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Experimental Determination of the Density Dependence of Electron-Hole Correlation in Electron-Hole Liquid

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We report measurements of total lifetime and e-h pair density of electron-hole liquid in Ge under ⟨111⟩ uniaxial stress. We show that the density dependence of electron-hole correlation can be estimated semiquantitatively from our data. Our result gives the first experimental evidence that electron-hole correlation increases rapidly as the e-h pair density is lowered. We compare our results with calculations based on different many-body theoretical approximations.

The discovery of electron-hole droplets (EHD) in semiconductors has prompted several many-body calculations of the ground-state properties of this unique quantum liquid system.[1-3] The ground-state energies and densities obtained from these calculations are in reasonable agreement with each other and with experimental results. These calculations, however, make significantly different predictions of electron-hole correlation as measured by the enhancement factor $g_{eh}(0)$, which denotes the value of the e-h pair correlation function $g_{eh}(r)$ for zero interparticle separation. The more sophisticated calculation[3] based on a self-consistent theory of Singwi *et al.*[4] (STLS) predicts a much larger value of $g_{eh}(0)$ than that given by using either the Hubbard or the RPA approximation.[2,3] The discrepancy becomes even greater as the density is lowered. Since the enhancement factor is a direct measure of the wave function, it provides a more sensitive test of the theoretical approximations than does the calculation of the ground-state energy. In principle, the enhancement factor can be determined experimentally by comparing the measured radiative decay rate of e-h pairs in EHD with that measured for exciton[5] or uncorrelated plasma.[6] However, it is difficult to obtain unambiguously an absolute value of the enhancement factor using these two methods. This has, hitherto, prevented any meaningful comparison between theory and experiment as regards $g_{eh}(0)$. Such a comparison would be very valuable in that

it would help in judging the relative merits of different many-body approximations.

In this Letter, we report measurements of the total decay rate of e-h droplets in Ge under uniform ⟨111⟩ uniaxial stress. We show that the density dependence of the enhancement factor can be estimated semiquantitatively from our data. Our result gives the first experimental evidence (1) that $g_{eh}(0)$ increases rapidly as the density is lowered and (2) that its density dependence is in good agreement with the predictions of the STLS approximation.[4]

The samples used in our experiments were rectangular solids of ultrapure, dislocation-free Ge. We found that both EHD luminescence intensity and lifetime increased with increasing ⟨111⟩ stress when stress was applied to the sample at room temperature. Otherwise, when the stress was applied at He temperature, the luminescence intensity and lifetime decreased.[7] In the former case, our time-resolved luminescence spectra showed the free-exciton and EHD luminescence lines shift to lower energy during the initial 10 μs. From this shift,[8] we determined the variation of stress across our samples to be less than 0.5 kg/mm^2 even when the applied stress reached 15 kg/mm^2. This small stress gradient is also consistent with the long drifting time of 10 μs in our case.[8] Thus the former way of applying stress probably created a shallow strain well which moves e-h droplets[8] away from the sample surface where complicated nonradiative processes can occur, especially when the sample is subjected to uniaxial stress. The small stress gradient in our samples is further evidenced by the continuous narrowing and shifting to lower energy of our EHD luminescence line with stress, the independence of our luminescence spectra and lifetime on laser power[9] used (maximum ≈ 0.5 mJ per pulse), and the reproducibility of our results obtained with Ge samples of different sizes and from different boules. All the results reported here were obtained under such conditions.

We first determined the density of EHD as a function of ⟨111⟩ stress by measuring the EHD

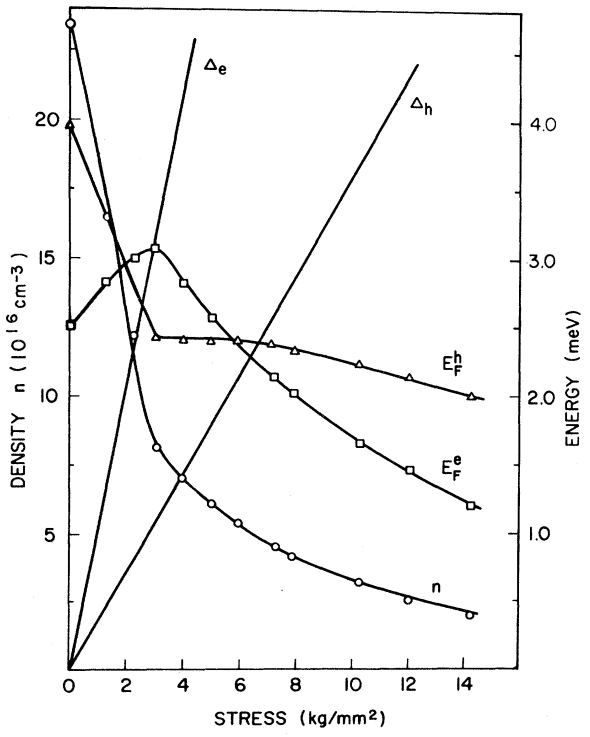

FIG. 1. EHD density, electron Fermi energy, and hole Fermi energy as a function of ⟨111⟩ uniaxial stress. Solid lines indicated by Δ_e and Δ_h are electron and hole band splittings determined with parameters given in Ref. 11.

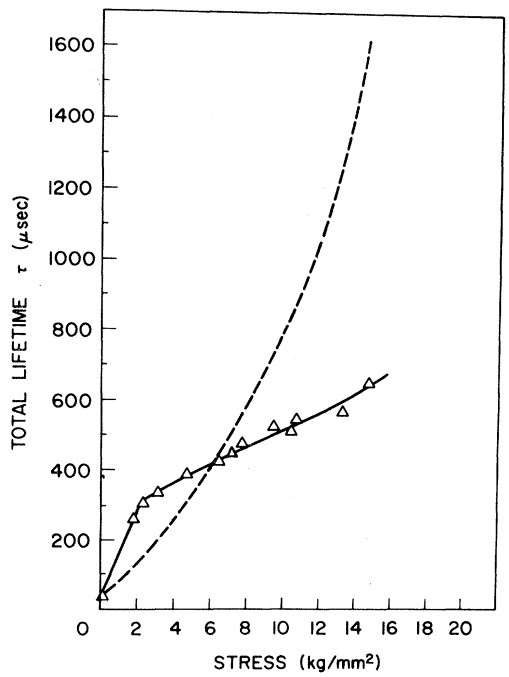

FIG. 2. Experimentally measured total lifetimes of EHD as a function of ⟨111⟩ uniaxial stress. The solid curve through the data points is drawn for visual guide only. The dashed curve is calculated from Eq. (1) by assuming $A\, g_{eh}(0)$ and $B\, g_{eh}^2(0)$ are independent of stress.

luminescence spectra at various stresses. In obtaining the e-h droplet densities shown in Fig. 1, the nonparabolicity of the valence band[10] has been taken into account in the standard theoretical analysis[11] of EHD luminescence line shapes. The e-h droplet density decreases rapidly as a function of ⟨111⟩ stress because the density of states of the conduction bands is reduced by the lifting of the valley degeneracy. The conduction- and valence-band splittings obtained from the deformation potentials[12] are also shown in Fig. 1 as Δ_e and Δ_h. Above 2.8 kg/mm^2, Δ_e exceeds the electron Fermi energy $E_F{}^e$ and the degeneracy of the conduction band is completely lifted. Likewise, the degeneracy of the valence band is lifted at 6.8 kg/mm^2.

In Fig. 2, we show the measured total lifetimes of EHD at 2°K. For all stresses, the EHD luminescence decay curve is exponential over a wide range, indicating that the decay of EHD is dominated by recombination of e-h pairs in the bulk of EHD. The bulk recombination can proceed through radiative recombination and nonradiative (Auger) recombination.[6] Hence, the total lifetime τ is given by[6]

$$\tau^{-1} = Ag_{eh}(0)n + Bg_{eh}{}^2(0)n^2, \quad (1)$$

where n is EHD density, $g_{eh}(0)$ is the enhancement factor, and A, B are constants proportional to the radiative rate and Auger rate, respectively. In arriving at the Auger term in Eq. (1), we have used the Kirkwood superposition approximation to approximate the three-particle correlation by $g_{eh}{}^2(0)g_{ee}(0)$ for electron-electron-hole processes and $g_{eh}{}^2(0)g_{hh}(0)$ for electron-hole-hole processes. Since $g_{ee}(0)$ and $g_{hh}(0)$ are not expected to be sensitive to the density,[3] they have been absorbed into B. At zero stress, $Ag_{eh}(0)$ and $Bg_{eh}{}^2(0)$ can be determined from our measured total lifetime of 46 μs, EHD density of 2.34×10^{17} cm^{-3}, and the best available radiative efficiency of 25%.[6] If $Ag_{eh}(0)$ and $Bg_{eh}{}^2(0)$ are assumed to be independent of stress, Eq. (1) allows us to calculate the total lifetime as a function of ⟨111⟩ stress by using the stress dependence of the EHD density shown in Fig. 1. The results are shown as dashed curve in Fig. 2. One sees that the agreement between the calculated and the experimental lifetimes is far from satisfactory. This indicates that $Ag_{eh}(0)$ and $Bg_{eh}{}^2(0)$ have significant stress dependence.

Above 6.8 kg/mm^2 the degeneracies of the conduction and valence bands are completely lifted and A, B can reasonably be taken to be independent of stress. The validity of this assumption and its effect on our analysis will be discussed later. Under this assumption, we can write Eq. (1) as

$$1/\tau' = Qn'g_{eh}{}'(0) + (1-Q)n'^2g_{eh}{}'^2(0), \quad (2)$$

where τ', n', and $g_{eh}{}'(0)$ are the total lifetime, e-h pair density, and enhancement factor normalized to their corresponding values at a stress of 7.2 kg/mm^2; Q is the radiative efficiency of EHD luminescence at 7.2 kg/mm^2. With the densities and lifetimes shown in Figs. 1 and 2, Eq. (2) allows us to calculate $g_{eh}{}'(0)$ as a function of density. The circles in Fig. 3 are the calculated $g_{eh}{}'(0)$ for $Q = 71\%$. As we shall discuss later,

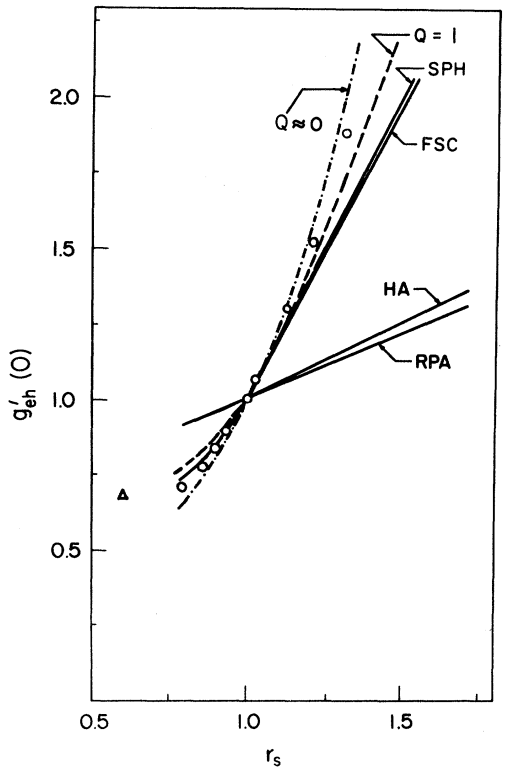

FIG. 3. Open circles are the density dependence of the relative enhancement factor determined for $Q = 71\%$. r_s is the usual dimensionless parameter defined by $r_s = (1/a_x)(3/4\pi n)^{1/3}$, where n is the density and a_x is the excitonic Bohr radius calculated with zero-stress hole mass. The dashed curves are drawn to indicate the maximum range of variation in our deduced values of $g_{eh}{}'(0)$ due to possible uncertainty in Q. Δ is the deduced relative enhancement factor at zero stress. The solid curves are the normalized theoretical enhancement factor obtained from Fig. 7 of Ref. 3. The value of r_s at which the curves intersect corresponds to the EHD density at 7.2 kg/mm^2.

the choice of $Q = 71\%$ is based on a radiative efficiency of 25% at zero stress.[6] The dashed curves in Fig. 3 show the density dependence of $g_{eh}'(0)$ obtained for $Q \simeq 0$ and $Q = 1$. One sees the irrespective of the exact value of Q, $g_{eh}'(0)$ increases rapidly as the density is lowered. Assuming A and B to be constant for all stress, we have also calculated $g_{eh}'(0)$ for stress below 7.2 kg/mm^2. The results are also shown in Fig. 3.

The datum point shown as Δ in Fig. 3 is the value of $g_{eh}'(0)$ at zero stress. In obtaining this value, we have taken into account the following consideration. At zero stress the EHD drop size (2–4 μm)[13] is comparable to the distance that the energetic Auger particle can travel before it relaxes to the Fermi level of the condensate by emission of phonons and plasmon.[14] Hence, practically all the Auger particles can escape from the droplet.[15] For Ge there is good evidence that the Auger process is dominated by electron-hole-hole processes[16] and the droplet will become negatively charged, resulting in a rapid decrease in the work function for thermionic emission of electrons. The resultant increase in thermionic emission of electrons is expected to balance the emission of Auger holes.[17] Thus at zero stress four carriers will be lost for each Auger recombination. However, only two carriers will be lost as the drop size becomes much larger than a few microns. At 3 kg/mm^2, the drop size should already be sufficiently large[18] so that only two carriers are lost. Considering this effect and taking a radiative efficiency of 25% at zero stress,[6] we used Eq. (1) to determine Q, the radiative efficiency at 7.2 kg/mm^2, to be 71%. Applying the same consideration to Eq. (2), we obtain the zero-stress $g_{eh}'(0)$ as Δ in Fig. 3.

To make a comparison with different theories, we normalized the theoretical value of the enhancement factor to its value at the e-h pair density of EHD at 7.2 kg/mm^2. The results obtained for different theoretical approximations[3] are shown in Fig. 3 as solid curves. These theoretical curves depend slightly on the hole- to electron-mass ratio.[3] The curves shown in Fig. 3 are the ones corresponding to a mass ratio of 1. One sees that our experimental results compare more favorably with the calculations based on STLS approximation.[4]

We now return to examine the assumption that A and B are independent of stress. From the studies on the temperature dependence of TO- and LA-phonon–assisted exciton recombination luminescence in Ge,[19] we estimated that the radiative recombination rate for an electron with $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ hole states is smaller than that for an electron with $|\frac{3}{2}, \pm\frac{3}{2}\rangle$ hole states by only about 24%. Hence, one does not expect any significant stress dependence in A. At any rate, since under $\langle 111 \rangle$ uniaxial stress $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ becomes the higher-energy hole state,[12] this 24% decrease in radiative recombination rate would make $g_{eh}'(0)$ increase even more rapidly as the density is lowered. As for the Auger process in Ge, it is expected that the dominant Auger recombination would be a four-particle process involving three carriers and a phonon. There does not seem to be any theoretical reason that such a process should have significant stress dependence, especially after the valence-band degeneracy is removed.

In conclusion, we have shown that the stress dependence of EHD lifetimes in Ge can be used to determine the density dependence of the enhancement factor. Our result gives the first experimental evidence that the enhancement factor increases rapidly with decreasing density as predicted by the STLS approximation.

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Extensive Energy Transfer in a Nearly One-Dimensional Crystal: The Emisson Spectrum of $CsMnBr_3$ Doped with Nd^{3+}

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Crystals of a nearly one-dimensional salt, $CsMnBr_3$, which have been doped with trivalent neodymium, exhibit emissions from both Nd^{3+} and Mn^{2+}. At room temperature the emission from Nd^{3+} is strikingly intense, much stronger than the Mn^{2+} emission. It appears that the manganese excitation migrates rapidly along the anionic chains in $CsMnBr_3$ and is trapped by the Nd^{3+} impurities. As the temperature is lowered, the rate of energy migration decreases and the Mn^{2+} emission grows at the expense of the Nd^{3+} emissions. There appears to be little energy transfer below 40 K.

Crystals of $CsMnBr_3$ which have been doped with trivalent neodymium luminesce strongly when excited by visible or uv radiation ($\lambda < 600$ mm). The emission spectrum contains features which can be attributed to both Nd^{3+} and Mn^{2+}. Although the concentration of Nd^{3+} in the doped crystals is relatively low (on the order of 1 part per 1000), the neodymium emission is quite intense even for exciting wavelengths which are absorbed only by Mn^{2+}. At room temperature the neodymium emission is considerably stronger than the manganese emission, which suggests that there is an efficient transfer of excitation energy from Mn^{2+} to Nd^{3+}. This communication describes the emission spectrum of the doped crystals and the temperature dependence of the energy transfer.

The host material, $CsMnBr_3$, is one of a number of AMX_3 halides which adopt linear-chain structures closely related to that of $CsNiCl_3$.[1] The $CsNiCl_3$ structure can be described as an array of parallel linear chains of MX_6^{4-} octahedra sharing opposite faces. The linear chains are anionic and have the stoichiometry $[MX_3^-]_n$. The univalent cations occupy positions between chains and balance the anionic charge. Since the intrachain separation between adjacent divalent metal ions is significantly shorter than the interchain separation, crystals of this type have a distinctly one-dimensional character. The magnetic properties of a number of AMX_3 salts have been extensively investigated.[2,3] The manganese salt $(CH_3)_4NMnCl_3$ (TMMC) has attracted particular attention since it behaves as a nearly perfect one-

