Oscillatory dynamics in light-induced drift

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The light-induced drift velocity of optically active atoms immersed in a buffer gas sometimes changes sign at some temperature. It is pointed out that the density of the active atoms and the temperature of the system form a nonlinear dynamical system that may exhibit instabilities. These may show up as an oscillatory behavior of these quantities as a function of time. A possible realization in the case of alkali-metal atoms is discussed.

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I. INTRODUCTION

Light-induced drift (LID) is a mechanical action of resonant light on optically active gas particles immersed in a buffer gas [1]. It arises when these active particles undergo both velocity-selective excitation by the light and state-dependent friction by the buffer gas. It has been extensively studied both experimentally [2–4] and theoretically [5,6]. These investigations mainly concerned the particle drift in nearly stationary conditions. Since it has been noted that the macroscopic equations describing LID are essentially nonlinear [7], one may expect nontrivial dynamical behavior of a gas mixture when the effect is operative. In fact, dramatic effects of LID such as the optical piston [5,8] or dissipative solitons [6,9] are a direct result of nonlinear behavior.

In this paper we show that LID can give rise to a nonlinear coupling of the density and the temperature of a molecular gas excited by infrared radiation. We demonstrate that this coupling may give rise to an oscillatory behavior of the concentration of the absorbing vapor and the temperature of the gas mixture as a whole. Qualitatively, this behavior is possible near a critical temperature T_c, where the drift velocity u changes sign. Such a sign change is known to occur for some alkali-metal–noble-gas mixtures [10]. In general, one might expect to create such a sign change when one takes for the buffer gas a mixture of two gases at appropriate proportions, where each component separately would lead to a drift velocity of opposite sign. In the neighborhood of this critical temperature we may write

$$u=\alpha(T-T_c)\ . \tag{1}$$

In this temperature region, also the frequency dependence of the drift velocity behaves in an anomalous fashion [11,12], as has been observed for C_2H_4 at room temperature [13].

We restrict ourselves to a one-dimensional configuration, where the physical quantities only vary in the x direction, which is the direction of the drift velocity u. The particle density n of the absorbing vapor obeys the continuity equation

$$\frac{\partial}{\partial t}n+\frac{\partial}{\partial x}j=0\ , \tag{2}$$

where the particle flow density j is given by

$$j=un-D\frac{\partial}{\partial x}n\ , \tag{3}$$

with D the diffusion coefficient for the absorbing particles in the buffer gas. Strictly speaking, the diffusion coefficient may be slightly affected by the difference in gas-kinetic cross sections for the two states of the active particles [14]. This effect can safely be neglected in the neighborhood of the critical temperature. When the temperature of the irradiated gas is equal to the critical temperature T_c, the drift velocity u is zero, and a stationary solution of Eq. (2) occurs for a uniform density n_0. We introduce δn as the (x-dependent) deviation of the density n from the value n_0. In a molecular gas, the absorbed radiation energy is dissipated in the gas mainly by collisions. In a region where δn is positive, the absorbed energy is larger than in the stationary case, so that the temperature will rise, whereas the gas is cooled in regions where δn is negative. Since according to Eq. (1) the drift velocity u depends on T, the density and the temperature form a pair of coupled dynamical variables. In this paper we wish to investigate the stability of this system.

II. MODEL DESCRIPTION

The physical sample we consider is an elongated capillary cell along the x direction. The cell contains a dilute vapor of optically active molecules immersed in a buffer gas. A beam of light traverses the cell, and light-induced drift occurs. The temperature is close to T_c, so that the

drift velocity is well described by Eq. (1). We assume that the heat exchange between the gas in the cell and the cell wall is rapid, so that they can be described by the same temperature $T(x)$. The cell can exchange heat with its surroundings, which is kept at a fixed temperature T_s. The equation for the temperature T follows from the heat balance of the cell. We denote as E the internal energy of the cell per unit length, and q is the one-dimensional heat flow, which is the heat that passes a given position x of the cell per unit time. (Note that the combined system of gas mixture and cell wall is simply called the cell.) The internal energy E is supposed to vary linearly with the temperature, so that we may write

$$E = CT \ , \tag{4}$$

where C is the heat capacity per unit length. The heat flow q obeys an effective one-dimensional heat conductivity equation

$$q = -\kappa \frac{\partial}{\partial x} T \ . \tag{5}$$

Furthermore, heat is created in the cell by absorption of the light intensity, and heat is exchanged with the surroundings. The net heat balance equation can be put in the simple form

$$\frac{\partial}{\partial t} E = -\frac{\partial}{\partial x} q + pn\sigma I - b(T - T_s) \ , \tag{6}$$

where the last term describes the transfer of heat per unit length from the cell to its surroundings. The third term indicates the heat production in the gas by absorption of light. Here I is the power of the light passing a given position x, and σ is the absorption cross section. The constant p denotes the fraction of the absorbed light energy that is turned into heat. We consider the situation of an optically thin system, so that in Eq. (6) the light power I may be taken as independent of x.

The two dynamical variables n and T are found to obey coupled nonlinear equations of motion. The diffusion equation for n follows from Eqs. (1)–(3), and we obtain

$$\frac{\partial}{\partial t} n = D \frac{\partial^2}{\partial x^2} n - \alpha \frac{\partial}{\partial x} (T - T_c) n \ . \tag{7}$$

The equation for T follows from Eqs. (4)–(6):

$$C \frac{\partial}{\partial t} T = \kappa \frac{\partial^2}{\partial x^2} T + pn\sigma I - b(T - T_s) \ . \tag{8}$$

A typical time scale is determined by the damping rate for the temperature, and a length scale follows then from the diffusion. We introduce a dimensionless time τ and length ξ by the definitions

$$\tau = t \frac{b}{C} \ , \quad \xi = x \left[\frac{b}{CD} \right]^{1/2} \ . \tag{9}$$

Then the effect of light-induced drift on the diffusion equation is measured by the reduced dimensionless temperature

$$\Theta = \alpha (T - T_c) \left[\frac{C}{bD} \right]^{1/2} \ . \tag{10}$$

Furthermore, we introduce the reduced density of active particles

$$N = \frac{n}{n_0} \ , \tag{11}$$

with n_0 the equilibrium density in the absence of light. Equations (7) and (8) take the reduced form

$$\frac{\partial}{\partial \tau} N = \frac{\partial^2}{\partial \xi^2} N - \frac{\partial}{\partial \xi} (N\Theta) \ , \tag{12}$$

$$\frac{\partial}{\partial \tau} \Theta = \chi \frac{\partial^2}{\partial \xi^2} \Theta + \varepsilon N - (\Theta - \Theta_s) \ . \tag{13}$$

In Eq. (13) we introduced the reduced thermometric conductivity

$$\chi = \frac{\kappa}{CD} \ , \tag{14}$$

and the reduced intensity

$$\varepsilon = p\sigma I \alpha n_0 \left[\frac{C}{b^3 D} \right]^{1/2} \ . \tag{15}$$

The parameter Θ_s is the reduced temperature of the surrounding. Equations (12) and (13) describe the coupled dynamics of the density of the active particles and the temperature of the vapor cell.

III. STABILITY ANALYSIS

A. Analytical treatment for long cell

An obvious homogeneous stationary solution of Eqs. (12) and (13) is given by

$$N = N_0 \equiv 1 \ , \quad \Theta = \Theta_0 \equiv \Theta_s + \varepsilon \ . \tag{16}$$

When this solution applies, the temperature of the cell is higher than the temperature of the surrounding by an amount $p\sigma I n_0 / b$. Hence the drift velocity does not vanish in general. This implies that the solution can only exist in an infinite cell, or in a cell consisting of a closed loop in which the light traverses a finite rectilinear arm, as sketched in Fig. 1. We consider the case that the stationary temperature is equal to the critical temperature,

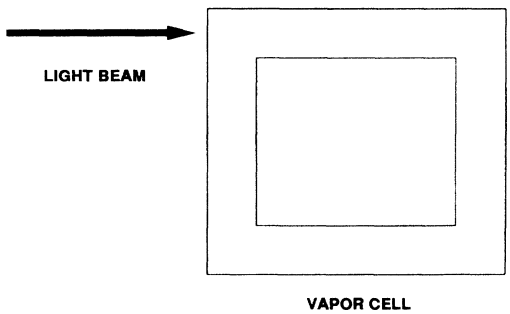

FIG. 1. Sketch of a cell in the form of a closed loop. In this way, an open cell with a finite effective length is realized, where the drift velocity does not have to vanish in the steady state.

so that $\Theta_0=0$. We wish to analyze the stability of this stationary solution in the case of a cell with a length L that is large compared with the characteristic length $\sqrt{CD/b}$. For this purpose we take a trial solution with a small deviation from the stationary density and temperature proportional to $\exp(iK\xi-i\Omega\tau)$, so that instability occurs when Ω has a positive imaginary part. Hence we seek solutions for N and Θ of the general form $p=p_0+p_1\exp(iK\xi-i\Omega\tau)$, and we linearize in the two amplitudes N_1 and Θ_1. After elimination of these amplitudes, we arrive at the dispersion relation between the reduced frequency Ω and the reduced wave number K:

$$\Omega^2+i\Omega[K^2(\chi+1)+1]-K^2(1+\chi K^2)-iK\varepsilon=0 . \tag{17}$$

For a given real value of K, this yields two complex solutions for $\Omega=\Omega'+i\Omega''$. We assume that $K\ll 1$, which implies that the deviation from the stationary solution varies little over the characteristic length $\sqrt{CD/b}$. We expand the solutions to fourth order in K. We introduce a characteristic reduced wave number K_c, defined by

$$K_c^2=\frac{\varepsilon^2-1}{3\chi+2} , \tag{18}$$

and consider the situation where $K_c^2\ll 1$. Then Eq. (17) has a solution for Ω given by

$$\begin{aligned}\Omega'&=\varepsilon K[1-K^2(1+\chi)] ,\\ \Omega''&=-\varepsilon^2(3\chi+2)K^2(K^2-K_c^2) .\end{aligned} \tag{19}$$

Instability of the stationary solution will occur when Ω'' is positive. Hence the instability condition is

$$|K|<K_c \quad \text{and} \quad \varepsilon^2>1 . \tag{20}$$

The behavior of the instability parameter Ω'' as a function of K is sketched in Fig. 2.

For a finite cell, when Ω'' is positive, it is possible that a finite wave packet grows in time, while moving out of the cell. This is called convective instability [15]. Here we restrict study to the stronger case of absolute instability, which occurs when a wave packet increases at every point in the cell. As explained in Ref. [15], the system

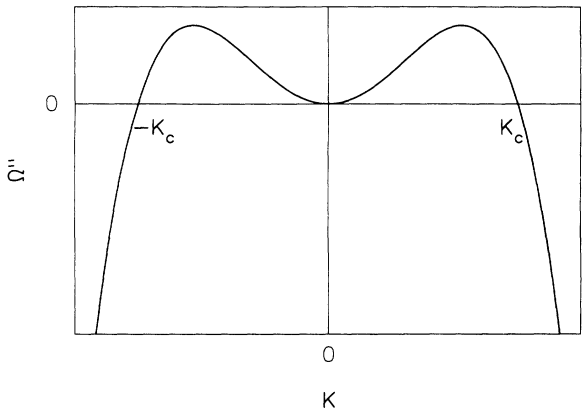

FIG. 2. Qualitative plot of the instability parameter Ω'' as a function of the reduced wave number K. This plot corresponds to Eq. (19). Instability occurs when Ω'' is positive.

can have a region of absolute instability when the complex solution $K(\Omega)$ of the dispersion relation (17) has a branch point for values of Ω_0 of Ω with positive imaginary part Ω_0''. This means that Eq. (17) has two coinciding roots $K_1=K_2$ at Ω_0. Furthermore, in order that instability occurs, these two coinciding roots must move to opposite half-planes of the complex K plane when we let Ω_0'' go to infinity. The value of Ω for which two roots of (17) coincide is obtained after rewriting (17) as

$$-\chi(K-K_1)^2(K-K_3)(K-K_4)=0 . \tag{21}$$

Equating powers of K in (17) and (20) then leads to four equations for the three roots K_1, K_3, and K_4, so that a solution exists only for a specific value of Ω_0. This value is the solution of the equation for Ω which results after elimination of the three roots. Absolute instability occurs for a range of values of ε and χ. This region is characterized by its border line in the ε-χ plane, which is termed the neutral curve. The equation describing this neutral curve follows if we impose the condition $\Omega_0=0$. The result can be put in the parameter form

$$\begin{aligned}&y^3(\chi-1)^4+2y^2(\chi-1)^2(2\chi-5)+y[5-4\chi+\varepsilon^2\{9-\tfrac{33}{4}(1+\chi)^2\}]+\frac{\varepsilon^2}{4}(1-\tfrac{27}{4}\varepsilon^2\chi)=0 ,\\ &y^2(\chi-1)^3(5-\chi)+y[2(\chi-1)(3\chi-5)+\frac{\varepsilon^2}{4}(\chi+1)(1-34\chi+\chi^2)]-1+\tfrac{3}{4}\varepsilon^2(11\chi-1)=0 .\end{aligned} \tag{22}$$

The relation between ε and χ describing the neutral curve is determined after elimination of $y=\Omega^2$ from this pair of equations. The result in the ε^2-χ plane is sketched in Fig. 3. For limiting values of χ, Eqs. (22) can be solved analytically, with the result

$$\begin{aligned}&\varepsilon^2=8 , \quad \Omega^2=1 \quad \text{for } \chi=0 ,\\ &\varepsilon^2=8-\frac{40}{\chi} , \quad \Omega^2=\frac{2}{\chi}-\frac{40}{\chi^2} \quad \text{for } \chi\gg 1 .\end{aligned} \tag{23}$$

B. Numerical result for limited cell length

When the length L of the cell is of the same order as the critical length $L_c=D/u$, the particle flux

$$j=\left[\frac{bD}{C}\right]^{1/2} n_0\left[\Theta N-\frac{\partial}{\partial\xi}N\right] \tag{24}$$

must obey the appropriate boundary conditions

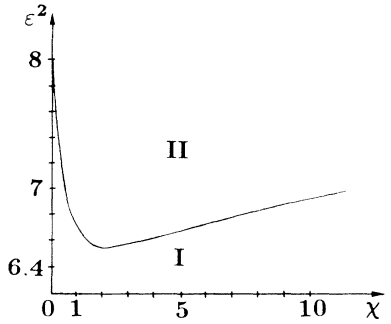

FIG. 3. Condition for absolute instability on the values for the reduced conductivity χ and the reduced intensity ε. (I) Region of stability; (II) region of absolute instability.

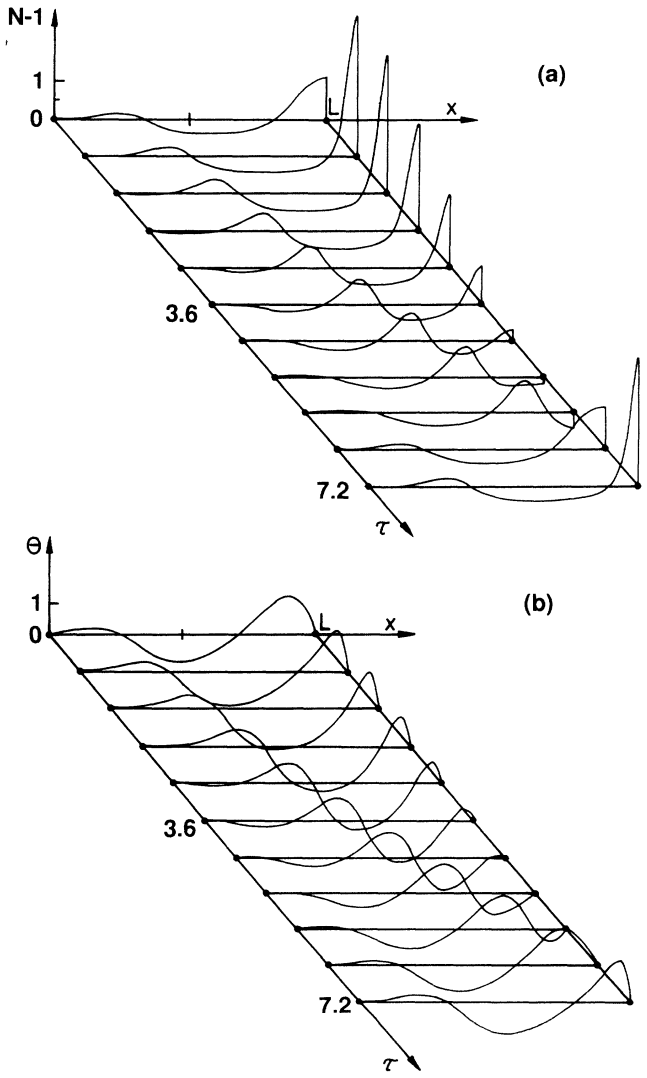

FIG. 4. Oscillatory behavior of the reduced density N and the reduced temperature Θ as a function of time and position. The boundary condition (26) holds with the parameter values $\beta=1$, $\gamma=0$, and $Q=0$. The reduced time τ is defined in Eq. (9), and x denotes the distance from the entrance window. The reduced time difference between successive curves is equal to 0.72. The intensity and the heat conductivity are specified by the parameters given in Eq. (27). (a) Reduced density N; (b) reduced temperature Θ.

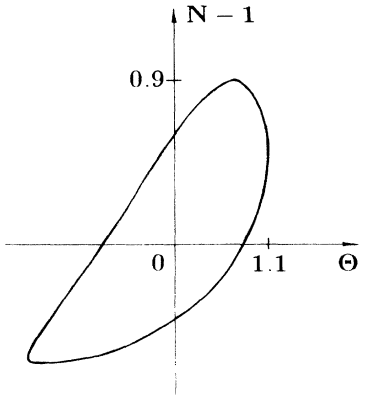

FIG. 5. Limit cycle of the oscillating density N and temperature Θ, in the middle of the cell ($x=L/2$).

$$j=0 \tag{25}$$

at the cell ends. The reduced temperature Θ obeys a boundary condition

$$\beta\Theta+\gamma\frac{\partial}{\partial\xi}\Theta=Q \tag{26}$$

at the cell ends, where the values of β, γ, and Q depend on the experimental conditions. We have solved the nonlinear equations (12) and (13) for $Q=0$, and various values of β and γ. In particular, we used the value $\beta=0$, $\gamma=1$, and $\beta=1,\gamma=0$. The intensity and the heat conductivity are specified by the parameters

$$\varepsilon^2=20\ ,\quad \chi=1\ , \tag{27}$$

which fall in the region of absolute instability (Fig. 3). The cell length is taken as

$$\tilde{L}\equiv L\left[\frac{b}{CD}\right]^{1/2}=13.4\ . \tag{28}$$

Figure 4 displays the oscillatory behavior described by Eqs. (12) and (13), which is reached after some transition time t_0. This transition time depends on the initial values of N and Θ. The oscillatory behavior only shows up when $\tilde{L}$ is larger than the reduced wavelength

$$\tilde{L}_c\equiv\frac{2\pi}{K_c}\ , \tag{29}$$

which has a value $\cong 3$ for the values (27). In the steady-state regime in the infinite cell, the reduced critical length $\tilde{L}_c$ is the dimensionless width of a peak in the density. In the conditions of Fig. 4, two peaks show up. Generally, the number of peaks increases for increasing values of the ratio $\tilde{L}/\tilde{L}_c$. The oscillations sketched in Fig. 4 can also be characterized by the limit cycle indicated in Fig. 5. The phase trajectory of the solution of Eqs. (12) and (13) is attracted to this limit cycle in a transition time t_0.

IV. POSSIBLE REALIZATION OF OSCILLATORY BEHAVIOR

In this section we discuss the possibility of observing the oscillatory behavior of light-induced drift in the case

of alkali-metal atoms. We recall that an essential ingredient of the effect is the conversion of absorbed light energy into heat. Since the conversion of optical excitation energy into translation energy is unlikely in the case of alkali-metal atoms immersed in a noble gas, this standard system is unfavorable for the effect. However, this situation may change radically when we add a dilute molecular vapor to the mixture [16] at a density of about 10% of the noble-gas pressure. Then we may safely assume that the drift velocity as a function of the intensity will not be affected. Also, the critical temperature T_c will not be changed. We specifically consider the systems Li-Ne, for which the value of T_c falls typically in the region of 600–1000 K, depending also on the radiation frequency [12]. The fraction p of the absorbed radiation energy that is converted into heat is assumed to be 10^{-1}. At a noble-gas pressure of 10 torr, the diffusion coefficient $D \cong 20$ cm^2 s^{-1}, and the photoabsorption length is $(n_0\sigma)^{-1}=10$ cm. For an intensity of 0.1 W cm^{-1}, the parameter α in Eq. (1) is about 10^{-1} cm s^{-1} K^{-1}. For a copper cell with internal and external radii equal to 0.2 and 0.3 cm, the heat exchange time C/b is estimated to be 10^2 s. Under these conditions, the parameter ε reaches the critical value 1 at the critical light intensity of about 2 W cm^{-1}. The critical length

$$L_c \equiv \tilde{L}_c \left[\frac{CD}{b} \right]^{1/2} \tag{30}$$

is then equal to 10 cm. In the linear regime for the drift velocity, the parameter ε varies quadratically with the intensity.

These estimates indicate that for the system Li-Ne, with a dilute molecular vapor added, the oscillatory behavior of the light-induced drift is within reach.

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