Theory of light-induced drift. V. Roles of accommodation of normal and tangential momenta in surface light-induced drift

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Surface light-induced drift (SLID) 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 model rate equations, which have not appeared before, have been tailored specifically in order to study the roles played in SLID, in a physically realistic setting, by both the tangential momentum and the normal momentum molecule-surface accommodation coefficients. Because the model equations are new (to the best of our knowledge), the results are generally different from those of previous work, although emphasis is placed on obtaining relations between the present and previous results; applications to experiments may be made by means of those relations.

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I. INTRODUCTION

In Papers I–IV of this series of papers [1–4], the phenomenon of light-induced drift (LID) was discussed, and exact treatments of models of surface and bulk LID (SLID and BLID, respectively), in both one dimension (1D) and three dimensions (3D), were presented and compared. In order to understand the material here (''here'' means ''in the present paper''), it is necessary for readers to have read and understood Papers I and II, which considered 3D SLID in, respectively, flat-plate (FP) and circular-cylindrical (CC) geometries, and at least a partial understanding of Papers III and IV would be beneficial. As in the previous Papers [1–4] where SLID is discussed, the limit of large Knudsen number (the ratio of molecular mean free path to cell width) is understood. Dimensionless variables are formed as in Papers I and II.

With the exception of the model due to Streater and Vaksman [5] earlier work [6–10] on SLID has considered Maxwellian-type molecule-surface tangential momentum accommodation coefficients (TMACs α_{jt}), with $j\equiv(g,e)\equiv$(ground,excited) state, incorporated into the Maxwell-Boltzmann rate equations (MBREs), whereas Papers I and II used ''overall'' accommodation coefficients (ACs α_j) in the MBREs [1,2]. The author [11] has expressed the opinion that, with a physically realistic model of molecule-surface collisions that takes proper account of the differences between the ACs for tangential momentum and normal momentum (NMACs α_{jn}), the important results for SLID, namely, the three ratios I_{xs}/I_{qd}, I_{1e}/I_{qd}, and I_{qd}/q_0 [1,2], should depend essentially only on the difference $\Delta\alpha_t$ between the TMACs, and should be essentially independent of the difference $\Delta\alpha_n$ between the NMACs, with the obvious notation

$$\Delta\alpha_k=\alpha_{gk}-\alpha_{ek}:k\equiv(n,t). \tag{1.1}$$

The quantities I appearing above are integrals over the molecular velocity distributions, first defined in Appendix A of Ref. [1]. Vaksman [11] has tried, without success, to convince the author that $\Delta\alpha_n$ does, in fact, play a role in SLID. The problem is that Streater and Vaksman's paper [5] on SLID, in which it is claimed that $\Delta\alpha_n$ plays an important part, is not relevant to the models of SLID of the type [1,2] under the present discussion because it makes assumptions, about the roles of the accommodation coefficients, which are quite different from those made here. It is the purpose of this paper to make, and to work out the consequences of, a model of SLID that takes proper account of the $\Delta\alpha_k$, and to try to resolve the issue discussed above. To be specific, we concentrate on exact calculation, in 3D, of the ratios I_{xs}/I_{qd}, I_{1e}/I_{qd}, and I_{qd}/q_0 with $\gamma=0$ for small q_0, for both FP and CC geometries; γ is a rate parameter that models the spontaneous relaxation of molecules from excited state to ground state, and the reason we set $\gamma=0$ is explained in Sec. II C below. The model is described, in the context of FP geometry, in Sec. II, and the changes necessary for CC geometry are documented in Sec. III. Section IV contains discussion and conclusion.

II. THE MODEL IN THE CONTEXT OF FP GEOMETRY

A. General considerations

The geometry is as described in Paper I, with the laser beam running in the x direction, the plates perpendicular to the z direction, and with Cartesian coordinates (v_x,v_y,v_z) used for the molecular velocity space. In steady state, the molecular velocity distribution is piecewise Maxwellian in v_x and, at tangential momentum accommodation, molecules are redistributed into a (one-piece) Maxwellian distribution in v_x; also, at normal momentum accommodation, molecules are redistributed into a Maxwellian distribution in v_z. For these reasons, the analysis is greatly simplified if velocity variables (s_1,s_2) are defined such that the bulk Maxwellian distribution $m(s_1,s_2)$ is constant, that is,

$$(s_1,s_2)=(\mathrm{erf}\,v_x,\mathrm{erf}\,|v_z|), \tag{2.1a}$$

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$$m(s_1,s_2)=\frac{1}{2}:-1<s_1<1,\quad 0<s_2<1. \tag{2.1b}$$

The third velocity component v_y is trivially integrated out as in Paper I.

We define intervals $\Delta s^{(u)}$ of s_1 as follows. The interval of s_1 inside which the laser excitation operates is denoted by $\Delta s^{(i)}$ and the remaining interval of s_1 by $\Delta s^{(o)}$, with $u \equiv (i,o) \equiv$ (inside,outside) in an obvious notation. Thus, we have

$$\Delta s^{(i)}+\Delta s^{(o)}=2. \tag{2.2}$$

In practice, $\Delta s^{(i)}$ will be a single interval (s_a,s_b) of s_1, but it could consist of several disjoint intervals; if we use the language of a single interval, then

$$\Delta s^{(i)}=s_b-s_a=d(\mathrm{erf}), \tag{2.3}$$

in which $d(\mathrm{erf})$ stands for $(\mathrm{erf}\, v_b-\mathrm{erf}\, v_a)$ as in earlier Papers [1–4].

In Paper I, only two distribution functions f_j entered the analysis, but here, because of the redistribution of molecules over s_1 at tangential accommodation, we have four functions $f_j^{(u)}$; for example, $f_e^{(i)}(s_1,s_2)$ is the distribution function for excited molecules (e) inside (i) the laser excitation interval, and is independent of s_1 in that interval. The two concentrations c_j of Paper I are replaced by four $c_j^{(u)}$ here, that is,

$$c_j^{(u)}=\int ds_1^{(u)}\int ds_2 f_j^{(u)}=\Delta s^{(u)}\int ds_2 f_j^{(u)}, \tag{2.4}$$

where $\int ds_1^{(u)}$ stands for the integral with respect to s_1 over the interval $\Delta s^{(u)}$ and $\int ds_2$ stands for the integral with respect to s_2 over the interval (0,1). The normalization condition on $c_j^{(u)}$ is as follows:

$$c_g^{(i)}+c_g^{(o)}+c_e^{(i)}+c_e^{(o)}=1. \tag{2.5}$$

The following definitions help in simplifying the analysis. Modified (indicated by mod) α_{jk} are defined by

$$\alpha_{jn\mathrm{mod}}=\alpha_{jn}(1-\alpha_{jt}), \tag{2.6a}$$

$$\alpha_{jt\mathrm{mod}}=\alpha_{jt}(1-\alpha_{jn}), \tag{2.6b}$$

and combinations (indicated by comb) of α_{jk} by

$$\alpha_{j\mathrm{comb}}=\alpha_{jn}+\alpha_{jt}-\alpha_{jn}\alpha_{jt}. \tag{2.7}$$

We note that $\alpha_{jn\mathrm{mod}}$ is the probability of accommodation of normal momentum, but not of tangential momentum, during a molecule-surface collision, with the analogous meaning of $\alpha_{jt\mathrm{mod}}$, and that $\alpha_{j\mathrm{comb}}$ is the probability of *some* accommodation (normal momentum or tangential momentum or both) during a collision, that is, $\alpha_{j\mathrm{comb}}=1-(1-\alpha_{jn})(1-\alpha_{jt})$. The analogous probability of accommodation of both normal and tangential momenta during a collision is given by $\alpha_{jn}\alpha_{jt}$. We note the following "closure" relations that follow from Eqs. (2.6) and (2.7):

$$\alpha_{j\mathrm{comb}}=\alpha_{jn\mathrm{mod}}+\alpha_{jt\mathrm{mod}}+\alpha_{jn}\alpha_{jt}=\alpha_{jn\mathrm{mod}}+\alpha_{jt}$$
$$=\alpha_{jt\mathrm{mod}}+\alpha_{jn}. \tag{2.8}$$

For future use, we define the following differences, products, and sum involving the α_{jk} and the $\alpha_{j\mathrm{comb}}$:

$$\Delta\alpha_k=\alpha_{gk}-\alpha_{ek}:k\equiv(n,t), \tag{2.9a}$$

$$\Delta\alpha_{\mathrm{comb}}=\alpha_{g\mathrm{comb}}-\alpha_{e\mathrm{comb}}=(1-\alpha_{gt})\Delta\alpha_n+(1-\alpha_{gn})\Delta\alpha_t$$
$$+\Delta\alpha_n\Delta\alpha_t, \tag{2.9b}$$

$$\Pi\alpha_k=\alpha_{gk}\alpha_{ek}:k\equiv(n,t), \tag{2.9c}$$

$$\Pi\alpha_{\mathrm{comb}}=\alpha_{g\mathrm{comb}}\alpha_{e\mathrm{comb}}, \tag{2.9d}$$

$$\Sigma\alpha_{\mathrm{comb}}=\alpha_{g\mathrm{comb}}+\alpha_{e\mathrm{comb}}. \tag{2.9e}$$

B. The Maxwell-Boltzmann rate equations

1. Contributions to the MBREs

With the notation $\dot{f}\equiv\partial f/\partial t$, there are four different types of contribution to $\dot{f}_j^{(u)}$. The spontaneous relaxation from excited state to ground state, modeled [1] by the rate parameter γ, gives contributions $\gamma f_e^{(u)}$ to $\dot{f}_g^{(u)}$ and $-\gamma f_e^{(u)}$ to $\dot{f}_e^{(u)}$. The laser excitation, modeled [1] by the rate parameter function $q^{(i)}=q_0$, $q^{(o)}=0$, gives contributions $q_0(f_g^{(i)}-f_e^{(i)})$ to $\dot{f}_e^{(i)}$ and $-q_0(f_g^{(i)}-f_e^{(i)})$ to $\dot{f}_g^{(i)}$, with zero contributions to $\dot{f}_j^{(o)}$. During molecule-surface collisions, there are also "annihilation" and "creation" contributions to $\dot{f}_j^{(u)}$.

The annihilation contribution, denoted by $\mathcal{A}_j^{(u)}$, is of the form $-\nu p f_j^{(u)}$, where ν stands for the average frequency of the collisions and p stands for the probability of accommodation, given that a collision occurs; as $\nu=|v_z|$ and $p=\alpha_{j\mathrm{comb}}$, we have

$$\mathcal{A}_j^{(u)}=-v\,\alpha_{j\mathrm{comb}}f_j^{(u)}, \tag{2.10}$$

where v stand for $|v_z|$ $(=\mathrm{erfi}\, s_2)$ in Eq. (2.10), and in the remainder of Sec. II, in order to simplify the notation.

Each creation contribution is of the form νpF, where ν,p are the analogs of the corresponding annihilation quantities listed in the previous paragraph, but where F is more complicated than simply $f_j^{(u)}$. F is given by $\langle f\rangle v/\langle v\rangle_\ell$, where $\langle f\rangle$ is an appropriate average of $f_j^{(i)}$ or $f_j^{(o)}$, over s_1 if tangential momentum is accommodated and over s_2 if normal momentum is accommodated; $\langle v\rangle_\ell$ is the value of v averaged over the Maxwellian distribution ($\ell\equiv m$) if normal momentum is accommodated, and over $f_j^{(i)}$ or $f_j^{(o)}$ ($\ell\equiv f_j^{(i)}$ or $f_j^{(o)}$) otherwise. The factor $v/\langle v\rangle_\ell$ is, in fact, a gas kinetic theory "streaming correction" to F [12,13]. If normal, but not tangential, momentum is accommodated, we say we have a "basic contribution" of type N; if tangential, but not normal, momentum is accommodated, we have types $T^{(u)}$; if both normal and tangential momenta are accommodated, we have types $NT^{(u)}$. Table I contains the information necessary to construct the five basic contributions to $\dot{f}_j^{(u)}$. The term

TABLE I. Information necessary to construct the five basic creation contributions to $\dot{f}_j^{(u)}$. The abbreviations $\int ds_1^{(u)}$ and $\int ds_2$ are defined just after Eq. (2.4). Additional factors of z and $(1-z)$ must be inserted in appropriate places when forming the MBREs, as discussed in Sec. II B 1.

Type	Origin of contribution	ν	p	$\langle f\rangle$	k
N	$f_j^{(u)}$	$\langle\lvert v_z\rvert\rangle_j^{(u)}$	$\alpha_{jn\mathrm{mod}}$	$\int ds_2 f_j^{(u)}$	m
$T^{(i)}$	$f_j^{(i)}$	$\langle\lvert v_z\rvert\rangle_j^{(i)}$	$\alpha_{jt\mathrm{mod}}$	$\frac{1}{2}\int ds_1^{(i)} f_j^{(i)}$	$f_j^{(i)}$
$T^{(o)}$	$f_j^{(o)}$	$\langle\lvert v_z\rvert\rangle_j^{(o)}$	$\alpha_{jt\mathrm{mod}}$	$\frac{1}{2}\int ds_1^{(o)} f_j^{(o)}$	$f_j^{(o)}$
$NT^{(i)}$	$f_j^{(i)}$	$\langle\lvert v_z\rvert\rangle_j^{(i)}$	$\alpha_{jn}\alpha_{jt}$	$\frac{1}{2}\int ds_1^{(i)}\int ds_2 f_j^{(i)}$	m
$NT^{(o)}$	$f_j^{(o)}$	$\langle\lvert v_z\rvert\rangle_j^{(o)}$	$\alpha_{jn}\alpha_{jt}$	$\frac{1}{2}\int ds_1^{(o)}\int ds_2 f_j^{(o)}$	m

basic contribution is used because, in forming the MBREs, additional factors of z and $(1-z)$ must be inserted in appropriate places to account for the modifications to the probabilities which result from the collision-relaxation processes that are modeled by the probability parameter z. Here, we assume that those processes occur only in collisions in which both normal and tangential momenta are accommodated, that is, they occur only in collisions of types NT [14].

It is convenient to define the following analogs of the quantities $c_j\beta_j$ which appear in the earlier parts:

$$C_j^{(u)}\equiv c_j^{(u)}\beta_j^{(u)}=\langle v\rangle_m^{-1}\int ds_1^{(u)}\int ds_2 v f_j^{(u)}, \quad (2.11)$$

where it follows from Table I and earlier definitions that the basic creation contributions, denoted by $\mathcal{C}_j^{(u)}$, are given by

$$\mathcal{C}_{Nj}^{(u)}=v\,\alpha_{jn\mathrm{mod}}C_j^{(u)}/\Delta s^{(u)}, \quad (2.12a)$$

$$\mathcal{C}_{Tj}^{(u)}=\frac{1}{2}v\,\alpha_{jt\mathrm{mod}}(f_j^{(i)}\Delta s^{(i)}+f_j^{(o)}\Delta s^{(o)}), \quad (2.12b)$$

$$\mathcal{C}_{NTj}^{(u)}=\frac{1}{2}v\,\alpha_{jn}\alpha_{jt}(C_j^{(i)}+C_j^{(o)}). \quad (2.12c)$$

2. The MBREs for FP geometry

As this is the first appearance of these equations, and because it is essential for readers to understand, and hopefully to criticize, them, we write them in full.

$$\dot{f}_g^{(i)}=v\Big[\frac{1}{2}\alpha_{gt\mathrm{mod}}(f_g^{(i)}\Delta s^{(i)}+f_g^{(o)}\Delta s^{(o)})+\alpha_{gn\mathrm{mod}}C_g^{(i)}/\Delta s^{(i)}+\frac{1}{2}\alpha_{gn}\alpha_{gt}(C_g^{(i)}+C_g^{(o)})+\frac{1}{2}z\alpha_{en}\alpha_{et}(C_e^{(i)}+C_e^{(o)})-\alpha_{g\mathrm{comb}}f_g^{(i)}\Big]-q_0(f_g^{(i)}-f_g^{(o)})+\gamma f_e^{(i)}, \quad (2.13a)$$

$$\dot{f}_g^{(o)}=v\Big[\frac{1}{2}\alpha_{gt\mathrm{mod}}(f_g^{(i)}\Delta s^{(i)}+f_g^{(o)}\Delta s^{(o)})+\alpha_{gn\mathrm{mod}}C_g^{(o)}/\Delta s^{(o)}+\frac{1}{2}\alpha_{gn}\alpha_{gt}(C_g^{(i)}+C_g^{(o)})+\frac{1}{2}z\alpha_{en}\alpha_{et}(C_e^{(i)}+C_e^{(o)})-\alpha_{g\mathrm{comb}}f_g^{(o)}\Big]+\gamma f_e^{(o)}, \quad (2.13b)$$

$$\dot{f}_e^{(i)}=v\Big[\frac{1}{2}\alpha_{et\mathrm{mod}}(f_e^{(i)}\Delta s^{(i)}+f_e^{(o)}\Delta s^{(o)})+\alpha_{en\mathrm{mod}}C_e^{(i)}/\Delta s^{(i)}+\frac{1}{2}(1-z)\alpha_{gn}\alpha_{gt}(C_e^{(i)}+C_e^{(o)})\Big]+q_0(f_g^{(i)}-f_e^{(i)})-\gamma f_e^{(i)}, \quad (2.13c)$$

$$\dot{f}_e^{(o)}=v\Big[\frac{1}{2}\alpha_{et\mathrm{mod}}(f_e^{(i)}\Delta s^{(i)}+f_e^{(o)}\Delta s^{(o)})+\alpha_{en\mathrm{mod}}C_e^{(o)}/\Delta s^{(o)}+\frac{1}{2}(1-z)\alpha_{gn}\alpha_{gt}(C_e^{(i)}+C_e^{(o)})\Big]-\gamma f_e^{(o)}. \quad (2.13d)$$

A crucial check on these, or any MBREs of this type, is the "unitarity check," that is,

$$\sum_j\sum_u \dot{c}_j^{(u)}=\sum_j\sum_u\int_{-1}^{1}ds_1\int_0^1 ds_2\dot{f}_j^{(u)}=0. \quad (2.14)$$

C. Steady-state solution to the MBREs

The steady-state solution for the $f_j^{(u)}$, denoted hereafter by simply $f_j^{(u)}$, is obtained by solving the four (linear) equations, which result from setting $\dot{f}_j^{(u)}=0$ in Eq. (2.13), for the four functions $f_j^{(u)}$. The general form of the results may be written as follows:

$$f_j^{(u)}=\frac{(vA+q_0B)v+(vE+F)\gamma}{(vC+q_0D)v+(vG+H)\gamma}, \quad (2.15)$$

where the eight quantities A–H, which are to be regarded as abbreviations of $A_j^{(u)}$–$H_j^{(u)}$, are polynomials in the several parameters which appear in the MBREs (2.13), but are independent of v.

Thus, in the general case, the $f_j^{(u)}$ are ratios of two quadratic polynomials in v, which renders the remainder of an exact analysis exceedingly complicated, if not intractable. Previous Papers [1–4] of this series of papers have developed methods that are able to handle cases in which the $f_j^{(u)}$ are ratios of *linear* polynomials in v [15], and we keep that restriction here. It follows from the form of Eq. (2.15) that linear polynomials are obtained if $\gamma=0$, and we now restrict discussion to that case. This restriction has been commonly made before [1,2,16,17], but not for the reason here, particularly when applications to experiments have been considered [1,2].

With $\gamma=0$, the $f_j^{(u)}$ may be written in the following form, which is a simplified version of Eq. (2.15):

$$f_j^{(u)}=\frac{vA_j^{(u)}+q_0B_j^{(u)}}{vC+q_0D}, \tag{2.16}$$

which serves to define the ten quantities $A_j^{(u)},B_j^{(u)},C,D$, the last two of which are chosen to be independent of j,u.

D. Exact analytical solution

Proceeding by analogy with the working in previous Papers, we make the definition

$$X_j^{(u)}=B_j^{(u)}/D-A_j^{(u)}/C, \tag{2.17}$$

where the $f_j^{(u)}$ may be written

$$f_j^{(u)}=A_j^{(u)}/C+aX_j^{(u)}/(v+a), \tag{2.18}$$

where a is the important laser excitation quantity, given in terms of q_0 here by

$$\frac{a}{q_0}=\frac{(\alpha_{g\mathrm{comb}}\alpha_{et\mathrm{mod}}\alpha_{gn}+\alpha_{e\mathrm{comb}}\alpha_{gt\mathrm{mod}}\alpha_{en})d(\mathrm{erf})/2+\Pi\alpha_n\Sigma\alpha_{\mathrm{comb}}}{\Pi\alpha_{\mathrm{comb}}\Pi\alpha_n}. \tag{2.19}$$

With the integration written now in terms of v rather than s_2 (2.1a), and with $\langle v\rangle_m=\pi^{-1/2}$, it follows from Eqs. (2.4) and (2.11) that [18]

$$c_j^{(u)}/\Delta s^{(u)}=\int_0^\infty dv\,|ds_2/dv|f_j^{(u)}=A_j^{(u)}/C+X_j^{(u)}G_a, \tag{2.20}$$

$$C_j^{(u)}/\Delta s^{(u)}=\langle v\rangle_m^{-1}\int_0^\infty dv\,|ds_2/dv|f_j^{(u)}$$
$$=A_j^{(u)}/C+\pi^{1/2}X_j^{(u)}aH_a, \tag{2.21}$$

where the functions $G_a\equiv G(a)$, $H_a\equiv H(a)\equiv 1-G(a)$ are defined in Appendix D of Paper I.

The exact solution is now found by solving the system of nine equations, four from each of Eqs. (2.20) and (2.21) and one from Eq. (2.5), for the eight quantities $c_j^{(u)},C_j^{(u)}$. The integrals over 1, $q(v_x)$, v_x, analogous to those in Paper I are as follows:

$$I_{1j}=c_j^{(i)}+c_j^{(o)}, \tag{2.22}$$

$$I_{qj}=q_0\int ds^{(i)}\int ds_2 f_j^{(i)}=q_0c_j^{(i)}, \tag{2.23}$$

$$I_{xj}=\int ds^{(i)}v_x\int ds_2 f_j^{(i)}+\int ds^{(o)}v_x\int ds_2 f_j^{(o)}$$
$$=(c_j^{(i)}/\Delta s^{(i)}+c_j^{(o)}/\Delta s^{(o)})d(\mathrm{exp})/\pi^{1/2}, \tag{2.24}$$

in which $d(\mathrm{exp})$ stands for $(e^{-v_a^2}-e^{-v_b^2})$ as in earlier Papers [1–4], and where the $c_j^{(u)}$ are already calculated in Eq. (2.20). The general exact results for the important quantities are readily obtained from the procedure described above, but are too long to be usefully presented here [19]. However, interest lies in the special case of small laser excitation parameter q_0, that is, Eq. (2.19) of small a, and we now present the exact leading terms for this case [20]. (It turns out that the results for FP and CC geometries have the same form when written in this manner.)

$$\frac{d(\mathrm{erf})}{d(\mathrm{exp})}\frac{I_{xs}}{I_{qd}}=\frac{\Pi\alpha_{\mathrm{comb}}\Delta\alpha_t+\lambda\Pi\alpha_t\Delta\alpha_{\mathrm{comb}}}{\Pi\alpha_t\Pi\alpha_{\mathrm{comb}}}+O(|\tau\ln q_0|), \tag{2.25}$$

$$\pi^{-1/2}\frac{I_{1e}}{I_{qd}}=\frac{1+\lambda z\alpha_{et}}{z\alpha_{en}\alpha_{et}}+O(|\tau\ln q_0|), \tag{2.26}$$

$$I_{qd}/q_0=d(\mathrm{erf})/2+O(|\tau|), \tag{2.27}$$

where the parameters λ,τ are given, for the present case of FP geometry, by

$$\lambda_{\mathrm{FP}}=-1-(\sigma+2\ln a)/\pi, \tag{2.28}$$

$$\tau_{\mathrm{FP}}=q_0\ln q_0, \tag{2.29}$$

where $\sigma(\approx 0.58)$ is Euler's constant [1]. Thus, for FP geometry, unpleasant logarithmic behavior in the term of largest size, for small q_0, found also in Paper I, seems to persist in the present model. However, that logarithmic behavior in the present model may be negligible in practice, making its predictions very different from those of the model in Paper I, as discussed in Sec. IV below.

Results (2.25)–(2.29) were obtained by solving the system of equations (2.5), (2.20), and (2.21) exactly, and then expanding in terms of increasing order in the small parameter a (or q_0); this is perhaps the best procedure, because making small-a simplifications before solving the system is dangerous, easily leading to results that look reasonable but which are wrong. However, *a posteriori*, it turns out that Eqs. (2.25)–(2.29) are still obtained if terms of order larger than a are neglected in the system of equations before it is solved. The aH_a $(=a-aG_a)$ in Eq. (2.21) contains a term of the order of a and one of the order of $|a^2\ln a|$, and the second one may be safely neglected; that is, the aH_a may be safely replaced by a in Eq. (2.21). The G_a must be kept in Eq. (2.20), as it is of the order of $|a\ln a|$; it is helpful to replace G_a by ag_a with $g_a\equiv G_a/a$ after solution in order that the a may cancel nicely at simplification.

For completeness, exact expansions of the FP results with $\gamma=0$, both of the present model and of that in Paper I, including all terms of order smaller than $|a \ln a|$, are collected in Appendix A.

III. THE MODEL IN THE CONTEXT OF CC GEOMETRY

The analysis follows that for FP geometry in Sec. II, with changes as follows. The geometry is as described in Paper II, with cylindrical polar coordinates (v_x, V, ζ) used for the molecular velocity space. At normal momentum accommodation, molecules are redistributed into a Maxwellian distribution in V, and Eq. (2.1a) is replaced by

$$(s_1, s_2) = (\mathrm{erf}\, v_x, e^{-V^2}), \tag{3.1}$$

the third velocity component ζ being trivially integrated out as in Paper II. The bulk Maxwellian distribution $m(s_1, s_2)$ is again given by Eq. (2.1b). Now, in Sec. III, the v used in Section 2 stands for $V[=(-\ln s_2)^{1/2}]$ instead of $|v_z|$, with the Maxwellian average $\langle v \rangle_m = \pi^{1/2}/2$ instead of $\pi^{-1/2}$.

The frequency parameter ν of Sec. II B 1 is now [2] given by $\nu = 2V/\pi$, which means that the MBREs for CC geometry may be obtained from those (2.13) by means of the following replacements, in an obvious notation:

$$\dot{f}_j^{(u)} \to (\pi/2)\dot{f}_j^{(u)}, \tag{3.2a}$$

$$q_0 \to Q_0 \equiv (\pi/2) q_0, \tag{3.2b}$$

$$\gamma \to \Gamma \equiv (\pi/2)\gamma. \tag{3.2c}$$

The replacement of Eq. (3.2b) is also made in Eqs. (2.15) and (2.16). In Eq. (2.18), the replacement

$$a \to A \equiv (\pi/2) a \tag{3.3}$$

must be made, with A/Q_0 $(=a/q_0)$ given via Eq. (2.19).

Results (2.20) and (2.21) undergo more severe changes because of the difference between the substitutions (2.1a) and (3.1) and the difference between the values of $\langle v \rangle_m$ $(=\pi^{1/2}/2$ now); we get [21]

$$c_j^{(u)}/\Delta s^{(u)} = A_j^{(u)}/C + \pi^{1/2} X_j^{(u)} A H_A, \tag{3.4}$$

$$C_j^{(u)}/\Delta s^{(u)} = A_j^{(u)}/C + 2\pi^{-1/2} X_j^{(u)} A (1 - \pi^{1/2} A H_A). \tag{3.5}$$

The "final" results (2.25)–(2.27) remain of the same form [20], with the parameters λ, τ given for CC geometry by

$$\lambda_{\mathrm{CC}} = \pi/2 - 1, \tag{3.6}$$

$$\tau_{\mathrm{CC}} = q_0. \tag{3.7}$$

Thus, for CC geometry, logarithmic behavior in the term of largest size is pleasingly absent, as was the case in Paper II.

Remarks analogous to those made at the end of Sec. II, concerning safe simplifications in the system of equations before it is solved, may be made now: the AH_A in Eq. (3.4) and that in Eq. (3.5) may be safely replaced by A and zero, respectively.

For completeness, exact expansions of the CC results with $\gamma=0$, both of the present model and of that in Paper II, including all terms of order smaller than a, are collected in Appendix A.

IV. DISCUSSION AND CONCLUSION

It is clear from Eq. (2.25) that I_{xs}/I_{qd} does in fact depend on both $\Delta\alpha_t$ and $\Delta\alpha_n$, on the latter through $\Delta\alpha_{\mathrm{comb}}$ Eq. (2.9b). However, the dependence on $\Delta\alpha_n$ is likely to be negligible in the context of current experimental capability for the following reason. With the molecule-surface systems in current use, the α_{jk} are sure to be close to unity [7–10,22], which implies that the $\Delta\alpha_k$ and $(1-\alpha_{jk})$ are "small." In fact [1,2,7–10], $|\Delta\alpha_k|$ is probably no larger than about 10^{-2} and $(1-\alpha_{jk})$ no larger than about 10^{-1}. This in turn implies that $|\Delta\alpha_{\mathrm{comb}}|$ is probably no larger than about 10^{-3} (indeed, $\Delta\alpha_{\mathrm{comb}}=0$ if either of α_{gk} and either of α_{ek} are equal to unity). It follows that the $\Delta\alpha_{\mathrm{comb}}$ term may be safely neglected in Eq. (2.25) provided that λ is not too large. Of course, λ_{CC} (≈ 0.57) is of the order of unity, but λ_{FP} would be large for sufficiently small a. With the α_{jk} close to unity, Eq. (2.19) which relates a to q_0 may be safely [23] simplified to read

$$a \approx 2 q_0, \tag{4.1}$$

just as before, [1,2,24]. It follows that, where a_1 stands for the parameter a used in Paper I, we have $a \approx a_1$ in practice. It turns out from experiment [1] that $a \approx 10^{-2}$, giving λ_{FP} (≈ 1.7) also of the order of unity. For the $\Delta\alpha_{\mathrm{comb}}$ term to be of the same order as the $\Delta\alpha_t$ term in FP geometry, λ_{FP} must probably be of the order of 10, implying that a is less than about 2×10^{-8}, that is [1] the laser radiation intensity r^* absorbed by the gas is of the order of nW/mm^2 or less, which is entirely outside, by a factor of the order of 10^6, the range of current experimental capability (in current experiments [7–11], r^* is of the order of mW/mm^2). Thus, in the context of current experiments, Eq. (2.25) may be safely [23] simplified to read

$$\frac{I_{xs}}{I_{qd}} \approx \frac{d(\exp)}{d(\mathrm{erf})} \Delta\alpha_t, \tag{4.2}$$

for both FP and CC geometries.

Now we relate results here to those of Papers I and II. It follows from Eq. (2.25), with the $\Delta\alpha_{\mathrm{comb}}$ term neglected, and the analogous result from Paper I, which comes from Eqs. (5.5c) and (5.9b) therein, that, in an obvious notation,

$$\left(\frac{\Delta\alpha_t}{\Delta\alpha}\right)_{\mathrm{FP}} \approx \frac{\alpha_{gt}\alpha_{et}}{\alpha_g \alpha_e} \frac{(-2\ln a_1 - \sigma)}{\pi}, \tag{4.3}$$

where a_1 is given from [1]

$$\frac{a_1}{q_0}=\frac{(\alpha_g+\alpha_e)}{\alpha_g\alpha_e}, \tag{4.4}$$

where $\Delta\alpha,\alpha_g,\alpha_e$ refer to the quantities in Paper I. The analog of Eq. (4.3) for CC geometry is

$$\left(\frac{\Delta\alpha_t}{\Delta\alpha}\right)_{\rm CC}\approx\frac{\alpha_{gt}\alpha_{et}}{\alpha_g\alpha_e}\frac{\pi}{2}. \tag{4.5}$$

For FP geometry, it now follows, using Eqs. (6.8) and (6.9) of Paper I with Eq. (4.3), that $\Delta\alpha_t$ here is related to $\widetilde{\Delta\alpha}$, which stands [1] for $\Delta\alpha_t$ quoted in previous work, for example, Ref. [9], by

$$(\Delta\alpha_t/\widetilde{\Delta\alpha})_{\rm FP}\approx(1+\varepsilon)\left[\ln(2X)-\frac{1}{2}\right]\Big/\ \pi, \tag{4.6}$$

where we have set the α_{jk} to unity on the right-hand side (RHS). The analog of Eq. (4.6) for CC geometry is

$$(\Delta\alpha_t/\widetilde{\Delta\alpha})_{\rm CC}\approx 1. \tag{4.7}$$

The explanation for the peculiar-looking RHS of Eq. (4.6) is that it is the ratio, $\tilde{\kappa}_{\rm FP}/\kappa_{\rm FP}$, of two free-molecule-flow parameters [25]: in Paper I, $\kappa_{\rm FP}$ was chosen as $2\pi^{1/2}/[\ln(2X)-\frac{1}{2}]$, where X is the dimensionless cell length, whereas, in Ref. [9], for example, $\tilde{\kappa}_{\rm FP}$ was chosen as $2(1+\varepsilon)/\pi^{1/2}$, where ε is "a small correction" depending on the dimensionless cell width Y. Peculiar-looking quantities are absent from the RHS of Eq. (4.7), consistently with both $\kappa_{\rm CC}$ in Paper II and $\tilde{\kappa}_{\rm CC}$ in Ref. [9], for example, having been chosen as $3\pi^{1/2}/4$.

A conclusion is that the exact calculations that are made here may well be unnecessary in order to interpret existing experimental results, except perhaps in helping to put earlier work on a firmer footing, and this seems to be most clearly evident from Eq. (4.7), which indicates that the previous calculations, giving $\widetilde{\Delta\alpha}$ for the difference between the TMACs, are adequate.

In the same context of existing experimental work, our calculations indicate that $\Delta\alpha_n$ may, after all, play no measurable part in SLID. This is an interesting and important point in view of the fact that, under other conditions, it has been claimed [5,11] that $\Delta\alpha_n$ does play an important part.

It would, of course, be very interesting if SLID experiments, which need the full form of Eq. (2.25) for their interpretation, could be done, for example, using systems in which the α_{jk} are not close to unity [22]. The difference between the forms of $\lambda_{\rm FP}$ and $\lambda_{\rm CC}$ clearly emphasizes the conclusion [2] that CC geometry is more preferable than FP geometry for SLID experiments.

As with the models [1–4] presented in Papers I–IV, numerical results may be obtained by iteration of the analytical steady-state equations, and, independently, by integration of the MBREs with respect to time, as thoroughly described in Paper I.

The models that have been presented here are physically quite different from the earlier models (Papers I and II) [1,2]. For example, it is not possible to choose the four ACs α_{jk} in the present models in order to reproduce the earlier ones. As given in Table I, molecule-surface collisions of types N,T,NT occur with respective probabilities $\alpha_{jn\rm mod}$, $\alpha_{jt\rm mod}$, $\alpha_{jn}\alpha_{jt}$ here, as opposed to respective probabilities $0,0,\alpha_j$ in Papers I and II; it is worth emphasizing that the choice $\alpha_{jn}=\alpha_{jt}=\alpha_j$ here does not reproduce earlier models, as it give the respective probabilities $\alpha_j(1-\alpha_j)$, $\alpha_j(1-\alpha_j)$, α_j^2. It is for future experiments to decide which of the two sets of models is more realistic physically, and, more importantly, whether or not either set is adequate.

Although a clear distinction between the NMACs and the TMACs, as is made here, renders the treatment relatively straightforward, it should be borne in mind that the various accommodation coefficients are in fact related; that is, they may be considered as being different weighted averages of the same relaxation probability function. These considerations are discussed in the theory presented in Ref. [26], a good summary of which is given in Ref. [27].

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APPENDIX A: EXACT EXPANSIONS, FOR $\gamma=0$ AND FOR SMALL q_0, OF THE RESULTS FROM THE MODELS HERE AND FROM THOSE IN PAPERS I AND II

The expansions of I_{qd}/q_0 are given by Eq. (2.27) in all cases. The parameter a used in Paper I is named a_1 here and is given from Eq. (4.4). The a used in Paper II could be named a_2 here, with [24] $a_2=(\pi/2)a_1$. However, we do not need to introduce a_2 because, in the results below, we are able to use the result

$$a_2\ln a_2=(\pi/2)a_1\ln a_1+O(q_0). \tag{A1}$$

Where $(e,p)\equiv$ (earlier,present), the expansions of the quantity Q, defined by

$$Q\equiv\pi^{-1/2}I_{1e}/I_{qd}, \tag{A2}$$

may be written, in an obvious notation, as follows:

$$\alpha_e Q^{({\rm FP}_e)}=\frac{-2\ln a_1}{\pi}+\left(\frac{1}{z}-1-\frac{\sigma}{\pi}\right)+\frac{4}{\pi^{3/2}}a_1\ln^2 a_1 + O(|q_0\ln q_0|), \tag{A2a}$$

$$\alpha_{\rm en}Q^{(\mathrm{FP}p)}=\frac{-2\ln a}{\pi}+\left(\frac{1}{z\alpha_{et}}-1-\frac{\sigma}{\pi}\right)+\frac{4}{\pi^{3/2}}a\ln^2 a + O(|q_0\ln q_0|), \tag{A2b}$$

$$\alpha_e Q^{(\mathrm{CC}e)}=\left(\frac{1}{z}+\frac{\pi}{2}-1\right)+\frac{\pi^{3/2}}{2}a_1\ln a_1+O(q_0), \tag{A2c}$$

$$\alpha_{\rm en}Q^{(\mathrm{CC}p)}=\left(\frac{1}{z\alpha_{et}}+\frac{\pi}{2}-1\right)+\frac{\pi^{3/2}}{2}a\ln a+O(q_0). \tag{A2d}$$

We define quantities $\delta\alpha_\xi$ by

$$\delta\alpha_\xi=\Delta\alpha_\xi/\Pi\alpha_\xi, \tag{A3}$$

where $\xi\equiv(t,\mathrm{comb})$ in the present models and where ξ is absent in the earlier ones, when $\Delta\alpha\equiv(\alpha_g-\alpha_e)$ and $\Pi\alpha\equiv\alpha_g\alpha_e$. The expansions of the quantity R, defined by

$$R\equiv\frac{d(\mathrm{erf})}{d(\exp)}\frac{I_{xs}}{I_{qd}}, \tag{A4}$$

may be written as follows:

$$R^{(\mathrm{FP}e)}=\frac{(-2\ln a_1-\sigma)}{\pi}\delta\alpha+\frac{4}{\pi^{3/2}}\delta\alpha a_1\ln^2 a_1+O(|q_0\ln q_0|), \tag{A5a}$$

$$R^{(\mathrm{FP}p)}=-\frac{2}{\pi}\delta\alpha_{\rm comb}\ln a+\left[\delta\alpha_t-\left(1+\frac{\sigma}{\pi}\right)\delta\alpha_{\rm comb}\right]+\frac{4}{\pi^{3/2}}\delta\alpha_{\rm comb}a\ln^2 a+O(|q_0\ln q_0|), \tag{A5b}$$

$$R^{(\mathrm{CC}e)}=\frac{\pi}{2}\delta\alpha+\frac{\pi^{3/2}}{2}\delta\alpha a_1\ln a_1+O(q_0), \tag{A5c}$$

$$R^{(\mathrm{CC}p)}=\left[\delta\alpha_t+\left(\frac{\pi}{2}-1\right)\delta\alpha_{\rm comb}\right]+\frac{\pi^{3/2}}{2}\delta\alpha_{\rm comb}a\ln a+O(q_0). \tag{A5d}$$

The similarities and differences among results (A2) and (A5) are obvious and somewhat pleasing. The results for FP_e are consistent with, and extend to a higher order, the results obtained from Eq. (5.5c) and (5.9) of Paper I, and those for CC_e are consistent with, and again extend to a higher order, those obtainable from Eq. (3.2) of Paper II. The FP_p and CC_p results are consistent with, and extend to a higher order, results (2.25)–(2.29), (3.6) and (3.7) here.

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[23] Of course, there may always be nagging doubts in cases such as this; the best procedure is probably to try to check each case on its own merits.
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cause of his having used the same symbols a and g in both Papers I and II; those in Paper II are equal to $\pi/2$ times those in Paper I.

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