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DENSITY DEPENDENCE OF AN ELECTRON-HOLE LIQUID
CORRELATION FACTOR IN Ge: EXPERIMENT*

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Density Dependence of an Electron-Hole Liquid Correlation Factor in Ge: Experiment

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(Received )

We report the first absolute measurement of the density dependence of the enhancement factor $g_{eh}(o)$ for the electron-hole liquid (EHL) in Ge. This factor $g_{eh}(o)$ is a measure of the electron-hole spatial correlation function, and provides a valuable and sensitive test for the predictions of various many body approximations. Our data show a steep rise in $g_{eh}(o)$ at lower EHL densities as does the most sophisticated theoretical calculation to date. Quantitatively, however, there is disagreement with theory.

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A uniquely useful physical system for testing the results of many body theory approximation schemes is the electron-hole liquid (EHL). The EHL is a two-component (electron-hole) Fermi liquid existing in optically excited semiconductors at liquid helium temperatures. Other Fermi liquids such as neutron stars, nuclear matter, and electrons in metals suffer from various disadvantages when used to test many body theory approximations: the experimenter cannot alter parameters; the number of constituent particles is small; many body effects are small corrections; or the system's characteristics are not known exactly for the purpose of theoretical calculation. In this experiment we determine the enhancement factor $g_{	ext{eh}}(0)$ of the EHL as a function of EHL density. A free exciton (FE) gas and at most one EHL droplet are confined to a strain induced potential well at temperature $T = 2.16\,\text{K}$ in an ultrapure Ge crystal. The EHL density is varied by stressing the crystal. The enhancement factor is the electron-hole (e-h) spatial correlation function evaluated at zero e-h separation and normalized to the average plasma density. This measurement of the correlation function provides a sensitive and valuable test for the predictions of many body approximation schemes. Several calculations for the EHL have yielded varying predictions for the density dependence of $g_{	ext{eh}}(0)$ while still being in reasonable agreement on EHL densities and ground state energies.

One experiment relevant to this work has been published. To estimate semiquantitatively the density dependence of a quantity proportional to $g_{	ext{eh}}(0)$, the authors assume a sample independent model for EHL decay. At a stress and density at which they measure an EHL lifetime
\( \tau_2 \approx 0.5 \text{ ms} \) we measure \( \tau_2 \approx 1 \text{ ms} \) in clear contradiction to the expectations of a sample independent model. Their decay model is also inconsistent with previous work.\(^7\)

Our method of determining \( g_{e\text{h}}(o) \) has the advantage of being independent of EHL recombination models. The enhancement factor is related to the probability of an electron being at the site of a hole and thus to the radiative decay rate. From a treatment\(^8\) of the LA phonon assisted FE and EHL recombination rates we have

\[
 g_{e\text{h}}(o) = \frac{\tau_{rX}}{\tau_{rL}} \frac{\left| \psi_X(o) \right|^2}{n_L} \tag{1}
\]

where \( \tau_{rX} \) and \( \tau_{rL} \) are the LA phonon assisted FE and EHL radiative lifetimes and \( \psi_X(o) \) is the FE wave function evaluated at zero e-h separation.

Radiative lifetimes are difficult to measure so we introduce the FE and EHL radiative efficiencies \( \varepsilon_{rX} = \tau_x/\tau_{rX} \) and \( \varepsilon_{rL} = \tau_L/\tau_{rL} \) into Eq. (1):

\[
 g_{e\text{h}}(o) = \left( \frac{\varepsilon_{rL}}{\varepsilon_{rX}} \right) \frac{\tau_x}{\tau_L} \frac{\left| \psi_X(o) \right|^2}{n_L} \tag{2}
\]

All terms on the right-hand side of this equation are accessible either experimentally or theoretically. From effective mass theory using s-wave energy bands \( \left| \psi_X(o) \right|^2 = 1/(\pi a_X^3) = 5.7 \times 10^{16} \text{ cm}^{-3} \) where \( a_X \) is the exciton Bohr radius in the high stress limit. This is the appropriate value because our lowest stress data were taken at \( \sigma = 6.5 \text{ kgf/mm}^2 \).\(^9\) The lifetimes \( \tau_x \) and \( \tau_L \) are measured from luminescence decay (see Fig. 1), the EHL density \( n_L \) from luminescence lineshape fits,\(^10\) and the ratio of radiative efficiencies from the experiment described below.

We perform a steady state experiment measuring both FE and EHL luminescence intensity, \( I_x \) plus \( I_L \), as a function of e-h pair generation
rate $G$ (see Fig. 2). Below EHL threshold the number of free excitons $N_x$ is determined by the steady state equation $G = N_x/\tau_x$. The measured FE luminescence intensity is related to $N_x$ by $I_x = (N_x/\tau_x)\varepsilon_{cx}$ where the FE collection efficiency $\varepsilon_{cx}$ is the ratio of the luminescence collected and detected to that emitted. An EHL collection efficiency $\varepsilon_{cl}$ is similarly defined. Combining equations, the slope $dI/dG$ below EHL threshold is $\varepsilon_{rx}\varepsilon_{cx}$. Above EHL threshold both FE and EHL exist, and the number of e-h pairs $N_x$ in FE and $N_1$ in the EHL are related to the generation rate $G$ by the steady state equation $G = N_x/\tau_x + N_1/\tau_1$. In a manner similar to the FE case, $I_1 = (N_1/\tau_1)\varepsilon_{cl}$. Combining equations, with the experimental observation (see Figs. 1 and 2) that $N_x$ = constant just above EHL threshold, we find the slope $dI/dG$ just above threshold is $\varepsilon_{rl}\varepsilon_{cl}$. Taking the ratio of these slopes just above to just below threshold we have

$$\frac{(dI/dG)_{\text{above}}}{(dI/dG)_{\text{below}}} = \frac{\varepsilon_{rl}}{\varepsilon_{rx}} \frac{\varepsilon_{cl}}{\varepsilon_{cx}}$$

(3)

An optical hysteresis in EHL droplet formation which would complicate the above discussion has not been observed for the strain confined FE, EHL droplet system we use. Upon separately measuring the ratio of the FE and EHL collection efficiencies, we obtain the desired ratio of radiative efficiencies. A pumping efficiency relating excitation power to e-h pair generation rate $G$ has been found to be constant over the range of excitation powers used and is thus neglected in the above discussion.

An assumption inherent in the use of both steady state and decay measurements for the terms in Eq. (2) is the existence of a thermal and diffusive quasi-equilibrium within the FE, EHL system. To compare steady
state and decay measurements, these measurements must be made on the same physical system. To ensure quasi-equilibrium three criteria must be satisfied: (a) The e-h pairs in the EHL must be in thermodynamic equilibrium. (b) The FE gas must be in thermodynamic equilibrium with itself. (c) The FE gas must be in thermodynamic equilibrium with the EHL droplet. If any of these conditions are not met, then equilibrium thermodynamics is not sufficient, and transport theory must be considered.

Given a carrier-phonon scattering time $\tau_p \sim 1$ ns the FE, EHL system is well characterized by the lattice temperature for the low excitation powers ($\mu W$) and long time scales (ms) of this experiment. For the FE gas to be in spatial equilibrium with itself, the FE diffusion length $L_x = \sqrt{D_x \tau_x}$ must be large compared to the spatial extent of the FE gas. If we approximate the bottom of the strain induced potential well with a parabola $U = \alpha r^2$, the spatial extent of the FE gas is characterized by $\alpha (\Delta r)^2 = k_B T$ or $\Delta r = \sqrt{k_B T/\alpha}$. Taking worst case values of $\alpha$ and $\tau_x$ from our data, we find $L_x/\Delta r \approx 450$ so the FE gas is in spatial equilibrium with itself to a very good approximation.

To ensure thermodynamic equilibrium between the EHL droplet and the FE gas, the net flux of FE out of (for decays) or into (for steady state) the EHL droplet must be small compared to the equilibrium flux of e-h pairs back and forth across the EHL droplet surface. Simple thermodynamic and kinetic arguments$^{11}$ give this equilibrium flux

$$J_\infty = 4\pi m_x/h^3 (k_B T)^2 S \exp(-\phi/k_B T)$$

for the infinite lifetime limit where $J_\text{in} = J_\text{out} = J_\infty$. Here $m_x$ is the FE translational mass; $\phi$ is the EHL ground state binding energy per e-h pair; and $S$ is the absorption probability for a FE incident on the EHL droplet. For decays the EHL droplet
acts as a FE source which keeps the number of FE approximately constant for small drop sizes, thus \(4\pi R_x^2 J_{\text{out}} - J_{\text{in}} = N_x/\tau_x\) where \(R_x\) is the droplet radius. For steady state excitation a FE flux inward counters the droplet's bulk recombination loss of carriers, thus \(4nR_1 (J_{\text{out}} - J_{\text{in}}) = (4/3)\pi R_x^3 n_x/\tau_x\). To treat safely these systems as in quasi-equilibrium, we must have \(|(J_{\text{out}} - J_{\text{in}})/J_\infty| << 1\). Worst case steady state and decay parameters from our data yield \(|(J_{\text{out}} - J_{\text{in}})/J_\infty| \leq 5 \times 10^{-3}\) which justifies the use of decay and steady state measurements together in Eq. (2) for \(g_{\text{eh}}(o)\).

Finally, we note that the \(g_{\text{eh}}(o)\) measured here are not altered by the compression\(^1\) in the strain well since a worst case (highest stress) estimate yields a 5% average compression, and most cases are much better.

The enhancement factors determined from our data are plotted with error bars in Fig. 3 as a function of \(r_s\) where \(r_s = \left[\frac{3}{(4\pi n_x)}\right]^{1/3}/a_x\). The solid curves are the results of several many body approximations\(^5\) for a model system assuming isotropic electron and hole bands. The lower two curves show the Hubbard and RPA predictions. The upper two curves are the results of more sophisticated approximations including multiple scattering between the plasma components. These last two curves come closest to the fast monotonic increase of our data with \(r_s\).

We propose several possible explanations for the lack of a closer agreement. There are details of the energy band structure, to which \(g_{\text{eh}}(o)\) may be sensitive, that have not been included in the most sophisticated theoretical calculations of the density dependence of \(g_{\text{eh}}(o)\). Energy band anisotropy is one example. If we include the experimental\(^13\) \(g_{\text{eh}}(o) = 4.4 \pm 1.6\) for unstressed Ge \((r_s = 0.57)\) with our \(g_{\text{eh}}(o)\) data, the
$g_{eh}(o)$ vs $r_s$ curve is seen to have a dip. No theoretical calculation to date predicts this qualitative behavior; this suggests further work is necessary.

The discrepancies could also be due to the use of effective mass theory in the calculations for both $g_{eh}(o)$ and $|\psi_x(o)|^2$. The effective mass approximation can do quite well in calculating energies, but poorly in determining wave functions as is illustrated by Ivey and Mieher's work on shallow donors in Si. Quantities such as $g_{eh}(o)$ and $|\psi_x(o)|^2$ characterizing spatial probabilities are especially sensitive to this approximation. Our data can be interpreted as measurements of $g_{eh}(o)/|\psi_x(o)|^2$ as a function of EHL density. However, the analysis given below supports our interpretation of the data as a measure of $g_{eh}(o)$.

A consistency check between the theoretical $g_{eh}(o)$ and the experimental values of $n_x$ and $\tau_x$ is informative. The radiative lifetimes for different density EHL are related by $\tau_{rL}(n_{x1})/\tau_{rL}(n_{x2}) = [n_{x}g_{eh}(o)]_2/[n_{x}g_{eh}(o)]_1$. Letting one subscript refer to the unstressed case where $n_{x} = 2.3 \times 10^{-17}$ cm$^{-3}$, $\tau_{rL} = \tau_x/c_{rL} = 160$ ms, and, from theory, $g_{eh}(o) = 2.3$ this equation can be used to estimate radiative lifetimes. Since $\tau_x \leq \tau_{rL}$ this provides an upper bound on $\tau_x$. Our $n_x$ and $\tau_x$ data are found to be inconsistent with the $g_{eh}(o)$ from the FSC approximation (see Fig. 3), the measured $\tau_x$ being up to 40% larger than the predicted upper bound on $\tau_x$. Using experimental $g_{eh}(o)$ instead we find our data are self consistent.

In summary, our experiment provides the first absolute determination of the density dependence of $g_{eh}(o)$. We have pointed out several difficulties in the interpretation of the one published experiment relevant to this work. Finally, more refined many body calculations appear necessary to completely describe this system.
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15 Using $\varepsilon_{\text{rx}} = 0.25$ from M. S. Skolnick and D. Bimberg, Phys. Rev. B 21, 4624 (1980).

FIGURE CAPTIONS

Fig. 1. Luminescence intensity is plotted semilog vs time for the electron-hole liquid droplet (a), free exciton gas (b), and total luminescence (c). The electron-hole liquid has disappeared by 4 ms. Curve (a) shows a transition from bulk to surface decay for the droplet.

Fig. 2. Luminescence intensity is plotted vs excitation power for the electron-hole liquid droplet (a), free exciton gas (b), and the total luminescence (c). The electron-hole liquid threshold is clearly visible in (a) and (b). All curves are scaled vertically to have the same height. No hysteresis in electron-hole liquid formation is seen within experimental resolution.

Fig. 3. The enhancement factor $g_{eh}(o)$ is plotted vs $r_s$ where $r_s = [3/(4\pi n_2)]^{1/3}/a_x$, $a_x$ being the free exciton Bohr radius and $n_2$ the electron-hole liquid density. Our data are plotted with error bars. The solid curves are theoretical results from Ref. 5.
Fig. 1

Luminescence intensity (lab units) vs time (ms)

(a) △ (b) □ (c)
Fig. 2

- Absorbed power (µW)
- Luminescence intensity (arb. units)

(a)
(b)
(c)
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