tory on the diagram depends on the mode of preparation.[3] A single short saturating pulse prepares the system at X on the diagram ($\beta_Z = \alpha_r = 0$). The subsequent relaxation is at a rate $A_Z + A_r$ parallel to one of the principal axes of relaxation, and at a much slower rate (~0) parallel to the other. Magnetic relaxation measurements observe the projection of this trajectory on the α_Z axis and this has the form of a sum of two exponential decay terms as is indeed observed experimentally.[6]

Other ways of preparing the system give different results. A long saturating pulse prepares the system at Y and relaxation occurs at the slow rate. A sudden rotation of the crystal through 180° about an axis perpendicular to the external field has the effect of switching the identities of E^a and E^b states and therefore of inverting the rotational polarization. Thus a system prepared at Y can be transferred to Y' when most of the relaxation would occur at the more rapid rate.

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Determination of the Optical Properties and Absolute Concentrations of Electron-Hole Drops in Germanium

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We report measurements of the scattering and absorption of 3.39-μm light by electron-hole drops in Ge. An unexpected Fabry-Perot effect in our samples allows us to measure not only the drop size but also their absolute concentration and the optical indices of refraction of the electron-hole condensate.

The existence of a dense liquid phase of a nonequilibrium electron-hole gas in a cold intrinsic semiconductor was predicted by Keldysh[1] and has been verified, at least in Ge and Si, by several different experiments. Analysis of the shape and separation of certain bands of recombination luminescence[2] has supported the hypothesis that in Ge the liquid phase is a two-component plasma with pair density $\sim 2\times10^{17}$ cm^{-3} and binding energy per pair ~1.8 meV. These conclusions are consistent with theoretical predictions of a stable liquid-plasma phase.[3]

Measurements of photocurrent noise[4] and infrared light scattering[5] in germanium have established that the liquid phase condenses as droplets with sizes in the 1–10-μm range. Luminescence-decay and dimensional-resonance experiments[6] suggesting droplet sizes on the order of hundreds of micrometers are, in our opinion, less direct, and subject to reinterpretation; though it is conceivable that these large drops are produced in some crystals under excitation conditions different from ours.

In our attempts to reproduce the light-scattering measurements of Pokrovskii and Svistunova,[5] we have discovered, to our surprise and delight, that we have been able to make use of a variable Fabry-Perot effect in our crystals to measure not only the droplet size but also the concentration of droplets and their infrared properties (the complex index of refaction of the liquid phase). Previous analyses of scattered and absorbed light have had to rely on assumed and derived values for the optical indices of the liquid.

Our experimental setup is similar to that reported by Pokrovskii and Svistunova.[5] A rectangular germanium crystal (n type, $\rho \geq 45\ \Omega$ cm) is immersed in superfluid helium at ~1.8 K.

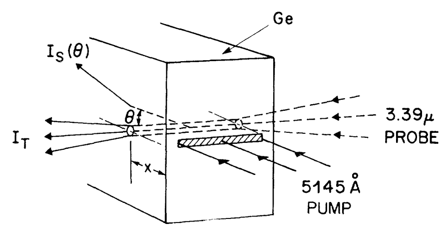

FIG. 1. Experimental configuration. The focused 3.39-μm beam probes the droplet distribution at a variable distance x behind the pumped strip.

Electron-hole pairs are produced at or near a Syton-polished surface by the absorption of pump light from a cw argon-ion laser (5145 Å). This pumped region is a strip approximately 2 mm by 0.1 mm as shown in Fig. 1. At a variable distance x behind this pumped surface, and parallel to it, we place a probe beam, obtained from a He-Ne 3.39-μm laser, and focused to a beam diameter of approximately 70 μm. There are two measurements to be made: the intensity of the directly transmitted beam, I_T; and the scattered intensity as a function of angle, $I_S(\theta)$. We measure both of these in the same apparatus, refocusing the probed region on an InAs detector with a coaxial system of germanium lenses, and defining the scattering angles with a set of circular masks. We further chop the pump beam at 1000 Hz and detect the scattering, $I_S(\theta)$, and the change in transmission, ΔI_T, with a phase-sensitive detector. These experiments can be carried out at various depths x and as a function of temperature and pump power. For brevity, we shall here limit ourselves to discussion of measurements at a single temperature ~1.8 K, and a single pump power ~100 mW. In later publications we shall deal with the application of this technique to different pump powers and configurations,[7] and also with the detailed justification of our analysis.[8]

The angular distribution of scattered intensity, $I_S(\theta)$, can be used to deduce an average droplet radius a. According to the Rayleigh-Gans theory of scattering,[9]

$$I_S(\theta)/I_T = (1/3\pi) l k\eta\, |\Delta n|^2 (ka)^3 G^2(ka\theta)\Delta\Omega, \quad (1)$$

where l is the length of the probed region, k is the optical wave vector, η is the fractional volume occupied by spherical scatterers of radius a and optical refractive index differing from that of the host medium by Δn, and $\Delta\Omega$ is the effective solid angle defined by the masks. $G^2(ka\theta)$ is a simple analytic function[9] which contains the angular dependence. The Rayleigh-Gans theory is valid when $ka \gg 1$ and $ka|\Delta n| \ll 1$. In all formulas we normalize the refractive index $n + i\kappa$ in the droplet to that of germanium ($n_{Ge} = 4$) so that $\Delta n = n + i\kappa - 1$, and compensate by using the wave vector k of the probe light in Ge ($k_{3.39} = 7.4\ \mu m^{-1}$). Scattering angles and solid angles are measured internally.

Equation (1) can be integrated over solid angle to give the total scattering efficiency:

$$I_S/I_T = \tfrac{3}{2} lk\eta(ka)|\Delta n|^2. \quad (2)$$

Our measurements of $I_S(\theta)$ basically verify those of Pokrovskii and Svistunova; we find scattering patterns consistent with Eq. (1), and droplet radii in a restricted range of values from about 2.5 to 4.0 μm. Radii seem to be independent of pump power but have a tendency to be slightly larger closer to the pumped surface.

It is clear that since both Eq. (1) and Eq. (2) contain the product $\eta|\Delta n|^2$, without a knowledge of Δn we cannot go much further. From scattering experiments alone, we can only show that η varies approximately exponentially with distance from the pumped surface, with a $1/e$ depth of ~0.3 mm. The following sections of the paper show how we can measure both Δn and η.

The optical absorption coefficient of our distribution of droplets is easily calculated in the Rayleigh-Gans approximation[9]:

$$\alpha = 2k\eta\kappa. \quad (3)$$

The total extinction coefficient contains both this absorption and the scattering losses.[10] Thus the imaginary part of the apparent macroscopic index, κ_M, in the droplet-infested region can be written as

$$\kappa_M = \eta[\kappa + \tfrac{3}{4}(ka)|\Delta n|^2]. \quad (4)$$

Our attempts to measure this attenuation were frustrated by an unforeseen Fabry-Perot effect, which, as we shall see, when correctly analyzed, gives us not only κ_M but also the real part of the apparent macroscopic index, $n_M - 1$. The real macroscopic index is very simply related to the real liquid index:

$$n_M - 1 = \eta(n-1). \quad (5)$$

With this new information, we are able to evaluate all the parameters in the theory directly from our experiments.

Figure 2 shows both I_T and ΔI_T as a function of depth behind the pumped surface. ΔI_T measures

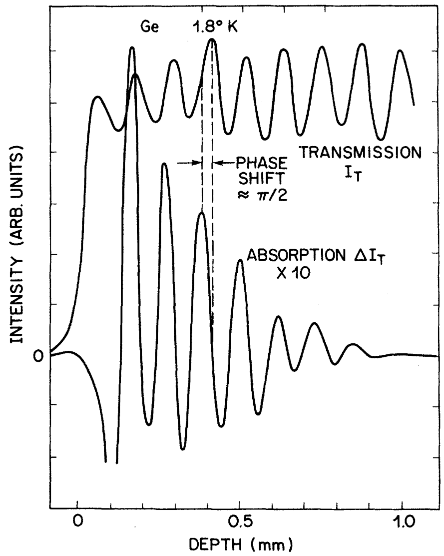

FIG. 2. Fabry-Perot oscillations in the transmitted intensity I_T and in the "absorbed" intensity ΔI_T as a function of the depth x behind the pumped strip.

the decrease in transmission when the pump beam is on. But we find that the "attenuation" oscillates violently about some depth-dependent average value and even becomes negative! Further, the period (in x) of the oscillation is consonant with that of the unmodulated intensity $I_T(x)$, and the phases differ by approximately $\pi/2$.

We explain and analyze this phenomenon as follows. The slightly nonparallel faces of the sample form an imperfect Fabry-Perot cavity whose order changes with x to give the transmission function $I_T(x)$ shown in Fig. 2. When the electron-hole drops are formed in the probed region, the apparent macroscopic index of refraction changes to $n_M + i\kappa_M$. The change κ_M causes a decrease in transmission, while the change $n_M - 1$ changes the Fabry-Perot order fractionally. κ_M therefore shifts the transmission curve down, while $n_M - 1$ shifts it laterally. $\Delta I_T(x)$ measures the result of these two changes, and is also shown in Fig. 2. In the limit of a low-finesse Fabry-Perot cavity, i.e., large wedge angle $[\alpha \gg (\sigma k)^{-1}$, where σ is the beam width and k the wave vector of the light], and with small index modulations, we can simplify the analysis and show that (1) the magnitude of the phase shift φ is given by $|\tan\varphi| = |n_M - 1|/2\kappa_M$; (2) the sign of the phase shift is determined by the sign of $n_M - 1$ and the sign of the wedge angle; (3) the mean value of $\Delta I_T(x)$, averaged locally over the Fabry-Perot oscillations, is related to κ_M by $\langle \Delta I_T(x)\rangle_x = 2kl\kappa_M \langle I_T(x)\rangle_x$; and (4) the amplitude of the Fabry-Perot excursions in $\Delta I_T(x)$ is related linearly to the amplitude of the excursions in I_T by

$$\{\Delta I_T(x)\}_{\text{amp}} = 2kl[(n_M - 1)^2 + 4\kappa_M^2]^{1/2}\{I_T(x)\}_{\text{amp}}.$$

Using these relations, we have analyzed the data shown in Fig. 2 to draw the following conclusions. (1) $n_M - 1$ is negative and varies exponentially with depth: $-2.2\times10^{-5}\exp[-x/(0.3\text{ mm})]$; (2) κ_M is about 16 times smaller, and has the same depth dependence.

We must now assume that substantially all the electron-hole pairs are condensed into droplets, so that the macroscopic-refractive-index changes can be related to the refractive indices of the liquid as shown in Eqs. (4) and (5). In order to complete the analysis, the scattering measurements must be invoked. Angular dependence gives us a droplet radius of approximately 3 μm, while the total scattering efficiency, Eq. (2), varies roughly exponentially with depth, and extrapolates to a surface value of 1.5%. Now, from the measured values of $n_M - 1$, κ_M, and the scattering efficiency, taking $l = 2$ mm, and using Eqs. (2), (4), and (5), we can calculate $n - 1$, κ, and $\eta(x)$: $n - 1 \cong -8\times10^{-4}$, $\kappa \cong 4\times10^{-5}$, and $\eta(x) \cong 3\times10^{-2}\exp[-x/(0.3\text{ mm})]$.

We have assumed that virtually all the electron-hole pairs are condensed into droplets. Measurements[11] of the equilibrium concentration of pairs in the vapor phase indicate that at 2 K this concentration is below 10^{+13} cm^{-3}. Our deduced values of condensed-pair concentrations range from 6×10^{15} cm^{-3} down to perhaps 6×10^{14} cm^{-3}, and so the assumption is reasonable.

Our measurement of $n - 1$ implies that the liquid in the droplets is responding at the infrared laser frequency as a plasma, with a dielectric constant less than unity, and a plasma frequency of 120 cm^{-1}. The plasma frequency obtained here compares very well with that obtained from direct infrared measurements by Vavilov, Zayats, and Murzin[12] (130 cm^{-1}), and also with that calculated from the accepted liquid density, 2×10^{17} cm^{-1}, and an optical effective mass[13] of $\frac{1}{20}$.

The value of κ we find here implies that the plasma is rather heavily damped at this frequency: Since $\omega\tau = |n-1|/\kappa \sim 20$, the implied scattering time τ is only 4×10^{-14} sec. Alternatively, we can interpret this lossy index in terms of a cross section per hole for inter-valence-band transitions. This interpretation gives an absorption cross section of 3×10^{-17} cm^2 which is about 3 times smaller than that observed in doped p-type germanium at 3.4 μm by Kaiser, Collins, and Fan.[14]

Finally, we discuss the density distribution $\eta(x)$. We believe that the spatial dependence can probably be understood in a model in which pairs diffusing out from the surface are captured by existing droplets, with very little decay in transit. If we assume that the droplets are distributed in a hemicylindrical cloud in which the concentration decays with exponential decay length 0.3 mm in all directions from the center of the cylinder, then we have approximately 5×10^{12} electron-hole pairs. If 50% of our pump laser power, or 50 mW, is absorbed, the electron-hole generation rate is $\sim1.5\times10^{17}$ sec^{-1}. The ratio of steady-state population to generation rate implies a lifetime of 30 μsec, which is in close agreement with measured values of the pair lifetime in the liquid state.[15]

At lower pump power levels the exponential decay depth decreases while the density of droplets near the pumped region is unchanged. Preliminary measurements with a spot-focused pump beam indicate that the droplet density is not exponential, but is quite uniform in a hemispherical cloud with rather sharp edges.[7]

We have made use of a Fabry-Perot interference effect in probing the scattering and absorption of electron-hole droplets in germanium. We have been able to deduce values for the optical constants of the liquid state which are in good agreement with other measurements and theories, and to find the absolute concentration of electron-hole pairs as a function of depth behind the optically pumped surface. We believe that this technique will be useful, not only in probing the optical properties of the liquid at other frequencies, but also in rapidly measuring the spatial variation of droplet concentrations in other pumping configurations, both optical and injection.[16]

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