

Capillary-Wave Resonances of Electron-Hole Droplets

B. Etienne, L. M. Sander,* C. Benoît à la Guillaume, and M. Voos

Groupe de Physique des Solides de l'École Normale Supérieure,† Université Paris 7, 75005 Paris, France

and

J. Y. Prieur

Laboratoire d'Ultrasons,† Université Paris 6, 75005 Paris, France

(Received 8 July 1976)

We have studied in the gigahertz range the ultrasonic attenuation by electron-hole droplets in Ge as function of their size. A broad bump is attributed to the resonant excitation of nonresolved capillary modes. From a detailed analysis of the line shape we derive a value of 3×10^{-4} erg/cm² for the surface tension of droplets. This is the first mechanical measurement of this quantity.

Investigations of the electron-hole liquid (EHL) in Ge have revealed a detailed picture of its ground-state and thermodynamic properties.¹ However, relatively little work has been done on the collective motions of the liquid: Infrared absorption due to plasmons² and Alfvén-wave resonances³ essentially exhaust the list of oscillations observed until now. In this Letter we report on an experiment on the collective modes of the EHL which may provide an entry into the potentially fascinating field of the *hydrodynamics* of the droplets, i.e. motions analogous to ordinary fluid flow. Our observation is of ultrasonic attenuation by the EHL in Ge due to shape oscillations of droplets; we find a broad bump which we identify with nonresolved capillary resonances.

Capillary resonances are a well-known classical phenomenon.⁴ For a spherical drop of fluid with surface tension σ , a series of normal modes of oscillation exist, which involve changes of shape without changes of volume. The normal mode frequencies are

$$\nu_l = [l(l-1)(l+2)\sigma/4\pi^2\rho R^3]^{1/2}, \quad (1)$$

where $l=2, 3, \dots$. Here ρ is the mass density, and R the radius of a drop. The lowest-frequency oscillation, $l=2$, can be visualized as an oscillation between a pancake-shaped and a cigar-shaped object. The surface tension, σ , for the EHL in Ge has been calculated several times,^{5,6} and measured by rather indirect means.⁷ It is in the range of $(1-4) \times 10^{-4}$ erg/cm². The frequencies which can be estimated from Eq. (1) are thus in the gigahertz range for ordinary drops. Note that a measurement of ν_l would provide a *mechanical* measurement of σ as was first suggested some years ago.⁵

We observe the capillary waves by looking for resonant absorption of ultrasound. A particular-

ly favorable aspect of sound as a probe is that for frequencies corresponding to the first few ν_l 's the wavelength of the sound, λ , is on the order of R . This maximizes the coupling to a deformation of the drop. Other ultrasound experiments have used sound such that $\lambda \gg R$; the result is a movement of the drop as a whole.⁸

Our experimental setup is as follows. A high-purity Ge sample ($5 \times 5 \times 3$ mm³) is maintained at low temperature. The excitation is provided by an Ar laser (5145 Å) focused on an etched surface of the crystal. The laser power is reasonably large (~ 100 mW) so that we can consider⁹ that the size distribution of the drops is sharply peaked near 2 μ m. Longitudinal ultrasonic waves propagate along a $\langle 100 \rangle$ axis parallel to the etched surface. Some pains were taken to produce a very thin ultrasonic beam about 250 μ m in diameter which propagates as close to the etched surface as possible (~ 500 μ m). Standard echo pulse techniques are used to measure the attenuation. In all the results reported below the sound frequency, ν , is between 1 and 2 GHz. The temperature is usually fixed at 1.8 K.

We measure the difference between the sound absorption with and without droplets. Our goal is to search for a resonance. In order to sweep the resonance frequency we exploit its dependence on R . The laser is pulsed with pulse length 300 μ sec and rise time 1 μ sec. During the first 150 μ sec (roughly), droplets attain their steady-state radius R_0 (~ 2 μ m); after the light is turned off the radius decreases while the number of drops remains constant.¹⁰ Thus we expect ν_l to *pass through* the frequency of the sound, ν , if the drops are originally too large to resonate: Resonant absorption should be delayed with respect to the extinction of the light.

We observe the ultrasonic attenuation as a func-

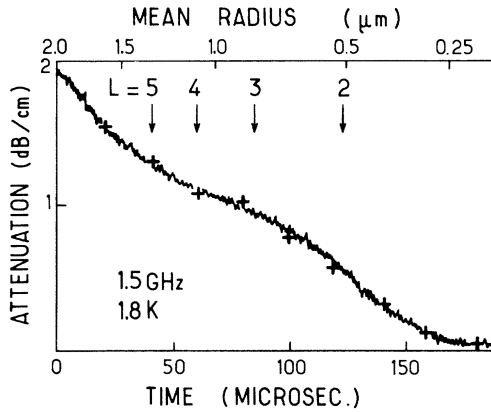


FIG. 1. Ultrasonic attenuation α as a function of time or droplets mean radius after the light is cut off. The solid line corresponds to the experimental results and the crosses to the fit obtained from Eq. (7). We sum from $l=1$ to $l=10$ with $\sigma=3 \times 10^{-4}$ erg/cm², $\gamma=1.4 \times 10^8$ sec⁻¹, $R_0=2.0$ μ m, and $\text{var}(R_0)=0.5$ μ m. In the interest of clarity only a few representative crosses are shown. The arrows indicate the position of the different capillary modes.

tion of time after cutting off the laser. As shown in Fig. 1, we do not observe a smooth decrease of attenuation but rather a bump delayed by, typically, 80–100 μ sec after the end of the pulse. This bump we identify as the capillary resonances, occurring at radii such that $\nu_l = \nu$, for the first few l 's (see below). Alternative explanations of our observations must of course be carefully explored. We do not believe that there are other collective resonances in our frequency regime which could give rise to the structure observed. The plasmon, for example, is at a much higher frequency.² The breathing modes, corresponding to density fluctuations of the drop, should occur at about 10 GHz for a 1- μ m drop if one accepts the current value of the EHL compressibility.¹¹ Incidentally, the fact that the lowest breathing mode of the drop is at much higher frequency than ν justified our use of Eq. (1) which is valid for incompressible fluids.

Our identification thus far is rather qualitative. We believe it useful to reinforce it with a more precise theory for the ultrasonic absorption which we now outline. A more detailed presentation will be published elsewhere.¹² A Lagrangian for-

malism is convenient for this calculation. We may write the Lagrangian as

$$\mathcal{L} = \int \frac{1}{2} \rho v^2 d^3r - \sigma \int dS - \int \mu n d^3r, \quad (2)$$

where μ is the chemical potential and n is the liquid density. We discuss now the first term, i.e., the kinetic energy; it provides us with a definition of the mass density to be used in this problem. From the many masses appropriate to the Ge band structure we must form combination which expresses the extra kinetic energy acquired by a portion of the fluid moving (electrons and holes together) with velocity v . A brief calculation¹² gives a "hydrodynamic" effective mass per pair which is $m = (m_l + 2m_t)/3 + (m_{lh}^{5/2} + m_{hh}^{5/2})/(m_{lh}^{3/2} + m_{hh}^{3/2})$. Here m_l and m_t are the longitudinal and transverse electron masses; m_{lh} and m_{hh} are the light-hole and heavy-hole masses. This yields $m=0.92m_0$. The first two terms of Eq. (2), the kinetic and surface energies, yield the classical theory of free capillary waves. The deformation is written $r = R + \xi Y_{lm} + O(\xi^2)$, where Y_{lm} is a spherical harmonic. The velocity field is derived from a harmonic velocity potential satisfying the boundary condition at the surface $v_r = d\xi/dt$. These two terms become $mn_0 r^3 \xi^2 / 2l - \sigma(l+2)(l-1)\xi^2 / 2$, where n_0 is the bulk density of electron-hole pairs in the EHL. The capillary-wave amplitude, ξ , is coupled to the external sound field via the third term in Eq. (2). The chemical potential may be written

$$\mu = \mu_0 + D \text{Tr}(\epsilon), \quad (3)$$

where μ_0 is the chemical potential in the absence of strain, D is a deformation potential, and ϵ the strain field. D is known to be of the order of 10 eV for Ge. The density inside a drop can be approximated by

$$n(r) = n_0 \theta(-r + R + \xi Y_{lm}) \approx n_0 [\theta(-r + R) - \xi Y_{lm} \delta(r - R_0)], \quad (4)$$

where θ is a step function. For a longitudinal sound wave

$$\text{Tr} \epsilon = \epsilon_{zz} \exp[i(kz - 2\pi\nu t)], \quad (5)$$

where $k = 2\pi/\lambda$. Combining Eqs. (3), (4), and (5) yields a coupling term from Eq. (2) of the form

$$i^l \delta_{m0} [4\pi(2l+1)]^{1/2} \xi \epsilon_{zz} D n_0 R^2 j_l(kR) \exp(-i2\pi\nu t) = F \xi, \quad (6)$$

where j_l is a spherical Bessel function and F is a generalized force. It is now straightforward to derive the equation of motion for ξ and the absorbed power, δP , which is given as usual by $\text{Re}(F^* \dot{\xi})/2$. Our final result may be expressed in terms of the absorption coefficient α per unit length and per

droplet,

$$\alpha = \sum_l [4\pi(2l+1)lD^2Rn_0j_l^2(kR)/c_{11}Sm] \gamma \nu^2 [4\pi^2(\nu^2 - \nu_l^2)^2 + \gamma^2 \nu^2]^{-1}, \quad (7)$$

where c_{11} is an elastic constant of Ge and S the sound velocity. The coefficient γ is a damping constant for the modes which we introduce phenomenologically. It can be easily shown¹² that if scattering with the Ge lattice is the dominant form of damping, our γ is the same as that measured in Ref. 8, of the order of 10^8 sec^{-1} . With this value of γ , the absorption estimated from Eq. (7) is a few decibels per centimeter, i.e., in the observed range. At this stage, there are no adjustable parameters in the estimate.

We have attempted to fit Eq. (7) to the form of the experimental results. From the luminescence decay measured in the same conditions¹³ we have taken for the mean radius of the droplets at a given time the following law:

$$R(t) = R_0 [\exp(-t/3\tau) - \exp(-t_c/3\tau)] [1 - \exp(-t_c/3\tau)]^{-1}. \quad (8)$$

Here τ is the bulk decay time ($\sim 40 \mu\text{sec}$ for our samples) and t_c is the cutoff time for droplet decay ($\sim 240 \mu\text{sec}$ at 1.8 K).¹⁴ In our fit we allow σ and γ to be parameters and we were also led to introduce a Gaussian distribution for the initial radius with a variance $[\text{var}(R_0)]$ compatible with our present knowledge.⁹ The best fit to our data is obtained with $\sigma = 3 \times 10^{-4} \text{ erg/cm}^2$, $\gamma = 1.4 \times 10^{-8} \text{ sec}^{-1}$, and $\text{var}(R_0) = 0.5 \mu\text{m}$. The agreement is excellent in our experimental range of frequencies (1–2 GHz), and we present in Fig. 1 the results for 1.5 GHz. The bump arises from an overlap of the first few l modes.

Our value of σ is within the range of current theoretical predictions. There is no particular reason for our γ to be identical to that of Ref. 8 because processes of internal friction (i.e., the EHL viscosity) could damp the capillary modes but not the translational motion. Our results indicate that the viscosity does not, in any case, give a very large additional damping. Our γ has the same general temperature dependence as that of Ref. 8, i.e., the peaks broaden with rising T . A viscosity-dominated damping would decrease as T is increased.¹⁵

Finally, we should make some further remarks about the absorption coefficient α : (i) Each term in the summation of Eq. (7) is the product of a term containing ν_l , leading to the capillary resonance, by the function $j_l^2(kR)$ whose maxima would give "geometrical resonances" for the appropriate values of kR . After summing over l these resonances become "washed out" and for large R the absorption is proportional to R^2 , i.e., the droplet surface area. (ii) For $l=1$ ($\nu_1=0$) Eq. (7) gives us exactly the results of Ref. 8 suitably generalized for the case $\lambda \sim R$. Although this "mode" does not correspond to a shape oscillation but to a translation of drops in the strain

field, we took it into account in our fit; thus the sum in Eq. (7) begins at $l=1$.

One of us (L.M.S.) would like to acknowledge helpful conversations with H. B. Shore.

*Permanent address: Physics Department, University of Michigan, Ann Arbor, Mich. 48104. Supported by the National Science Foundation.

†Laboratoire associé au Centre National de la Recherche Scientifique.

¹See, for example, C. D. Jeffries, *Science* **189**, 955 (1975); M. Voos and C. Benoît à la Guillaume, in *Optical Properties of Solids, New Developments*, edited by B. O. Seraphin (North-Holland, Amsterdam, 1976), p. 143.

²V. S. Vavilov, V. A. Zayats, and V. N. Murzin, *Pis'ma Zh. Eksp. Teor. Fiz.* **10**, 304 (1969) [*JETP Lett.* **10**, 192 (1969)].

³R. S. Markiewicz, J. P. Wolfe, and C. D. Jeffries, *Phys. Rev. Lett.* **32**, 1357 (1974).

⁴See for example L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon, Oxford, 1963).

⁵L. M. Sander, H. B. Shore, and L. J. Sham, *Phys. Rev. Lett.* **31**, 533 (1973).

⁶H. Buttner and E. Gerlach, *J. Phys. C* **6**, L433 (1973); T. M. Rice, *Phys. Rev. B* **9**, 1540 (1974); T. L. Reinecke and S. C. Ying, *Solid State Commun.* **14**, 38 (1974); J. H. Rose and H. B. Shore, *Bull. Am. Phys. Soc.* **21**, 223 (1976); P. Vashista, R. K. Kalia, and K. S. Singwi, to be published.

⁷V. S. Bagaev, N. N. Sibeldin, and V. A. Tsvetkov, *Pis'ma Zh. Eksp. Teor. Fiz.* **21**, 180 (1975) [*JETP Lett.* **21**, 80 (1975)]; R. M. Westervelt, J. L. Staehli, and E. E. Haller, *Bull. Am. Phys. Soc.* **21**, 224 (1976); B. Etienne, C. Benoît à la Guillaume, and M. Voos, *Phys. Rev. B* **14**, 712 (1976).

⁸A. S. Alekseev, T. I. Galkina, V. N. Maslennikov, R. G. Khakimov, and E. P. Shebnev, *Pis'ma Zh. Eksp. Teor. Fiz.* **21**, 578 (1975) [*JETP Lett.* **21**, 271 (1975)]; L. V. Keldysh and S. G. Tikhodeev, *Pis'ma Zh. Eksp. Teor. Fiz.* **21**, 582 (1975) [*JETP Lett.* **21**, 273 (1975)].

⁹M. Voos, K. L. Shaklee, and J. M. Worlock, *Phys. Rev. Lett.* **33**, 1161 (1974).

¹⁰This assumption is supported by the results of C. D. Jeffries, J. P. Wolfe, S. M. Kelso, R. S. Markiewicz, and J. E. Furneaux, *J. Lumin.* **12/13**, 659 (1976).

¹¹G. A. Thomas, T. G. Phillips, T. M. Rice, and J. C. Hensel, *Phys. Rev. Lett.* **31**, 386 (1973).

¹²L. M. Sander, to be published.

¹³J. C. Hensel, T. G. Phillips, and T. M. Rice, *Phys. Rev. Lett.* **30**, 227 (1973); C. Benoît à la Guillaume,

M. Capizzi, B. Etienne, and M. Voos, *Solid State Commun.* **15**, 1031 (1974); R. M. Westervelt, T. K. Lo, J. L. Staehli, and C. D. Jeffries, *Phys. Rev. Lett.* **32**, 1051, 1331(E) (1974).

¹⁴There are some difficulties with the interpretation of the cutoff time as presented in Ref. 13. These do not concern us here because only the measured value enters in Eq. (8).

¹⁵A. A. Abrikosov and I. M. Khalatnikov, *Rep. Prog. Phys.* **22**, 329 (1959).

Microwave-Enhanced Critical Currents in Point Contacts of Superconducting Aluminum

B. R. Fjordbøge, T. D. Clark, and P. E. Lindelof

Physics Laboratory I, H. C. Ørsted Institute, University of Copenhagen, Copenhagen, Denmark

(Received 14 May 1976; revised manuscript received 5 October 1976)

Enhancement of the critical current of clean point contacts made of bulk superconducting aluminum is measured when the contacts are in a 1–10 GHz microwave field. This enhancement interferes with the ac Josephson effect.

There has been much recent interest in the influence of microwave fields on the critical current of constricted superconductors. In particular the enhancement of the critical current, which is observed for a number of experimental arrangements, has received considerable attention. In this Letter we report the observation of a microwave-enhanced critical current (Dayem-Wyatt effect) in point-contact Josephson junctions.

The Dayem-Wyatt effect was first observed in thin-film constrictions.^{1–3} It was found that very small bridges³ of the order of the coherence length or smaller has a maximum relative enhancement of only 10% or less but in a temperature region which extends at least down to a reduced temperature $t = T/T_c = 0.5$, whereas larger bridges^{1,2} sometimes have very large relative enhancements but only in a temperature region very close to T_c . The Dayem-Wyatt effect has been observed also in proximity bridges of tin with gold underlay⁴ and in long thin-film strips of superconducting aluminum.⁵

It is obviously of interest to see whether the enhancement can be observed in other weak-link geometries as well. We have studied the occurrence of the Dayem-Wyatt effect in point-contact Josephson junctions. Earlier results for thin-film microbridges indicated that the enhancement was largest in films with a large residual resistivity ratio. We therefore included in our efforts also a study of clean point contact.⁶ We studied tin, indium, and aluminum contacts. The micro-

wave frequencies were in the range 1–10 GHz. Whereas we did not observe any enhancements in tin or indium, we did observe the enhancement in the aluminum point contact as evidenced in Figs. 1 and 2.⁷

In order to observe the enhancement in aluminum point contacts we indeed found that these had to be clean, i.e., with little or no oxide or dirt in the contact region. We obtained these clean contacts either by breaking a constricted wire or by passing a very large current through the constricted wire and thereby melting the constriction when immersed in the helium bath. In both cases the two halves of the wire were very accurately brought together again to form a clean contact. The point contacts were difficult to adjust and very sensitive to mechanical vibration because the clean contacts must be very small in order to have a sensible supercurrent. In order to avoid effect of Earth's magnetic field the sample holder was encased in a superconducting lead shield and the Dewar surrounded by two concentric Mumetal cans. To exclude the possibility of trapped magnetic flux the point contacts were occasionally heated for a short time to above the critical temperature, and the measurements were repeated. The microwaves were either coupled to the point contact by a loop antenna or conducted directly through the point contact, using microwave capacitors. The temperature was determined by pressure above the He bath. Thus in the case of aluminum, the temperature was not