Theory of light-induced drift. VI. Roles of accommodation of normal and tangential momenta in surface light-induced drift with positive spontaneous relaxation parameter

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Surface light-induced drift of a rarefied gas in cells with flat-plate and circular-cylindrical geometries is studied, and exact analytical solutions to the model rate equations are obtained in the limit of large Knudsen number. The work is a continuation of part V of this series of papers, in which the model rate equations were tailored specifically in order to study the roles played by both the tangential and normal momentum molecule-surface accommodation coefficients. In part V, the "spontaneous decay" parameter, γ , was set equal to zero, and the case of general "laser excitation" parameter, q, was studied. In the present paper, we get exact analytical results for the case of arbitrary γ in the limit of small q; we also get accurate numerical results for arbitrary γ and q. In passing, one serious, and other less serious, typographical errors in part V are corrected.

DOI: 10.1103/PhysRevA.71.033412

PACS number(s): 34.50.Rk

I. INTRODUCTION

This paper is the last of a series of papers [1-5] on the phenomenon of light-induced drift. It is on surface lightinduced drift (SLID), and is a continuation of part V, it being necessary for readers to have read and understood part V; an understanding of parts I and II would be highly beneficial also. In the previous papers of the series it was shown how exact results may be obtained for reasonably nontrivial models of both SLID and BLID (the bulk analogue of SLID), and how meaningful applications to experiment may be made. In part V it was shown that both the normal and tangential momentum molecule-surface accommodation coefficients play roles in SLID, whereas the author's intuition was that the normal momentum coefficient would be irrelevant. However, a problem with part V is that the spontaneous decay rate, from excited state to ground state, denoted by γ , was set equal to zero in order to simplify the working therein, and so the role of γ is still unclear. A main purpose of this paper is to clarify the role of γ in this problem.

There is one serious typographical error, and a few less serious ones, in part V, and these are corrected here ("here" means "in the present paper") in Appendix A. The notation used here is the same as that used in part V, except that the laser excitation frequency parameter q_0 , defined from Eqs. (I.2.3) and (I.2.4), is abbreviated to q. Additional definitions analogous to some of those made in Eqs. (V.2.9) are as follows:

$$\Delta \alpha \equiv \alpha_g - \alpha_e, \tag{1.1a}$$

$$\Pi \alpha \equiv \alpha_g \alpha_e, \tag{1.1b}$$

where $\alpha_{g,e}$ are the accommodation coefficients for ground, excited state molecules used in parts I and II.

The starting point is the set of Maxwell-Boltzmann rate equations, the first appearance of which, for the present model, is Eqs. (V.2.13 a-d) for flat-plate (FP) geometry together with the replacements in Eqs. (V.3.2) and (V.3.3) for circular-cylindrical (CC) geometry.

The "active speed" of the molecules is denoted here by v; for the flat-plate (FP) case [1,5], $v \equiv |v_z|$, and for the CC case [2,5], $v \equiv V$. With $\gamma > 0$, for both the FP and CC cases, the four molecular velocity distribution functions $f_{ju} \equiv f_j^{(u)}$ ($j \equiv g, e; u \equiv i, o$) are of the form [5]

$$\frac{f_{ju}}{2m} = \frac{v^2 A_{ju} + v J_{ju} + K_{ju}}{v^2 + v B + C},$$
(1.2)

where *m* is the (bulk) Maxwellian distribution, with $2m(s_1, s_2)=1$ where $s_{1,2}$ are the velocity variables defined by Eq. (V.2.1a); Eq. (1.2) defines the fourteen constants, which depend on the four (as yet unknown) quantities $C_{ju} \equiv C_j^{(u)}$ and on the case (FP or CC) considered [5]. The notation $u \equiv i, o$ used here refers to molecules whose components v_x of velocity along the cell are inside, outside of the laser excitation interval (v_a, v_b) defined by Eq. (I.2.4); thus $u \equiv i$ if $v_a \leq v_x \leq v_b$, and $u \equiv o$ otherwise. In general, all of the fourteen constants in Eq. (1.2) depend on q, γ , and the normal and tangential momentum accommodation coefficients for ground-state and excited-state molecules, denoted by α_{gn} , α_{gt} , α_{en} , α_{et} in the obvious notation [5].

In parts I-IV, methods were developed to get exact analytical results in cases in which the analogues (f_i) of f_{iu} are ratios of *linear* polynomials in v; for $\gamma=0$, Eq. (1.2) becomes just such a ratio [5], which is the reason why only the case $\gamma=0$ was studied in part V. For $\gamma>0$, the author is still unable to get exact analytical results for arbitrary q in the cases considered in part V [5]. However, using ideas which stem from the work in part V, such exact results have been obtained for the case $\gamma > 0$ in the important limit of small q. These results, together with a description of the method by which they are obtained, are presented and discussed here. The author is able to get numerical results, correct to arbitrary accuracy, for arbitrary values of the parameters, including q now; those numerical results indicate that the important results, as far as SLID is concerned, do not depend significantly on q.

II. METHOD OF SOLUTION

We define B_{ju} , C_{ju} , M_{ju} , P_{ju} by

$$B_{ju} \equiv J_{ju} - BA_{ju}, \qquad (2.1)$$

$$C_{ju} \equiv K_{ju} - CA_{ju}, \qquad (2.2)$$

$$(v_p - v_m)(M_{ju}, P_{ju}) \equiv (C_{ju} - v_m B_{ju}, v_p B_{ju} - C_{ju}),$$
 (2.3)

with $v_{m,p}$ ($m, p \equiv$ minus, plus) given by

$$\begin{pmatrix} v_p \\ v_m \end{pmatrix} \equiv \frac{B}{2} \pm \left(\frac{B^2}{4} - C\right)^{1/2}, \qquad (2.4)$$

in order to expand Eq. (1.2) into partial fractions in the form

$$f_{ju} = A_{ju} + \frac{M_{ju}}{(v + v_m)} + \frac{P_{ju}}{(v + v_p)}.$$
 (2.5)

We use the notation

$$H_k \equiv H(v_k) \equiv 1 - G_k, \quad k \equiv m, p, \qquad (2.6)$$

where the functions G(u) and $H(u) \equiv 1 - G(u)$ are defined and discussed in Appendix D of part I.

The concentrations $c_{ju} \equiv c_i^{(u)}$ are given by [5]

$$c_{ju} = \int ds_{1u} \int ds_2 f_{ju}, \qquad (2.7)$$

where $ds_{1u} \equiv ds_1^{(u)}$, and the quantities $C_{ju} \equiv C_j^{(u)}$ are given by [5]

$$C_{ju} = \langle v \rangle_m^{-1} \int ds_{1u} \int ds_2 v f_{ju}, \qquad (2.8)$$

where $\langle v \rangle_m$ is the average speed in the relevant Maxwellian distribution, given from

$$\pi \langle v \rangle_m^{(FP)} = 2 \langle v \rangle_m^{(CC)} = \pi^{1/2}, \qquad (2.9)$$

and where s_1 , s_2 are given in terms of v_x , v by

$$s_1 = \operatorname{erf} v_x, \qquad (2.10a)$$

$$s_2^{(FP)} = \operatorname{erf} v, \qquad (2.10b)$$

$$s_2^{(CC)} = e^{-v^2}$$
. (2.10c)

The integrands in Eqs. (2.7) and (2.8) are independent of s_{1u} , and we get [5]

$$\int ds_{1u} = \Delta s_u \equiv \Delta s^{(u)}. \tag{2.11}$$

In converting the integrals over s_2 to integrals over v, the Jacobians are given from Eqs. (2.10a) and (2.10b) by

$$|ds_2/dv|^{(FP)} = (2/\pi^{1/2})e^{-v^2},$$
 (2.12a)

$$|ds_2/dv|^{(CC)} = 2ve^{-v^2}.$$
 (2.12b)

Use of the results (2.7)–(2.12) gives, in an obvious notation,

$$\frac{c_{ju}^{(FP)}}{\Delta s_u} = \left[A_{ju} + \frac{G_m}{v_m} M_{ju} + \frac{G_p}{v_p} P_{ju} \right]^{(FP)}, \qquad (2.13)$$

$$\frac{\mathcal{C}_{ju}^{(FP)}}{\Delta s_u} = [A_{ju} + \pi^{1/2} (H_m M_{ju} + H_p P_{ju})]^{(FP)}, \quad (2.14)$$

$$\frac{c_{ju}^{(CC)}}{\Delta s_u} = [A_{ju} + \pi^{1/2} (H_m M_{ju} + H_p P_{ju})]^{(CC)}, \qquad (2.15)$$

$$\frac{\mathcal{C}_{ju}^{(CC)}}{\Delta s_u} = \left[A_{ju} + 2(\pi^{-1/2} - H_m \upsilon_m)M_{ju} + 2(\pi^{-1/2} - H_p \upsilon_p)P_{ju}\right]^{(CC)}.$$
(2.16)

The "unitary condition" is

$$c_{gi} + c_{go} + c_{ei} + c_{eo} = 1, \qquad (2.17)$$

and exact analytical results, for arbitrary q, would be found for the FP and CC cases if the nine relevant equations were solved for the four quantities C_{ju} in each case; for the FP case, there are four equations from each of Eqs. (2.13) and (2.14), together with Eq. (2.17); for the CC case, there are four equations from each of Eqs. (2.15) and (2.16), together with Eq. (2.17). As stated in Sec. I, the author has failed to get exact analytical results for arbitrary q, and that is because the resulting algebraic expressions are too complicated for the author's computing resources to handle; however, he has obtained such exact results in the limit of small q, using the procedure described in the next paragraph, which gives great simplification of the calculations. Accurate numerical results are discussed at the end of Sec. IV below.

We expand all necessary quantities in powers of q, keeping only terms of orders zero and one, and use the notation that, for any quantity X(q), X_o and X_p are defined by

$$X_o \equiv X(0), \tag{2.18a}$$

$$X_p \equiv X'(0), \qquad (2.18b)$$

where the prime ' and the subscript p denote derivative. We get

$$M_{ju} = qM_{jup}, \qquad (2.19a)$$

$$P_{iu} = q P_{iup}, \qquad (2.19b)$$

$$A_{ju} = A_{juo} + qA_{jup}, \qquad (2.20)$$

$$\mathcal{C}_{ju} = \mathcal{C}_{juo} + qC_{jup}, \qquad (2.21)$$

where

$$A_{guo} = \mathcal{C}_{guo} = \frac{1}{2}, \qquad (2.22)$$

$$A_{euo} = \mathcal{C}_{euo} = 0. \tag{2.23}$$

From Eqs. (2.13)–(2.16), (2.19a), (2.19b), (2.20), and (2.21) we get

$$\frac{c_{ju}^{(FP)}}{\Delta s_u} = \left[A_{ju} + q \left(\frac{G_{mo}}{v_{mo}} M_{jup} + \frac{G_{po}}{v_{po}} P_{jup} \right) \right]^{(FP)}, \quad (2.24)$$

$$\frac{c_{ju}^{(CC)}}{\Delta s_u} = [A_{ju} + \pi^{1/2} q (H_{mo} M_{jup} + H_{po} P_{jup})]^{(CC)}, \quad (2.26)$$

$$\frac{C_{jup}^{(CC)}}{\Delta s_u} = [A_{jup} + 2(\pi^{-1/2} - H_{mo}v_{mo})M_{jup} + 2(\pi^{-1/2} - H_{po}v_{po})P_{jup}]^{(CC)}.$$
 (2.27)

It follows from the working described above that the quantities v_{mo} and v_{po} , that is the values of v_m and v_p with q=0, are given from

$$v_{mo}\alpha_{e \ comb} = v_{po}\alpha_{en} = g, \qquad (2.28)$$

with the notation of Eq. (V.2.7) for $\alpha_{e \ comb}$, where $g^{(FP)}, g^{(CC)}$ are defined by

$$g^{(FP)} \equiv \gamma, \qquad (2.29a)$$

$$g^{(CC)} \equiv \Gamma, \qquad (2.29b)$$

with Γ defined by [5]

$$\Gamma \equiv \pi \gamma / 2. \tag{2.30}$$

Exact analytic results, in the limit of small q, are found by solving the nine equations for the four quantities C_{jup} in each case: for the FP case, there are four equations from each of Eqs. (2.24) and (2.25), together with Eq. (2.17); for the CC cases, there are four equations from each of Eqs. (2.26) and (2.27), together with Eq. (2.17). The c_{ju} may now be calculated from Eqs. (2.24) and (2.26), and the quantities I_{1e} , I_{xs} , and I_{qd} from [5]

$$I_{1e}(q) = c_{ei} + c_{eo}, \qquad (2.31)$$

$$I_{xs}(q) = \left[\frac{(c_{gi} + c_{ei})}{\Delta s_i} - \frac{(c_{go} + c_{eo})}{\Delta s_o}\right] \frac{d(\exp)}{\pi^{1/2}}, \quad (2.32)$$

$$I_{qd}(q) = q(c_{gi} - c_{ei}), \qquad (2.33)$$

where the notation $d(\exp)$ is defined by Eq. (I.E4), and where we note that a typographical error in Eq. (V.2.24) is pointed out in Appendix A.

It turns out that I_{1e}/q and I_{xs}/q are now independent of q, as desired, but I_{qd}/q is not, so, consistently with our small-q analysis, we calculate a revised $I_{qd}(q)$ from

$$I_{qd}(q) = qI_{qdp}, \qquad (2.34)$$

using the notation of Eq. (2.12b) for the derivative. Our results for I_{1e}/I_{qd} , I_{xs}/I_{qd} , and I_{qd}/q are exact (for $\gamma > 0$) in the limit $q \rightarrow 0$, and are presented in Appendix B.

III. RECOVERY OF RESULTS, FOR THE LIMIT OF SMALL γ, FROM EARLIER PARTS

Using the results in Appendix B, together with the analytical properties [1] of the functions G(u), H(u) in the limit

of small u, the results for the limit $\gamma \rightarrow 0$ may be written as follows:

$$\frac{-1}{\ln \gamma} \left(\frac{I_{1e}}{I_{qd}} \right)^{(FP)} \to \frac{2}{\pi^{1/2} \alpha_{en}}, \tag{3.1}$$

$$\frac{-1}{\ln \gamma} \left(\frac{I_{xs}}{I_{qd}} \right)^{(FP)} \to \frac{2}{\pi} \frac{d(\exp)}{d(\operatorname{erf})} \frac{\Delta \alpha_{comb}}{\Pi \alpha_{comb}},$$
(3.2)

$$\left(\frac{I_{1e}}{I_{qd}}\right)^{(CC)} \rightarrow \frac{\pi^{1/2} [1 + (\pi/2 - 1)z\alpha_{et}]}{z\alpha_{et}\alpha_{en}}, \qquad (3.3)$$

$$\left(\frac{I_{xs}}{I_{qd}}\right)^{(CC)} \to \frac{d(\exp)}{d(\operatorname{erf})} \left[\frac{\Delta\alpha_t}{\Pi\alpha_t} + \left(\frac{\pi}{2} - 1\right)\frac{\Delta\alpha_{comb}}{\Pi\alpha_{comb}}\right], \quad (3.4)$$

where the notation d(erf) is defined by Eq. (I.4.10). All four of these results are the exact analogues of those obtained, for the limit $q \rightarrow 0$, in part V, in which γ was set equal to zero *a priori*. To "recover" the analogous results in parts I and II, it is sufficient to make the following formal direct substitutions in the results (3.1)–(3.4):

$$\alpha_{et} \to 1, \quad \alpha_{en} \to \alpha_{e}, \quad \alpha_{t} \to \alpha, \quad \alpha_{comb} \to \alpha.$$
(3.5)

For example, the analogue of result (3.4) appears in part II as

$$\left(\frac{I_{xs}}{I_{qd}}\right)^{(CC)} \to \frac{\pi d(\exp)}{2} \frac{\Delta \alpha}{d(\operatorname{erf})} \frac{\Delta \alpha}{\Pi \alpha}.$$
(3.6)

While the physical significance of the last three substitutions in Eq. (3.5) is clear, the author does not understand why the first substitution works; maybe the fact that α_{et} appears only in the product $z\alpha_{et}$ is relevant.

IV. DISCUSSION AND CONCLUSION

The effects of nonzero γ may of course be studied, for general values of the several parameters, by comparing the results in Appendix B with those in Sec. III and in Appendix V A. However, as discussed in Sec. IV of part V, in the context of current experimental capability, the α_{jk} are sure to be close to unity [6–10], and this fact allows considerable simplification of the results. Let us briefly consider such simplification for one of the important quantities in the analysis. The three important integrals are (i) I_{1e} , that is the concentration of excited molecules, given by [1]

$$I_{1e} = \int \int \int d^3 \mathbf{v} f_e(\mathbf{v}), \qquad (4.1a)$$

(ii) I_{xs} , that is the sum of the molecular fluxes (which is proportional to the pressure difference across the cell), given by [1]

$$I_{xs} = \int \int \int d^3 \mathbf{v} [f_g(\mathbf{v}) + f_e(\mathbf{v})] v_x, \qquad (4.1b)$$

and (iii) I_{qd} , that is the laser contribution to the rate of increase in the concentration of excited molecules, given by [1]



$$I_{qd} = \int \int \int d^3 \mathbf{v} [f_g(\mathbf{v}) - f_e(\mathbf{v})] q(\upsilon_x).$$
(4.1c)

The important quantity we study here is the ratio I_{xs}/I_{qd} , which is the quantity most directly related to experimental measurements [1]. With the α_{jk} close to unity, it follows from the results in Appendix B, bearing in mind results such as

$$G_{mo}^{(FP)} \equiv G(\gamma/\alpha_{e \ comb}) \approx G(\gamma), \qquad (4.2)$$

which follows from Eq. (2.28), that

$$\frac{d(\text{erf})}{d(\text{exp})} \left(\frac{I_{xs}}{I_{qd}}\right)^{(FP)} \approx G(\gamma) \left(\Delta \alpha_t + \frac{\Delta \alpha_{comb}}{\pi^{1/2} \gamma}\right), \quad (4.3)$$

$$\frac{d(\text{erf})}{d(\text{exp})} \left(\frac{I_{xs}}{I_{qd}}\right)^{(CC)} \approx J(\Gamma) \Delta \alpha_t, \qquad (4.4)$$

where Γ is defined by Eq. (2.30) and the function J(u) is defined, for u > 0, by

$$J(u) \equiv 1 - \pi^{1/2} u H(u)$$
 (4.5a)

$$=1 - \pi^{1/2}u + 2u^2 \int_0^\infty dx \frac{e^{-x^2}}{(x+u)},$$
(4.5b)

with $J(0) \equiv 1$ [1]. We show J(u), together with its expansions for small and large u [11], up to quartic terms, in Fig. 1.

In deriving the approximation (4.4), we have assumed that $\Delta \alpha_t \neq 0$ and have dropped a term proportional to $\Delta \alpha_{comb}$, which should [5] be negligible. The $\Delta \alpha_{comb}$ term may not necessarily be dropped from the approximation (4.3) because γ may be of order $|\Delta \alpha_{comb}/\Delta \alpha_t|$ or smaller; of course, if this is not the case, then the term may be dropped.

The effects of nonzero γ are most clearly seen by studying Eqs. (4.3) and (4.4) and the functions G(u) and J(u).

FIG. 1. Our function J(u) defined by Eq. (4.5), and its expansions, up to quartic terms, for small and large u, where $\sigma(\approx 0.58)$ is Euler's constant.

Relevant to the FP case Eq. (4.3), G(u)/u becomes [1] proportional to $\ln u$ for very small u, which leads to interesting behavior if $\Delta \alpha_{comb} \neq 0$. In any case, the strong dependence on γ in both the FP and CC cases is now clear. Importantly, the introduction of a nonzero γ does not affect the qualitative conclusions in part V concerning the roles of the normal and tangential momentum accommodation coefficients in SLID.

The strong dependence on γ , and the weak dependence on q, mentioned in Sec. I above, are illustrated in Fig. 2, in which numerical results, in the CC case, for the quantity I_{xs}/I_{qd} are shown as functions of both γ and q. The parameters chosen for the calculations are shown in the caption to Fig. 2. The calculations are consistent with the approximation (4.4) in all cases.

APPENDIX A: TYPOGRAPHICAL ERRORS IN PART V

The only potentially misleading, and hence serious, error in part V occurs in the second line of Eq. (V.2.24), in which the + sign should be a - sign. A less serious error, because it is easier to find, occurs in the first line of Eq. (V.2.21): a factor v is missing from the integrand therein.

Three trivial, easy to find, errors occur in Eq. (V.2.23) and the first line of Eq. (V.2.24), in which $ds^{(i)}$ and $ds^{(0)}$ should be $ds_1^{(i)}$ and $ds_1^{(0)}$, respectively. Finally, although it could be argued that these are not errors, v should be V in three places in Ref. [21] of part V.

APPENDIX B: EXACT ANALYTICAL RESULTS, FOR $\gamma > 0$ IN THE LIMIT $q \rightarrow 0$

All of the results presented in this appendix are derived from Eqs. (2.31)–(2.33).

1. FP geometry

With the notations defined by Eqs. (V.2.6) to (V.2.9), the results are given from



$$\frac{\gamma}{G_{po}}\frac{I_{1e}}{I_{qd}} = \frac{z\alpha_{et}\alpha_{en} + \pi^{1/2}(1 - z\alpha_{et})\gamma}{z\alpha_{et}\alpha_{en} + \pi^{1/2}(1 - z\alpha_{et})\gamma G_{po}},$$
(B1)

$$\frac{\gamma \alpha_{gt} \alpha_{g \ comb}}{G_{mo}} \frac{d(\text{erf})}{d(\text{exp})} \frac{I_{xs}}{I_{qd}} = \frac{(Y_t \Delta \alpha_t - Y_n \Delta \alpha_n) \gamma + Y_c \Delta \alpha_{comb}}{\alpha_{et} \alpha_{e \ comb} + \pi^{1/2} \alpha_{en \ mod} \gamma G_{mo}},$$
(B2)

$$I_{ad}/q = d(\text{erf})/2, \tag{B3}$$

with $Y_{t,n,c}$ defined by

$$Y_t \equiv \alpha_{en} + (1 - \alpha_{gt}) [(1 - \alpha_{en})(\alpha_{et} + \alpha_{gt}) - (1 - \alpha_{gn})(\alpha_{gt} + \alpha_{e \ comb})], \qquad (B4a)$$

$$Y_n \equiv (1 - \alpha_{gt}) \alpha_{gt}^2, \tag{B4b}$$

$$Y_c \equiv \pi^{-1/2} \alpha_{e \ comb} \Pi \alpha_t. \tag{B4c}$$

2. CC geometry

The analogues of Eqs. (B1)-(B4) are

$$\frac{1}{\pi^{1/2}} \frac{I_{1e}}{I_{qd}} = \frac{\alpha_{en} + z\alpha_{et}\alpha_{en}(\pi H_{po}/2 - 1) - \pi^{1/2}(1 - z\alpha_{et})H_{po}\Gamma}{z\alpha_{et}\alpha_{en}^2 + 2(1 - z\alpha_{et})(\pi^{-1/2}\alpha_{en} - H_{po}\Gamma)\Gamma},$$
(B5)

FIG. 2. Numerical results, in the CC case, for the quantity I_{xs}/I_{qd} as a function of q, for several values of γ . The other parameters are $\alpha_{gn} = \alpha_{gt} = 0.998$, $\alpha_{en} = \alpha_{et} = 0.996$, d(erf) = d(exp) = 1.0, and z = 0.5. All quantities are dimensionless, with characteristic dimensional quantities chosen as in parts I and II.

$$\alpha_{gt}\alpha_{g\ comb}\frac{d(\text{erf})}{d(\text{exp})}\frac{I_{xs}}{I_{qd}} = \frac{Z_{t}\Delta\alpha_{t} + Z_{c}\Delta\alpha_{comb}}{\alpha_{et}\alpha_{e\ comb}^{2} + 2\alpha_{en\ mod}(\pi^{-1/2}\alpha_{e\ comb} - H_{mo}\Gamma)\Gamma},$$
(B6)

$$I_{ad}/q = d(\text{erf})/2, \tag{B7}$$

with $Z_{t,c}$ defined by

$$Z_t \equiv (\alpha_{e \ comb} - \pi^{1/2} H_{mo} \Gamma) \Pi \alpha_{comb}, \qquad (B8a)$$

$$Z_c \equiv \left[\alpha_{e \ comb}(\pi H_{mo}/2 - 1) + \pi^{1/2} H_{mo}\Gamma\right] \Pi \alpha_t. \quad (B8b)$$

Readers are reminded that G_{ko} and H_{ko} $(k \equiv m, p)$ are given from [Eqs. (2.6), (2.18a), (2.28), (2.29a), and (2.29b)]

$$H_{mo} = H(g/\alpha_{e \ comb}) = 1 - G_{mo}, \tag{B9a}$$

$$H_{po} = H(g/\alpha_{en}) = 1 - G_{po}.$$
 (B9b)

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