

Theoretical Aspects of Superconductivity in Boron-Doped Diamond

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The theoretical aspects of the electronic states and superconductivity in boron (B)-doped diamond are reviewed. Starting from the explanation of the basic notions of the electronic structure of diamond and the impurity states of a doped B atom, we first discuss the electronic states of B-doped diamond by placing particular emphasis on the effects of disorder on the electronic states induced by the B impurities. We then consider the superconductivity of B-doped diamond by calculating the impurity states of B atoms and superconducting critical temperature by the coherent potential approximation. We show that the effects of disorder play a crucial role in the electronic structure and superconducting critical temperature of B-doped diamond.

1. Introduction

The discovery of superconductivity in boron (B)-doped diamond^(1,2) was a major surprise in the basic science of superconductivity because the effects of disorder induced by B atoms are so strong that the coherent motion of Cooper pairs, and therefore the superconductivity, was thought to be strongly suppressed. A considerable number of experimental and theoretical studies⁽¹⁻²⁴⁾ have been performed over the past three years on the mechanism of this unusual superconductor, but to our understanding, adequate descriptions even of the basic electronic states of this highly disordered system have not yet been given. In this paper, we will first discuss the basic electronic structure of diamond and then review our research^(23,24) on the electronic states of this highly disordered electron system.

Our research^(23,24) is based on a simplified model for the valence band of diamond using coherent potential approximation (CPA) to consider the effects of the substitutional disorder of B ions. We have calculated the density of states, lifetime, and evolution of single-particle spectra due to doping, the results of which have been compared

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with available experimental data including angle-resolved photoemission spectra (ARPES).⁽¹²⁾ We have also calculated the superconducting pairing susceptibility on the basis of the ladder approximation under the assumption of instantaneous interaction and have estimated the doping dependence of the critical temperature T_c . We have thereby clarified the effects of disorder on the superconductivity in such unusual situations. In particular, we have compared our results with those for supercell calculations (i.e., the periodic arrangement of B ions) to determine the role of disorder, which turns out to be of crucial importance.

This paper is organized as follows. In §2, we explain the basic electronic states of diamond and the effects of B doping. We then discuss the effects of disorder in the metallic regime of B-doped diamond in §3. In §4, we present our theoretical model and briefly discuss the method of calculation of the electronic states and pairing susceptibility by CPA. In §5, we present our theoretical results for the superconductivity of B-doped diamond. We discuss the results in §6, and we conclude this paper in §7.

2. Basic Electronic States

First, let us discuss the basic electronic states of diamond and the effects of B doping. An isolated C atom has the electronic configuration of six valence electrons as $2s^2 2p^4$. The energy levels of the one $2s$ and three $2p$ orbitals of a C atom are shown in Fig. 1(a). The C atoms are bonded to form bulk diamond in the three-dimensional diamond structure. The largest energy scale in the electronic states of diamond is the hopping integral between the nearest-neighbor C atoms, or the overlap of the wave functions of the $2s$ and $2p$ orbitals of C atoms, which forms the covalent bond; i.e., a C atom shares two electrons with each of the four neighboring C atoms in the diamond lattice, where

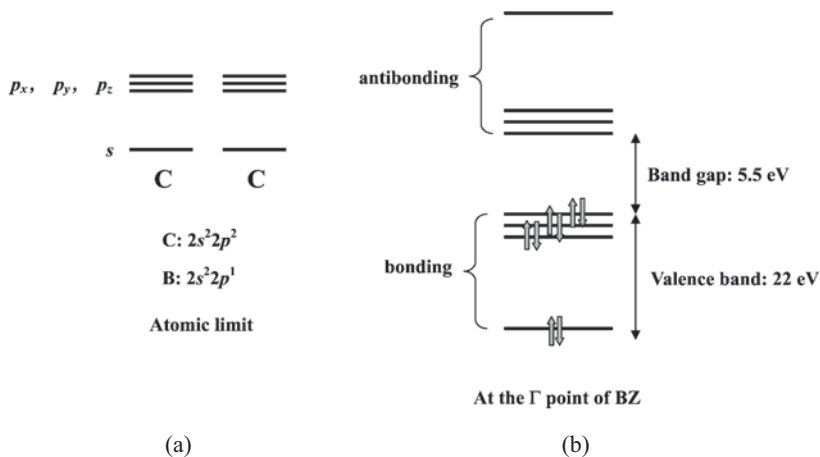


Fig. 1. (a) Schematic representation of the energy levels of the $2s$, $2p_x$, $2p_y$, and $2p_z$ orbitals of a pair of isolated C atoms without covalency. (b) Schematic representation of the energy levels of diamond at the Γ point of the Brillouin zone.

two electrons reside in the bonding orbitals to stabilize the structure. Because of the large hopping integrals, the atomic levels split into bonding and antibonding levels, as shown in Fig. 1(b), and form wide valence and conduction bands. The band gap formed between the valence and conduction bands is very large, having a size of 5.5 eV. The valence band has a width of 22 eV. This strong covalency causes the strong bonding of C atoms in diamond, which results in very high frequencies of the bond-stretching phonon mode⁽³⁾ as well as its exceptionally high Debye temperature of ~ 2300 K. Note that the top of the valence band is located at the Γ point of the Brillouin zone and is triply degenerate.

Now let us suppose that one of the C atoms in diamond is replaced by a B atom. The electronic configuration of an isolated B atom is $2s^2 2p^3$ and it has five valence electrons per atom, so that one hole is introduced by this substitution of a B atom. The hole is bound by the B impurity, the radius of which is several Å (see Fig. 2). The bond length between the B atom and its four neighboring C atoms shrinks slightly and uniformly and the energy level of the impurity increases to above the valence band continuum, as shown in Fig. 3. It has been well established^(25,26) that a single B impurity in otherwise pure diamond has a deep energy level in the band gap at 0.37 eV above the top of the valence band of diamond. Note that the impurity energy level is also triply degenerate.^(25,26)

Upon increasing the concentration of B impurities, the system becomes metallic. The mechanism of this metallization may be either the overlap of the wave functions of the B impurities keeping the impurity state basically unchanged due to strong impurity potential, or the rigid-band shift of the Fermi level due to doping (which has been obtained by the virtual crystal approximation based on first-principles local-density approximation (LDA) band calculations⁽¹⁶⁻¹⁸⁾ using the adjusted chemical potential). Note that the simpler latter picture may not necessarily be justified from experiments; spectroscopic experimental data⁽¹⁹⁻²¹⁾ seem to suggest the presence of non-rigid-band-like

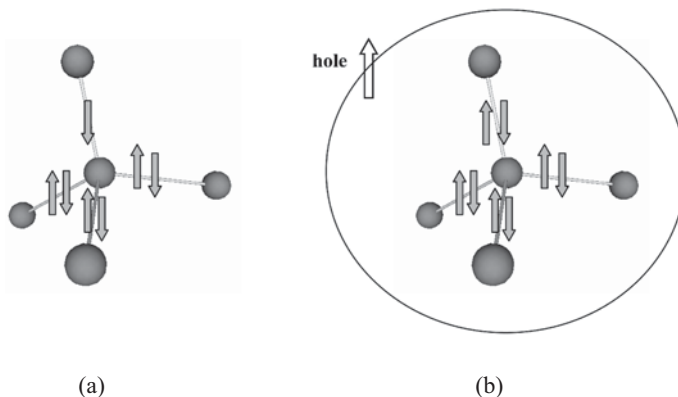


Fig. 2. Schematic representation of the local configuration around a B impurity in diamond. The central atom is B, which is surrounded by four C atoms. Arrows represent the electron spins. An electron is missing in one of the four covalent bonds in (a), but in reality, a hole is loosely bound by the impurity B atom, as shown in (b).

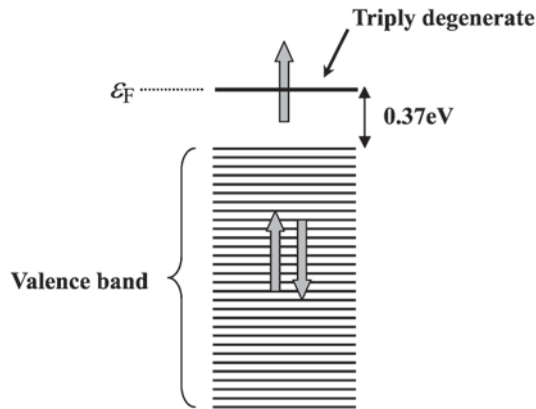


Fig. 3. Schematic representation of the energy level of a single impurity in diamond. The energy level of the impurity, which is triply degenerate, increases to above the valence-band continuum of diamond.

features reminiscent of impurity states even in the metallic regime of B-doped diamond. In this respect, we may point out that the first-principles LDA band calculations by the supercell method cannot reproduce the separated impurity energy level established experimentally at the low doping limit of diamond.

Another important feature of doping is the effects of hydrogen (H) ions introduced in the doping process of carriers by B doping. A recent analysis of the nuclear magnetic resonance (NMR) spectra⁽⁶⁾ suggests that the real doping rate of carriers (or the number of carriers per site) is at most only $\sim 1\%$, which differs considerably from the nominal doping rate of $\sim 5\%$, and that this is due to the formation of a boron-hydrogen (BH) complex; i.e., H atoms inevitably introduced during the syntheses of the materials absorb the holes introduced by B impurities. This possibility has been suggested recently both theoretically and experimentally; an LDA calculation⁽²²⁾ has shown that the B ions occupy the carbon sites substitutionally and that the H ions sit on the interstitial positions between B ions and the neighboring carbon ions. Assuming the presence of these two types of B ions, the observed NMR spectra can be analyzed consistently.⁽⁶⁾ We should therefore reexamine the electronic states of B-doped diamond, in particular, in the regime of the low carrier concentration of $\sim 1\%$, starting from the dilute limit of B doping.

3. Effects of Disorder

Next, let us discuss the effects of disorder. The holes introduced by doping behave as hole carriers at low doping levels in B-doped diamond, where the doping rate is at most $\sim 1\%$ in superconducting samples. The C atoms are substituted randomly with the B atoms, and therefore, the doped B impurities not only yield hole carriers but also introduce spatial disorder in the system because of their random spatial distribution. This

disorder causes the strong scattering of hole carriers, the effect of which is evident in the observed temperature dependence of the electronic conductivity in B-doped diamond as well as in the line shape of quasi-particles observed by the ARPES experiment,⁽¹²⁾ indicating that the lifetime of electrons τ is very short with the characteristic parameter $\varepsilon_F\tau$ of order 1 (throughout this paper we set $\hbar = k_B = 1$), which violates the Ioffe-Regel criterion for coherent Bloch-like transport.^(14,15) This small value of $\varepsilon_F\tau$ implies that the Fermi surface is not well-defined in momentum space but is blurred by scattering (see Fig. 4); the superconductivity in B-doped diamond may therefore be referred to as “superconductivity without a Fermi surface.”⁽¹³⁾

The strong scattering potential of the impurity B ions may be responsible not only for such disorder-induced anomalous features but also for the basic electronic states. The energy level of a single B impurity in diamond is very deep, located at 0.37 eV above the top of the valence band of diamond,^(25,26) and in the presence of such a strong scattering potential, the simple picture of the “rigid-band shift” of the Fermi level due to doping (which can be calculated by the virtual crystal approximation based on LDA band calculations^(16–18) using the adjusted chemical potential) may not necessarily be justified. In fact, some spectroscopic experimental data^(19–21) seem to suggest the presence of non-rigid-band-like features reminiscent of impurity states even in the metallic regime of B-doped diamond, which has been focused upon by Baskaran.^(14,15) Hence, evolution of the impurity states due to doping should be considered with more care.

In the following, we adopt a simplified model for the electronic structure of B-doped diamond and present our calculated results for the evolution of the impurity states by doping as well as for the effects of disorder on the pairing susceptibility and superconducting critical temperature.

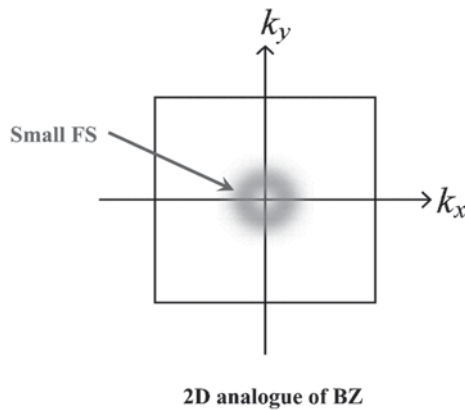


Fig. 4. Schematic representation of the Fermi surface in the two-dimensional analogue of the simple-cubic Brillouin zone for hole-doped diamond. The Fermi surface is blurred by impurity scattering.

4. Model and Method

Our model is as follows. We assume, for the sake of simplicity, the tight-binding model based on a simple cubic structure in order to determine the effects of disorder on the electronic states near the top of the valence bands of diamond. By this simplification, the degeneracy of the three bands at the top of the valence bands of diamond is ignored. However, we believe that the essential features of the effects of disorder can be taken into account in this simple model. Adding the random potential Δ at B sites, our model may be expressed by the Hamiltonian

$$\mathcal{H} = \sum_{k\sigma} (\mathcal{E}_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \Delta \sum_{i \in B} \sum_{\sigma} n_{i\sigma} \quad (1)$$

with

$$\mathcal{E}_k = t(\cos k_x a + \cos k_y a + \cos k_z a), \quad (2)$$

using standard notation. $n_{i\sigma}$ is the number operator at site i and spin σ , and $c_{k\sigma}^\dagger$ is the creation operator of an electron with momentum k and spin σ . μ is the chemical potential. To allow for superconductivity, we include a term

$$-V \sum_i n_{i\uparrow} n_{i\downarrow} \quad (3)$$

for the on-site attractive interaction $-V$ ($V > 0$) in the Hamiltonian. Here, we do not pursue the processes of attractive interaction in detail, although we assume that it is due to the electron-phonon interaction associated with bond-stretching modes with a very high frequency, as has been indicated both experimentally and theoretically.^(3,16-18)

We assume $t = 1$ as the unit of energy unless otherwise indicated; to reproduce the width of the valence band of diamond, 22 eV, this choice corresponds to assuming 3.7 eV as the unit of energy. The effective mass of the top of the valence band of diamond is roughly reproduced using this value. We determine the value of Δ so as to reproduce the energy level of B ions in the dilute limit, which is experimentally known to be located at 0.37 eV above the top of the valence band of pure diamond;^(25,26) we obtain the value $\Delta = 2.30$, which is used throughout the present work. The density of states of this model is shown in Fig. 5 along with the energy level of a single B impurity.

The sites at which Δ has this nonzero value are distributed randomly over the lattice sites of the simple cubic structure; this substitutional disorder is treated by CPA. Using CPA,⁽²⁷⁻³¹⁾ we can take into account the effects of the multiple scattering of an electron due to B impurities at a mean-field level (see Fig. 6) and calculate the self-energy Σ , so that we can obtain the lifetime of electrons due to impurity scattering. Calculation of the superconducting pairing susceptibility is performed at the same level of CPA. We here do not present further details of our theory, but readers can refer to our original paper⁽²⁴⁾ for this purpose.

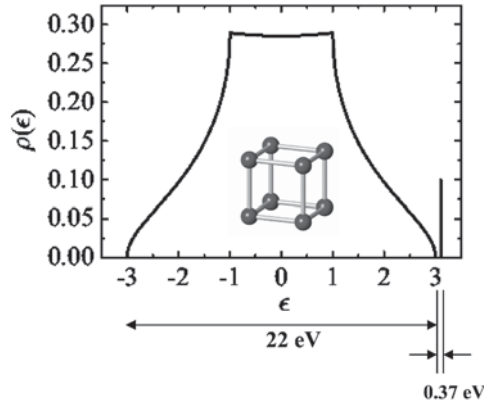


Fig. 5. Model density of states $\rho(\epsilon)$ (in units of 1/atom/spin) for the tight-binding band on the three-dimensional simple cubic lattice without disorder. The energy level of a single impurity is also shown.

$$G = G^{(0)} + G^{(0)} \sum_j t_j G^{(0)} \\ + G^{(0)} \sum_j t_j G^{(0)} \sum_{l(\neq j)} t_l G^{(0)} + \dots$$



Fig. 6. Green's function G defined by the multiple scattering of a hole by impurities. $G^{(0)}$ and t are the free propagator and t -matrix, respectively.

5. Superconductivity

Now, let us calculate the pairing susceptibility within CPA and discuss the effects of disorder on the superconducting critical temperature. We use the ladder approximation to consider the attractive interaction V and derive the CPA expression for the pairing susceptibility as⁽²⁴⁾

$$\chi(T) = \Pi \frac{\Pi(0,0)}{1 - V\Pi(0,0)} \quad (4)$$

with

$$\Pi(0,0) = \frac{1}{2} \int d\epsilon \rho(\epsilon) \frac{1}{\epsilon - \mu} \tanh\left(\frac{\beta(\epsilon - \mu)}{2}\right), \quad (5)$$

where $\Pi(0,0)$ is the pairing susceptibility at the vanishing momentum and the frequency in the absence of the attractive interaction V . $\rho(\varepsilon)$ is the density of states in the presence of the disorder determined by CPA, and $\beta = 1/k_B T$. The critical temperature T_c is determined as the temperature at which $\chi(T)$ diverges or $V\Pi(0,0) = 1$. Note that this expression can be considered as an extension of Anderson's theorem,⁽³²⁾ which states that T_c for an isotropic s-wave superconductor is not affected by nonmagnetic impurities if the density of states is unchanged by scattering. Our extension is that, within CPA, even a strong disorder that deforms the density of states does not affect the validity of the Bardeen-Cooper-Schrieffer (BCS) expression (see the above expression for $\Pi(0,0)$) for the pairing susceptibility.

The calculated results for the doping dependence of T_c are compared with experimentally obtained results in Fig. 7. We define the concentration of B ions as $c_B = N_B/N$, where N_B is the number of B sites and N is the total number of sites in the system. We find that if we assume an appropriate value of V , e.g., $V \sim 6$ (which corresponds to ~ 20 eV), our result of $T_c \sim 7$ K at $c_B = 1\%$ is in reasonable agreement with the results of the LDA calculations⁽¹⁶⁻¹⁸⁾ and therefore with those of the experiment. In particular, we find that the rapid increase in T_c with increasing doping rate, which is observed in the experiment, is reproduced well.

Improvements of our calculations presented in this paper may be possible by considering the following points. First, note that our calculated values of T_c are not of sufficient quantitative accuracy since the calculations are based on the single-band model of the simple-cubic lattice and the instantaneous interaction for the attractive force. For a more accurate estimation of T_c , we may need to make the following improvements: (i) The nearly degenerate three-band structure around the top of the valence band of diamond in the actual lattice structure should be included. (ii) The retardation effects for the electron-phonon coupling should be taken into account. (iii) The dynamical aspect of the Coulomb interaction between holes should be considered; because of the effects

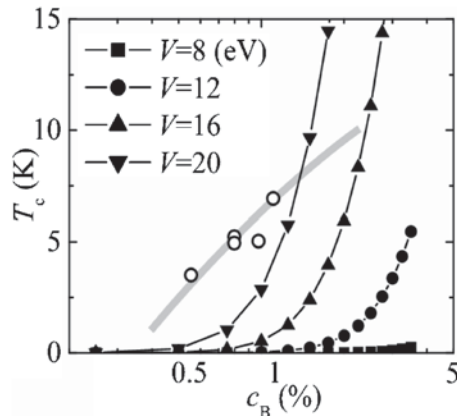


Fig. 7. Calculated superconducting critical temperature T_c compared with that obtained from the experiment in ref. 6.

of diffusion brought about by randomness, the Coulomb pseudopotential parameter μ^* effectively increases.^(33,34) (iv) Approximations superior to CPA are desirable. In CPA, both the width and height of the density of states of the impurity states vary as $\sim(c_B)^{1/2}$ (hence, the area varies as $\sim c_B$), whereas in reality, there is a tendency that the width varies as $\sim c_B$ and the height remains almost constant at ~ 1 . This discrepancy may be partly due to the insufficiency (or mean-field nature) of CPA. Points (i)–(iv) merit further study.

6. Discussion

Next, let us consider what would happen to T_c if there were no effects induced by the disorder of B ions. To determine this, we carry out supercell calculations for the same set of parameter values used in the CPA calculations. The B ions are arranged periodically using $6\times 6\times 6$, $5\times 5\times 5$, $4\times 4\times 4$, and $3\times 3\times 3$ supercells of the simple-cubic lattice (see Fig. 8), which correspond to 0.46%, 0.80%, 1.56%, and 3.7% doping rates, respectively. The calculated results for the density of states are compared with the results of CPA at the same doping rates. We find that a narrow density of states for the impurity states appears above the top of the valence band, which form bands (impurity bands), and that the density of states at the Fermi energy is much higher than the result obtained from the CPA calculation. The separation between the impurity band and the top of the valence band is ~ 1 eV, which is significantly larger than the energy of the impurity level in the dilute limit, 0.37 eV, due to repulsion between impurity energy levels. We then calculate the values of T_c for these supercell systems using the same expressions as used for the pairing susceptibility. We find that the obtained values of T_c are nearly an order of magnitude higher than the results obtained from the CPA calculations for the same attractive strength. Thus, we may conclude that, if impurity B ions could be arranged periodically, one would have a critical temperature of approximately ~ 100 K. This value of T_c exceeds $T_c \sim 30\text{--}40$ K, which is considered to be the theoretical limit according to BCS theory for conventional superconductors; this is not surprising because

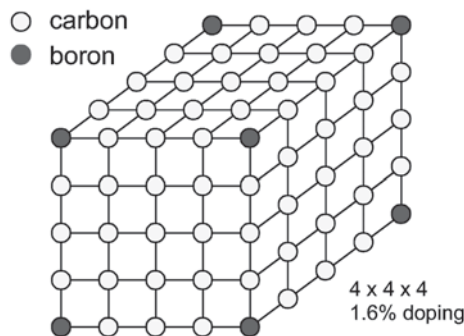


Fig. 8. Hypothetical periodic arrangement of B ions in the simple-cubic lattice of C ions.

of the exceptionally high Debye temperature of diamond $\omega_D = 2300$ K, together with the presence of the strong electron-phonon coupling due to the hole doping into the covalent bonds.⁽¹³⁾ In other words, in actual B-doped diamond, the effect of disorder strongly suppresses the value of T_c .

Together with reducing the effects of disorder, increasing the hole concentration is obviously another method of raising T_c . By conducting actual experiments, one may try to discover a method of removing or reducing the number of H atoms introduced during the syntheses of the materials; i.e., to reduce the passivation rate.

Finally, we consider the possibility of the occurrence of superconductivity in similar covalent-bonded materials. On the basis of the present study, we suggest that the doping of carriers in boron nitride (BN) and silicon carbide (SiC) may produce interesting results; these materials with the cubic (zinc blende) structure not only have a valence-band structure with a large band gap similar to that of diamond but also have a bond-stretching phonon mode with a very high frequency.⁽³⁵⁻⁴¹⁾ To be more specific, it is known that BN has a Debye temperature of 1700 K and a bond-stretching phonon mode of frequency ~ 130 meV,⁽³⁵⁾ and can be doped with C with a donor level of 0.28 eV (the carriers are electrons).⁽³⁶⁾ The bottom of the conduction band is nondegenerate,⁽³⁷⁾ so that the model used in our study can be directly applied. For SiC, it is known that the Debye temperature is 1200 K, the bond-stretching phonon frequency is ~ 100 meV,⁽³⁸⁾ and the material can be doped with either N, with a donor level of 0.06 eV,⁽³⁹⁾ or B, with an acceptor level of 0.735 eV.⁽⁴⁰⁾ The top of the valence band is triply degenerate, as in B-doped diamond, but the bottom of the conduction band is nondegenerate.⁽⁴¹⁾ We therefore suggest that the doping of carriers in these materials should be attempted, in order to find new superconducting materials with appreciable T_c for a variety of doped semiconductors.

7. Conclusions

We have reviewed our research^(23,24) on the superconductivity in B-doped diamond, where we have used CPA to consider the substitutional disorder of B ions and have treated the attractive force between holes using the ladder approximation under the assumption of instantaneous interaction. Our main findings are as follows:

- (i) The effects of disorder are essential for understanding the electronic state of B-doped diamond. The broadening of the density of states and single-particle spectra near the Fermi energy as well as the very short lifetime and mean free path are due to the effects of disorder, i.e., the large imaginary part of the self-energy induced by the disorder of B ions.
- (ii) The pairing susceptibility derived by CPA is in agreement with BCS theory, where the effects of disorder are renormalized solely into the density of states, even in the presence of a strong disorder that deforms the shape of the density of states. The vertex correction is found to be essential here; the susceptibility becomes too small if we only take into account the effect of the self-energy, but it is recovered by the vertex correction, leading to the BCS expression for the pairing susceptibility.

- (iii) The superconducting critical temperature T_c in B-doped diamond is strongly suppressed by the effects of disorder. It will therefore be very challenging to discover a method of removing or reducing the disorder in B doping, e.g., by the regular spatial arrangement of B ions or by another method with similar results, so that superconductivity with higher T_c is realized.

We believe that doped semiconductors are promising materials for producing a new class of superconductors and therefore hope that our work presented here will encourage further experimental studies on doped semiconductors.

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