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LIFETIME AND RADIATIVE EFFICIENCY VS DENSITY IN THE STRAIN-CONFINED ELECTRON-HOLE LIQUID IN Ge

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ABSTRACT

We have performed two experiments on the electron-hole liquid (EHL) in inhomogeneously stressed Ge in order to obtain information on the lifetime and radiative efficiency as a function of e-h pair density in the strain-confined electron-hole liquid (SCEHL). Our data can only be explained if a density-independent recombination mechanism is the most important decay process. We show that our result is consistent with other experiments indicating that such a mechanism has a negligible effect on the recombination time in unstressed Ge.

The electron-hole liquid (EHL) in semiconductors has been the subject of much experimental and theoretical study over the last decade.[1] This Fermi fluid is formed when photoexcited electrons, holes, and free excitons (FE) condense at low temperatures. In unstressed Ge the liquid phase consists of a fog of electron-hole drops (EHD) each several μm in size.[1] By suitably stressing a Ge crystal, a three-dimensional energy minimum for the EHL is formed in the interior of the crystal.[2] In this case, photoexcited carriers, FE, and EHD are accelerated into this strain well and coalesce into a single large electron-hole drop; the liquid is then a strain-confined electron-hole liquid (SCEHL). The drop size varies with excitation level[3,4] and can be varied from $R \lesssim 50$ μm up to the size of the well, $R \approx 700$ μm in some cases.

It has been found[5,6] that in the SCEHL the electron-hole pair density varies with position. At first this is a surprising result, since a liquid is generally considered to have a constant density. However, the density variation in the SCEHL can be understood rather simply[7] by noting the following: the strain energy is parabolic[2] with the distance from the center of the well, $E_S \propto \alpha r^2$; the chemical potential is uniform throughout the liquid volume; and the gas pressure outside the liquid and the surface pressure can both be neglected at low temperature. These assumptions give to first order[7] a density distribution

$$n(r) = n_o[1 + \hat{\alpha}(R^2 - r^2)] \quad . \tag{1}$$

Here $n(r)$ is the density at the position r, R is the drop radius, $\hat{\alpha} = \alpha/(n_o^2 E_o'')$, $E_o'' = \left.\frac{d^2E}{dn^2}\right|_{n_o}$, and n_o is the equilibrium density. A more

exact theoretical treatment[6] gives a result similar to Eq. (1) with α a function of R. Thus the SCEHL is compressed by the strain gradient, resulting in an increased density at the center of the drop which falls off to the equilibrium value at the surface. For relatively small drop size, $R \lesssim 150$ μm, the density variation is $\lesssim 20\%$, depending on the parameters of the experiment, particularly the strain parameter α. For these small drop sizes, then, the equilibrium properties of the SCEHL can be studied.[3] For the largest drop sizes, $R \approx 700$ μm, density increases of up to a factor of three have been observed by directly measuring the density profile.[5,6] The agreement between experimental and theoretical density profiles is good.

At first, the density variation may seem like a complication in understanding simple experimental results such as the total luminescence decay time and lineshape vs excitation level, because the observed luminescence represents a superposition from liquid in all parts of the strain well. However, once the density variations are understood, these experiments can be simulated theoretically, and different models can be tested. In fact, the changes in density with drop size can be utilized in studying the properties of the SCEHL over an exceptionally wide range of densities simply by varying the drop size, i.e. the excitation level

A second method to vary the density at a fixed temperature is to apply an external magnetic field. Experiments on unstressed Ge showed that the density oscillates with magnetic field as electron Landau levels pass through the Fermi level.[8] For the strain-confined EHL, if a sufficiently small drop size is chosen, a uniform density can be varied directly without the complications of compression. In this type of

experiment density oscillations of ~ 10% can be obtained for $H \lesssim 20$ kOe.

In this paper we will discuss the dependence of the various recombination processes on density. The total recombination rate is assumed to vary with density as follows:

$$\tau^{-1}(n) = \tau_{di}^{-1} + \tau_{rad}^{-1}(n) + \tau_{Aug}^{-1}(n) \tag{2}$$

$$= A + Bn + Cn^{s} \quad .$$

Here the first term represents processes independent of density; the second term represents the usual radiative recombination processes; and the third term represents Auger processes, where s is expected to be 2 or 3. Efficiences for these processes can be defined as follows:

$$\begin{aligned} \varepsilon_{di}(n) &= \tau(n)/\tau_{di} \\ \varepsilon_{rad}(n) &= \tau(n)/\tau_{rad}(n) \\ \varepsilon_{Aug}(n) &= \tau(n)/\tau_{Aug}(n) \end{aligned} \tag{3}$$

The experiments were performed on a 4x4x3 mm^3 sample of ultra-pure, $N_A \leq 10^{11}$ cm^{-3}, dislocation-free Ge grown by Hansen and Haller.[9] The sample was cut along crystallographic axes and etched in $3HNO_3$:HF. The stress was applied from above along a ⟨111⟩ axis by a rounded nylon plunger in the inhomogeneous geometry described in Refs. (2) and (3).

The experimental setup was standard: a mechanically chopped Argon-ion laser or a pulsed GaAs laser was focussed onto a (110)-face of the Ge sample immersed in pumped liquid helium within a superconducting solenoid. Luminescence from the LA phonon replica at about 707 meV was collected from the opposite face, focussed onto the

front slit of a spectrometer, and detected by a cooled high-sensitivity Ge photodiode. A signal averager or boxcar was used to obtain data as a function of time with 10 μsec resolution.

Two types of experiment will be discussed here. In the first experiment, the luminescence was recorded as a function of time after laser cutoff for a series of different excitation levels. The luminescence was collected from the entire drop, without spatial or spectral selection. Because the average density increases with drop size, the decay is non-exponential (see for example Ref. (3)), and therefore the initial decay time τ_i is of interest. In addition, in order to eliminate complications arising from variations in production efficiency with power, the initial decay time was recorded as a function of initial drop size. The drop size was estimated as follows: the focussed luminescence image of the crystal was translated past a spectrometer slit, thus producing a spatial slit scan of the luminescence. The full width at half maximum (FWHM) of the slit scan is then used as an estimate of the drop size.[3,6]

The results of this experiment are shown as the dots in Fig. 1, where the initial decay time τ_i of the total EHL luminescence is plotted as a function of the initial FWHM drop size. It should be noted that the equilibrium density and lifetime for this sample

$$n_o \approx 0.50 \times 10^{17}\ \mathrm{cm}^{-3}$$
$$\tau(n_o) \approx 500\ \mu\mathrm{sec} \qquad (4)$$

are typical of the SCEHL in dislocation-free Ge.[3,4,10,11,12]

The curves in Fig. 1 represent calculations of the initial luminescence decay time vs drop size, using calculated density distributions[6]

which are in good agreement with the measured profiles,[6] and using different values for the parameters in Eq. (2). The dot-dashed curve represents $\varepsilon_{di} = 0.$, $\varepsilon_{rad}(n_o) = 0.75$, $\varepsilon_{Aug}(n_o) = 0.25$, and $s = 2$. This model was formulated using experimental indications[3] that

$$\varepsilon_{rad}(\text{SCEHL}) \approx 3\ \varepsilon_{rad}(\text{unstressed EHL}) \tag{5}$$

and the evidence obtained by Betzler et al.[8] that A is negligible and that $\varepsilon_{rad} \approx 25\%$ for unstressed Ge. This model is clearly unsatisfactory; the initial decay time decreases too much as the average density increases, indicating that the Auger term is too large. Thus an Auger process is not dominant here, as is believed to be the case in unstressed Ge.[8,13] The dashed curve represents $\varepsilon_{di} = 0.$, $\varepsilon_{rad}(n_o) = 1.0$, and $\varepsilon_{Aug}(n_o) = 0$. In this model, the change in τ_i^{-1} with drop size corresponds to the change in a kind of average density, as can be seen qualitatively from Eq. (2). This model is also unsatisfactory. The solid curve represents $\varepsilon_{di} = 0.75$, $\varepsilon_{rad}(n_o) = 0.24$, $\varepsilon_{Aug}(n_o) = 0.01$, and $s = 2$, which is an approximate fit to the data. It is evident that a dominating density independent recombination mechanism is necessary to explain this data.

Additional evidence of the importance of the density independent recombination mechanism can be obtained from a second experiment. In this experiment the luminescence intensity from the entire drop was monitored as a function of magnetic field at discrete times after a pulse from a GaAs laser. Under these conditions the drop grows very rapidly[12] to a maximum size and then decays in the same way as a drop formed by cw excitation. The total energy per pulse was chosen to produce a drop with radius $R \approx 125\ \mu m$. The magneto-oscillations of the luminescence at

several delay times are shown in Fig. 2, for $H \leq 18$ kOe. It can be seen that at $t = 1$ msec the oscillations are reduced in amplitude but have not changed sign, where $t = 1$ msec is approximately twice $\tau(n_o)$. This is in contrast to the case for unstressed Ge, where the oscillations change sign after approximately 1/3 of the zero-field decay time.[8]

To understand the significance of this result it is necessary to consider the kinetics of a pulsed experiment. Because the drop lifetime is much longer than the laser pulse width, ≈ 100 nsec, the drop initially contains a constant number $N(t=0)$ of e-h pairs, assuming the pumping efficiency is independent of magnetic field. The luminescence intensity as a function of time is given by

$$i(H,t) = \frac{N(t)}{\tau_{rad}(n)} = BnN(0)e^{-t/\tau(n)} \quad . \tag{6}$$

At a magnetic field for which the equilibrium density is higher than the zero field value n_o, the initial luminescence intensity is greater than $i(0,0)$ and the lifetime $\tau(n)$ is shorter than $\tau(n_o)$. Therefore at some later time t_T

$$i(0,t_T) = i(H,t_T) \quad ; \tag{7}$$

at this turnover time the magneto-oscillations change sign. Assuming $n(H) - n_o << n_o$ a straightforward analysis yields

$$t_T^{-1} = Bn_o + sCn_o^s \quad . \tag{8}$$

This result combined with equation (2) yields

$$\frac{\tau(n_o)}{t_T} = \epsilon_{rad}(n_o) + s\epsilon_{Aug}(n_o) \equiv \xi \ . \tag{9}$$

The quantities ξ and

$$1-\xi = \varepsilon_{di} - (s-1)\ \varepsilon_{Aug}(n_o) \tag{10}$$

appear throughout the analysis of magneto-oscillatory phenomena. Other combinations of recombination efficiencies cannot easily be obtained from these experiments. If $\xi < 1.0$, it can be seen from Eq. (10) that the density independent recombination mechanism is more important than the Auger mechanism. Conversely, if $\xi > 1.0$, the Auger mechanism is more important than the density independent mechanism (for $s = 2$). The data displayed in Fig. 2 give direct information about ξ. Since the turnover time

$$t_T \gtrsim 2\tau(n_o) \quad , \tag{11}$$

we conclude that

$$\xi \lesssim 1/2 \quad . \tag{12}$$

In addition, from Eq. (10) we find that

$$\varepsilon_{di} \gtrsim .50 \quad , \tag{13}$$

in agreement with the results of Fig. 1. We find from the data of Betzler et al.[8] that $\xi \sim 2$ for the EHL in unstressed Ge. This indicates that the Auger mechanism dominates for this case.

Betzler et al.[8] concluded that the density independent recombination efficiency was less than 10% of the Auger efficiency and therefore set $\varepsilon_{di} = 0$. However, we note that if $\varepsilon_{di} = 0.05$ and $\tau = 36$ μsec for unstressed Ge this corresponds to a value of

$$\tau_{di} = \tau/\varepsilon_{di} = 720\ \mu\text{sec} \quad . \tag{14}$$

For the strain-confined EHL, the same density independent decay time corresponds to

$$\varepsilon_{di} = \tau(n_o)/\tau_{di} = 0.69\ (\text{SCEHL}) \tag{15}$$

using $\tau(n_o) = 500\ \mu\text{sec}$. It is clear that such a process which can be neglected in unstressed Ge can at the same time dominate the decay time for the SCEHL.

We believe that a density independent recombination time of several hundred μsec is reasonable for Ge. Possible recombination sites include shallow traps and deep traps. A detailed calculation is beyond the scope of this paper. However, crude estimates can be made of the required concentrations of shallow or deep recombination centers. First, we note that several groups [14-16] have measured the EHL decay time in doped (unstressed) Ge. While there is some variation in the results, the lifetime generally starts to decrease when the impurity concentration N_i is greater than $\sim 10^{15}$-10^{16} cm^{-3}. For example, Zhurkin et al.[16] report $\tau = 24\ \mu\text{sec}$ for $N_i = 2 \times 10^{16}\ cm^{-3}$ As impurities. If the e-h pair density remains constant at these doping levels, the decrease in lifetime may be attributed to impurity-induced recombination. The change in lifetime then corresponds to $\tau_{di} \sim 70\ \mu\text{sec}$ for $N_i = 2 \times 10^{16}\ cm^{-3}$. Assuming further that this recombination rate is proportional to the impurity concentration, $\tau_{di} \sim 700\ \mu\text{sec}$ would correspond to $N_i \sim 2 \times 10^{15}\ cm^{-3}$ of shallow impurities. An estimate can also be made for deep levels, where nonradiative

recombination could take place via multiphonon emission. Experimental values for nonradiative capture cross-sections at room temperature have been tabulated for Ge[17] and range from $\sigma_{cap} \sim 10^{-14}$-$10^{-16}$ cm^2. A simple model predicts that the capture rate is $\sim \sigma_{cap} \times v \times N_i$, where v is the carrier velocity which can be taken to be the Fermi velocity for carriers in the EHL. In this case $\tau_{di} \sim 1$ msec would correspond to $N_i \sim 10^{18}$ cm^{-3}. Our samples have not been fully characterized to this level, although it is known that the concentration of shallow impurities is $\lesssim 10^{11}$ cm^{-3} and that levels are present associated with hydrogen.[18] Thus the study of the SCEHL in differently prepared samples may prove to be a sensitive test of impurity-induced recombination.

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FIGURE CAPTIONS

Fig. 1. Initial decay time τ_i for the total luminescence intensity at $\approx$ 707 meV plotted vs drop size estimated by the full width at half maximum (FWHM) of a slit scan, for the strain-confined electron-hole liquid (SCEHL) in Ge. Sample CR50 was inhomogeneously stressed along a $\langle 111 \rangle$ axis. T = 1.9 K. The dots are the experimental results; the three curves are calculations discussed in the text.

Fig. 2. Total luminescence intensity as a function of magnetic field H for several different delay times after an excitation pulse, for the same sample as in Fig. 1. $H \parallel \langle 1\bar{1}0 \rangle$, T = 1.6 K. The vertical scale is offset from zero by an arbitrary amount. The symbols at the right indicate 10% of the total intensity.

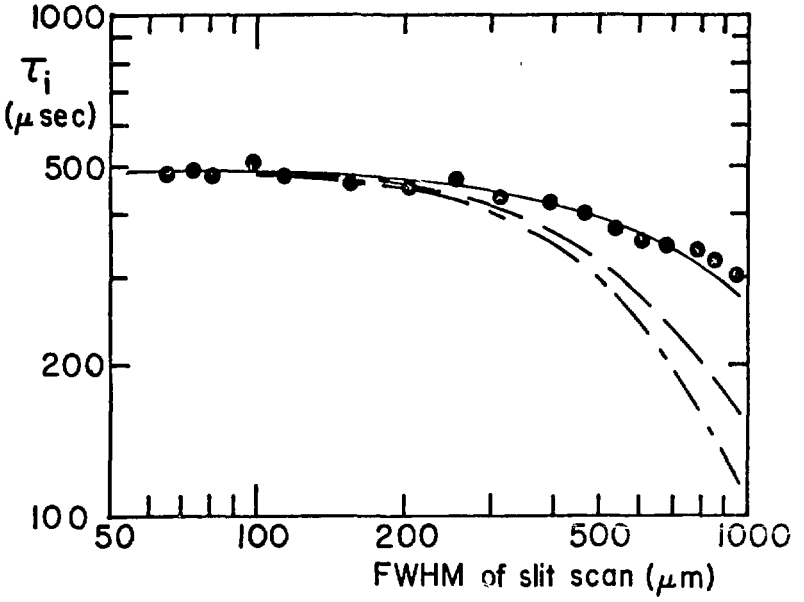
1000
τ_i
(μsec)
500
200
100
50
100
200
500
1000
FWHM of slit scan (μm)

Fig. 1

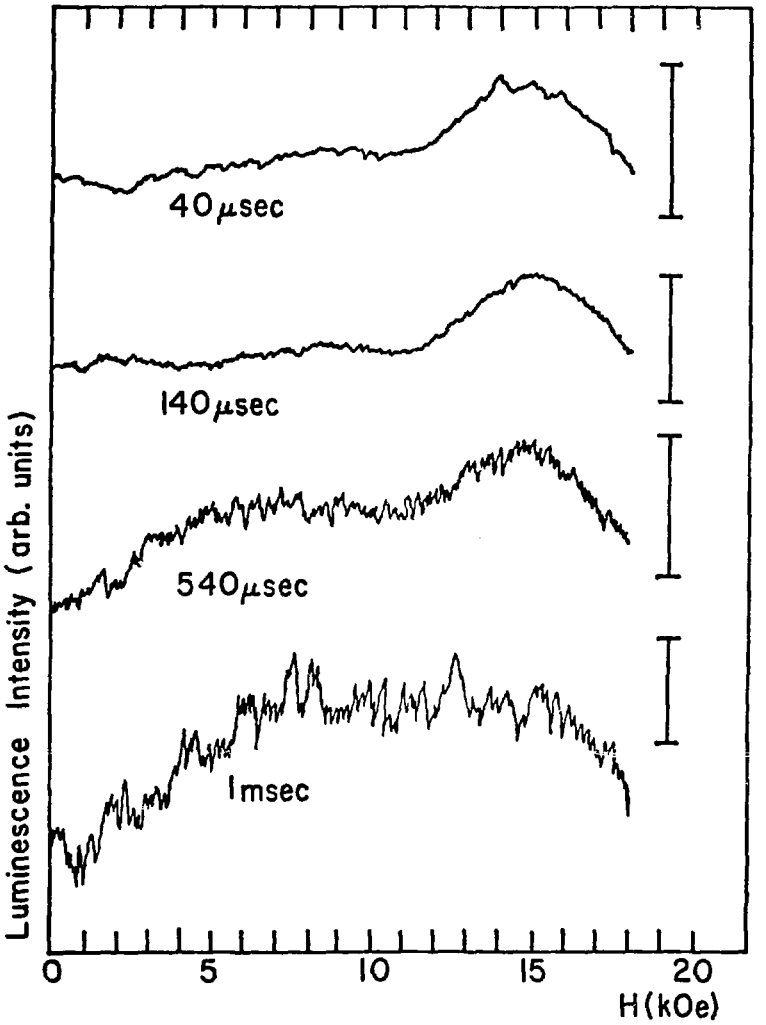
40μsec
140μsec
540μsec
1msec
Luminescence Intensity (arb. units)
0
5
10
15
20
H(kOe)

Fig. 2