

is not clear, but it suggests that the cascade may be composed of sequences of two- or three-step related transitions based on occasional intrinsic states which scatter around a straight line reminiscent of the Fermi-gas model. No  $\Delta J \geq 3$  transition (yrast trap) was observed.

In summary, we have found in  $^{154}\text{Er}$  a series of yrast levels from spin (35, 36) which, on an  $E$  vs  $I(I+1)$  plot, fall on straight segments of a broken line with side feeding occurring mainly at the break points of the line (Fig. 3), resulting in transitions of equal intensities between states on straight segments of the yrast line. Transitions within the segments are either all quadrupole or all dipole. Angular-distribution data indicate an abrupt reduction of alignment at one of these break points so that it is expected that isomers might exist at these points. This cascade differs from a quasi-ground-state band up to spin  $18^+$  which shows side feeding at all levels, as is the usual case in the deformed region of nuclei.

This research was supported by the Division of Basic Energy Sciences, U. S. Department of Energy, under Contract No. EY-76-C-02-0016.

<sup>1</sup>A. Bohr and B. R. Mottelson, Phys. Scr. **10A**, 13 (1974).

<sup>2</sup>T. Døssing, K. Neergård, K. Matsuyanagi, and Hsi-

Chen Cheng, Phys. Rev. Lett. **39**, 1395 (1977).

<sup>3</sup>P. Auger *et al.*, Z. Phys. A **285**, 59 (1978).

<sup>4</sup>T. L. Khoo *et al.*, Phys. Rev. Lett. **41**, 1027 (1978).

<sup>5</sup>O. C. Kistner, A. W. Sunyar, and E. der Mateosian, Phys. Rev. C **17**, 1417 (1978).

<sup>6</sup>A. W. Sunyar, E. der Mateosian, O. C. Kistner, A. Johnson, A. H. Lumpkin, and P. Thieberger, Phys. Lett. **62B**, 283 (1976).

<sup>7</sup>The 674-keV peak has an admixture of the 675-keV peak in it so that our ordering of the 554- and 674-keV transitions is subject to more error than in the other pair of transitions from the spin-9 state. This does not affect the assignment of spin 9 to the level below the isomeric state which is established by the 432-796-keV pair. The acceptance of spin 7 for the level fed by the 554-keV transition is supported as well by systematics of the  $N=86$  isotones of  $^{154}\text{Er}$ .

<sup>8</sup>*Table of Isotopes*, edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978), 7th ed.

<sup>9</sup>D. R. Haenni and T. T. Sugihara, Phys. Rev. C **16**, 120 (1977).

<sup>10</sup>Equal-intensity sequences of  $\gamma$  transitions have been reported in this region of nuclei before. See M. Piiparinen *et al.*, Phys. Scr. **17**, 193 (1978).

<sup>11</sup>T. Yamazaki, Nucl. Data, Sect. A **3**, 1 (1967).

<sup>12</sup>For an interpretation of the high-spin cascade in terms of collective bands, see L. K. Peker, J. O. Rasmussen, and J. H. Hamilton, in Proceedings of the International Conference on Nuclear Interactions, Canberra, Australia, 1978 (to be published).

<sup>13</sup>*Proceedings of the Topical Conference on Problems of Vibrational Nuclei, Zagreb, Croatia, Yugoslavia*, edited by G. Alaga, V. Parr, and L. Šips (North Holland, New York, 1975), p. 320.

<sup>14</sup>R. J. Liotta and R. A. Sorensen, Nucl. Phys. **A297**, 136 (1978).

## New Infrared Absorption Bands of Alkali Vapors

N. D. Bhaskar, E. Zouboulis, T. McClelland, and W. Happer

*Columbia Radiation Laboratory, Department of Physics, Columbia University, New York, New York 10027*

(Received 26 December 1978)

We have identified two new infrared absorption bands in potassium vapor. The band between 1.1 and 1.6  $\mu\text{m}$  is attributed to the  $K_2$  ( $^3\Sigma_u^+ \rightarrow ^3\Sigma_g^+$ ) transition, the absorption analog of the intense emission continuum of  $H_2$ . The absorption at wavelengths longer than 1.6  $\mu\text{m}$  is probably due to trimers.

In this paper we report on a new region of absorption of infrared radiation by alkali-metal vapors. Recently, Chertoprud<sup>1</sup> has reported that absorption exists in saturated potassium vapor at wavelengths well beyond the edge of the A band (1.1  $\mu\text{m}$ ), and the absorption extends at least as far as 2.5  $\mu\text{m}$ . Chertoprud<sup>1</sup> assigns this absorption to the intercombination transition ( $X^1\Sigma_g^+ \rightarrow ^3\Sigma_u^+$ ). Our recent experiments confirm that ab-

sorption does exist in the alkali vapors potassium, rubidium, and cesium at wavelengths at least as long as 2.5  $\mu\text{m}$ . However, our observations differ considerably from those of Chertoprud<sup>1</sup> since we find at least two distinct absorption bands, a much smaller attenuation coefficient at low temperatures, a complex dependence on wavelength, and a temperature dependence which unambiguously rules out the assignment of

this absorption to the transition ( $X^1\Sigma_g^+ \rightarrow {}^3\Sigma_u^+$ ). As we shall show below, the most likely assignment of the shorter-wavelength absorption (1.1–1.6  $\mu\text{m}$ ) is the previously unobserved transition ( ${}^3\Sigma_u^+ \rightarrow {}^3\Sigma_g^+$ ) in the alkali dimer molecule. This is the analog of the powerful ultraviolet emission continuum of  $\text{H}_2$  molecules, a transition whose apparent absence in the emission spectrum<sup>2</sup> of alkali dimers has long been a puzzle. The longer-wavelength absorption (1.6–2.0  $\mu\text{m}$ ) cannot be assigned to dimers, and it seems consistent with electronic absorption bands in clusters of three potassium atoms or trimers. These new absorption bands lie in a broad spectral region which was previously considered to be transparent and they add a new and important element to considerations of alkali-metal vapors for laser media or heat-transfer media.

The experimental apparatus used in our work is shown in Fig. 1. The effective length  $z$  (about 20 cm) of a vapor column in a heat-pipe oven<sup>3</sup> is determined from the measured outside temperature profile along the length of the heat pipe. The uncertainty in  $z$  is about 5%. The saturated vapor pressure of the alkali metal is controlled with an

inert buffer gas of argon, neon, or helium. At gas pressures above 300-Torr neon and argon are found to produce dense clouds of alkali-metal fog. We found helium to be free of this problem, presumably because of the high kinematic viscosity and heat conductivity of the helium. Our measurements of vapor pressure  $P$  and temperature  $T$  for potassium were consistent to within about 1% with the accepted empirical formula<sup>4</sup>

$$\log_{10} P_{\text{mm}} = 7.18 - (4435/T). \quad (1)$$

The Clausius-Clapeyron equation (1) implies that  $l$ , the latent heat of vaporization of potassium, is 20.3 kcal/mole.

The experimental procedure is as follows.  $I(\lambda, T)$ , the intensity of light of wavelength  $\lambda$  transmitted through the vapor at temperature  $T$  and  $I(\lambda, T_r)$  the transmitted intensity at room temperature  $T_r$  are measured, the latter measured at the beginning and end of each run. For a vapor column of length  $z$ , the attenuation coefficient  $\alpha(\lambda, T)$  is given by

$$\alpha(\lambda, T) = z^{-1} \ln [I(\lambda, T_r)/I(\lambda, T)]. \quad (2)$$

Representative data for  $\ln \alpha(\lambda, T)$  are plotted as a function of  $1/T$  in Fig. 1. Note that the data can be fitted very well with a straight line in the temperature interval covered by our experiments. Thus, we may represent  $\alpha$  by the empirical formula

$$\alpha(\lambda, T) = \exp[A(\lambda) - E(\lambda)/RT]. \quad (3)$$

A summary of our experimental data is shown in Fig. 2 where the measured attenuation coefficient  $\alpha(\lambda, T)$  and the activation energy  $E(\lambda)$  are plotted as a function of wavelength  $\lambda$  for  $T = 943^\circ\text{K}$ .

In the temperature range involved in our experiments the potassium vapor consists predominantly of free potassium atoms. Bound dimers  $\text{K}_2$  also exist at about 6% of the monomer concentration at  $943^\circ\text{K}$ . In addition to the bound dimers, unbound pairs of potassium atoms are present at concentrations large enough to cause substantial optical absorption. Trimers and higher polymers of potassium are also important at the relatively high vapor pressures of our experiments. The attenuation coefficient of the potassium vapor will therefore consist of contributions from various clusters of  $n$  potassium atoms, each of which will contribute a term

$$\alpha_n(\lambda, T) = b(\lambda) N_K^n \exp[-V(\lambda)/RT] \quad (4)$$

to the total attenuation coefficient of the vapor. Here  $V(\lambda)$  is the potential energy of association

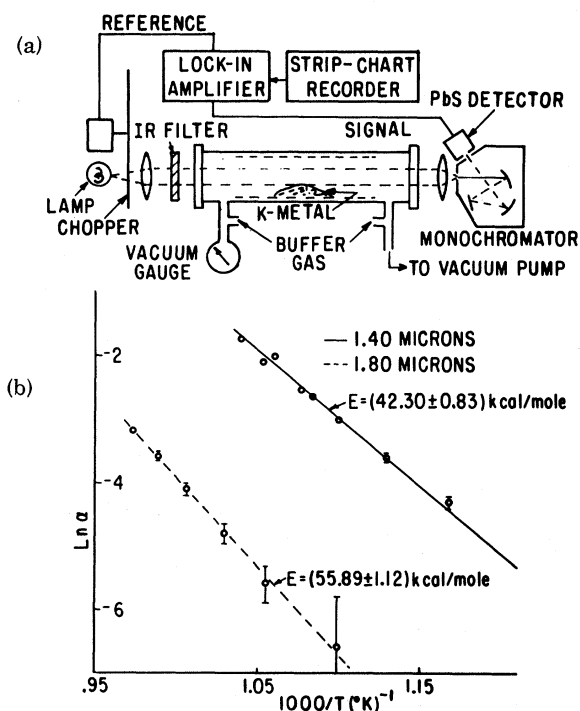


FIG. 1. (a) Apparatus. (b) Typical data for attenuation of infrared radiation by saturated potassium vapor. The straight line fits and the standard deviation of the slope were obtained by least-squares error analysis.

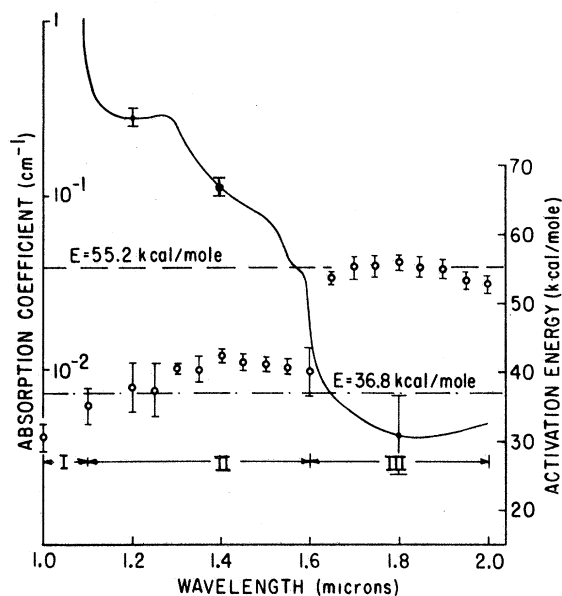


FIG. 2. Activation energies (points) and attenuation coefficient (solid line) for saturated potassium vapor as a function of wavelength. Error estimates are obtained from least-squares fits to experimental data like that in Fig. 1(b). The error estimates for the absorption coefficient are also shown. Monochromator resolution is  $50 \text{ \AA}$  (full width at half-maximum). Bands I, II, and III are indicated at the bottom of the figure.

of  $n$  potassium atoms into a configuration for which an electronic transition of wavelength  $\lambda$  is possible, and  $R$  is the gas constant. The factor  $b(\lambda)$  includes oscillator strengths and phase-space factors, and  $N_K$  is the number density of potassium atoms. In the case of potassium dimers (4) can be written in the well known form<sup>5</sup>

$$\alpha_2(\lambda, T) = 2\pi^2 r_e c f (g/4) r^2 |dr/d\nu| N_K^2 \times \exp[-V(\lambda)/RT], \quad (5)$$

where  $r_e$  is the classical electron radius,  $c$  is the speed of light,  $f$  is the oscillator strength of the electronic transition,  $g$  is the statistical weight of the lower electronic potential curve, and  $\nu = c/\lambda$  is the frequency of a photon which can resonantly excite the molecule between two electronic states at an internuclear separation  $r$ .

Saturated potassium vapor consists predominantly of free potassium atoms, and we may therefore approximate the vapor pressure with the partial pressure of the free atoms. Using

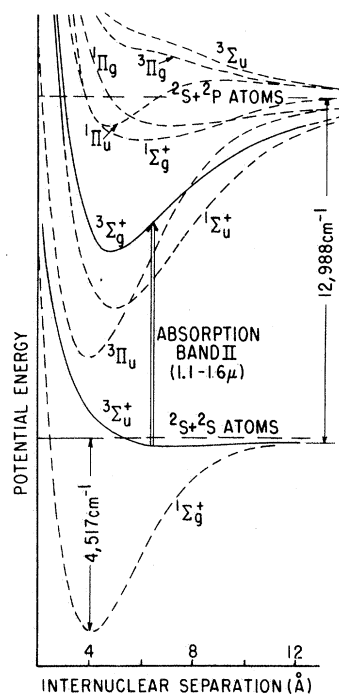


FIG. 3. Qualitative sketch of the electronic potential curves of  $K_2$  based on the recent calculations (Ref. 6) for  $Li_2$ . The assignment of the new infrared band II is indicated.

(1) and the ideal-gas law we write

$$\ln(N_K/N_0) = \ln P - \ln(RT) \cong \ln[P_0/(RT_0)] - (\epsilon/RT) - 1, \quad (6)$$

where the latent internal energy of vaporization  $\epsilon$  is related to the latent heat  $l$  by  $\epsilon = l - RT_0 = 18.4 \text{ kcal-mole}$ .  $P_0$  is a constant,  $N_0$  is Avogadro's number, and  $T_0 = 943^\circ\text{K}$  is a representative temperature which characterizes the conditions of our experiments. Thus, Eq. (4) can be written as

$$\alpha_n(\lambda, T) = c(\lambda) \exp[-(n\epsilon + V)/RT]. \quad (7)$$

Comparing (7) with (3) we conclude that the activation energy  $E(\lambda)$  is equal to the sum of the association energy  $V$  and  $n$  factors of the vaporization energy  $\epsilon$ ,

$$E(\lambda) = n\epsilon + V. \quad (8)$$

For reference we have indicated the energies  $2\epsilon = 36.8$  and  $3\epsilon = 55.2 \text{ kcal/mole}$  in Fig. 2.

Three distinct regions of absorption, designated as regions I, II, and III, are indicated in Fig. 2. In band I ( $\lambda < 1.1 \text{ }\mu\text{m}$ ) the absorption coefficient has values in excess of  $1 \text{ cm}^{-1}$  and activa-

tion energies of  $\sim 25$  kcal/mole. Band I is the red edge of the well known A band, the strongest absorption band of  $K_2$  dimers. The association energy of  $\sim -12$  kcal/mole corresponds approximately to the known dissociation energy of the  $K_2(^1\Sigma_g^+)$  ground-state molecule. The potential energy curves for  $K_2$  are shown in Fig. 3.

Band II ( $1.1 \mu\text{m} < \lambda < 1.6 \mu\text{m}$ ) is a new absorption band which is considerably weaker than the A band and which has a considerably higher activation energy. The activation energy of band II is of the order of 40 kcal/mole while the vaporization energy  $2\epsilon$  of two potassium atoms is 36.8 kcal/mole. The difference, 3.2 kcal/mole, can be interpreted as a repulsive interaction potential between a pair of ground-state K atoms on the  $^3\Sigma_u^+$  potential curve. Theoretical calculations<sup>6</sup> for  $Li_2$  suggest that the transition ( $^3\Sigma_u^+ \rightarrow ^3\Sigma_g^+$ ) gives rise to a band to the red of the corresponding A band. The band should have a satellite, corresponding to the distance of closest approach of the potential curves (see Fig. 3) at a wavelength which is somewhat greater than twice the wavelength of the first resonance line of the free alkali atom (7665–7699 Å for K). It is reasonable to identify the observed knee at  $1.58 \mu\text{m}$  in Fig. 2 with the predicted satellite.

The observed magnitude of the attenuation coefficient of band II is also consistent with the value expected for the transition ( $^3\Sigma_u^+ \rightarrow ^3\Sigma_g^+$ ). Using Eq. (5), and substituting therein the following reasonable values  $f=1$ ,  $g=3$ ,  $r=8 \text{ \AA}$ ,  $dv/dr=5 \times 10^{21} \text{ sec}^{-1} \text{ cm}^{-1}$ ,  $N_K=3 \times 10^{18}$ , and  $V=E(1.2 \mu\text{m}) - 2\epsilon = 37.5 - 36.8 = 0.7 \text{ kcal/mole}$ , we find  $\alpha_2(1.2 \mu\text{m}, 943^\circ\text{K}) = 0.6 \text{ cm}^{-1}$  a number in satisfactory agreement with the measured attenuation coefficient of  $0.3 \text{ cm}^{-1}$  from Fig. 2.

In summary, the observed wavelength, activation energy, absorption-coefficient magnitude, and satellite of band II all support the assignment of this band to the transition ( $^3\Sigma_u^+ \rightarrow ^3\Sigma_g^+$ ).

Band III ( $\lambda > 1.6 \mu\text{m}$ ) is about a factor of 70 weaker than band II and it has a substantially higher activation energy ( $E \approx 56 \text{ kcal/mole}$ ). If we attempt to assign band III to a dimer transition we must interpret the high activation energy,  $E(1.8 \mu\text{m}) = 56 \text{ kcal/mole}$ , as a repulsive potential energy  $V = 19.2 \text{ kcal/mole}$ . For example, one might try to assign band III to the transition of electronic excited  $^3\Pi_u$  state to  $^3\Sigma_g^+$ . However, the measured magnitude of the absorption coefficient in band III is about 300 times too big to be consistent with dimer absorption and any reasonable values of the parameters in (5).

It is interesting to note that the activation energy of band III, 56 kcal/mole, is very close to the energy of vaporization of three potassium atoms  $3\epsilon = 55.2 \text{ kcal/mole}$ . Thus, loosely bound alkali trimers are a possible source of the absorption in band III. The "number density"  $e$  of oscillator electrons involved in the absorption band is related to the strength,  $S = \int \alpha d\nu$ , of that band by the well known formula  $e = S/\pi r_e c$ . For band III (integrating from  $1.6$ – $2.0 \mu\text{m}$ ) we estimate  $e$  to be about  $5.6 \times 10^{12} \text{ cm}^{-3}$ . The total number density of valence electrons in the vapor is very nearly equal to the potassium-atom number density  $N_K = 3 \times 10^{18}$ . The fraction of valence electrons involved in band III is therefore  $1.9 \times 10^{-6}$ . This is a crude estimate of the ratio of trimers (associated with band III to monomers. Since the trimer association energy is comparable to  $RT$  (see Fig. 2), the trimer number density is related to the monomer number density by  $N_{K_3} \approx e \approx a^6 N_K^3$  where  $a$  is a characteristic internuclear separation for the cluster of three potassium atoms. Using the estimates  $e = 1.9 \times 10^{-6}$  and  $N_K = 3 \times 10^{18}$  we find  $a$  to be  $7.8 \text{ \AA}$ . In view of the known range of interatomic potentials between alkali atoms this is an entirely reasonable value for the characteristic separation of potassium atoms in a loosely bound trimer. We should also point out that the closely analogous trimer  $H_3$  is known from theoretical calculations<sup>7</sup> to have low-lying electronic potential surfaces between which fully allowed electronic transitions are possible; for example, the  $^2\Sigma_u^+$  and  $^2\Sigma_g^+$  states of  $D_{\infty h}$  symmetry. Note that the shorter-wavelength trimer absorption spectrum is completely masked by the powerful dimer absorption bands below  $1.6 \mu\text{m}$ . We are therefore unable to draw any conclusions about the population of trimers (perhaps the great majority)<sup>8</sup> with absorption bands below  $1.6 \mu\text{m}$ .

We have observed analogous absorption bands in rubidium and cesium vapors, and similar experiments for sodium and lithium vapors are also underway. A more extensive paper describing our experimental results for all of the alkali vapors is in preparation.

This work was supported by the U. S. Army Research Office (Durham) under Contract No. DAAG29-77-G-0015, with supplemental support by the Joint Services Electronics Program (U. S. Army, U. S. Navy, and U. S. Air Force) under Contract No. DAAG29-77-C-0019.

<sup>1</sup>V. E. Chertoprud, *Teplofiz. Vys. Temp.* **14**, 216

(1976).

<sup>2</sup>P. P. Sorokin and J. R. Lankard, *J. Chem. Phys.* **55**, 3810 (1971).<sup>3</sup>C. R. Vidal and J. Cooper, *J. Appl. Phys.* **40**, 3370 (1969).<sup>4</sup>T. B. Reed, *Free Energy of Formation of Binary Compounds* (MIT Press, Cambridge, Mass., 1971).<sup>5</sup>L. K. Lam, A. Gallagher, and M. M. Hessel, *J. Chem. Phys.* **66**, 3350 (1977).<sup>6</sup>M. L. Olson and D. D. Konowalow, *Chem. Phys.* **21**, 393 (1977); D. K. Watson, C. J. Cerjan, S. Guberman, and A. Dalgarno, *Chem. Phys. Lett.* **50**, 181 (1977).<sup>7</sup>E. Frenkel, *Z. Naturforsch.* **25a**, 1265 (1970).<sup>8</sup>C. H. Wu, *J. Chem. Phys.* **65**, 3181 (1976).

## Raman Side-Scatter Instability in Nonuniform Plasma

Michael A. Mostrom<sup>(a)</sup> and Allan N. Kaufman

*Department of Physics and Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720*  
(Received 1 August 1978)

For the Raman instability in nonuniform plasma, the linear space-time asymptotic response consists of *two* parts, convectively growing wave packets and temporally growing localized side-scattered eigenmodes. Eigenmodes dominate the response only *after* the growth of the side-scattered wave packets is terminated by refraction from their resonance zones. A finite pump-beam diameter typically ends all growth before eigenmodes appear, except near quarter-critical density. These considerations may reconcile simulations, previous theories, and experiments.

The theory of Raman side-scatter instability<sup>1-5</sup> has developed in a confusing way (examples to be given later). This has not seemed to generate much worry, possibly because the instability has not been observed in experiments<sup>6</sup> exceeding the threshold predicted by theory<sup>4</sup> and computer simulation.<sup>5</sup> This should have created even more worry because a potentially violent reflective instability is hiding behind some saturation mechanism that may not operate or may be detrimental under laser-fusion conditions.

In this Letter, our first goal is to present a more complete theory of this instability, in order to resolve the confusion and paradoxes associated with the incomplete nature of the several previous theories. We view this as more than of academic interest or tying up loose ends, because of the necessity to have a clear understanding of the basic features of this potentially important instability. Moreover, the theory presented here provides a possible simple explanation for the lack of experimental evidence for this instability. This linear (i.e., convective) saturation mechanism is not effective for a single incident laser beam of power greater than about 3 TW, and thus could possibly be tested by the new laser systems that are now becoming available. Most important, this mechanism is not very effective for spherically illuminated pellets under typical proposed reactor parameters, and this would necessitate the instability saturating under a nonlinear me-

chanism that may have detrimental results (e.g., fast-electron generation). This disturbing prediction motivates our second goal, which is to encourage experiments designed specifically to observe and determine the saturation properties of this instability.

Above a threshold intensity, an electromagnetic pump wave (frequency  $\omega_0$ , wave vector  $\vec{k}_0 = k_0 \hat{z}$ ) undergoes stimulated decay into lower-frequency scattered electromagnetic waves ( $\omega_1 < \omega_0$ ,  $\vec{k}_1$ ) and Langmuir waves ( $\Omega = \omega_0 - \omega_1$ ,  $\vec{K} = \vec{k}_0 - \vec{k}_1$ ). For given  $\omega_1$ , this process is localized to a narrow resonance zone<sup>1</sup> where  $\Omega$  is near the local plasma frequency  $\omega_p(z)$ . Thus side-scattered electromagnetic waves (i.e., waves with coincident turning point and resonance position) might grow to a level sufficient to prevent the pump wave from reaching and heating higher-density plasma regions.<sup>2,3</sup>

Indeed, temporally growing eigenmodes have been predicted,<sup>4</sup> and computer simulation<sup>5</sup> (with periodicity assumed perpendicular to the density gradient) has clearly shown strong growth until termination by electron trapping and heating and consequent severe damping of the Langmuir waves. But, as mentioned earlier, this instability has not yet been seen at all in the many laser-fusion experiments<sup>6</sup> apparently exceeding the theoretical threshold intensity. One possible explanation,<sup>7</sup> a linear (i.e., convective) saturation due to wave propagation out of the finite-diameter