if the cross sections are divided by the degeneracies of the product states, the probability per state of producing a $5^2D_{3/2}$ atom in comparison to a $5^2D_{5/2}$ becomes as great as a factor of 2.7 larger for the peaks in Fig. 2.

The surprising detail found with this new multiphoton technique in the cross sections for the photolysis of Cs_2 attests to the importance of experimental measurements of these quantities. It is clear that the highly selective photodissociation of Cs_2 into $5D_{3/2}$ atoms for wavelengths between 470 and 485 nm could not have been anticipated from existing theoretical curves. It appears that the technique introduced here holds the promise of contributing substantially toward the detailed characterization of the repulsive states of simple molecules that are becoming of critical interest and importance in a variety of applications.

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[1] J. E. Mentall and P. M. Guyon, J. Chem. Phys. 67, 3845 (1977).

[2] M. LuVan, G. Mainfray, C. Manus, and I. Tugov, Phys. Rev. A 7, 91 (1973).

[3] C. B. Collins, B. W. Johnson, M. Y. Mirza, D. Popescu, and I. Popescu, Phys. Rev. A 10, 813 (1974).

[4] C. B. Collins, S. M. Curry, B. W. Johnson, M. Y. Mirza, M. A. Chellehmalzadeh, J. A. Anderson, D. Popescu, and I. Popescu, Phys. Rev. A 14, 1662 (1976).

[5] N. M. Shen and S. M. Curry, Opt. Commun. 20, 392 (1977).

[6] M. Y. Mirza and W. W. Duley, Proc. Roy. Soc. London, Ser. A 364, 255 (1978).

[7] M. Y. Mirza and W. W. Duley, Opt. Commun. 28, 179 (1979).

[8] R. Gupta, W. Happer, J. Wagner, and E. Wennmyr, J. Chem. Phys. 68, 799 (1978).

[9] J. Anderson, Ph.D. dissertation, The University of Texas at Dallas, 1980 (unpublished).

[10] D. Popescu, M. L. Pascu, C. B. Collins, B. W. Johnson, and I. Popescu, Phys. Rev. A 8, 1666 (1973).

[11] Supplementary data were obtained by exciting the $5^2D_{5/2} \to 25^2F_{5/2,7/2}$ transition with the delayed pulse. As the detection efficiency of the product populations was expected to be proportional to n^{-3} for $n \geq 20$, where n was the principal quantum number of the Rydberg state, the ionization signals obtained from this transition were multiplied by $(25/20)^3$. They were then included in the set from which the relative cross sections shown in Fig. 2 were taken.

Possibility of Double Sound Propagation in Disparate-Mass Gas Mixtures

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Disparate-mass neutral gases are here predicted to support two different forced-sound modes at moderately high frequencies, a fast and a slow wave. Specific predictions are given for mixtures of xenon and helium.

Forced sound propagation in gases may be studied by considering small disturbances from absolute equilibrium of the form $\exp[i(kx - \omega t)]$, where the frequency ω is constrained to be real and positive. The dispersion relation which one subsequently obtains for the complex wave number $k = k(\omega)$ in general has several roots. Usually, one root of the equation will correspond to sound propagation in the $+x$ direction. That root is identified by its low-frequency behavior: As $\omega \to 0$, $\mathrm{Im}(k)$ (proportional to the absorption coefficient) goes to zero, and $\mathrm{Re}(k)$ (proportional to the dispersion) goes to a positive known constant.

An exception to this simple situation can arise, however. Consider the Navier-Stokes description of a simple monatomic gas. The relevant equations may be written in linearized, nondimensional form, for deviations from equilibrium of the form $\exp i(kx - \omega t)$, as follows: Conservation equations,

$$n = zU \quad \text{(number-density deviation)}, \tag{1a}$$

$$U = z(n + T + P) \quad \text{(flow velocity)}, \tag{1b}$$

$$3T = 2n + 2zq \quad \text{(temperature deviation)}; \tag{1c}$$

constitutive equations

$$P = -(i\omega z)mU \text{ (pressure deviator)}, \tag{1d}$$

$$q = -(i\omega z)lT \text{ (heat flux)}; \tag{1e}$$

the reference standards are equilibrium values of number density (n_0), pressure (p_0), temperature (T_0), and speed $c \equiv (p_0/\rho_0)^{1/2}$, where ρ_0 is the equilibrium mass density. The reduced wavenumber z is

$$z \equiv kc/\omega. \tag{2}$$

Here

$$\tilde{P} \equiv \{P\}_{xx}/p_0, \quad \tilde{q} \equiv q/p_0 c,$$

where $\{P\}$ is the symmetric traceless part of the pressure tensor; and other small quantities, e.g.,

$$n = n_0[1 + \tilde{n}\exp i(kx - \omega t)],$$

are also denoted by tildes; finally, all tildes have been omitted from Eqs. (1a)–(1e). The nondimensional coefficients of viscosity (m) and thermal conductivity (l) are related to their usual, dimensional forms (μ, λ, respectively) by

$$l \equiv \lambda(T_0\omega_0/p_0c^2), \quad m \equiv \mu(4\omega_0/3p_0), \tag{3}$$

where ω_0 is an arbitrary reference frequency, hereafter chosen so that $l = 1$.

The dispersion relation following from Eqs. (1a)–(1e) is a quadratic in $y \equiv z^2$, of the form

$$ay^2 + by + 3 = 0, \tag{4}$$

where

$$b = -5 + (2i\omega l + 3i\omega m), \quad a = -2i\omega l - 2\omega^2 lm. \tag{5}$$

Usually, one root of Eq. (4) corresponds to sound propagation in the $\pm x$ direction; this root may be identified by the behavior

$$\begin{aligned} \mathrm{Re}[(\tfrac{5}{3})^{1/2}z] &= \text{Dispersion } (V_0/V) \underset{\omega\to 0}{\to} 1, \\ \mathrm{Im}[(\tfrac{5}{3})^{1/2}z] &= \text{Absorption } (\alpha) \underset{\omega\to 0}{\to} 0, \end{aligned} \tag{6}$$

for propagation in the $+x$ direction, where V_0 is the equilibrium speed of sound $[V_0 = (\frac{5}{3})^{1/2}c]$.

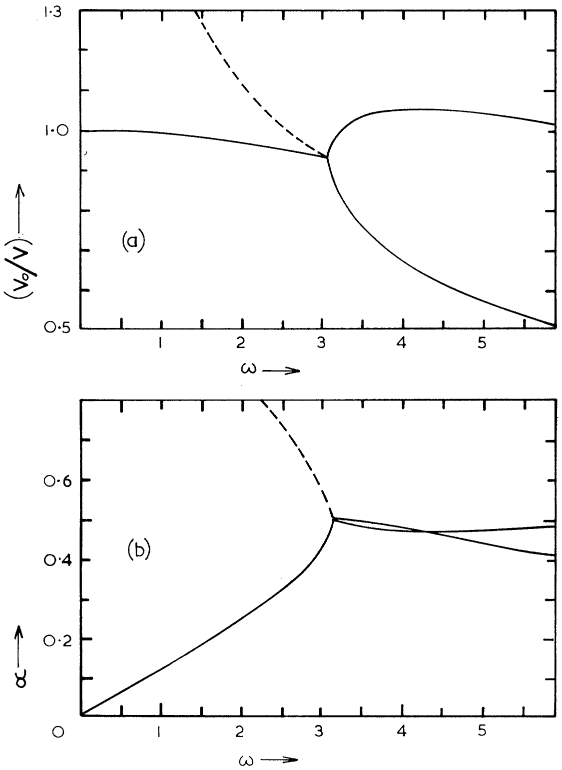

FIG. 1. (a) Dispersion and (b) absorption in a pure monatomic gas at critical viscosity $m_c = \frac{2}{15}$. For $\omega < \omega_c$, the solid curve shows the contribution of the sound root, the dashed curve that of the other root, of the dispersion relation. For $\omega > \omega_c$, both curves describe "sound propagation". Units of ω are chosen so that $l = 1$.

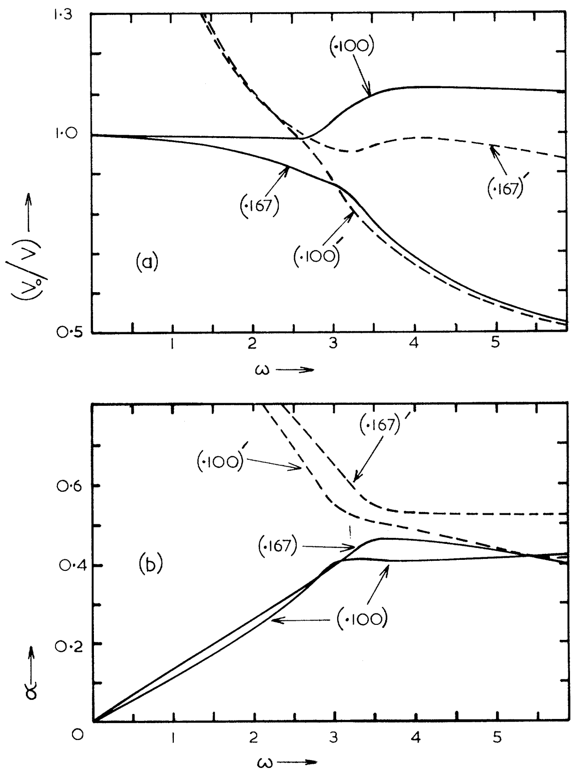

FIG. 2. (a) Dispersion and (b) absorption for a pure monatomic gas of viscosity $m = 0.167 > m_c$ and $m = 0.100 < m_c$. Solid and dashed curves refer to the sound mode and the interfering mode, respectively. Units of ω as in Fig. 1.

However, an exception to this simple situation occurs when

$$\omega = \omega_c \equiv \tfrac{25}{8}\,, \quad m = m_c \equiv \tfrac{2}{15}\,; \tag{7}$$

at these critical values of the parameters ω and m (with l set equal to unity), the two roots of Eq. (4) coincide. Figure 1 shows dispersion and absorption in forced sound as ω varies, for $m = m_c$. When $\omega > \omega_c$, it is evident that two propagating modes are predicted, of comparable absorption but very different sound speeds.[1]

Equally interesting, for m near m_c the behavior of the sound root shows clear signs of the interference of the second root, and indeed cannot be understood without reference to the latter. Figure 2 shows dispersion and absorption curves for values of m somewhat higher, and somewhat lower, than critical. One sees that the sound root changes its behavior *qualitatively* as m goes through the critical value. It therefore seems physically reasonable to call the general effect, for m in the *neighborhood* of m_c, and $\omega \gtrsim \omega_c$, *double sound propagation.*

It happens that for a pure monatomic gas, double sound propagation does not correspond to an observable effect because it occurs only for an artificially low viscosity.[2] For disparate-mass gas mixtures, however, double sound propagation is here predicted (apparently for the first time) as a realizable physical effect.[3]

A (binary) disparate-mass gas mixture is one in which the molecules of one component are very much lighter than those of the other $(m_1 \ll m_2)$. It has been shown[4-9] that, because of the slow exchange of kinetic energy between species of very different molecular masses, there are circumstances in which the usual equations of hydrodynamics do not give a correct description of such mixtures even though they correctly describe other more ordinary gases in the same circumstances. For forced sound propagation in disparate-mass gases, for instance, the equations of hydrodynamics are inapplicable when frequencies approach values of the order of $(m_1/m_2)^{1/2}$ of frequencies at which the hydrodynamic equations are usually supposed to become invalid (the latter frequency being of order V_0/L, where V_0 is the equilibrium speed of sound and L a mean free path). Correct equations, which necessarily involve separate species temperatures in part of the continuum regime $\omega \ll V_0/L$, have been developed independently by several authors.[4-11] Present calculations are based upon the work of Goebel, Harris, and Johnson.[5-8,12]

The relevant two-temperature continuum equations for a disparate-mass gas are Eqs. (24)–(31) of Ref. 7. The linearized nondimensional form of these equations, for small deviations from equilibrium of the form $\exp[i(kx-\omega t)]$, is as follows:

Conservation equations,

$$\left.\begin{aligned} n_1 &= z(U-W) \\ n_2 &= zU \end{aligned}\right\}\ \text{(number density deviations)}, \qquad \begin{aligned}&(8a)\\&(8b)\end{aligned}$$

$$U = z\left[\textstyle\sum_i x_i n_i + T + P\right] \ \text{(flow velocity)}, \tag{8c}$$

$$T = (\tfrac{2}{3})\textstyle\sum_i \left[x_i n_i + z q_i\right] \quad \text{(temperature deviation)}; \tag{8d}$$

Constitutive equations

$$W = (i\omega z)d[n_1 + T - x_2\Delta] \quad \text{(diffusion velocity } W \equiv U_2 - U_1\text{)}, \tag{8e}$$

$$P = -(i\omega z)mU \ \text{(pressure deviator)}, \tag{8f}$$

$$\left.\begin{aligned} q_1 &= -(i\omega z)l_1[T - x_2\Delta] \\ q_2 &= -(i\omega z)l_2[T + x_1\Delta] \end{aligned}\right\}\ \text{(heat fluxes)}, \qquad \begin{aligned}&(8g)\\&(8h)\end{aligned}$$

$$\Delta = -(i\omega z)\tau[W - (q_1/x_1)] \quad \text{(temperature separation } \Delta \equiv T_2 - T_1\text{)}; \tag{8i}$$

where most definitions and reference parameters are similar to those used in obtaining Eqs. (1a)–(1e); x_i is the volume fraction of species i $(i = 1, 2)$. The reference frequency is here taken to be the relaxation frequency ω_Δ for separation of temperature between the species,

$$\omega_\Delta = 2c^2 x_2/D\,, \tag{9}$$

where D is the usual coefficient of diffusion.[13] The nondimensional transport coefficients are related to those in common use by the formulas

$$d = D\omega_\Delta/c^2 x_2 = 2 \ \text{(diffusion)}, \tag{10a}$$

$$m = 8\mu_{20} x_2/\rho_0 D \ \text{(viscosity)}, \tag{10b}$$

$$l_i = \lambda_{i0} T_0 \omega_\Delta/p_0 c^2 \ \text{(thermal conductivity)} \tag{10c}$$

$$\tau = (\tfrac{2}{3})[1 - i\omega]^{-1} \ (\Delta \text{ transport}), \tag{10d}$$

where μ_{i0} and λ_{i0} are given by Ref. 7, Eqs. (27a) and (27b), respectively.

The dispersion relation following from Eqs. (8a)–(8i) has been obtained numerically for real ω, for mixtures of xenon and helium at standard temperature and pressure, with the pure-gas viscosities and the mixture coefficient of diffusion as experimental input.[14] The results of these calculations are presented in Fig. 3 for

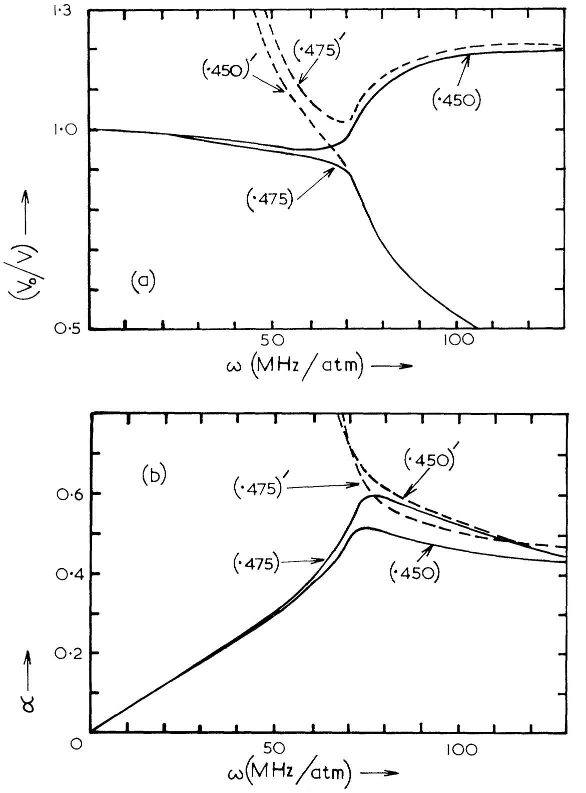

FIG. 3. (a) Dispersion and (b) absorption in mixtures of Xe-He for He volume fractions $x_1 = 0.450$ and $x_1 = 0.475$. Solid and dashed curves refer to the sound mode and the interfering mode, respectively. In (a), the dispersion of the sound mode for $x_1 = 0.475$ overlaps that of the interfering mode with $x_1 = 0.450$ to within graphical accuracy, for $\omega \gtrsim 75$ MHz/atm.

two different but similar compositions. It is evident that there are a critical composition x_{1c} of helium ($0.450 < x_{1c} < 0.475$), and a critical frequency ω_c ($\omega_c \approx 73$ MHz/atm, corresponding to a rarefaction parameter $r = p_0/\mu\omega \approx 10.5$), which characterize double-sound-propagation effects analogous to those already described. The effect of increasing the helium fraction x_1 in this case is very similar to that of increasing the viscosity m in the predictions following from Eqs. (1). At frequencies higher than ω_c, for compositions near the critical, such as those shown in Fig. 3, two propagating modes are predicted, of comparable absorption but very different speed of propagation. For He compositions less (greater) than critical, the low-frequency sound mode goes over into the slower (faster) of the two predicted waves.[15]

Calculation shows that standard hydrodynamics (with use of Maxwell forces, and simplified transport coefficients[5-7]) also predicts double sound propagation in Xe-He, with a similar critical frequency, but a critical composition $0.225 < x_c < 0.250$.

Finally, it should be emphasized that the predictions presented here can be tested by experiment at the present time.[16] The frequencies of interest are well within experimental capability, as well as being low enough that mean-free-path corrections should be unimportant.

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[1] C. Truesdell, J. Rational Mech. Anal. 5, 55 (1956), notes the existence of a critical (m/l), but fails to draw the *physical* conclusion of double sound propagation.

[2] The relation $\lambda = 5\mu c_v/2$, where c_v is the specific heat at constant volume, gives $m = \frac{16}{45}$; for polyatomic gases, m is larger still.

[3] Physical conditions for the existence of multiple sound propagation in gas mixtures are discussed by R. L. Liboff, J. Acoust. Soc. Am. 36, 661 (1964).

[4] L. D. Tsendin, Pis'ma Zh. Eksp. Teor. Fiz. 56, 929 (1969) [JETP Lett. 29, 502 (1969)].

[5] C. J. Goebel, S. M. Harris, and E. A. Johnson, Phys. Fluids 19, 627 (1976).

[6] E. A. Johnson, Arch. Mech. (Warsaw) 28, 803 (1976).

[7] C. J. Goebel, S. M. Harris, and E. A. Johnson, *Rarefied Gas Dynamics*, edited by J. L. Potter (American Institute of Aeronautics and Astronautics, New York, 1977), Vol. 51, Pt. 1, p. 109.

[8] E. A. Johnson, Phys. Fluids, 21, 1239 (1978).

[9] J. P. Petit and J. S. Darrozes, J. Mec. 14, 745 (1975).

[10] E. Goldman and L. Sirovich, Phys. Fluids 10, 1928 (1967), and 12, 245 (1969). Mistakes in this work were corrected in Ref. 5.

[11] E. B. Goldman, J. Acoust. Soc. Am. 44, 708 (1968).

[12] The associated restriction to Maxwell molecules (intermolecular forces proportional to r^{-5}, where r is the intermolecular separation) must be improved upon, but is useful for obtaining initial results.

[13] S. Chapman and T. G. Cowling, *Mathematical Theory of Non-Uniform Gases* (Cambridge Univ. Press, Cambridge, England, 1970), Chap. 8.

[14] See Ref. 13, Chaps. 12 and 14.

[15] Model calculations for disparate-mass gases by L. D. Tsendin, Akust. Zh. 13, 617 (1967) [Sov. Phys.-Acoust. 13, 521 (1968)] predict only the slow wave, for $\omega \gg \omega_\Delta$ and $x_1 \sim x_2$. Calculations of Ref. 11 for Xe-He, which should agree with those herein, predict only the fast wave, for $\omega \sim \omega_\Delta$ and $x_1 = x_2$.

[16] A calculation incorporating more realistic intermolecular potentials might be expected to produce slight changes in the predicted values of the critical parameters.