Stationary states and their stability for a mixture of molecular gases under velocity-selective excitation

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We consider a mixture of resonant two-level molecules under velocity-selective excitation and a buffer gas. The corresponding kinetic equations are augmented by a strong-collision model with free parameters. We obtain the equilibrium velocity distribution functions of the resonant and buffer components. We then use the Boltzmann kinetic equations to derive the H-theorem analog for this mixture (valid for arbitrary values of the collision rates ν and the spontaneous decay rate γ) and discuss the implications for the stability of the stationary solutions. [S1050-2947(96)07010-2]

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I. INTRODUCTION

For more than a decade, kinetic phenomena arising in gases under velocity-selective excitation have been the subject of many studies (see, in particular, Refs. [1–10]). When the momentum relaxation rates ν_e for the excited and ν_g for the ground-state molecules are different, the macroscopic state of the gas changes entirely. Macroscopic gas flows can arise, as well as pressure and temperature anisotropy and spatial temperature and density nonuniformity. Recently, the possibility of the existence of nonstationary regimes and the oscillatory dynamics in these conditions has been discussed [11,12]. These studies make the problem of the stability of the stationary solutions obtained under velocity-selective excitation quite important. This problem has been partially resolved for a single-component resonant gas in the case when the rate of spontaneous relaxation γ is much higher than the frequency ν of velocity-changing collisions, $\gamma \gg \nu$. In this situation, it is possible to show [4,5] that a relation similar to the usual H theorem [13] holds, which provides a criterion of stability. However, for the case $\gamma \leqslant \nu$, the problem remains unsolved. Meanwhile, the latter situation is most typical for molecular vibrotational transitions where many of the experiments on the velocity-selective excitation are being done. The main purpose of the present work is to provide a consistent description of the stationary state of a mixture of a resonant molecular gas and a buffer gas and derive a criterion for its stability.

Many different approaches have been used to model the collision kernel in the Boltzmann kinetic equations to describe the kinetic effects occurring due to velocity-selective excitation. Besides the strong-collision model [3], those include, in particular, the Keilson-Storer model [7] and the Bhatnagar-Gross-Krook model [9]. In the present study we use the model of strong collisions with free parameters (the FPSC model) [4,5]. A particular reason why, in spite of the availability of results obtained in more sophisticated models [7,9], the FPSC model seems to be worth examining is its physical simplicity. Thus results obtained in this model can provide a basis and a test for any approximate and qualitative derivations.

Light-induced kinetic phenomena are very sensitive to the changes in the collisional cross sections and can therefore be used to study the effect of external fields on the molecular collisions. Also, recently it has been demonstrated that light-induced kinetic effects can play a significant role in astrophysical processes [14], where gravitation is important. In these situations, external fields could affect the resulting spatial distributions of particles. Thus it is important to take into account also the possible acceleration of molecules in an external field. In order to further broaden the applicability of our results, we take into consideration the quenching collisions and collisions in which molecules are promoted into the excited state, in addition to the elastic collisions.

The organization of the paper is as follows. In Sec. II we introduce the Boltzmann kinetic equations and the collision model and then we obtain the equilibrium velocity distribution functions of the resonant and buffer components. In Sec. III we use the above kinetic equations to derive the H-theorem analog for the case $\gamma \lesssim \nu$ and discuss the implications for the stability of the stationary solutions. In Sec. IV we give a brief conclusion.

II. KINETIC EQUATIONS AND THE MODEL OF STRONG COLLISIONS WITH FREE PARAMETERS

In the present work we use the FPSC model [4,5]. In this model, it is assumed that collisions shape the velocity distributions $f_e(\mathbf{v})$ of excited molecules and $f_g(\mathbf{v})$ of nonexcited resonant molecules towards a nonequilibrium Maxwellian

$$W(\mathbf{v}) = \left(\frac{\alpha'}{\pi}\right)^{3/2} \exp[-\alpha'(\mathbf{v}-\mathbf{V}')^2]. \tag{1a}$$

Similarly, the velocity distribution $f_b(\mathbf{v})$ of the buffer gas is shaped towards another nonequilibrium Maxwellian

$$W_b(\mathbf{v})=\left(\frac{\alpha_b'}{\pi}\right)^{3/2}\exp[-\alpha_b'(\mathbf{v}-\mathbf{V}')^2]. \tag{1b}$$

Here the momentary values of the velocity shift and energy parameters $\mathbf{V}'$, α', and α_b' should be determined assuming particle-number, momentum, and translational energy conservation (neglecting the momentum and energy transferred from the radiation field). In this model, the Boltzmann kinetic equations are of the form

$$\begin{aligned}\frac{df_{g,e}}{dt}&\equiv\frac{\partial f_{g,e}}{\partial t}+\mathbf{v}\cdot\frac{\partial f_{g,e}}{\partial\mathbf{r}}+\mathbf{a}\cdot\frac{\partial f_{g,e}}{\partial\mathbf{v}}\\&=(\nu_{g,e}'N_{g,e}'+\nu_{q,T}'N_{q,T}')\left(\frac{\alpha'}{\pi}\right)^{3/2}e^{-\alpha'(\mathbf{v}-\mathbf{V}')^2}\\&\quad-[\nu_{g,e}'+\nu_{T,q}'+\gamma_{g,e}'+Q(\mathbf{v})]f_{g,e}\\&\quad+[\gamma_{e,g}'+Q(\mathbf{v})]f_{e,g}\end{aligned} \tag{2a}$$

$$\begin{aligned}\frac{df_b}{dt}&\equiv\frac{\partial f_b}{\partial t}+\mathbf{v}\cdot\frac{\partial f_b}{\partial\mathbf{r}}+\mathbf{a}\cdot\frac{\partial f_b}{\partial\mathbf{v}}\\&=\nu_bN_b'(\alpha_b'/\pi)^{3/2}e^{-\alpha_b'(\mathbf{v}-\mathbf{V}')^2}-\nu_bf_b.\end{aligned} \tag{2b}$$

Here $\mathbf{a}\equiv\mathbf{F}/m$ is the acceleration of a molecule in the external field, indices e and g correspond to the excited and the nonexcited resonant molecules, respectively, while the index b corresponds to the buffer-gas molecules. Equation (2a) contains a velocity-selective optical pumping term, with the velocity-dependent excitation rate

$$Q(\mathbf{v})\equiv\int BM(\omega)\{\Gamma^2/[\Gamma^2+(\omega-\omega_0-\mathbf{k}\cdot\mathbf{v})^2]\}d\omega$$

[where B is the Einstein coefficient, $M(\omega)$ is the laser spectral intensity distribution, ω_0 and Γ are the resonance frequency and homogeneous absorption linewidth, and ω is the radiation frequency], and the relaxation terms, where $\nu'_{g,e}(\mathbf{v})$ are the rates of velocity-changing collisions that proceed without the change in the internal state of the molecules, $\nu_q'(\mathbf{v})$ is the rate of quenching velocity-changing collisions, $\nu_T'(\mathbf{v})$ is the rate of velocity-changing collisions in which molecules are promoted to the excited state; $\gamma_e'(\mathbf{v})$ is the rate of decay of the excited state resulting from spontaneous emission and other processes that are not accompanied by the change in the molecular velocity (such as resonant energy transfer) and $\gamma_g'(\mathbf{v})$ is the rate of molecular excitation in the latter processes. Equation (2b) for the buffer gas has only the collision term on its right-hand side.

Distribution functions f_e, f_g, and f_b must satisfy the following conditions resulting from the laws of conservation of number of particles, momentum, and energy:

$$\int(f_g+f_e)d^3\mathbf{v}=N, \tag{3a}$$

$$\int\mathbf{v}\left[(f_g+f_e)+\frac{m_b}{m}f_b\right]d^3\mathbf{v}=\left(N+\frac{m_b}{m}N_b\right)\mathbf{V}; \tag{3b}$$

$$\int v^2\left[(f_g+f_e)+\frac{m_b}{m}f_b\right]d^3\mathbf{v}=N(3v_T^2+V^2)+\frac{m_b}{m}N_b(3v_{bT}^2+V_b^2), \tag{3c}$$

$$\int f_bd^3\mathbf{v}=N_b. \tag{3d}$$

Here N and N_b, $\mathbf{V}$ and $\mathbf{V}_b$, and $v_T\equiv(2\alpha)^{-1/2}$ and $v_{Tb}\equiv(2\alpha_b)^{-1/2}$ are the number densities, macroscopic velocities, and mean thermal velocities of the resonant molecules and the buffer-gas molecules, correspondingly, and α and α_b are the parameters of the Maxwell distributions of the resonant and buffer gases before the laser was switched on. In addition to the conditions Eqs. (3a)–(3d), parameters $N'_{g,e,b}$ and $N_{q,T}$ must satisfy the conditions of conservation of number of particles in collisions:

$$\int\nu'_{g,e,b}[N'_{g,e,b}W(\mathbf{v})-f_{g,e,b}]d^3\mathbf{v}=0, \tag{4a}$$

$$\int\nu'_{q,T}[N'_{q,T}W(\mathbf{v})-f_{e,g}]d^3\mathbf{v}=0. \tag{4b}$$

Thus, for *eight* free parameters ($N'_{g,e,b}$, $N_{q,T}$, $\mathbf{V}'$, α', and α_b') we have a system of *nine* self-consistency equations (3) and (4), which might therefore seem incompatible. However, if we add together the right-hand side and left-hand side of Eqs. (2a), integrate over velocities, and use the continuity equation, we obtain the equality

$$\begin{aligned}&\sum_{i=e,g}\int\nu_i'[N_i'W(\mathbf{v})-f_i]d^3\mathbf{v}\\&\quad+\sum_{i=e,g;\ j=q,T}\int\nu_j'[N_jW(\mathbf{v})-f_i]d^3\mathbf{v}\\&\quad=\frac{dN}{dt}\\&\quad=0.\end{aligned} \tag{5}$$

According to Eq. (5), Eqs. (4a) and (4b) are linearly dependent and thus the system of Eqs. (3) and (4) is compatible (the number of linearly independent equations is equal to the number of unknowns).

Equation (2a) yields the stationary, spatially homogeneous solution

$$f_{g,e}=\frac{(\nu'_{e,g}\nu'_{q,T}+Q)\nu'_{g,e}N'_{g,e}+(\gamma'_{e,g}+Q)(\nu'_{T,q}N'_{T,q}+\nu'_{e,g}N'_{e,g})+(\nu'_{e,g}+\nu'_{q,T}+\gamma'_{e,g}+Q)\nu'_{q,T}N'_{q,T}}{(\nu'_e+\nu'_q)(\nu'_g+\nu'_T)+(\nu'_e+\nu'_q)(\gamma'_g+Q)+(\nu'_g+\nu'_T)(\gamma'_e+Q)}\,W(\mathbf{v}). \tag{6}$$

In the case when

$$\frac{\nu'_q(\mathbf{v})}{\nu'_e(\mathbf{v})}=\text{const},\quad \frac{\nu'_T(\mathbf{v})}{\nu'_g(\mathbf{v})}=\text{const} \tag{7}$$

one can see that $N'_T=N'_g$ and $N'_g=N'_e$. In this case Eq. (6) yields

$$f_g=\frac{(1+\theta+\kappa)N'_g+\beta(1+\kappa)N'_e}{1+\theta+\beta\beta_\gamma+(1+\beta)\kappa}\,W(\mathbf{v}), \tag{8a}$$

$$f_e=\frac{(\beta_\gamma+\kappa)N'_g+(\beta\beta_\gamma+\theta+\beta\kappa)N'_e}{1+\theta+\beta\beta_\gamma+(1+\beta)\kappa}\,W(\mathbf{v}), \tag{8b}$$

where

$$\kappa\equiv\frac{Q(\mathbf{v})}{\gamma_e},\quad \beta\equiv\frac{\nu_e}{\nu_g},\quad \beta_\gamma\equiv\frac{\gamma_g}{\gamma_e},$$

$$\theta\equiv\frac{\nu_e}{\gamma_e}-\frac{\nu_g}{\gamma_e}-\beta\,\frac{\nu_T}{\nu_e}\equiv\frac{\nu_g\nu_e-\nu_g\nu_q-\nu_e\nu_T}{\nu_g\gamma_e}\equiv\frac{\nu^2}{\nu_g\gamma_e}$$

$$\nu_{g,e}\equiv\nu'_{g,e}+\nu'_{T,g},\quad \gamma_{e,g}\equiv\gamma'_{g,e}+\nu'_{T,q},\quad \nu_{T,q}\equiv\nu'_{T,q}.$$

Subsequent analysis in this section will be performed for the case when

$$\nu_{g,e,T,q}(\mathbf{v})=\text{const}. \tag{9}$$

If the conditions of Eq. (9) are met, Eqs. (4) yield

$$N'_{T,q}=N'_{g,e}=N_{g,e}\equiv\int f_{g,e}d^3\mathbf{v}, \tag{10}$$

where $N_{g,e}$ are the total number densities of the ground-state and excited molecules. Thus, in order to determine the constants $N'_{g,e}=N_{g,e}$ for the determination of $f_{g,e}$ from Eqs. (8a) and (8b), it is sufficient to find their ratio $n\equiv N_e/N_g$. From Eq. (6) we find

$$\int\frac{(\gamma_e+Q)N_e-(\gamma_g+Q)N_g}{\nu_g(\gamma_e+Q)+\nu_e(\gamma_g+Q)+\nu_g\nu_e-\nu_g\gamma_q-\nu_e\nu_T}\,W(\mathbf{v})d^3\mathbf{v}=0. \tag{11}$$

Equation (11) yields

$$\int\frac{(\beta-\theta-\beta\beta_\gamma)n+1+\theta-\beta_\gamma}{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa}}\,W(\mathbf{v})d^3\mathbf{v}=n-\beta_\gamma. \tag{12}$$

From Eq. (12),

$$\frac{1+\dfrac{\beta_\gamma}{n}}{\beta-\theta-\beta\beta_\gamma+\dfrac{1+\theta-\beta_\gamma}{n}}=\int\frac{W(\mathbf{v})}{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa}}\,d^3\mathbf{v}. \tag{13}$$

Therefore,

$$n=\frac{\beta_\gamma\left(1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa_0}\right)+(1+\theta-\beta_\gamma)\zeta}{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa_0}+(\beta\beta_\gamma+\theta-\beta)\zeta}, \tag{14}$$

where the x axis is collinear to the laser beam direction,

$$\kappa_0\equiv\frac{Q_0}{\gamma_e},\quad \int_{-\infty}^{\infty}\frac{(\alpha'/\pi)^{1/2}e^{-\alpha'(v_x-V'_x)^2}dv_x}{\nu_g+\nu_e+\dfrac{\nu_g\gamma_e+\nu_e\gamma_g+\nu^2}{Q(v_x)}}\equiv\frac{\zeta}{\nu_g+\nu_e+\dfrac{\nu_g\gamma_e+\nu_e\gamma_g+\nu^2}{Q_0}};$$

$$\zeta\equiv\int_{v_0-\Delta v/2}^{v_0+\Delta v/2}\sqrt{\alpha'/\pi}e^{-\alpha'(v_x-V'_x)^2}dv_{x'}$$

where v_0 is the center of the excited velocity interval and Δv is its width. Using the results of Eqs. (8) and (14), we can derive the following expression for the velocity distribution function of the resonant gas $f(\mathbf{v})\equiv f_e+f_g$:

$$f=N_{ef}[1-\phi(\mathbf{v})]W(\mathbf{v}),$$

$$\phi(\mathbf{v})\equiv\frac{(\beta-1)(1-\beta_\gamma)}{(1+\beta_\gamma)\left(1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa_0}\right)+2\theta\zeta}\times\frac{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa_0}}{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa(v_x)}}. \tag{15}$$

Here N_{ef} is a constant to be found from the self-consistency conditions (3).

Meanwhile, Eq. (2b) yields the following expression for the buffer-gas velocity distribution f_g:

$$f_b=N'_b\left(\frac{\alpha'_b}{\pi}\right)^{3/2}e^{-\alpha'_b(\mathbf{v}-\mathbf{V}')^2}. \tag{16}$$

In the case when $\nu_b(\mathbf{v})=\text{const}$, Eqs. (3d) and (16) yield

$$N_b' = N_b \equiv \int f_b d^3\mathbf{v}. \tag{17}$$

Substituting the result of Eq. (15) into the self-consistency conditions (3), we arrive at the equations

$$N_{ef}\left(1-\int \phi(\mathbf{v})W(\mathbf{v})d^3(\mathbf{v})\right)=N, \tag{18a}$$

$$\left(N_{ef}+\frac{m_b}{m}N_b\right)V' - N_{ef}\int \mathbf{v}\ \phi(\mathbf{v})W(\mathbf{v})d^3\mathbf{v} = \left(N+\frac{m_b}{m}N_b\right)\mathbf{V}, \tag{18b}$$

$$N_{ef}\left(\frac{3}{2\alpha'}+V'^2\right)+\frac{m_b}{m}N_b\left(\frac{3}{2\alpha_b'}+V'^2\right) - N_{ef}\int v^2\ \phi(\mathbf{v})W(\mathbf{v})d^3(\mathbf{v}) = N\left(\frac{3}{2\alpha}+V^2\right)+\frac{m_b}{m}N_b\left(\frac{3}{2\alpha_b}+V^2\right). \tag{18c}$$

According to Eqs. (18),

$$N_{ef}=\frac{N}{1-\epsilon}, \quad \mathbf{V}'=\frac{\mathbf{V}\left(N+\frac{m_b}{m}N_b\right)+N_{ef}V_\epsilon}{N_{ef}+\frac{m_b}{m}N_b}$$

$$\frac{1}{\alpha'}=\frac{2}{3}(N_{ef}+N_b)^{-1}\left[\frac{3}{2}(N+N_b)\frac{1}{\alpha}+\left(N+\frac{m_b}{m}N_b\right)V^2 - \left(N_{ef}+\frac{m_b}{m}N_b\right)V'^2+N_{ef}v_\epsilon^2\right].$$

Here

$$\epsilon \equiv \int \phi(\mathbf{v})W(\mathbf{v})d^3\mathbf{v}$$

is the relative area of the collision-induced ''dip'' in the velocity distribution function $f(\mathbf{v})$;

$$\mathbf{V}_\epsilon \equiv \int \mathbf{v}\ \phi(\mathbf{v})W(\mathbf{v})d^3\mathbf{v},$$

$$v_\epsilon^2 \equiv \int v^2\phi(\mathbf{v})W(\mathbf{v})d^3\mathbf{v}.$$

In the case when the excitation is nearly monochromatic and homogeneous broadening is small, so that $\Delta v \ll v_T$ and the light-induced effects are small ($\epsilon \ll 1$),

$$N_{\rm eff}=(1+\epsilon)N; \quad V'=\frac{N}{N+\frac{m_b}{m}N_b}\,\epsilon v_0;$$

$$\alpha'^{-1}=\alpha^{-1}+\frac{2}{3}\,\epsilon\,\frac{N}{N+N_b}\left(v_0^2-\frac{3}{2\alpha}\right). \tag{19}$$

where v_0 is the center of the excited velocity interval.

For spectroscopic applications, the value of $f_g - f_e$ that determines the profile of the absorption can be important. From Eqs. (8a) and (8b) we find

$$f_g - f_e = \frac{N(\alpha'/\pi)^{3/2}e^{-\alpha'(\mathbf{v}-\mathbf{V}')^2}}{1+\beta_\gamma+\dfrac{(\beta-1)(\beta_\gamma-1)+2\theta}{1+\beta-\dfrac{1+\theta+\beta\beta_\gamma}{\kappa_0}}\,\zeta} \times\left[1-\frac{1+\beta}{1+\beta+\dfrac{1+\theta+\beta\beta_\gamma}{\kappa(\mathbf{v})}}\right]. \tag{20}$$

Monitoring the absorption profile can provide an alternative way of searching for nonstationary regimes.

III. *H*-THEOREM ANALOG AND THE STABILITY OF STATIONARY STATES

In this section we will analyze the stability of the stationary solution. We will derive the criterion of stability that is valid not only for the systems with rapid spontaneous decay ($\gamma \gg \nu$) and a pure resonant gas [4,5], but for any γ and ν and a gas mixture. We start by introducing the analog of the gas entropy S_{ef},

$$S_{ef}=\int\int\left(f\ln\frac{e}{f_{ef}}+f_b\ln\frac{e}{f_b}\right)d^3\mathbf{v}\,d^3\mathbf{r}, \tag{21}$$

where

$$f \equiv f_g+f_e, \quad f_{ef}\equiv f_g+\beta f_e \equiv \frac{f}{\psi}, \quad \beta \equiv \frac{\nu_e}{\nu_g}.$$

Taking the time derivative of the first (resonant-gas related) term in S_{ef}, we obtain

$$\frac{dS_{ef}}{dt}=\int\int\left[\left(\mathbf{v}\cdot\frac{\partial f}{\partial \mathbf{r}}+\mathbf{a}\cdot\frac{\partial f}{\partial \mathbf{v}}\right)\ln\frac{f}{\psi}+f\frac{\partial\ln\psi}{\partial t} - St_{g+e}\ln\frac{f}{\psi}\right]d^3\mathbf{v}\,d^3\mathbf{r}. \tag{22}$$

Here

$$St_{g+e}\equiv \nu_g[N_{ef}W'-f_{ef}].$$

The first two terms on the right-hand side of Eq. (22) can be transformed using the following relations:

$$\int\int \mathbf{v}\cdot\frac{\partial f}{\partial \mathbf{r}}\ln\frac{f}{\psi}\,d^3\mathbf{v}\,d^3\mathbf{r}=\int\int\left[-\frac{\partial}{\partial\mathbf{r}}\cdot\left(\mathbf{v}f\ln\frac{e\psi}{f}\right)+f\mathbf{v}\cdot\frac{\partial\ln\psi}{\partial\mathbf{r}}\right]d^3\mathbf{v}\,d^3\mathbf{r}, \tag{23a}$$

$$\int\int \mathbf{a}\cdot\frac{\partial f}{\partial \mathbf{v}}\ln\frac{f}{\psi}\,d^3\mathbf{v}\,d^3\mathbf{r}=\int\int\left[-\frac{\partial}{\partial\mathbf{v}}\cdot\left(\mathbf{a}f\ln\frac{e\psi}{f}\right)+f\left(\ln\frac{e\psi}{f}\right)\frac{\partial\mathbf{a}}{\partial\mathbf{v}}+f\mathbf{a}\cdot\frac{\partial\ln\psi}{\partial\mathbf{v}}\right]d^3\mathbf{v}\,d^3\mathbf{r}. \tag{23b}$$

The last term on the right-hand side of Eq. (22) can be represented as

$$St_{g+e}\ln\frac{f}{\psi}=-\nu_q N_{ef}W'(Y'-1)\ln Y'+\nu_g[f_{ef}-N_{ef}W_{ef}]\times\{\ln[N_{ef}(\alpha'/\pi)^{3/2}]-\alpha'(\mathbf{v}-\mathbf{V}')^2\},\qquad(24)$$

with

$$Y'\equiv\frac{f_{ef}}{N_{ef}\left(\frac{\alpha'}{\pi}\right)^{3/2}e^{-\alpha'(\mathbf{v}-\mathbf{V}')^2}}.$$

The buffer-gas-related term in S_{ef} can be transformed in the same fashion. Using the Gauss theorem and assuming particle-number, momentum, and energy conservation in the collisions (3), we finally arrive at the H-theorem analog

$$\begin{aligned}\frac{dS_{ef}}{dt}=&\int\int f_{ef}\frac{d\psi}{dt}\,d^3\mathbf{v}\,d^3\mathbf{r}+\int\int\left(\frac{\partial}{\partial\mathbf{v}}\cdot\mathbf{a}\right)\psi f_{ef}\\&\times\ln\frac{e}{f_{ef}}\,d^3\mathbf{v}\,d^3\mathbf{r}-\int\mathbf{G}_{ef}d^2\mathbf{r}_\sigma\\&+\int\int\left(\frac{\partial}{\partial\mathbf{v}}\cdot\mathbf{a}\right)f_b\ln\frac{e}{f_b}\,d^3\mathbf{v}\,d^3\mathbf{r}-\int\mathbf{G}_b d^2\mathbf{r}_\sigma\\&+\int\int\nu_g N'\left(\frac{\alpha'}{\pi}\right)^{3/2}e^{-\alpha'(\mathbf{v}-\mathbf{V}')^2}(Y'-1)\\&\times\ln Y'\,d^3\mathbf{v}\,d^3\mathbf{r}+\int\int\nu_b N_b'\left(\frac{\alpha_b'}{\pi}\right)^{3/2}\\&\times e^{-\alpha_b'(\mathbf{v}-\mathbf{V}')^2}(Y_b'-1)\ln Y_b'\,d^3\mathbf{v}\,d^3\mathbf{r},\end{aligned}\qquad(25)$$

where

$$\mathbf{G}_{ef}\equiv\int\mathbf{v}f\ln\frac{e}{f_{ef}}\,d^3\mathbf{v},\quad\mathbf{G}_b\equiv\int\mathbf{v}f_b\ln\frac{e}{f_b}d^3\mathbf{v}$$

are the flows of ''entropy density''; $d^2\sigma$ is an element of the surface surrounding the gas;

$$Y_b'\equiv\frac{f_b}{N_b'\left(\frac{\alpha'}{\pi}\right)^{3/2}e^{-\alpha_b'(\mathbf{v}-\mathbf{V})^2}}.$$

For many force fields (for example, Lorentz force), $\partial F_i/\partial v_i=0$, so that the terms in Eq. (25) that contain $(\partial\mathbf{a}/\partial\mathbf{v})$ often vanish.

For a common (nonexcited) gas there is only one potential source for the instability of the stationary solution: entropy flux through the walls of the cell containing the gas. When the gas is excited in a velocity-selective fashion, then, according to Eq. (25), the nonzero flux of entropy will arise in a broader range of physical conditions than without the excitation. In particular, without the excitation there can be no flux through the walls that reflect the gas molecules in a specular fashion. In contrast, according to Eq. (25), in order for the entropy flux in an excited gas to be zero, one needs, even in the case of specular surface scattering, also the symmetry of ψ with respect to v_x. Besides, under velocity-selective excitation new sources of instability arise: spatial inhomogeneity of the radiation $(\partial\psi/\partial\mathbf{r}\neq\mathbf{0})$ and the external force fields $(\mathbf{a}\partial\psi/\partial\mathbf{v}\neq0)$. Thus it follows from Eq. (25) that in the case of vanishing of the flow of ''entropy'' through the cell surface, the absence of external fields, and $d\psi/dt=0$, the stationary solution is stable against any perturbations. If those conditions are not met, the problem of the stability of the stationary solution remains open.

Although the physics of surface-induced effects arising under velocity-selective excitation [11,15–21] is quite different from the physics of the ''bulk'' effects [1–10], the former can also be described using the strong-collision model [15]. Thus the present results can be applied also to analyze those effects. As we see, the H-theorem analogy derived here limits the scope of situations where the instabilities can arise and therefore provides guidance for their search.

IV. CONCLUSION

In conclusion, we have considered the use of the strong-collision model with free parameters to describe the kinetics of the mixture of the resonant and buffer gases excited in a velocity-selective fashion. We have obtained the equilibrium velocity distribution functions of the resonant and the buffer components. We have used the Boltzmann kinetic equations to derive the H-theorem analog for this mixture in the case when $\gamma\leqslant\nu$ and have discussed the implications for the stability of the stationary solutions.

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