FIG. 3. Longitudinal-mode spectra of GaAs-junction lasers showing simultaneous oscillation in two families of modes. In (a) only one family exhibits the coupled, pulsing behavior, while the single mode at 8424 Å is only weakly modulated; in (b) all modes are pulsing.

orthogonality of modes with different transverse distributions. The difference between the two types of behavior is a result of the variation among diodes in the strength of the dispersion, which determines whether or not Δ for each family is close enough to ω_R for strong coupling to occur. The possibility of having all modes pulsing is greatest for families whose center frequencies are close to each other.

While our experimental observations have been made on GaAs stripe-geometry injection lasers, it is reasonable to expect that similar phenomena can also occur in other semiconductor lasers. However in nonsemiconductor lasers, the dispersion is generally insufficient to produce the effect. In conclusion, we note that the high dispersion, which is of prime importance in the phe-

nomenon described above, is also one factor which greatly hinders spontaneous mode locking of semiconductor lasers.[12]

We wish to thank R. Roldan for communicating several of his observations and H. Haug for a preprint of his very interesting paper. We are also indebted to J. C. Dyment and L. A. D'Asaro for many valuable comments on the manuscript. Finally we wish to thank J. A. Richards and L. H. Drewes for their competent technical assistance.

[1]D. A. Kleinman, Bell System Tech. J. 43, 1505 (1964), and references contained therein.

[2]W. E. Lamb, Phys. Rev. 134, A1429 (1964).

[3]H. Haug, Z. Physik 194, 482 (1966), and 195, 74 (1966).

[4]T. L. Paoli, J. E. Ripper, and T. H. Zachos, to be published.

[5]Variations which go as $\cos[(q_3+q_4)\pi z/L]$ are substantially reduced by the motion of the carriers.

[6]D. T. F. Marple, J. Appl. Phys. 35, 1241 (1964).

[7]T. H. Zachos and J. E. Ripper, IEEE J. Quantum Electron. QE-5, 29 (1969).

[8]D. E. McCumber, Phys. Rev. 141, 306 (1966); H. Haug, to be published.

[9]L. A. D'Asaro, J. M. Cherlow, and T. L. Paoli, IEEE J. Quantum Electron. QE-4, 164 (1968).

[10]The current-temperature region for which Δ and ω_R coincide is a characteristic of each laser; but the multimode pulsing described in this paper was observed in nearly all laser diodes tested.

[11]Observed by R. Roldan, private communication.

[12]D. Gloge and R. Roland, Appl. Phys. Letters 14, 3 (1969).

COLLECTIVE OSCILLATIONS IN PURE LIQUID BENZENE*

M. W. Williams†
Physics Department, University of Tennessee, Knoxville, Tennessee 37916

and

R. A. MacRae,‡ R. N. Hamm, and E. T. Arakawa
Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
(Received 21 March 1969)

Measurements of the optical properties of pure liquid benzene in the vacuum ultraviolet are interpreted in terms of a molecular excitation of the π electrons and also collective, volume-plasma oscillations of these same electrons.

Benzene vapor exhibits an intense absorption band in the vacuum ultraviolet[1-4] with a maximum at about 1790 Å. This corresponds[5] to the allowed transition $^1A_{1g}{}^- - {}^1E_{1u}{}^+$ which is a molecular excitation of the π electrons in the benzene ring.

We have observed this same molecular excitation in pure liquid benzene,[6] shifted slightly in energy by the close proximity of other molecules in the liquid state. In addition our optical data show the existence of volume-plasma oscillations, presumably involving the π electrons acting collec-

tively in the liquid. These are the first reported values of the optical constants for pure liquid benzene over the wavelength region from 1200 to 2000 Å and also the first observation known to the authors of collective oscillations in a liquid insulator.

The reflectance was measured as a function of the angle of incidence over the wavelength range 1200 to 3200 Å. Pure liquid benzene[7] was placed in a sealed cell, one side of which consisted of a transparent semicylinder.[8-10] Light entered the semicylinder normal to its curved surface and was reflected at the plane semicylinder-benzene interface. For measurements from 1200 to 1700 Å the semicylinder was made of MgF_2, from 1300 to 2000 Å of CaF_2, and from 1800 to 3200 Å of quartz. When using the MgF_2 and CaF_2 semicylinders, observations were made as rapidly as possible to minimize errors resulting from the fact that these materials slowly dissolve in liquid benzene. The real and imaginary parts (n and k, respectively) of the complex refractive index of liquid benzene at each wavelength were determined from a two-parameter least-squares fit by Fresnel's equations of the measured reflectance at seven angles of incidence. The values obtained by this method for n and k as a function of wavelength are shown in Fig. 1. Absorption coeffi-
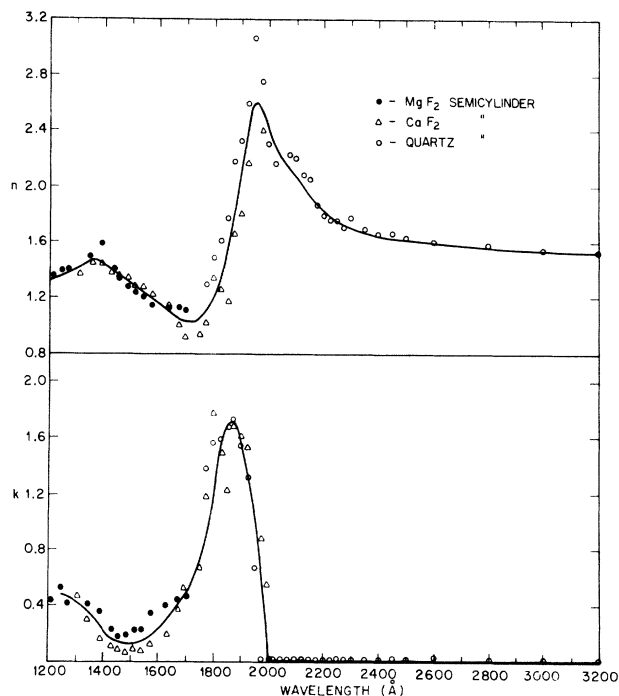
cients have previously been obtained for pure liquid benzene above 2000 Å by means of transmission measurements.[11,12] This absorption is weak and is barely detected by our techniques, which are more applicable to the region of strong absorption where transmission measurements are impossible. Some reflectance measurements have been reported[13] over the wavelength range 1400 to 2800 Å, but the optical constants were only calculated at one wavelength, and structure in the reflectance is far less pronounced than in the normal-incidence reflectance calculated from the n and k values in the present work.

The real and imaginary parts (ϵ_1 and ϵ_2, respectively) of the complex dielectric constant ϵ, where $\epsilon_1 = n^2 - k^2$ and $\epsilon_2 = 2nk$, and the calculated loss function, $-Im\epsilon^{-1}$, are shown in Fig. 2. The energy loss function $-Im\epsilon^{-1}$ describes the probability that high-energy charged particles traversing the material will lose energy, either by exciting single-electron transitions or collective volume-plasma oscillations.[14] For comparison
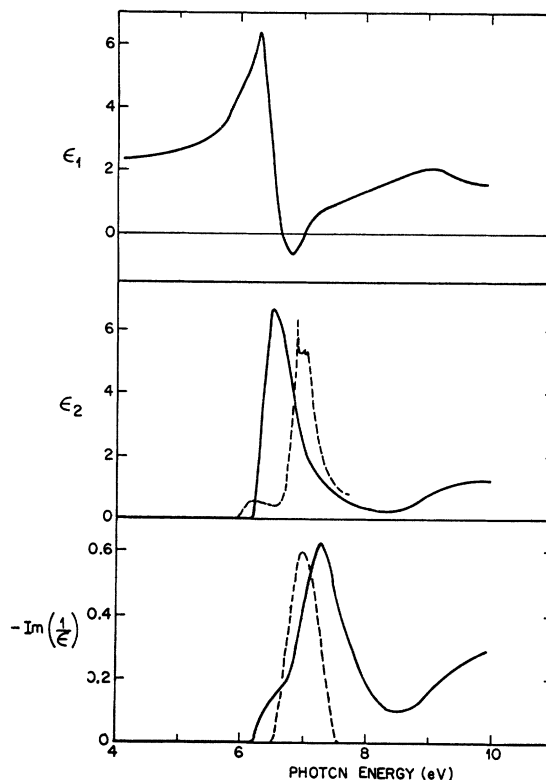

FIG. 1. Optical constants n and k of pure liquid benzene.

FIG. 2. Dielectric constants ϵ_1 and ϵ_2 and energy-loss function $-Im\epsilon^{-1}$ for pure liquid benzene (solid line). ϵ_2 for benzene vapor (dashed line) is from Ref. 2. For comparison with $-Im\epsilon^{-1}$ for the liquid, characteristic electron energy losses for benzene vapor are presented from Ref. 15. Both quantities for the vapor are on arbitrary ordinate scales.

purposes one typical set of data,[2] plotted on an arbitrary ordinate scale, is shown of ϵ_2 for the vapor. In addition, characteristic electron energy-loss measurements for benzene vapor,[15] again on an arbitrary ordinate scale, are compared with the loss function, $-\mathrm{Im}\epsilon^{-1}$, calculated for the liquid.

In general, both the characteristic electron energy-loss spectrum and the calculated loss function $[-\mathrm{Im}\epsilon^{-1} = \epsilon_2/(\epsilon_1^2 + \epsilon_2^2)]$ show structure associated with two absorptive processes: optical absorption or structure in ϵ_2; and the case when ϵ_1 goes through zero and ϵ_2 is small—that is, when the conditions are correct for the excitation of volume-plasma or collective oscillations.

In the vapor the maximum radiant absorptance occurs at about 1790 Å (6.9 eV, see Fig. 2) and has been attributed to the strongly allowed molecular excitation of the π electrons designated[5] by $^1A_{1g}^- \rightarrow {}^1E_{1u}^+$ using molecular-orbital notation and by $^1A \rightarrow {}^1B$ using Platt's free-electron–orbital model. The characteristic electron energy-loss spectrum for the vapor (Fig. 2) also shows a loss at 6.9 eV which has been associated with this molecular excitation of the π electrons.[15]

It is seen in Fig. 1 that there is a maximum in the radiant absorptance for pure liquid benzene at 1870 Å corresponding to the maximum in ϵ_2 at 6.5 eV, shown in Fig. 2. We attribute this absorption in the liquid to the same molecular excitation of the π electrons as occurs in the vapor, shifted from 6.9 eV by the perturbation of other molecules in the liquid state. The optical oscillator strength for this band in the liquid has been calculated using the sum rule

$$\int_0^{\omega_0} \omega \epsilon_2(\omega)d\omega = \tfrac{1}{2}\pi\omega_p^2 f(\omega_0).$$

This integral exhibits a plateau in the neighborhood of 8.2 eV, leading to an oscillator strength f of 2.1 ± 0.1. However, a dispersion analysis, which makes a correction (using a spherical cavity) for the polarization field due to the presence of neighboring molecules in the liquid, gives a value of 1.25 ± 0.15 in fair agreement with values reported for the same transition in the vapor.[2,4] The shoulder in the vicinity of 6.5 eV in the calculated loss function, $-\mathrm{Im}\epsilon^{-1}$, for the liquid corresponds to the structure in ϵ_2 at 6.5 eV and thus is associated with the molecular excitation of the π electrons.

The peak in $-\mathrm{Im}\epsilon^{-1}$ at 7.3 eV is thought to be associated with a volume-plasma oscillation since there is no corresponding structure in ϵ_2 and all of the conditions[16] for a plasma resonance

are satisfied at 7.3 eV. Presumably this volume-plasma oscillation involves the same electrons as are involved in the molecular excitation. Although the theoretical value of the plasma energy $(\hbar\omega_p)$ for a free-electron gas of the six π electrons per benzene molecule in the liquid is 7.5 eV, the close agreement between this and the observed resonance at 7.3 eV is coincidental, since the presence of the nearby bound state makes a free-electron approximation inappropriate.[17]

This is the first known observation of collective oscillations in a liquid insulator, although data very similar to those presented here for liquid benzene have been observed for crystalline graphite.[18] In graphite a peak in $-\mathrm{Im}\epsilon^{-1}$ near 7 eV is attributed to a collective excitation of the π electrons, while a stronger resonance near 25 eV is associated with plasma oscillations involving the combined π-plus-σ electrons. Similarly, it is anticipated that liquid benzene should show a strong collective resonance involving the π and σ electrons at some energy above the 10-eV limit of the present work.

We acknowledge R. D. Birkhoff, C. E. Klots, and H. C. Schweinler for many helpful discussions concerning the interpretation of these data.

[1]P. G. Wilkinson, Can. J. Phys. 34, 596 (1956).

[2]V. J. Hammond and W. C. Price, Trans. Faraday Soc. 51, 605 (1955).

[3]V. J. Hammond, W. C. Price, J. P. Teegan, and A. D. Walsh, Discussions Faraday Soc. 9, 53 (1950).

[4]L. W. Pickett, M. Muntz, and E. M. McPherson, J. Am. Chem. Soc. 73, 4862 (1951).

[5]J. B. Birks, L. G. Christophorou, and R. H. Huebner, Nature 217, 809 (1968).

[6]M. W. Williams, R. A. MacRae, and E. T. Arakawa, Bull. Am. Phys. Soc. 13, 1412 (1968).

[7]Reagent grade. Baker Chemical Co., Phillipsburg, N. J.

[8]L. Robinson Painter, R. N. Hamm, E. T. Arakawa, and R. D. Birkhoff, Phys. Rev. Letters 21, 282 (1968).

[9]L. Robinson Painter, thesis, University of Tennessee, 1968 (unpublished).

[10]L. Robinson Painter, R. D. Birkhoff, and E. T. Arakawa, to be published.

[11]G. N. Lewis and M. Kasha, J. Am. Chem. Soc. 67, 994 (1945).

[12]H. Damany, Bull. Am. Phys. Soc. 13, 989 (1968).

[13]C. L. Braun, S. Kato, and S. Lipsky, J. Chem. Phys. 39, 1645 (1963).

[14]R. H. Ritchie, Phys. Rev. 106, 874 (1957).

[15]A. Skerbele and E. N. Lassettre, J. Chem. Phys. 42, 395 (1965).

[16]H. Ehrenreich and H. R. Philipp, in Proceedings of the International Conference on the Physics of Semi-conductors, Exeter, England, 1962, edited by A. C.

Stickland (The Institute of Physics and The Physical Society, London, England, 1962), p. 367.

[17]H. Raether, in Springer Tracts in Modern Physics, Ergebnisse der exakten Naturwissenschaften, edited by G. Höhler (Springer-Verlag, Berlin, Germany, 1965), Vol. 38, p. 84.

[18]E. A. Taft and H. R. Philipp, Phys. Rev. 138, A197 (1965).

CRITIQUE OF A BOOTSTRAP THEORY OF PHASE TRANSITIONS

Peter J. Feibelman

Service de Physique Théorique, Centre d'Etudes Nucléaires de Saclay, 91 Gif-sur-Yvette, France

It is shown that the bootstraplike equation of Migdal's recent theory of phase transitions has a spurious solution if truncated improperly and must be handled with caution if physically sensible solutions are to be obtained.

In this note we point out that considerable care must be taken in attempting a solution of the bootstraplike equation for the three-point function which appears in Migdal's recent theory of phase transitions[1] and which is supposed to determine the two fundamental exponents of the scaling laws.[2] We show in particular that the truncated version[3] of this equation, whose first iteration Migdal evaluates, has an exact spurious solution and that in the full equation it is necessary to impose the condition that the system be at rest before one can find a unique solution. Finally, we remark that because the bootstrap equation contains less analytic information than the inhomogeneous equation to which it is an approximation, it is not clear that its solution will yield physically reasonable results.

The equation for the three-point function at a temperature T, slightly above the critical temperature T_C, of the Bose liquid is given diagrammatically in Fig. 1. Now the "bootstrap hypothesis," or in Migdal's theory the supposition that $(T-T_C)/T_C \ll 1$, is assumed in order to permit us to drop the inhomogeneous term in Fig. 1. We proceed to study the solutions of the remaining equation, which we call the bootstrap equation.

As a first approximation, we keep only the term on the right-hand side of Fig. 1, which is third order in τ. Defining

$$u(\vec{p}, \vec{q}) = G^{1/2}(\vec{p})\,\tau(\vec{p}, \vec{q})\,G^{1/2}(\vec{q})D^{1/2}(\vec{p}-\vec{q}), \qquad (1)$$

we then have simply

$$u(\vec{p}, \vec{q})$$
$$= \int d^3k\, u(\vec{p}, \vec{p}-\vec{k})u(\vec{p}-\vec{k}, \vec{q}-\vec{k})u(\vec{q}-\vec{k}, \vec{q}). \qquad (2)$$

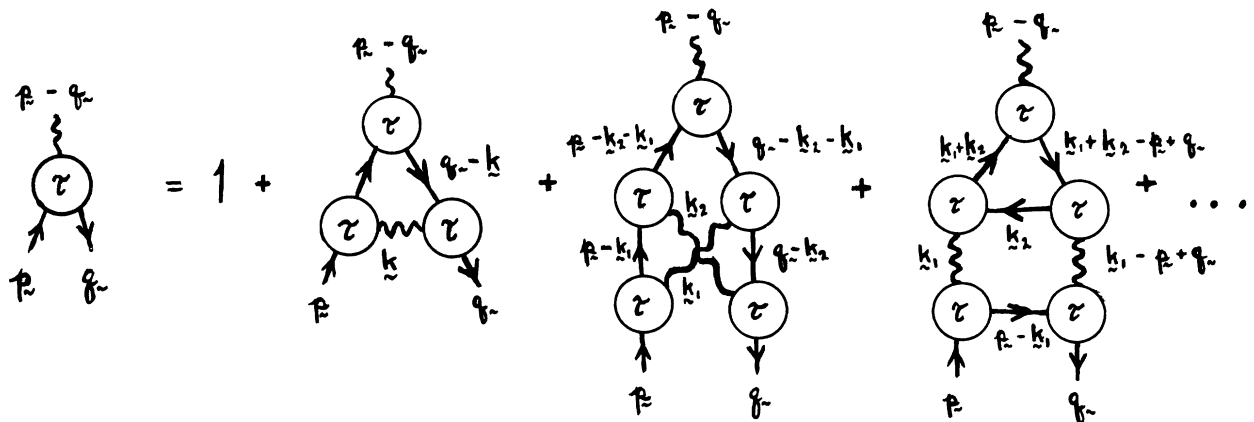Now it is easily seen that this truncated boot-

FIG. 1. Diagrammatic expression of the equation for the three-point function at a temperature T, where the solid lines represent renormalized propagators $G(\vec{p})$, the wavy lines represent the renormalized interaction $D(\vec{k})$, and τ is the unknown three-point function.