Strong Enhancement of Superconducting Correlation in a Two-Component Fermion Gas

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We study high-density electron-hole (e-h) systems with the electron density slightly higher than the hole density. We find a new superconducting phase, in which the excess electrons form Cooper pairs moving in an e-h BCS phase. The coexistence of the e-h and e-e orders is possible because e and h have opposite charges, whereas analogous phases are impossible in the case of two fermion species that have the same charge or are neutral. Most strikingly, the e-h order enhances the superconducting e-e order parameter by more than one order of magnitude as compared with that given by the BCS formula, for the same value of the effective e-e attractive potential $\lambda^{ee}$. This new phase should be observable in an e-h system created by photoexcitation in doped semiconductors at low temperatures.

KEYWORDS: doped electron-hole system, multiply ordered superconducting phase, superconductivity

It is expected that electron-hole (e-h) systems created through photoexcitation of semiconductors exhibit various phases depending on the material parameters and the densities $N^e$ and $N^h$ of electrons and holes, respectively.1) Some of the interesting phases have been successfully observed, including the e-h liquid2) and the Bose–Einstein condensation of excitons.3, 4) These successes are largely due to the careful control of both the material parameters (by choosing a semiconductor) and the e-h density (through the excitation intensity). By further increasing the e-h density, one should observe an “e-h BCS phase” at low temperatures, which is characterized by a nonzero e-h order parameter.5) Although these phases are realized when $N^e = N^h$, one can also use doped semiconductors. In this case, one can control $N^e$ and $N^h$ independently; $N^e - N^h \equiv N^x$ through the donor (or acceptor) density $N^d$, and $N^e + N^h$ through the excitation intensity.5) The additional parameter $N^x$ may lead to a new quantum phase(s) at low temperatures. When $0 < |N^x| \ll N^e \approx N^h$, in particular, we may expect a new superconducting phase, which we call a multiply ordered superconducting (MS) phase, in which doped electrons form Cooper pairs moving in the e-h BCS state. On the other hand, most of the previous studies on superconductivity in the doped e-h BCS state (or related systems such as doped exciton insulators and doped exciton systems) treated either the case where the doped electrons are located in a third band that is different from the bands of e-h pairs6, 7) or the case $N^h \ll N^e$ (or equivalently, $N^e \ll N^h$).8, 9) In these theories, the e-h BCS condensate (or excitons) work as polarizable media, which induce a large attractive interaction between electrons, whereas the superconducting order parameter $\Delta^{ee}$ as well as the superconducting transition temperature $T_c^{ee}$ is essentially given by substituting $\lambda^{ee}$ (effective dimensionless e-e attraction) and a cutoff parameter into the BCS formula or the McMillan formula.10) However, we expect the new MS phase when excess electrons (or holes) are doped in the (e-h) band where electrons (holes) forming the e-h BCS state are located, and when the e-h pairing is dominant, i.e., when $0 < |N^x| \ll N^e \approx N^h$ and $|\Delta^{ee}|, |\Delta^{hh}| \ll |\Delta^{eh}|$. Here, $\Delta^{ee}$ and $\Delta^{hh}$ are the superconducting e-e and h-h order parameters, respectively, and $\Delta^{eh}$ denotes the e-h order parameter.

In this Letter, we explore the possibility of such a new MS phase, by studying the phase diagram of high-density e-h systems as a function of $N^x$. It is shown that the MS phase, which has the order parameters $0 < \langle |\Delta^{ee}|, |\Delta^{hh}| \rangle \ll |\Delta^{eh}|$, can be realized when $0 < |N^x| \ll N^e \approx N^h$. Most strikingly, $|\Delta^{ee}|$ is enhanced in the MS phase by more than one order of magnitude in comparison with the value given by the BCS or McMillan formula, for the same value of $\lambda^{ee}$.

We assume a three-dimensional, high-density, isotropic e-h gas at zero temperature, so that BCS-like mean field approximations work well.5) We decompose the interaction Hamiltonian $H_{int}$ into the short- and long-range parts, $H_{int}^{SR}$ and $H_{int}^{LR}$, respectively. The latter is related to long-range charge fluctuations, and will be discussed later. For the moment, we consider a charge-neutral region of unit volume, in which $H_{int}^{LR}$ is irrelevant. In $H_{int}^{SR}$, the e-e, h-h and e-h interaction matrix elements are renormalized to effective values, $U^{ee}$, $U^{hh}$ and $U^{eh}$, respectively, due to the screening effect and negative (attractive) contributions from various bosonic excitations, such as lattice phonons and excitons. Since the bare value of $U^{eh}$ is negative, the bosonic excitations make it more negative, whereas (basically positive) $U^{ee}$ and $U^{hh}$ are reduced. Hence, $|U^{eh}|$ tends to be larger than

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It was suggested that $U^{ee}$ (and $U^{hh}$) can be negative for some parameter values. In the $e$-$h$ BCS phase, there is an additional negative contribution from the Goldstone mode of the $e$-$h$ order parameter. Since there is no reliable method of estimating $U^{ee}$ and $U^{hh}$, we here treat them as given parameters, assuming that $U^{ee}, U^{hh} < 0$ and $|U^{eh}| > |U^{ee}|, |U^{hh}|$, and explore the phase diagram as a function of them and $N^z (\geq 0)$. We find that the minimal form of $H_{\text{int}}^{\text{SR}}$ for the MS state is

$$H_{\text{int}}^{\text{SR}} = \sum_{k \neq k'} U^{eh}_{kk'} (c_{k\sigma}^\dagger h_{k'}^\dagger - h_{k'} c_{k\sigma}^\dagger) + \sum_{k \neq k'} U^{hh}_{kk'} (c_{k\sigma}^\dagger h_{k'}^\dagger - h_{k'} c_{k\sigma}^\dagger),$$

where $c_{k\sigma}$ ($h_{k\sigma}$) denotes the annihilation operator of $e$ ($h$) with momentum $k$ and spin $\sigma$, and

$$\begin{cases} U^{ee}_{kk'} = -V^{ee} e^{-i(k-k')}, & \text{if } |\xi_k|, |\xi_{k'}| < \omega_c, \\ U^{eh}_{kk'} = U^{eh}_{kk} = 0, & \text{otherwise}. \end{cases}$$

Here, $V^{ee}, V^{ee} > 0$ are constants, and $\omega_c (\ll \mu)$ is a cutoff of the interactions. We assume that $V^{eh} > V^{ee}$ so that $|\Delta^{ee}| > |\Delta^{eh}|$. Although we do not include an $h$-$h$ interaction $U^{hh}$ in $H$, we have confirmed that $U^{hh}$ can only modify the magnitude of the pair correlations by a factor of order unity, as long as $|U^{hh}| \lesssim |U^{ee}|$. We do not include terms of the form $ehh + hcc$ (such terms are important in the case of two-band superconductors) because in $e$-$h$ systems intermediate processes involving such terms cost a large amount of energy of the order of the energy gap $E_g (\gg |U^{ee}|)^5$. The total Hamiltonian without $H_{\text{int}}^{\text{SR}}$ is denoted by $H$, and we put

$$H = \mu^e N^e + \mu^h N^h$$

$$= \sum_{k\sigma} \left[ (\xi_k - \nu) e_{k\sigma} c_{k\sigma} + (\xi_k + \nu) h_{k\sigma}^\dagger h_{k\sigma} + H_{\text{SR}} \right],$$

with $\nu \equiv E_g/2 + \mu + \mu^h \equiv E_g/2 + \mu - \nu (\nu \geq 0)$ are the chemical potentials of $e$ and $h$, respectively, which are assumed to have the same energy dispersion $k^2/(2m) + E_g/2$. Moreover, $N^e \equiv \sum_{k\sigma} e_{k\sigma} c_{k\sigma}, N^h \equiv \sum_{k\sigma} h_{k\sigma}^\dagger h_{k\sigma}, \xi_k \equiv k^2/(2m) - \mu$, and we take $\hbar = 1$.

We apply the mean field approximation that assumes the $e$-$h$ correlation $\delta^{eh}_{k\sigma} \equiv \langle h_{-k\sigma} c_{k\sigma} \rangle$ and the $e$-$e$ correlation $\delta^{ee}_{k\sigma} \equiv \langle c_{-k\sigma} c_{k\sigma} \rangle$. We assume that $\langle h_{-k\sigma} c_{k\sigma} \rangle$. Using (2), we find that the order parameters, defined by $\Delta^{eh}_{k\sigma} \equiv \sum_{k'k} U^{eh}_{kk'} c_{k\sigma}^\dagger h_{k'\sigma}^\dagger$ and $\Delta^{ee}_{k\sigma} \equiv \sum_{k} U^{ee}_{kk} c_{k\sigma}^\dagger c_{k\sigma}^\dagger$, take simple forms: $\Delta^{eh}_{k\sigma} = \Delta^{eh} c_{k\sigma}, \Delta^{ee}_{k\sigma} = \Delta^{ee} c_{k\sigma}$ if $|\xi_k| < \omega_c; \Delta^{eh}_{k\sigma} = \Delta^{eh} c_{k\sigma}, \Delta^{ee}_{k\sigma} = \Delta^{ee} c_{k\sigma}$ otherwise. Here, $\Delta^{eh}$ and $\Delta^{ee}$ are constants, which are taken to be real without loss of generality. We then diagonalize $H$, and obtain the self-consistent equations. The system is characterized by $N^e$ and dimensionless effective coupling constants $\lambda^{eh} \equiv n_F V^{eh}, \lambda^{ee} \equiv n_F V^{ee}$, where $n_F$ is the density of states per spin at the Fermi surface.

We have solved the self-consistent equations numerically and found five solutions, which we denote by I–V: I: $|\Delta^{ee}| = |\Delta^{eh}| = 0$; possible for all $N^e$. The one-particle distribution functions of $e$ and $h$ are $n^{e}(\xi) = \theta(\nu - \xi)$ and $n^{h}(\xi) = \theta(\nu + \xi)$, respectively, where $\theta(x)$ is the step function.

II: $|\Delta^{eh}| = 0, |\Delta^{ee}| = 0$; possible for $N^e = 0$ and $\nu < |\Delta^{eh}|$. The wave function takes the same form as the BCS state, if $\langle c_{k\sigma}^\dagger h_{-k\sigma} \rangle$ is replaced by $\langle c_{k\sigma}^\dagger h_{-k\sigma} \rangle$. This is the ordinary $e$-$h$ BCS state in nondoped ($N^e = N^h$) semiconductors. The energy cost of adding an electron-like quasiparticle to this state is $E_k = \mu^e N^e - \mu^h N^h$.

III: $|\Delta^{eh}| = 0, |\Delta^{ee}| = 0$; possible for small but finite $N^e > 0$ and $\nu > |\Delta^{eh}|$. Formally, this solution (whose wave function is denoted by $|III\rangle$) is obtained from solution II (whose wave function $|II\rangle$) by adding electron-like quasiparticles (whose annihilation operator $c_{k\sigma}$) up to $E_k < \nu$, i.e., $|III\rangle = (\prod_{k\sigma} c_{k\sigma}^\dagger h_{k\sigma}^\dagger)|II\rangle$. The $\prod_{k\sigma}$ denotes the product over the range $E_k < \nu$. Direct calculation shows that $|III\rangle = (\prod_{k\sigma} c_{k\sigma}^\dagger h_{k\sigma}^\dagger)|II\rangle$, where $S_{k\sigma}$ annihilates an $(c_{k\sigma}, h_{-k\sigma})$ pair. Therefore, $n^e(\xi) = 1,$
\[ n^h(\xi) = 0, \quad \delta^h = 0 \quad (e \text{ and } h \text{ are unpaired}) \text{ in the region } [|\xi| < \sqrt{\nu^2 - |\Delta^{ch}|^2}] \equiv \xi^o. \] This unpairing is demonstrated in Fig. 1 by dotted lines, which have discontinuities (secondary “Fermi surfaces”) at \( \xi = \pm \xi^o \). We call this solution the partially unpaired \( e-h \) BCS state (PU state). As \( N^x \) increases, \( |\Delta^{ch}| \) diminishes gradually until it vanishes at a certain value of \( N^x \), where this solution changes into solution I continuously.

IV: \( |\Delta^{ch}| = 0, \quad |\Delta^{ee}| \neq 0 \); possible for all \( N^x \). Similar to solution I except that \( \delta^e \neq 0 \) for \( |\xi| \neq |\Delta^{ee}| \); this is an ordinary superconductor of electrons.

V: \( |\Delta^{ch}|, \quad |\Delta^{ee}| \neq 0 \quad \Rightarrow |\Delta^{ee}| \) because \( \lambda^{ch} > \lambda^{ee} \); possible for small but finite \( N^x > 0 \). Similar to solution III except that \( \delta^e \neq 0 \) if \( [|\xi| \pm \xi^e] \leq |\Delta^{ee}| \); i.e., the \( e \)-pair correlation exists around the secondary “Fermi surfaces” (see solid lines in Fig. 1). This is the only solution where \( \Delta^{ch} \) and \( \Delta^{ee} \) coexist.\(^{15} \) We call this solution the multiply ordered superconducting (MS) state.

To identify what solution is physically realized, we compare their energies \( E \equiv \langle H \rangle \) for all values of \( N^x \equiv N^e - N^h \). Note that we compare \( \langle H \rangle \) rather than \( \langle \mathcal{H} \rangle \), because the natural parameters controlled directly by the photoexcitation intensity and the doping are \( N^e \) and \( N^h \) rather than \( \mu^e \) and \( \mu^h \)\(^{5} \). If we compared \( \langle \mathcal{H} \rangle \), the discussion would be rather complicated because the relations between \( (N^e, N^h) \) and \( (\mu^e, \mu^h) \) are different for different solutions.] Figure 2 shows \( E \) as a function of \( N^x \), where \( N \equiv (N^e + N^h)/2 \). One recognizes that the solution V has the lowest energy for all values of \( N^x/(\pi v_\Delta^0) \lesssim 1.86 \). However, care should be taken because its curve is convex up, which might indicate a phase separation. We now show that the solution V is stable because \( e \) and \( h \) have opposite electrical charges.

To show the stability, we first consider the unstable case where \( e \) and \( h \) denote some fermions that have charges of the same sign (including the neutral case). All calculations so far are also applicable to such a general case (as long as we regard \( U^{eh} \) and \( I^{ee} \) as given parameters). Since the energy of a single phase of \( V \) is larger than the average of the energies of two phases II and IV, the system should undergo a phase separation into two phases, one with the excess density \( N^x = 0 < \xi^o \) (phase II) and the other with \( N^x > N^o_{\xi^o} \) (phase IV), where \( N^o_{\xi^o} \) denotes the average value of \( N^x \) of the total system. As \( N^o_{\xi^o} \) is decreased, the region(s) of phase IV becomes smaller, until the total system turns into a single phase II for \( N^o_{\xi^o} = 0 \). In a similar manner, one can also show the instability of phase III. This corresponds to the instability of the Sarma state\(^{16} \) that was discussed in the studies of superconductivity in a magnetic field.

On the other hand, the situation is totally different if we return to the electron-hole system, where \( e \) and \( h \) have opposite charges \( -q \) and \( q \) (\( q > 0 \), respectively). If the phase separation occurred, each phase would have global net charge of density \( -q \delta N \equiv -q(N^x - N^d) \) in phase II, and \( q \delta N \) in phase IV. The global charge would result in a large cost of the long-range part of the Coulomb energy \( E_{LR} \equiv (H_{LR}^0) \), which we neglected in \( H, \) eq. (3). Taking \( E_{LR} \) into account, the total energy of such a nonuniform state would be \( E_{tot} = \sum_i v_i E_i + E_{LR} + E_B \), where \( v_i \) and \( E_i \) denote the volume and the expectation value of \( H \) for phase \( i \), respectively, and \( E_B \) the boundary energy. Since \( E_B \geq 0, \quad E_{tot} \geq \sum_i v_i E_i + E_{LR} + E_{tot} \). When the system is in a single phase \( V \), then \( E_{LR} = 0 \) and \( E_{tot} \) equals \( E \) of phase \( V \) of Fig. 2. On the other hand, when the system is separated into cells of phases II and IV, one obtains a finite \( E_{LR} \), which would be dominated by the electrostatic energy. We find that \( E_{V} \) is smaller than \( E_{tot}^{II+IV} \) (\( \leq E_{tot}^{II+IV} \)), hence a single phase \( V \) should be realized, except for very small values of \( N^x \) if \( L^{ch} \gg L_c \).

This is the only solution where \( \Delta^{ch} \) and \( \Delta^{ee} \) coexist.\(^{15} \) We call this solution the multiply ordered superconducting (MS) state.

By performing calculations of the energies of various solutions as a function of \( \lambda^{ee} \) and \( N^x \), we obtain the phase diagram shown in Fig. 3. Phase \( V \) (MS phase), for which \( |\Delta^{ch}| \gg |\Delta^{ee}| \) is realized in the shaded region. This phase changes continuously into phase II as \( N^x \rightarrow 0 \). On the other hand, this phase changes into phase IV as \( N^x \) increases, because then the Fermi energies of \( e \) and \( h \) become separate, which prevents \( e-h \) pairing. To study the \( N^x \) dependence in more detail, we plot \( |\Delta^{ee}| \) in the MS phase in Fig. 4. One sees that \( |\Delta^{ee}| \) strongly depends on \( N^x \) and takes a maximum value \( \Delta^{ee}_{tot} \) at an optimum \( N^x \) (\( \equiv N^{opt} \)), although \( N^c = N + N^d^2/2 \) is almost constant (because \( N^c \approx N \)). This is in marked contrast with the single-component case, where the BCS theory gives \( |\Delta^{ee}| = 2\omega_c \exp[-1/\lambda^{ee}] \equiv \Delta^{ee}_{BCS} \), which is constant when \( N^c \) and \( \lambda^{ee} \) are constant. To compare \( \Delta^{ee}_{tot} \) with \( \Delta^{ee}_{BCS} \), we plot the enhancement factor \( Q \equiv \Delta^{ee}_{tot}/\Delta^{ee}_{BCS} \) in the inset of Fig. 4. Since \( \Delta^{ee}_{BCS} \) is the magnitude of \( |\Delta^{ee}| \) which would arise if there were no \( e-h \) interaction, the fact \( Q > 1 \) implies that \( |\Delta^{ee}| \) is enhanced by the presence of \( |\Delta^{ch}| \). It is found that an enhancement by one order of magnitude or larger occurs as \( \lambda^{ee} \) becomes smaller. As \( \lambda^{ee} \rightarrow 0 \), in particular, \( |\Delta^{ee}| \) diminishes much more slowly than \( \Delta^{ee}_{BCS} \), resulting in a very large enhancement factor \( Q \). Since the superconducting transition temperature \( T^{ee}_{c} \propto |\Delta^{ee}| \), the MS phase should have a \( T^{ee}_{c} \) which is much higher than the BCS transition temperature.\(^{18} \) The parameter region in which this enhancement occurs is hatched in Fig. 3, where the optimum doping line is also shown.

To reveal the physical mechanism leading to the enhancement of \( |\Delta^{ee}| \), we reexamine the \( e \) distributions of
the MS and PU states of Fig. 1(a), which is schematically plotted in Fig. 5. In the PU state, for which $|\Delta^{ee}| = 0$, excess electrons are concentrated in the region $|\xi| \leq \xi_F^e$, and the secondary (lower and upper) “Fermi surfaces” appear at $\xi = \pm \xi_F^e$. In the MS state, electrons form $e-e$ pairs to benefit from the attractive $e-e$ interaction and consequently $\delta E_{\text{kin}}$ develops around those “Fermi surfaces” [Fig. 1(d)]. However, this pairing necessarily broadens the “Fermi surfaces”, resulting in increase of the kinetic energy $E_{\text{kin}}$. Hence, the total energy is minimized for an optimum $\delta E_{\text{kin}}$. Although this mechanism is similar to the BCS instability in ordinary superconductors, there is an essential difference: In the BCS state, the broadening of the Fermi surface of width $\delta \xi$ increases the kinetic energy by $\delta E_{\text{kin}} \approx n_F|\delta \xi|^2 \approx n_F|\Delta^{ee}|^2$. In the MS state, on the other hand, the broadenings of the lower and upper “Fermi surfaces” change the kinetic energy by $\delta E_{\text{kin}} \approx -n_F|\Delta^{ee}|^2$ and $\delta E_{\text{kin}} \approx + n_F|\Delta^{ee}|^2$, respectively [when $\lambda^{ee}$ (thus $|\delta \xi|$) is small]. These two contributions cancel to give a relatively small increase in the kinetic energy $\delta E_{\text{kin}} = \delta E_{\text{kin}}^{\text{ee}} + \delta E_{\text{kin}}^{\text{ee}}$. This is the origin of the huge enhancement factor $Q$ for small $\lambda^{ee}$. Since the cancellation is not perfect, $|\Delta^{ee}|$ of course takes a finite value. As $\lambda^{ee}$ is increased, the cancellation becomes less complete, and $Q$ is reduced. The existence of an optimum doping can be understood in a similar manner: If $N^x$ is too small, no room for broadening is left between the two “Fermi surfaces”. Conversely, if $N^x$ is too large, the magnitude of the jump of $n^e(\xi)$ at the two “Fermi surfaces” differs, which makes the cancellation of contributions from the two “Fermi surfaces” less perfect, resulting in a smaller $|\Delta^{ee}|$. Therefore, $|\Delta^{ee}|$ takes a maximum value at an intermediate value of $N^x$.

Finally, we point out that although we have assumed the $e-h$ BCS state in semiconductors, the same mechanism for the enhancement of the $e-e$ correlation upon doping may be expected also in other ordered phases of other materials, if the electron distribution without the $e-e$ interaction and $e-h$ plasma for $N^x$ too large, the magnitude of the jump of $n^e(\xi)$ at the two “Fermi surfaces” differs, which makes the cancellation of contributions from the two “Fermi surfaces” less perfect, resulting in a smaller $|\Delta^{ee}|$. Therefore, $|\Delta^{ee}|$ takes a maximum value at an intermediate value of $N^x$.

5) We here use the word “$e-h$ BCS state” to avoid confusion with the “excitonic insulator” in semimetals, which has the same order parameter. The former is realized in semiconductors, for which $E_{\text{F}} \gg |U^{hh}|$, so that the rigid-band approximation (RBA) is good and $N^x$ and $N^h$ are independent of $U^{hh}$. For the latter, on the other hand, $|E_{\text{F}}| \sim |U^{hh}|$, so that the RBA breaks down and $N^x$ and $N^h$ strongly depend on $U^{hh}$.
11) For example, the Migdal approximation is only justified for the electron-phonon interactions in metals.
12) We take the same cutoff for both $U^{hh}$ and $U^{ee}$, because we found that the final results are insensitive to the cutoff of $U^{ee}$.
15) When an $h-h$ interaction $U^{hh} < 0$ is included in $H$ of eq. (3), $h-h$ order parameter $|\Delta^{hh}|$ also becomes finite.
17) When $r_s > 1$, our assumption $\omega_s \ll \mu$ no longer holds, and therefore the stability of the MS state for $r_s > 1$ is beyond the scope of this Letter.
18) If $\lambda^{ee}$ is very small, $T^{ee}_{\text{kin}}$ is low even when $Q \gg 1$. 

Fig. 3. Phase diagram of a high-density $e-h$ plasma for $\lambda^{eh} = 0.25$. The MS phase is shadowed. $|\Delta^{ee}|$ is enhanced in the hatched region, where $|\Delta^{ee}|$ takes maximum values with respect to $N^x$ on the line denoted as optimum doping. The vertical line at $N^x = 0$ corresponds to phase II. Dashed lines are guides for the eye (not calculated).

Fig. 4. The $N^x$ dependence of $|\Delta^{ee}|$ in the MS phase for $\lambda^{eh} = 0.25$. The inset shows the enhancement factor $Q$ vs $\lambda^{ee}$.

Fig. 5. Schematic illustration of the enhancement mechanism. The electron distributions $n^e(\xi)$ for the PU state (dotted line) and the MS state (solid line) are shown.