Decay of H atoms excited in small electric fields

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The branching ratios for radiative decay of H atoms excited in small electric fields (0-5 V/cm) have been calculated for the *nl* states up to n = 6. A simple computational procedure was employed, allowing only for Stark-effect mixing of levels with the same values of the quantum numbers (n, j, m_j) . The results are compared with more detailed calculations available for 3*l*-state decay made using the density-matrix formalism, and new calculations of this type reported here for 4*l*-state decay. In conjunction with theory, this allowed the domain of validity of the simple computational procedure to be established as a function of *n*. The results show that the branching ratios depend quite strongly on electric-field magnitude, pointing to the need to exercise caution in measurements of H emissions, and in application of the available data to other problems.

I. INTRODUCTION

Studies of the emissions from atomic hydrogen (H) have played a key role in the development of the field of atomic physics, and continue today as valuable tools in a wide variety of basic and applied scientific investigations. Because of this importance, the production of excited H atoms in various atomic interactions has received considerable attention. Indeed, the literature abounds with cross-section data for electron- and ion-impact excitation of H, or dissociative excitation of H_2 and other hydrogen-containing molecules. Reactions leading to (fast) excited-H-atom formation by projectile (H⁺, H, H^- , H_2^+ , etc.) impact on a variety of atomic and molecular targets and surfaces also have been examined extensively. Almost all of the available information about such processes has been obtained by measurements of the H emissions resulting from the interactions.

Our purpose here is not to review this literature, but to discuss a problem which, at least to some extent, may have influenced many of these measurements. This problem results from the presence of small electric fields in the experimental regions where these emissions originate. Such fields can come from a variety of sources including contact potentials, projectile-beam space-charge effects, fringe fields from nearby electron- and ion-gun electrodes (or their electrical leads and vacuum-feed through headers), fringe fields from photomultiplier dynodes, ionization pressure gauges, or ion vacuum pumps, and from small potentials sometimes applied to nearby surfaces to trap or otherwise control often troublesome secondary electrons and ions within the system. Thus, electric fields in the V/cm range can be present in many laboratory environments, and great care must be taken if such fields are to be reduced to levels near or below 0.1 V/cm.

In addition, for those cases where emissions from fast reaction-product H atoms are detected, the $\mathbf{v} \times \mathbf{B}$ Lorentz

force can give rise to electric fields in the atom's moving reference frame which can be quite large. For example, a 50-keV H atom moving across the Earth's magnetic field (say 0.5 G) will experience an electric field of about 1.5 V/cm. Furthermore, fringe magnetic fields from such sources as beam-analyzing magnets, magnetically confined discharge sources, and magnetic ion pumps, can be present at levels significantly larger than the Earth's field itself. Many measurements have been made without using Helmholtz coils or other magnetic shielding to cancel out such effects.¹

In this paper we examine how the branching ratios for decay of the various nl excited states of H for $n \le 6$ are changed by electric fields between 0 and 5 V/cm, values which should encompass many of the effects noted above. In addition, however, similar field magnitudes can be present in situations where various H emissions are being used to study and quantify numerous other phenomena, ranging all the way from their use as laboratory cross-section and gas-discharge emission standards to the examination of extraterrestrial sources of such emissions. In fact, the basic motivation for this work was to investigate how the various H-emission intensities observed during proton auroras would be influenced by the motion of the fast emitting H atoms across the Earth's magnetic field.

Our goal in this work was thus twofold. First, we wanted to extend such branching-ratio data as calculated by Rouze *et al.*² for decay of the 3l excited states of H to higher principal quantum numbers, with particular emphasis on electric fields in the range of 1 or 2 V/cm. Second, however, was our desire to develop and employ a sufficiently simple computational procedure that could easily be applied to the proton-auroral problem of our own interest,³ but could be used also by other workers for their own needs without having to resort to more sophisticated and time-consuming calculations.

37 4201

II. COMPUTATIONAL PROCEDURES

Two procedures were used for the branching-ratio calculations reported here. One of us (W.B.W.) employed the density-matrix formalism described by Rouze *et al.*² for decay of the 3*l* states of H to extend the calculations to the 4*l* states. While not reviewed here, this procedure (subsequently called method I) accounts properly for the post-excitation time evolution of all the excited states of H at a given *n* level in the presence of the electric field, and is not limited to the small electric-field magnitudes of interest here. The other procedure (method II) to be described below, has a more limited electric-field range of applicability (dependent on principal quantum number *n*, as will be discussed in Sec. III), but satisfies the goal of computational simplicity noted above, allowing for ease of extension of the calculations to the higher-*n* levels.

Consider the electric-field-induced mixing of two essentially degenerate H-atom states (identified by subscripts 1 and 2) having the same values of quantum numbers n, j, and m_j . (These might be, for example, the 3p and 3d states having $j = \frac{3}{2}$ and $m_j = \frac{1}{2}$.) Let N_1 and N_2 be the rates of excitation of these states (in sec⁻¹ cm⁻³) by some collisional mechanism. Let $4\beta_1$ and $4\beta_2$ be their transition probabilities (sec⁻¹) for radiative decay, and define $\beta = \beta_1 + \beta_2$.

For weak fields under most excitation conditions, according to Bethe and Salpeter,⁴ we can determine the time-averaged populations (cm^{-3}) of H atoms in these two unperturbed excited states from

$$n_{1} = \left[\frac{4\beta\beta_{2} + f^{2}}{4\beta(4\beta_{1}\beta_{2} + f^{2})}\right]N_{1} + \left[\frac{f^{2}}{4\beta(4\beta_{1}\beta_{2} + f^{2})}\right]N_{2} ,$$
(1)

$$n_{2} = \left[\frac{f^{2}}{4\beta(4\beta_{1}\beta_{2} + f^{2})}\right]N_{1} + \left[\frac{4\beta\beta_{1} + f^{2}}{4\beta(4\beta_{1}\beta_{2} + f^{2})}\right]N_{2} .$$
(2)

Note that the state population n_1 , for example, depends in general on both N_1 and N_2 through the parameter f, the matrix element between states 1 and 2 of the perturbation Hamiltonian due to the weak electric field. This fis shown by Bethe and Salpeter⁴ to be

$$f = 8.040 \times 10^6 \left| \pm \frac{3}{4} \left[n^2 - (j + \frac{1}{2})^2 \right]^{1/2} \frac{nm_j}{j(j+1)} F \right|, \quad (3)$$

except that the numerical constant 8.040×10^6 has been added here to convert f to the units of sec⁻¹ when the electric-field magnitude F is expressed in V/cm.

We can use Eqs. (1) and (2) to find $B_1^*(\lambda)$ and $B_2^*(\lambda)$, the effective branching ratios for decay of states 1 and 2 via a photon emission of wavelength λ in the presence of the electric field:

$$B_1^*(\lambda) = 4\beta_1 B_1(\lambda) \left[\frac{n_1(N_1)}{N_1} \right] + 4\beta_2 B_2(\lambda) \left[\frac{n_2(N_1)}{N_1} \right] ,$$
(4)

$$B_2^*(\lambda) = 4\beta_1 B_1(\lambda) \left\lfloor \frac{n_1(N_2)}{N_2} \right\rfloor + 4\beta_2 B_2(\lambda) \left\lfloor \frac{n_2(N_2)}{N_2} \right\rfloor .$$
⁽⁵⁾

Here, $B_1(\lambda)$ and $B_2(\lambda)$ are the branching ratios for decay of states 1 and 2 with no field present. The $n_1(N_1)$ and $n_2(N_1)$ in Eq. (4) are those parts of the state populations n_1 and n_2 which result from the N_1 excitation process, given by the first terms in Eqs. (1) and (2). Similarly, the $n_1(N_2)$ and $n_2(N_2)$ in Eq. (5) are the second terms in Eqs. (1) and (2). In other words, we can think of the result of the mixing of these (parent) states in an electric field as a change in their effective branching ratios for radiative decay.

Unfortunately, Bethe and Salpeter⁴ do not derive equations for the populations in states 1 and 2 when their Lamb-shift energy separation ω_L is included.⁵ However, while the algebra is tedious, we have found that the effect of including the Lamb shift in these calculations is equivalent to replacing the f in Eqs. (1) and (2) by

$$f_L = \frac{f}{\left[1 + (\omega_L / 2\beta)^2\right]^{1/2}} , \qquad (6)$$

where the f in Eq. (6) is that from Eq. (3). (This does not result from any real change in the matrix element f, but is convenient to work with mathematically.) It is essentially that this substitution be made when considering the mixing of $ns_{1/2}$ and $np_{1/2}$ states (because of their large Lamb-shift energy separations), although its effect on the mixing of other states is relatively small.

The use of Eqs. (4) and (5) to compute $B_1^*(\lambda)$ and $B_2^*(\lambda)$ is, of course, limited to the condition where only two states can be mixed by the electric field. However, for small fields, states with the same values of the quantum numbers n, j, and m_j should mix first because of their near energy degeneracy. Therefore, when the field-induced splitting of these states (or more properly, of the two new states produced via the mixing process⁴) is small compared to the fine-structure separation of states with different j values, these calculations should be valid. We will examine the limits of this condition in Sec. III.

Thus, in the state notation $nl(j, m_j)$, these method-II calculations provide only, for example, for mixing of the $4p(\frac{1}{2}, \frac{1}{2})$ state with the $4s(\frac{1}{2}, \frac{1}{2})$ state, while the $4p(\frac{3}{2}, \frac{1}{2})$ and $4p(\frac{3}{2}, \frac{3}{2})$ states mix only with the $4d(\frac{3}{2}, \frac{1}{2})$ and $4d(\frac{3}{2}, \frac{3}{2})$ states, respectively. We show in Table I how the branching ratios for decay for this 4*p*-state example depend upon the quantum numbers *j* and m_j for an electric field of 1.5 V/cm. Also shown are similar results from the method-I calculations, where the 4*p*-state sublevels are characterized by their m_l values.^{2,6} As can be seen, the individual branching ratios tabulated are functions of the specific quantum numbers employed by each computational method. As expected, however, assuming that both methods are valid, the sublevel-averaged branching ratios found by the two methods are the same.

In Sec. III we graphically compare the results of these average branching ratios determined by the two methods for all the 3l and 4l excited states of H as a function of

TABLE I. Branching ratios for decay of the 4p excited state of H via Lyman- γ , Balmer- β , and Paschen- α emission for electric fields of 0 and 1.5 V/cm.

Excited state	Branchi Lyman-γ emission	ng ratios for 4 <i>p</i> Balmer-β emission	-state decay Paschen-α emission
No electric field			
4p			
(all sublevels)	0.839	0.119	0.042
Method-II calculations (electric field is 1.5 V/cm) 4n(i.m)			
$4p(\frac{1}{2},\frac{1}{2})$	0.830	0.124	0.046
$4p(\frac{3}{2},\frac{1}{2})$	0.771	0.169	0.060
$4p(\frac{3}{2},\frac{3}{2})$	0.667	0.247	0.086
Average	0.756	0.180	0.064
Method-I calculations (electric field is 1.5 V/cm)			
4p(+1)	0.737	0.194	0.069
4p(0)	0.791	0.154	0.055
4p(-1)	0.737	0.194	0.069
Average	0.755	0.181	0.064

electric-field strength. We recognize, of course, that use of such average branching ratios for any data analysis contains the implicit assumption that all the sublevels of any given nl state are populated equally in whatever excitation process is operative, an assumption that is certainly not valid for all such processes. However, for excitation of H atoms to the nl states for $n \ge 4$, only rarely does one even have partial information about how the nl states themselves are populated,⁷ to say nothing about how the population is distributed among the sublevels. Thus, in most cases, the assumption that the sublevels are populated equally is probably as reasonable an approximation as any other that could be made.

For 3*l*-state excitation, of course, the situation is quite different, with numerous workers⁸ reporting cross sections for collisional population of the individual 3*l* states for various reactions. Even here, however, only such recent studies of the type reported by Havener *et al.*⁹ and Westerveld *et al.*¹⁰ have started to investigate seriously how the 3*l*-state sublevels are populated during the collisions of interest (here being electron capture by H⁺ on He). This type of work, in fact, has even begun examining the off-diagonal elements of the density matrix describing the excitation to determine the coherence properties of the sublevel-population process.

Thus, while there are a few exceptions where sufficient information is available to warrant a sophisticated data analysis beyond the scope of this work, we have not included here the influence of any such coherent-excitation process. This is equivalent to ignoring the off-diagonal coherence terms in the density-matrix formalism² of method I, and ignoring any phase relations⁴ between the state populations n_1 and n_2 in Eqs. (1) and (2) in method II. For the small electric fields of interest here this is quite a reasonable approximation.¹¹ In addition, for application of these data to many phenomena such as the proton aurora, any effects of these coherent-excitation processes that occur in individual collisional interactions will be washed out by the integration over the various directions of motion and velocities of the emitting H atoms.³ Similar arguments, of course, can be made about dissociative excitation of hydrogen-containing molecules by any projectile, or the study of H emissions from gas discharges, fast-projectile impact on surfaces, most extraterrestial sources of emission, etc.

III. RESULTS AND DISCUSSION

We show in Fig. 1 how the branching ratios for decay of the excited nl states of H to lower-n levels depend upon the electric-field magnitude for fields up to 5 V/cm. The data points shown for decay of the parent 3l and 4lstates are from the calculations made using method I, the 3l data being taken from the earlier work of Rouze et $al.^2$ The line curves are the results of the method-II calculations described in Sec. II. The branching ratios plotted here, as noted in Sec. II, are those that result from assuming a statistical population of the sublevels of each nl state. The branching ratios for the individual sublevels (of the type presented in Table I) can be calculated easily using the method-II procedures outlined in Sec. II.

As can be seen, for decay of the 3l states, the changes in the branching ratios with electric-field magnitude are not severe, although they could have an influence, for example, on otherwise precise emission-cross-section measurements. Note also that the results of the two computational methods are in excellent agreement.

However, even for the 4*l* states, the branching-ratio changes are already quite dramatic. For example, in a 5-V/cm field, the 4s state decays almost 60% of the time to the 1s state, with the emission of otherwise forbidden Lyman- γ radiation. The resulting Balmer- β and Paschen- α radiations are, of course, decreased accordingly, by more than a factor of two in a 5-V/cm field. While the changes in the branching ratios for decay of the other 4*l* states are not quite as large, they can clearly have a substantial impact on the relative emission intensities resulting from excitation of these states in small electric fields. Note also that, once again, the branching ratios calculated by the two methods employed here are in very satisfactory agreement.

For the 5l and 6l states, Fig. 1 shows that the branching-ratio changes with electric field are even more profound. Once again, the largest changes occur for decay of the *ns* states, which decay primarily via Lyman-line emissions even at very small electric fields. (While not shown, the branching ratios for decay of the 5g, 6g, and 6h states were calculated, and are available on request.)

When considered as a group, the results shown in Fig. 1 reveal some interesting trends. For example, for electric fields above 2 or 3 V/cm, the 6l-state branching ratios appear to have reached almost constant values, while those for 5l-state decay seem to be approaching similar



FIG. 1. Branching ratios for decay of the excited nl states of hydrogen to lower-n levels vs the electric field. The data points are the results obtained using method I and the line curves are those using method II.

plateaus near 5-V/cm electric field. Indeed, even the 4*l*-state branching ratios seem to be trending toward such plateaus at somewhat larger electric fields. This observation is consistent with the theory of such state-mixing phenomena which, according to Bethe and Salpeter,⁴

should scale with an n^5 dependence on principal quantum number.

To further explore this point, we show in Fig. 2 the branching ratios for $ns \rightarrow 1s$ decay as a function of "scaled electric field," defined here as n^5 times the actual



FIG. 2. Branching ratios for $ns \rightarrow 1s$ transitions in hydrogen vs the scaled electric field. The data points are the results obtained using method I and the line curves are those using method II.

electric field. As can be seen, these *ns*-state branching ratios exhibit essentially identical dependences upon scaled-electric-field magnitude, nicely consistent with the theoretically predicted n^5 dependence cited above.

Note, however, that the method-I data points shown in Fig. 2 agree with the method-II curves only up to a scaled electric field of about 20 000 V/cm. Beyond this value, these data points begin to diverge downward from the field-independent plateaus exhibited by the method-II curves. We thus conclude that this scaled-electric-field value marks the onset for mixing of states not accounted for by the method-II calculations, i.e., for mixing of states with different *j* values. (It is interesting to note that this information is not readily apparent from the more detailed method-I calculations themselves.) In other words, our method-II results for the $ns \rightarrow 1s$ branching ratios appear to be accurate for (actual) electric fields up to about 80 V/cm for 3s decay and 19 V/cm for 4s decay. By inference then, we expect the 5s-state data to be accurate up to about 6.2 V/cm, but the 6s-state data only up to about 2.5 V/cm.

Thus, the essentially-field-independent plateau exhibited by the $6s \rightarrow 1s$ branching ratio plotted in Fig. 1 for fields above about 2.5 V/cm reflects the onset for breakdown of our method-II calculations. (For this reason, this branching ratio and the others for 6*l*-state decay for electric fields above 2.5 V/cm are plotted as dashed-line curves in Fig. 1.) In fact, by analogy with the data shown in Fig. 2, we estimate that this branching ratio in a 5-V/cm field (a scaled electric field of 39 000 V/cm) is too large by 5 or 10%. However, even if this is so, it is clearly a much better approximation to use the value plotted in Fig. 1 than to totally ignore the effects of such state mixing altogether (i.e., to use a $6s \rightarrow 1s$ branching ratio of zero).

Basically similar conclusions can be reached by comparing the data obtained by the two methods for decay of the np and nd states. Again, the very rapid initial changes in these branching ratios with scaled electric field result from the mixing of states with the same (j, m_i) values, which are accounted for properly by the method-II calculations. These rapid changes are then (generally) followed by much slower variations at larger fields, where the goodness of the quantum numbers j becomes increasingly invalid. However, for some transitions, for example, 4p decay to n = 2 or 4d decay to n = 3, the method-I and method-II data are still in agreement to within better than 10% for scaled electric fields up to well above 50 000 V/cm. (In other words, some of the method-I branching ratios also exhibit rather extended field-independent plateaus.) In contrast, for others such as 4p decay to n = 1or 4d decay to n = 2, the results obtained by the two method are already different by close to 10% at a scaled electric field of 20 000 V/cm.

Thus, strictly speaking, the domains of validity of the np and nd branching ratios obtained using method II are dependent on the specific branching ratios of interest. On average, however, they appear to be about comparable to those given above for *ns*-state decay. In any case, use of data for even the 6p and 6d decays shown in Fig. 1. should be considerably more accurate for fields of a few V/cm than assuming that the field-free branching ratios are valid.¹²

We also attempted to crudely estimate the influence small magnetic fields might have on the method-II branching ratios calculated here. These fields, of course, remove the energy degeneracy of the m_j sublevels for each $nl(j,m_j)$ configuration. This was done simply by including these Zeeman-effect energy splittings as part of the Lamb-shift energy separations ω_L used in Eq. (6). In general, the resulting changes in the calculated (sublevelaveraged) branching ratios introduced by this effect were found to be quite small, so long as the magnetic-field intensity was kept below a few Gauss.¹³

Based upon the data shown in Fig. 1, it is clear that electric fields of only a few V/cm can have a significant influence on the branching ratios for decay of the excited states of H, particularly for the $n \ge 4$ levels. Quantitative measurements of H emissions from atomic interactions should be made only in systems where all electric fields can be kept to values well below 1 V/cm. The often-used technique of employing a previously measured emission cross section as a photon-detector-calibration standard can result in error if the electric fields in the two systems are different. Great care must be taken also during studies attempting to verify, for example, the n^{-3} scaling law for excitation by observing the relative intensities of the various Lyman- or Balmer-line emissions resulting from some excitation process, for the electric-field-induced changes in the branching ratios entering such analyses are strong functions of n.

In addition, as noted earlier, care must be exercised by those who apply H-emission data to practical situations where small electric (and sometimes magnetic) fields are present. Our own analysis of auroral emissions, for example, while still preliminary, suggests that the (popularly measured) Balmer- β emission observed during energetic proton precipitation into the atmosphere will be smaller than expected if this effect is not taken into account.¹⁴ Thus the auroral models¹⁵ attempting to relate such observed emissions to the incident proton flux will require modification.

In conclusion, it might seem at first thought that studies of Lyman- α emission from various phenomena would not be affected by electric-field-induced changes in branching ratios for decay of the *nl* states for $n \ge 3$. However, this is not true for those situations where a significant part of the total Lyman- α emission results from cascade population of the 2*p* state of H. For example, Van Zyl *et al.*¹⁶ have shown recently that much of the Lyman- α resulting from H⁺ impact on rare-gas-atom targets for H⁺ energies above 50 keV results from electron capture into the higher *ns* states, whose (cascade) de-

- ¹While our ion-beam apparatus at the University of Denver may not be typical, the fringe magnetic field in its collision region is nearly an order of magnitude larger than the Earth's magnetic field when its Helmholtz coil is not used.
- ²N. Rouze, C. C. Havener, W. B. Westerveld, and J. S. Risely, Phys. Rev. A **33**, 294 (1986).
- ³During aurora, protons enter the atmosphere with a variety of velocities and at a variety of pitch angles to the Earth's magnetic field, thus experiencing a continuum of $\mathbf{v} \times \mathbf{B}$ -induced electric fields. Analysis of this and other similar problems would be very difficult without having a simple computational procedure that could be repetitively and rapidly applied.
- ⁴H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One-and Two-Electron Atoms* (Springer-Verlag, Berlin, 1957). Eqs. (1) and (3) in this paper are Eqs. (67.17) and (67.11) in this reference.
- ⁵When the Lamb shift is included, the differential equations for the time-dependent wave-function coefficients in Eq. (67.10) of Ref. 4 must include the terms $\exp(\pm i\omega_L t)$, as shown by W. E. Lamb, Jr. and R. C. Retherford, Phys. Rev. **79**, 549 (1950); or G. Lüders, Z. Naturforsch. **5a**, 608 (1950).
- ⁶The branching ratios shown for these method-I results are for the case where the electric field is parallel to the H-atoms' direction of motion.
- ⁷See, for example, B. Van. Zyl, H. Neumann, H. L. Rothwell, Jr., and R. C. Amme, Phys. Rev. A 21, 716 (1980); or references to similar work in B. Van Zyl and M. W. Gealy, Phys. Rev. A 35, 3741 (1987).
- ⁸See, for example, A. H. Mahan, A. Gallagher, and S. J. Smith, Phys. Rev. A 13, 156 (1976); R. H. Hughes, C. A. Stigers, B. M. Doughty, and E. D. Stokes, *ibid.* 1, 1424 (1970); J. Lenormand, J. Phys. (Paris) 37, 699 (1976); F. B. Yousif, J. Geddes, and H. B. Gilbody, J. Phys. B 19, 217 (1986); or Refs. 7 above and 9, 10, and 16 below.
- ⁹C. C. Havener, N. Rouze, W. B. Westerveld, and J. S. Risely,

cays to the 2p state are clearly influenced by the presence of small electric fields. (This is also the type of reaction where the $\mathbf{v} \times \mathbf{B}$ -induced electric field can be quite large when suitable magnetic shielding is not used during an emission-cross-section measurement.) There thus appears to be no H-atom emission study that is totally immune to the type of potential problem discussed here.

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- ¹¹The average magnitude (in absolute value) of the 60 branching-ratio terms resulting from the off-diagonal density-matrix elements for decay of the n = 4 level was found from the method-I calculations to be only 0.013 for an electric field of 1.5 V/cm.
- ¹²The largest errors that result from use of the branching ratios shown in Fig. 1 for the n = 5 and 6 levels probably occur for the decay of nf states (or for the data not plotted for the 5g, 6g, and 6h states). These states will all have finite branching ratios for transitions to the 1s state in electric fields, which are not accounted for in the method-II calculations. However, the net effect of ignoring these transitions is probably quite small, for such states are not expected to be heavily populated in most common excitation processes.
- ¹³For this crude analysis, we have ignored the possibility of exhanced state mixing at Zeeman-splitting-induced level crossings (see Ref. 4). This could be a problem for the very-high-n, high-j, and high- m_j state mixings, because of the very small fine-structure energy separations between such states, and the large Landé-g factors for the high- m_j values. Again, however, as noted in Ref. 12, such states are unlikely to be heavily populated in most collisions, so the net effect of ignoring this problem is probably quite small.
- ¹⁴This results from the electric-field-induced decreases in the 4s and 4d branching ratios to n = 2 shown in Fig. 1.
- ¹⁵See, for example, M. H. Rees, Planet. Space Sci. **30**, 463 (1982); or B. Van Zyl, M. W. Gealy, and H. Neumann, J. Geophys. Res. **89**, 1701 (1984).
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