and CO_2. The hydroxyl radical is usually excluded from consideration but our quantal calculations establish that photodissociation of OH is a source of O(^{1}S) and O(^{1}D) atoms (cf. Table I). The production rates depend directly on the solar Lyman flux. At solar maximum, the rates per OH molecule are predicted to be 1.3×10^{-7} and 1.0×10^{-6} sec^{-1}, respectively, for O(^{1}S) and O(^{1}D). These rates are similar in magnitude to those for the production of O(^{1}S) and O(^{1}D) from H_2O (Festou, 1981b; Festou and Feldman, 1981), but because of contamination from the airglow it will not be easy to separate out the contribution from the photodissociation of OH.

PHOTODISSOCIATION OF OD

The destruction rate of the deuterated hydroxyl radical OD by absorption into the excited $A^2\Sigma^+$ state is much smaller than that of OH, raising the possibility of a substantial enhancement of the OD/OH abundance ratio in comets over the HDO/H_2O production ratio (Singh *et al.*, 1983). However, the additional channels, in particular the $1^2\Sigma^-$ channel, do not differentiate between OH and OD and they dominate the photodissociation of OD. Unlike OH, the photodissociation lifetimes of OD do not depend upon the velocity of the comet. They vary between 2.6×10^5 and 4.3×10^5 sec through the solar cycle, leading to a relative OD/OH enhancement of a factor between 2 and 3, depending on the velocity of the comet. The ratio may be further enhanced by chemical reactions such as

$$D + OH \rightarrow OD + H.$$

The reaction has a rate coefficient of 1.3×10^{-10} cm^3 sec^{-1} at 300°K (Margitan *et al.*, 1975) and is exothermic by 810°K.

The source of low-velocity deuterium atoms is negligible, but photodissociation of OD by absorption into the $1^2\Sigma^-$ state results in deuterium atoms with a mean velocity of 17.3 km sec^{-1}, and oxygen atoms with a mean velocity of 2.2 km sec^{-1}. The deuterium atoms, together with those from the

photodissociation of HDO, will be present as a small part of the hydrogen Lyman α profile.

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