bridge, Mass. 01518. The glass from which the array is constructed is Kimble EN1.

[18]Nuclepore Corporation, 7035 Commerce Circle, Pleasanton, Calif. 94566. This material is irradiated and etched to obtain holes.

[19]G. Sellers and A. C. Anderson, to be published.

[20]R. W. B. Stephens, Phil. Mag. 14, 897 (1932).

[21]R. Berman, Phys. Rev. 76, 315 (1949).

[22]A. C. Anderson, W. Reese, and J. C. Wheatley, Rev. Sci. Instrum. 34, 1386 (1963).

[23]R. A. Fisher, G. E. Brodale, E. W. Hornung, and W. F. Giauque, Rev. Sci. Instrum. 39, 108 (1968).

[24]At 10^7 Hz we obtain 3.19×10^5 cm/sec for the transverse acoustic velocity at 300 K, and 3.17×10^5 cm/sec at 77 K. For longitudinal waves we obtain 5.17×10^5 cm/sec at 300 K.

[25]R. O. Pohl, W. F. Love, and R. B. Stephens, in *Proceedings of the Fifth International Conference on Amorphous and Liquid Semiconductors, Garmisch-Partenkirchen, W. Germany, 1973,* edited by J. Stuke and W. Brenig (Taylor and Francis, London, 1974), p. 1121. Pohl, Love, and Stephens assumed diffusive reflection of phonons from the surface of a fiber. In similar work carried out in our laboratory, only specular reflection could be produced. This will be discussed in a future publication by L. Scherr, M. P. Zaitlin, and A. C. Anderson.

[26]G. S. Cieloszyk, M. T. Cruz, and G. L. Salinger, Cryogenics 13, 718 (1973).

[27]The calculation was carried only to ≈3 K for the sample without holes. In the model represented by Eqs. (1)–(3), the two-state systems saturate at high temperature leading to unrealistically long phonon mean free paths for low-frequency phonons. For example the calculated κ is a factor of 2.3 too large at 20 K. Other scattering mechanisms would need to be included, as discussed by A. S. Pine, Phys. Rev. 185, 1187 (1969) and J. Jackle, Z. Phys. 257, 212 (1972).

[28]A. C. Anderson and S. C. Smith, J. Phys. Chem. Solids 34, 111 (1973).

Measurement of the Spatial Distribution of Electron-Hole Drops in Ge

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We have determined the radius and spatial distribution of electron-hole droplets in Ge at 2 K excited by a 100-mW focused beam of 5145-Å light. The droplet distribution can be approximated by a uniform hemispherical cloud of radius ≈1 mm containing droplets with radii of 2.0 ± 0.5 μm. These results disagree with solutions of simple diffusion equations and with recent reports of the observations of single large droplets with radii ≈1 mm.

Since the theoretical prediction of Keldysh[1] in 1968 of the existence of electron-hole drops, and the interpretation of luminescence data in Ge in terms of this model by Pokrovskii and Svistunova[2] in 1969, there has been a considerable amount of experimental and theoretical effort expended on the problem of the gas-liquid phase transition in a nonequilibrium electron-hole plasma. Most of the work to date has been devoted to measuring and understanding quantities such as the binding energy[3-7] and the liquid density.[3,5,7-10] Although some efforts have been made to study the spatial distribution of electron-hole droplets in Ge using microwave[11] and luminescence techniques,[3,12-14] these techniques are not able to give absolute measurements of the electron-hole density, and hence are not able to provide a complete picture of the electron-hole droplet (EHD) distribution.

Recently Worlock, Damen, Shaklee, and Gordon[15] (WDSG) have shown that it is possible to determine the EHD concentration and the total density of nonequilibrium electron-hole excitation in Ge by measuring the attenuation and the scattering of light at 3.39 μm.

In this paper we report the results of measurements made using the technique of WDSG to study germanium at ≈2 K, excited by a 100-mW focused beam of 5145-Å light. We have measured (a) the radius r_0 of electron-hole drops and their spatial distribution $\gamma(\vec{R})$, and (b) the spatial distribution $\rho(\vec{R})$ of the total nonequilibrium electron-hole excitation. We find that $\rho(\vec{R})$ can be approximated by a uniformly dense hard hemisphere of radius $R_0=1.05$ mm centered on the pump spot and containing an average electron-hole density $\approx10^{15}$ cm^{-3}. From the light-scattering results we conclude that this hemisphere is a cloud of drops with radii $r_0=2.0\pm0.5$ μm and containing most of the nonequilibrium excitation. Such a spatial distribution is contrary to the predictions of simple diffusion theory[16] and is not understood

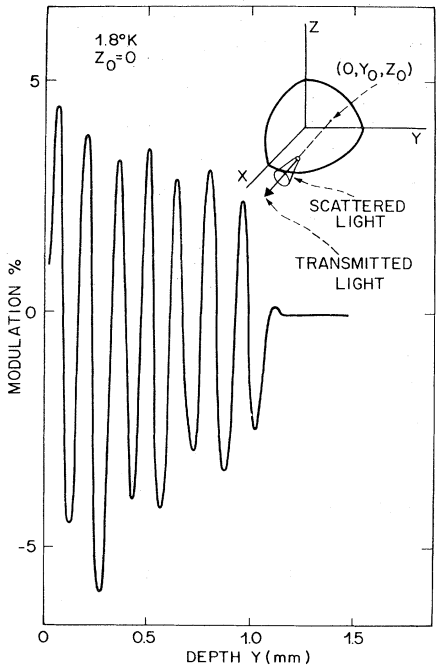

FIG. 1. Ratio of the change in sample transmission to the amplitude of the unperturbed Fabry-Perot oscillations when the pump source is shined on the sample, as a function of the depth of the probe beam in the sample. The insert shows schematically the experimental geometry. The sample surface is in the x-z plane, with the pump beam focused at the origin. One octant of the distribution is depicted by the solid curved lines, and the dashed line shows the path of the probe beam through the excited region.

at present. At lower excitation levels, the cloud shrinks in size rather than decreasing in density, and r_0 remains unchanged, while at higher temperatures (crystal not immersed in superfluid helium) the drops disappear entirely and the depth of penetration of nonequilibrium excitation is drastically reduced.

The experiments were carried out in a manner similar to that reported by WDSG, the major difference being that the exciting light is focused to a small spot (less than 50 μm in diameter) in the present case, as is indicated in the insert to Fig. 1. If we consider a coordinate system with the origin at the pump spot and x and z axes in the plane of the sample surface, with the x axis parallel to the probe beam as indicated in the insert in Fig. 1, then we have two degrees of freedom (y and z) for probing the excitation cloud.

Because the incident probe beam is affected locally and linearly by the contents of the excitation cloud, the measured signal (either scattering or attenuation) is proportional to an integral over the beam path of the local concentration:

$$S(y,z) \propto \int f(x,y,z)\,dx, \tag{1}$$

where the function f represents either the droplet concentration $\gamma(\vec{R})$ (scattering), or the total nonequilibrium electron-hole pair concentration $\rho(\vec{R})$ (attenuation).

Since the cloud has cylindrical symmetry about the y axis, it is possible to determine $f(x,y,z)$ from the measured values of $S(y,z)$ using an Abel transform.[17] However, for the present discussion, such a detailed analysis is not necessary.

The modulated transmission signal as a function of the depth y is shown in Fig. 1. As pointed out by WDSG, the modulated transmission signal has a periodic oscillatory nature due to the fractional order change of the sample transmission Fabry-Perot caused by the macroscopic refractive index of the charge cloud.

The ratio T_{real} of envelope of the modulation signal to the unperturbed Fabry-Perot modulation is a measure of the change of the real part of the macroscopic index of refraction, while the average signal (averaged over the Fabry-Perot oscillations) T_{imag} gives the change in the imaginary part. Both the envelope and the average decrease slowly with increasing y for $y<0.8$ mm and then decrease rapidly for $y>0.8$ mm.

In order to analyze this data, let us define

$$\eta' = \int_{-\infty}^{\infty} k\eta(x)\,dx,$$

where $\eta=\rho/\rho_c$ is the fractional volume occupied by the excitation if it is all condensed into droplets of density ρ_c, and k is the wave vector of the probe light beam in Ge. Then the total scattered light intensity M and the Fabry-Perot transmission T can be expressed in terms of η' as[15,18]

$$M = 7.5\eta'(kr_0)|\Delta n|^2,$$

$$T_{\text{real}} = 2\eta'\Delta n, \quad T_{\text{imag}} = 2\eta'\Delta k,$$

where Δn is the complex index of refraction of the liquid.

Figure 2 shows the values of η' obtained from both scattering measurements (M) and transmission measurements (T). The results agree with one another to better than a factor of 2, which is quite acceptable considering that M can only be reliably determined to within about a factor of 2.

The solid line in Fig. 2 is a fit to the data assuming that the droplets are uniformly distributed in a hemisphere of radius 1.05 mm, cen-

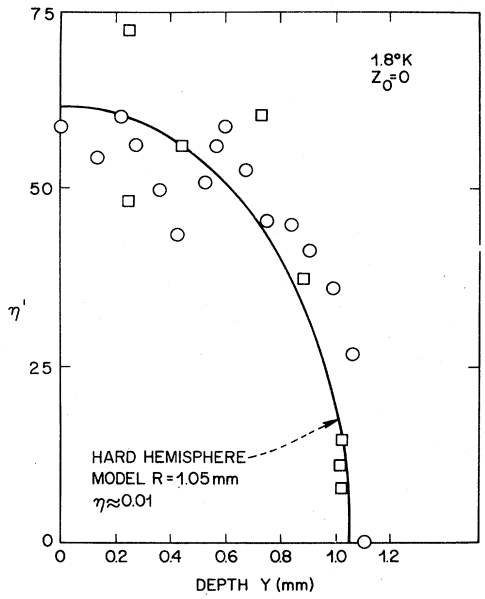

FIG. 2. η' as determined from transmission measurements (circles) and total scattered-light measurements (squares) as a function of the depth of the probe beam into the sample. The solid line shows the predicted values of η' if the droplets are assumed to be uniformly distributed in a hard hemisphere of radius 1.05 mm.

tered on the pumped spot, with $\eta \approx 1\%$. The fit is quite acceptable, and hence we conclude that at an excitation intensity of 100 mW, the droplets form a hemispherical cloud of uniform concentration, that most of the excited electron-hole pairs are probably condensed in droplets, and that the average density of electron-hole pairs in the cloud is $\approx 10^{15}$ cm^{-3}. From measurements of the angular dependence of the scattering, we find that the droplets in the cloud have a radius of 2.0 ± 0.5 μm.

We further find that with increasing excitation intensity, the radius R_0 of the cloud increases while the density in the cloud ρ and the drop size r_0 do not change appreciably. These results imply that there is a maximum number of droplets that can be supported by the excitation cloud (perhaps because of the available number of nucleation centers), that the drop radius is determined by some factor other than the excitation level (and perhaps even temperature to some degree), and that the cloud size is then determined by the drop radius and the maximum drop density.

These results disagree with the recent recent reports of observations of large single drops ($r \approx 1$ mm).[6,14,19] We feel that those experimental observations are probably consistent with our interpretation, and that the experimental techniques and theoretical models used in gathering and interpreting those data are perhaps not capable of differentiating between a uniform hemispherical cloud and one large drop. Measurements of the spatial distribution of the lumincescence[14] give only a value of R_0, but have no possibility of determining η. Dimensional resonance experiments[19] have been interpreted using approximations that are probably valid only for the case of a large single drop, and time-dependence measurements[6] have been interpreted using rate equations that do not realistically describe the system. Since the present experiments are capable of directly determining the electron-hole density, we are quite confident of the validity of our interpretation. Preliminary results[20] using pulsed excitation indicate that a similar cloud is formed following the excitation pulse.

In conclusion, we have measured the size of the cloud of electron-hole droplets for the case of point excitation by 100 mW of 5145-Å light. We find that the radius R_0 of the cloud is 1.1 ± 0.1 mm, that the cloud is filled with a nearly uniform concentration of droplets of radius $r_0 = 2.0 \pm 0.5$ μm, and that the concentration does not decay exponentially with increasing R as would be expected on the basis of a diffusion model. We further find that the macroscopic density of elementary matter in the cloud is $\simeq 10^{15}$ cm^{-3} and is relatively independent of excitation level in the range of excitation intensities of 50 to 100 mW, and that rather R_0 increases with increasing excitation intensity. And finally, the droplet radius appears to be independent of excitation intensity in the range 10 mW $< I <$ 100 mW.

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[1]L. V. Keldysh, in *Proceedings of the Ninth International Conference on the Physics of Semiconductors, Leningrad, 1967,* edited by S. M. Ryvkin and Yu. V. Shmartsev (Nauka, Leningrad, 1968), p. 1303.

[2]V. E. Pokrovskii and K. I. Svistunova, Pis'ma Zh. Eksp. Teor. Fiz. 9, 435 (1969) [JETP Lett. 9, 261 (1969)].

[3]Y. Pokrovskii, Phys. Status Solidi (a) 11, 385 (1972).

[4]J. C. Hensel, T. G. Phillips, and T. M. Rice, Phys. Rev. Lett. 30, 227 (1973); C. Benoît à la Guillaume and M. Voos, Solid State Commun. 12, 1257 (1973); T. K. Lo, B. J. Feldman, and C. D. Jeffries, Phys. Rev. Lett. 31, 224 (1973); J. C. McGroddy, M. Voos, and O. Christensen, Solid State Commun. 13, 1801

(1973); K. Fujii and E. Otsuka, Solid State Commun. 14, 763 (1974); C. Benoît à la Guillaume, M. Capizzi, B. Etrenne, and M. Voos, to be published.

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

[6]R. M. Westervelt, T. K. Lo, J. L. Staehli, and C. D. Jeffries, Phys. Rev. Lett. 32, 1051 (1974).

[7]W. F. Brinkman and T. M. Rice, Phys. Rev. B 7, 1508 (1973); M. Combescot and P. Nozières, J. Phys. C: Proc. Phys. Soc., London 5, 2369 (1972); P. Vashishta, P. Bhattacharyya, and K. S. Singwi, Phys. Rev. Lett. 30, 1248 (1973).

[8]C. Benoît à la Guillaume, M. Voos, and F. Salvan, Phys. Rev. B 5, 3079 (1972), and 7, 1723 (1973).

[9]V. S. Vavilov, V. A. Zayats, and V. N. Murzin, in *Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Massachusetts, 1970,* edited by S. P. Keller, J. C. Hensel, and F. Stern, CONF-700801 (U. S. AEC division of Technical Information, Springfield, Va., 1970), p. 509.

[10]J. C. Hensel and T. G. Phillips, in *Proceedings of the Eleventh International Conference on the Physics of Semiconductors, Warsaw, 1972,* edited by The Polish Academy of Sciences (PWN–Polish Scientific Publishers, Warsaw, 1972), p. 671.

[11]J. C. Hensel and T. G. Phillips, in Proceedings of the Twelfth International Conference on the Physics of Semiconductors, Stuttgart, 1974 (to be published).

[12]C. Benoît à la Guillaume, M. Voos, and F. Salvan, Phys. Rev. Lett. 27, 1214 (1971).

[13]R. W. Martin, Phys. Status Solidi (b) 61, 223 (1974).

[14]B. J. Feldman, Phys. Rev. Lett. 33, 359 (1974).

[15]J. M. Worlock, T. C. Damen, K. L. Shaklee, and J. P. Gordon, Phys. Rev. Lett. 33, 771 (1974).

[16]J. P. Gordon, private communication.

[17]R. S. Markiewicz, J. P. Wolfe, and C. D. Jeffries, Phys. Rev. Lett. 23, 1357 (1974).

[18]J. Irving and N. Mullineux, *Mathematics in Physics and Engineering* (Academic, New York, 1959), p. 628.

[19]J. M. Worlock, to be published.

[20]K. L. Shaklee, J. M. Worlock, R. W. Epworth, and T. C. Damen, to be published.

Nonlinear, Self-Consistent Theory of Proton Screening in Metals Applied to Hydrogen in Al and Mg†

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The density-functional formalism has been used to treat self-consistently the nonlinear screening of a proton in an electron gas. The results have been used to calculate activation energies for diffusion of hydrogen in Al and Mg. The theory is in good agreement with experimental results which are only available for Al. Hydrogen-vacancy trapping has also been investigated and was found to be a distinct possibility in the case of Al.

Apart from notable exceptions such as Pd and Ta, the solubility of H_2 in most metals is very low; for instance Al and Mg near their melting points dissolve about 1 atomic ppm[1] and 700 atomic ppm,[2] respectively, of H_2 at atmospheric pressure. In spite of this, the behavior of a proton and the surrounding electronic structure in simple metals such as these are of considerable interest since the proton is the simplest impurity, being a point ion with no complicating core-electron structure. The hydrogen molecule dissociates upon dissolving and because of the small solubilities, the screened protons may be regarded as independent impurities. We present here calculations for Al and Mg of the energy of the metal for various positions of the proton. Self-consistent, nonlinear theory has been used to treat the screening of the proton, and the lattice was taken into account using first-order perturbation theory. The results indicate that the proton at the octahedral site in Al, rather than the tetrahedral site, gives a lower energy but that the proton at a vacant site leads to a much lower energy than either of these, implying a strong tendency for proton-vacancy trapping. In the case of Mg the octahedral and tetrahedral sites had indistinguishable energies but binding to a vacancy was not indicated. The activation energies for proton diffusion were estimated and the result for Al is in good agreement with experi-