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Non-Adiabatic Effects of Superconductor Silane under High Pressure *

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We present the investigations of non-adiabatic effects by including vertex corrections in the standard Eliashberg theory and show that high phonon frequency is unfavorable to superconductivity in the regime of strong vertex correction. This means that it is hard to find high-transition-temperature superconductors in the compounds with light elements if the non-adiabatic effects are strong. The interplay interaction between non-adiabatic effect and Coulomb interaction makes the transition temperature of silane superconductor not so high as predicted by the standard Eliashberg theory.

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The recently found superconductor silane (SiH_4) at high-pressure[1] partially realizes the theoretical predictions of high-temperature superconductors of hydrogen dominant metallic alloy[2] and metallic hydrogen at high pressure.[3] However, the transition temperature (T_c) is significantly lower than the values predicted in previously theoretical calculations.[4−7] What is the underlaying mechanism of the lower T_c compared with the higher T_c predicted in strong-coupling theory. With increasing pressure the electronic structures of silane will evolve from molecule-like energy-levels to electron-bands when gaseous silane transforms into crystalline silane. The effective band widths of conducting electrons of metallic silane are narrow so that the non-adiabatic effects will be prominent. It is very desirable to study the influences of non-adiabatic effects on T_c.

The non-adiabatic effects are partially included in the vertex corrections in the theory of electron-phonon interaction. The Eliashberg theory[8−10] combined with the vertex corrections has been widely studied by the perturbation method.[11−13] We have performed a full parameter-space search based on the Eliashberg theory with and without vertex corrections.[13,14] In this Letter, we study the influences of the vertex corrections on T_c of superconductor silane at high pressure using reliable Eliashberg functions $\alpha^2F(\omega)$ obtained from the calculations of linear-response theory.[4] Our results indicate that the vertex correction can efficiently suppress T_c approaching to the values found in experiments. Additionally, we find that high phonon frequency is unfavorable to high-T_c if the strong vertex corrections are included. This means that it is hard to realize high-T_c superconductors in silane and other hydrogen-rich materials.

We have generalized the equation of energy gap in Ref. [11] by including the Coulomb interaction.[13] The calculations of vertex corrections are greatly simplified under isotropic approximation. The electron-phonon interactions are included in the vertex corrections only by the functions λ_n defined as $\lambda_n = 2\int_0^\infty d\nu\alpha^2F(\nu)\nu/(\nu^2+\omega_n^2)$. When temperature is very close to T_c, the energy-gap equation including the leading vertex correction from electron-phonon interaction is written as $\sum_{n'=-\infty}^{+\infty} K_{nn'}\Delta_{n'}/|\omega_{n'}| = 0$ with the kernel matrix

$$K_{nn'} = [\lambda_{n-n'}B_{nn'} - \mu^* + C_{nn'}]a_{n'} - \delta_{nn'}H_{n'}, \quad (1)$$

where the definitions of H_n, $A_{nn'}$, $B_{nn'}$, $C_{nn'}$ and a_n can be found in Refs. [11,13]. In order to calculate T_c, the matrix $K_{nn'}$ is symmetrized with the same manner as in Ref. [10]. E_B is the effective band width and the Coulomb pseudo-potential μ^* is defined as $\mu^* = \mu_0/(1+\mu_0\ln(E_B/\Omega_0))$, where $\mu_0 = N(0)U$ with $N(0)$ being the density of state at Fermi energy E_F, U the Coulomb interaction between electrons, and Ω_0 the characteristic energy of typical phonon correlated to superconductivity. Generally, the ratio Ω_0/E_B takes as the parameter to measure vertex correction and the larger Ω_0/E_B is corresponding to the stronger vertex correction. In this context, the vertex corrections are controlled by E_B, the smaller E_B (or larger Ω_0/E_B) for stronger vertex corrections. With development of method of electronic-structure calculation in solid materials, the parameters of electron-phonon interaction can be calculated using density functional theory combined with perturbing linear-response theory.[15,16] The Elaishberg functions $\alpha^2F(\omega)$ obtained by linear response theory are easily merged in the formulae used in this context.

In the standard Eliashberg theory, T_c will infinitely increase with phonon frequency or energy. It is widely expected that high-temperature superconductors should be found in compounds containing light

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elements such as hydrogen-rich material silane and metal hydrogen at high pressure. We will see that the non-adiabatic effects set bounds to infinitely increasing T_c. In order to obtain very general results, at the first step, we apply the simple phonon spectrum and Eliashberg function $\alpha^2F(\omega)$ used in Ref. [9]. Figure 1 shows how T_c changes with Ω_P and E_B. We can see that for large band width $E_B = 5\,\text{eV}$ (weak vertex correction), T_c increases monotonously with Ω_P just as in the standard Eliashberg theory. However for very strong vertex correction with small band width E_B=1 eV, T_c is non-monotonously dependent on Ω_P. When Ω_P is larger than a threshold value, T_c will decrease with Ω_P. This means that, if the vertex corrections are included, high phonon energy is unfavorable to superconductivity.

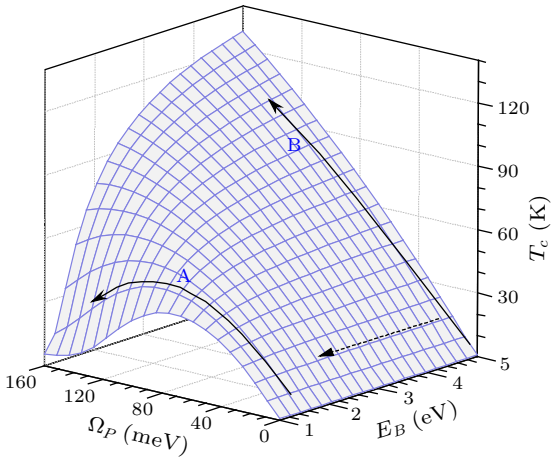

Fig. 1. T_c on $\Omega_P - E_B$ parameter plane. The two solid arrows A and B show the changes of T_c with Ω_P at fixed E_B (A) near 1 eV and (B) 5 eV, respectively. The dashed arrow shows the direction enhanced the vertex corrections.

We perform the calculations of the vertex corrections of real superconducting material silane containing the lightest element: hydrogen. The lowest order vertex correction can significantly reduce T_c when energy of phonon Ω_P is larger than 80 meV shown in Fig. 1. We adopt the Eliashberg functions that have been reported in Ref. [4] in the calculations of linear-response theory. The structures of crystal silane at high-pressures have not been completely defined. The metallic $P6_3$ structure has been found in experiment.[1] The theoretical layered structure with $Cmca$ space-group symmetry is more stable in 60–200 GPa,[4] other stable structures at higher pressures have already been reported.[17] The hydrogen-rich superconductor silane at higher pressure has very high phonon energies coming from the vibrations of hydrogen atoms. The effective phonon energies $\langle\omega\rangle_{ln}$ for silane at high pressures distribute from 50 meV to 75 meV, dependent on pressures. The parameters λ of electron-phonon interaction are about 1.17, 0.62, 0.75 for the $Cmca$ structure and 0.84, 0.87, 1.1 for the $C2/c$(2D) structure, respectively, when pressure increases from 70 GPa to 150 GPa and to 200 GPa. The calculated T_c using Allen-McMillan formula are distributed from 20 K to 80 K[4] that are greatly larger than experimental $T_c < 17$ K.

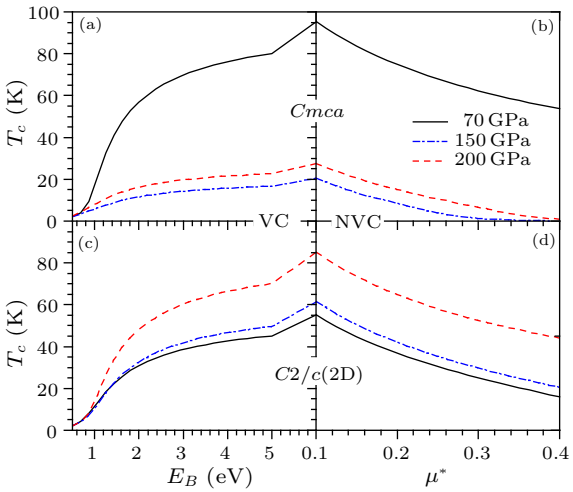

Fig. 2. Dependences of T_c on E_B and μ^*, based on the Eliashberg functions $\alpha^2F(\omega)$ obtained from linear-response theory at different pressures for the $Cmca$ structure (a,b) and $C2/c$(2D) structure (c,d) respectively.[4] Here (a) and (c) include vertex corrections (VC), and there are no vertex corrections (NVC) in (b) and (d).

As reported in Ref. [4], the pressure-dependence T_c of the $Cmca$ structure is different from the $C2/c$(2D) structure. The T_c of the $Cmca$ structure decreases when pressure increases from 70 GPa to 150 GPa, and slightly increases with increasing pressure further to 200 GPa. However for the $C2/c$(2D) structure, T_c monotonously increases with pressures from 70 GPa to 200 GPa. We first present the results of the $Cmca$ structure in detail, as shown in Figs. 2(a) and 2(b). If the vertex corrections are not included and $\mu^* = 0.1$, the values of T_c are close to 20 K at pressures 150 GPa and 200 GPa, which are smaller than those obtained from the McMillan formula. Thus our results indicate that for very broad distributed phonon spectrum, the standard strong-coupling calculations are needed to accurately calculate T_c beyond the simple McMillan and Allen T_c formulae.

At low pressure with larger inter-molecule distances, the crystalline silane is an insulator with narrow energy bands corresponding to discrete energy levels of a single molecule. The effective band widths will increase with increasing pressures and overlaps of molecular orbits of different molecules. If the vertex corrections are included in our calculations with the effective band width E_B of conducting electrons from 0.5 eV to 5 eV, then T_c at 150 GPa and 200 GPa are significantly reduced to the value smaller than 17 K as shown in Fig. 2(a). The effective band widths of conducting electrons obtained from the density functional calculations increase from very small values at

low pressure to about 1.0–3.0 eV at 100–200 GPa for the $C2/c$(2D) and Cmca structures.[4] In another case, the effects of Coulomb interaction are more significant because T_c decreases to very small values with increasing μ^* at 150 GPa and 200 GPa as shown in Fig. 2(b). If we want to know what is more important for the depression of T_c we should know real value of μ^* at 150 GPa and 200 GPa. At 70 GPa, the small band widths with E_B<1 eV guarantee T_c lower than 17 K. We find that, if without vertex correction, even larger μ^* (= 0.4) can not suppress T_c to a value smaller than 17 K at 70 GPa.

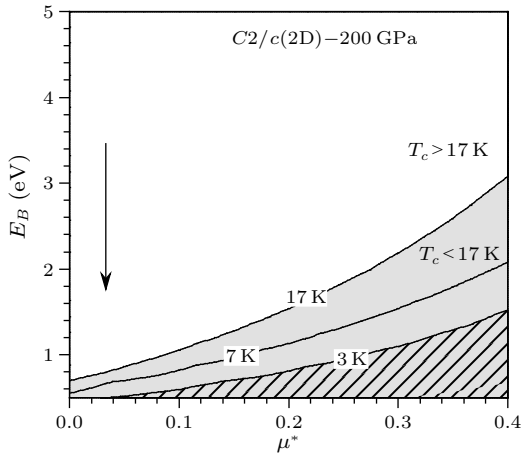

Fig. 3. T_c on μ^*-E_B plane using the Eliashberg functions $\alpha^2F(\omega)$ obtained from the linear-response theory.[4] The structure has $C2/c$(2D) space group symmetry at pressure 200 GPa. The shade areas are regions with T_c < 17 K. The arrow shows the directions of increasing the vertex correction.

The effects of vertex correction of the $C2/c$(2D) structure in Fig. 2(c) are similar to the $Cmca$ structure in Fig. 2(a), however the influences of the Coulomb interaction in Fig. 2(d) are different from the $Cmca$ in Fig. 2(b). The $C2/c$(2D) structure is less stable than the $Cmca$ structure. The stronger electron-phonon interaction induces its structure more unstable at high pressures from 150 GPa to 200 GPa. Theoretical values of T_c increase with pressures. This is because the enhancements of T_c induced by the increase of λ are larger than the depressions of T_c due to the vertex correction. For the $C2/c$(2D) structure, the Coulomb interaction depresses T_c to values which are still larger than 17 K as shown in Fig. 2(b). Therefore, Coulomb interaction individually can not explain why T_c smaller than 17 K in high-pressure experiments. The effective band widths E_B of conducting electrons in the $C2/c$(2D) structure are larger than 1 eV at 150 GPa and 200 GPa[4] so that the vertex correction individually can not explain low T_c at 100 GPa and 200 GPa as well, as shown in Fig. 2(c). Figure 3 illustrates T_c for the $C2/c$(2D) structure on μ^*-E_B plane at pressure 200 GPa. The shading region in the figure shows the effective region with parameters μ^* and E_B that can explain T_C smaller than 17 K, which is only small area of parameter-space with $\mu^* > 0.1$ and $E_B < 2.0$ eV. Especially, the hatched region shows the parameter-space to explain why experimental T_c is smaller 3 K at 200 GPa. Thus our results indicate that the interplay of vertex correction and Coulomb interaction can explain the low T_c of the $C2/c$(2D) structure. Our calculations can not fully explain pressure-dependent T_c in Ref. [1], especially, the higher T_c = 17 K near 100–120 GPa because of the lack of information of structural changes.

In summary, we have studied the effects of the vertex correction on T_c for superconductor of silane at high pressure. Our results indicate that the non-adiabatic effects are the barrier to prevent T_c from increasing infinitely with phonon frequency. This means that it is hard to realize high T_c to home temperature in high pressure rich-hydrogen materials. Our results also show that the interplay interaction between Coulomb interaction and vertex correction is essentially to explain the larger differences between theory and experiments.

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References

[1] Eremets M I, Medvedev I A, Tse J S and Yao Y 2008 *Science* **319** 1506
[2] Ashcroft N W 2004 *Phys. Rev. Lett.* **92** 187002
[3] Ashcroft N W 1968 *Phys. Rev. Lett.* **21** 1748
[4] Chen X J, Wang J L, Struzhkin V V, Mao H K, Hemley R J and Lin H Q 2008 *Phys. Rev. Lett.* **101** 077002 and supplemental materials
[5] Feng J, Grochala W, Jaroń T, Hoffmann R, Bergara A and Ashcroft N W 2006 *Phys. Rev. Lett.* **96** 017006
[6] Pickard C J and Needs R J 2006 *Phys. Rev. Lett.* **97** 045504
[7] Yao Y, Tse J S, Ma Y and Tanaka K 2007 *Eur. Phys. Lett.* **78** 37003
[8] Éliashberg G M 1960 *Sov. Phys. JETP* **11** 696
[9] Scalapino D J, Schrieffer J R and Wilkins J W 1966 *Phys. Rev.* **148** 263
[10] Allen P B and Dynes R C 1975 *Phys. Rev.* B **12** 905
[11] Kostur V N and Mitrović B 1994 *Phys. Rev.* B **50** 12774
[12] Grimaldi C, Pietronero L and Strässler S 1995 *Phys. Rev.* B **52** 10530
[13] Fan W 2009 *Physica* C **469** 177
[14] Fan W 2008 *Chin. Phys. Lett.* **25** 2217
[15] Savrasov S Y and Savrasov D Y 1996 *Phys. Rev.* B **54** 16487
[16] Baroni S, de Gironcoli S, Dal Corso A and Giannozzi P 2001 *Rev. Mod. Phys.* **73** 515
[17] Martinez-Canales M, Oganov A R, Ma Y M, Yan Y, Lyakhov A O and Bergara A 2009 *Phys. Rev. Lett.* **102** 087005