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Doped holes and/or electrons induced superconductivity domes explained with the free volume concept

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Abstract

The conductivity equation developed in our previous work without any restrictions to specific materials is employed to explore how superconductivity transition temperature T_c changes with the doped hole or electron concentrations based on the free volume concept. The predicted relationship is used to fit experimental data available in the literature and a good agreement with observations is achieved. Our findings may provide an alternative explanation for doping-induced domes and/or double domes with a-dip phenomena observed among many superconductors.

Keywords Superconductivity dome, Eyring's rate process theory, Free volume concept

Introduction

Dome-shaped relationships between superconductivity transition temperature T_c and doped electron or hole concentration in cuprates were observed right after the breakthrough work of high-temperature superconductivity found in cuprates (Bednorz and Müller 1986; Tokura et al. 1988). It has been an extensive research topic for the last 40 years (Ding et al. 1996; Li et al. 2019; Honma and Hor 2006; Lin et al. 2021; Chen et al. 2019). A similar dome-like correlation was found in other types of superconductors like iron-based (Cho et al. 2016), hydrides (Mazziotti et al. 2021), and metal alloys (Wan et al. 2023), etc., in both electron and hole-doped systems. It seems to be a universal phenomenon across broad superconductor materials, and theoretical investigations are performed as well (Wang et al. 2023; Langmann et al. 2019). A dome-shaped relationship can be theoretically projected in principle; however, an exact equation governing superconductivity transition temperature and the doped concentrations has not been elucidated. An empirical equation obtained from experimental data regression is shown below, and it works for many superconductors (Presland et al. 1991; Honma and Hor 2006):

$$T_c = T_{max}^c \left[1 - 82.6(p - 0.16)^2\right] \quad (1)$$

where T_c is the superconductivity transition temperature, T_{max}^c is the maximum value of T_c in dome-shaped curve, and p is the doped hole concentrations. Note that p is intrinsically defined as a two-dimensional (2D) quantity, the hole content per CuO_2 plane (Honma and Hor 2006). It can be determined by the excess oxygen content, the thermoelectric power at room temperature S^{290}, or the Hall coefficient. The number 0.16 in Eq. 1 is later experimentally found not to be a universal constant for all superconductors (Honma and Hor 2006; Honma et al. 2004). A modified version of Eq. 1 was proposed to replace the parameter p with a 3D quantity p_{3D} (Honma and Hor 2006):

$$T_c = T_{max}^c \left[1 - 83.63(p_{3D} \times 10^{-22} - 0.159)^2\right] \quad (2)$$

Equation 2 was claimed to have a universal nature of fitting capability (Honma and Hor 2006). Mathematically, both Eqs. 1 and 2 are very similar. Theoretically, an

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equation similar to empirical Eqs. 1 and 2 is still missing, even though intensive studies have been performed to elucidate the dome-shaped phenomena.

In this article, we will use the generic conductivity equation developed before to target this issue. We will continue to use the free volume as a main argument and assume that the free volume of the doped holes or electrons should be excluded when we consider that of the conduction electrons. In other words, doped electrons/holes will take some free volume that originally belonged to the conduction carriers in the system. We will go with the theoretical derivation first and then compare the obtained equations with the experimental data through data fitting practice. A discussion and summary will be at the end of the article.

Theoretical derivations

We will borrow the generic conductivity equation developed in 2015 (Hao 2015) to determine the superconductivity transition temperature first. This equation can be simplified as (Hao 2015, Hao 2023b, Hao 2019)

$$\sigma = AT\left[\exp\frac{B\alpha}{T} - \exp\frac{-B(1-\alpha)}{T}\right] \tag{3}$$

where $A = \frac{ek_BN_c}{hE}\exp\left(\frac{-\Delta G}{RT}\right)\lambda$, $B = \frac{eE}{k_B}\lambda$, $\lambda = \left[\frac{(9\pi N_v - 4)V_m}{9\pi N_v N_c}\right]^{1/3}$, the electron travel distance. e is the electron charge, k_B is the Boltzmann constant, T is the temperature, h is the Planck constant, R is the gas constant, E is the applied electric field, N_v is the number of valence electrons per unit cell, N_c is the number of conduction electrons in the whole system, V_m is the volume of a material under study, ΔG is the standard Gibbs free energy, and α is a parameter related to the packing structures of electrons and has a simple relationship with the coordinate number of an electron in the system, c_n, $\alpha = \frac{1}{c_n}$. As indicated in the articles (Hao 2015 and Hao 2023a), the parameter λ is temperature dependent, as V_m is dependent on temperature with a generic relationship $V_m = a + bT^n$, typically $n < 1.5$ (Kaptay 2015; Srivastava 2006). However, it can be reasonably assumed to be independent of temperature unless temperature approaches zero Kelvin, as demonstrated in the article (Hao 2023a). We want to find the relationship between T_c and λ, so approximately we will assume that λ is independent of temperature. There are several articles cited therein (Hao 2015) showing that λ can be assumed independent of temperature if the temperature variation range is not too wide. Differentiating Eq. 3 and assuming $d\sigma/dT = 0$, $e^{-x} \approx 1 - x + \frac{x^2}{2}$, we obtain:

$$T_c = \frac{\frac{e^2\lambda^2N_c}{h}\left(\frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{k_B}\right)}{2\left(\frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{2k_B}\right)} \tag{4}$$

$$= \frac{e^2\lambda^2N_c}{2h}\left(1 + \frac{1}{\frac{2k_B\Delta G}{eE\lambda R} - 2\alpha + 1}\right) \tag{5}$$

Equation 5 indicates that T_c has a complicated relationship with λ, and it also varies with the parameter α that has been demonstrated in our previous articles (Hao 2015, Hao 2019, Hao 2023a, Hao 2023b). However, this equation can be simplified if we assume $\frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{k_B} \approx \frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{2k_B}$. Hence, we can rewrite Eq. 5 as:

$$T_c = \frac{e^2N_c}{2h}\lambda^2 \tag{6}$$

Equation 6 indicates that superconductivity transition temperature has a quadratic relationship with λ. Note that Eq. 6 is obtained under several approximations to correlate to λ. The different expressions of T_c shown in the articles (Hao 2023a, Hao 2023b) are from the same original Eq. 3 and approximated for particular purpose. Now, we need to correlate T_c with the free volume of doped electrons or holes via the inter-particle spacing (IPS) concept developed for particulate dispersion systems. The inter-particle spacing is derived in the article (Hao and Riman 2006). For 3D systems, we assume that the particles can freely move in x, y, and z directions for a distance two times the inter-particle spacing and obtain the free volume of an individual particle as:

$$V_{if}^{3D} = 64r^3\left[\left(\frac{\phi_m}{\phi}\right)^{1/3} - 1\right]^3 \tag{7}$$

$$= \frac{3V_m}{4\pi N_d}\left[\phi_m^{1/3} - \phi^{1/3}\right]^3 \tag{8}$$

by considering that $\phi V_m = \frac{4\pi r^3N_d}{3}$, where r is the radius of the particle, V_m is the volume of the material, ϕ_m is the maximum packing fraction of particles, N_d is the number of holes or electrons doped in the system, and ϕ is the doped particle volume fraction. The free length, L_{if}^{3D}, can be defined as the basic scale of the free volume and thus can be expressed as:

$$L_{if}^{3D} = 4r\left[\left(\frac{\phi_m}{\phi}\right)^{1/3} - 1\right] \tag{9}$$

$$= \left(\frac{3V_m}{4\pi N_d}\right)^{1/3}\left[\phi_m^{1/3} - \phi^{1/3}\right] \quad (10)$$

We then may use Eq. 10 to build 2D situations by assuming that the particles can freely move only in x and y directions. Therefore, the free area/volume of an individual particle in a 2D system can be expressed as:

$$V_{if-r}^{2D} = 16r^2\left[\left(\frac{\phi_m}{\phi}\right)^{1/3} - 1\right]^2 \quad (11)$$

$$= \frac{16V_m\phi}{\pi N_d}\left[\left(\frac{\phi_m}{\phi}\right)^{1/3} - 1\right]^2 \quad (12)$$

$$= \frac{16V_m\phi^{1/3}}{\pi N_d}\left[\phi_m^{1/3} - \phi^{1/3}\right]^2 \quad (13)$$

if we use the relationship with the particle radius in 3D and replace r^2 with $V_m\phi/\pi N_d$ in 2D systems. Alternatively, we may use the relationship with the volume fraction in 3D to obtain the free area/volume:

$$V_{if-\phi}^{2D} = \left(\frac{3V_m}{4\pi N_d}\right)^{2/3}\left[\phi_m^{1/3} - \phi^{1/3}\right]^2 \quad (14)$$

Note that Eqs. 13 and 14 are slightly different, and we will show later that they are very similar in terms of data fitting. They are just derived from a different route. Equations 13 and 14 can be used to describe the free area/volume of a doped individual hole or electron in 2D systems. The free area/volume of conduction carriers that participate in the superconductivity must be excluded from the free area/volume of doped holes or electrons, i.e., these two items should be constrained by each other. The free volume of carriers in a conduction system is dependent on the temperature, packing structures of the carriers, the atomic lattice structures of the material, etc. In other words, on the condition that the material is under study, the free volume has a certain value. Once extra holes/electrons are introduced into the system, they will take some free volume from the original "majority" of the conduction carriers, as the doped holes/electrons will participate in conduction behaviors. This is the reason that they must be excluded from each other. The free volume of an individual conduction carrier may be expressed as a subtraction of the unit volume from that of a doped electron/hole:

$$V_{ifr}^{c-2D} = 1 - \frac{16V_m\phi^{1/3}}{\pi N_d}\left[\phi_m^{1/3} - \phi^{1/3}\right]^2 \quad (15)$$

if we start with Eq. 13. If we start with Eq.14, we may obtain another equation:

$$V_{if\phi}^{c-2D} = 1 - \left(\frac{3V_m}{4\pi N_d}\right)^{2/3}\left[\phi_m^{1/3} - \phi^{1/3}\right]^2 \quad (16)$$

As the free area/volume defined previously (Hao 2015), for 2D systems the free area/volume of an individual conduction carrier can be correlated with the travel distance λ as:

$$V_{ifr}^{c-2D} \text{ or } V_{if\phi}^{c-2D} = (2\lambda)^2 = 4\lambda^2 \quad (17)$$

Combining Eqs. 6, 15, 16, and 17 leads to the correlation between T_c and the doped hole or electron concentration for 2D system

$$T_c = \frac{e^2N_c}{8h}\left[1 - \frac{16V_m\phi^{1/3}}{\pi N_d}\left(\phi^{1/3} - \phi_m^{1/3}\right)^2\right] \quad (18)$$

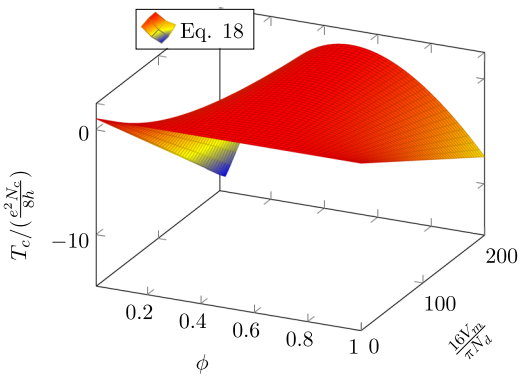
and

$$T_c = \frac{e^2N_c}{8h}\left[1 - \left(\frac{3V_m}{4\pi N_d}\right)^{2/3}\left[\phi_m^{1/3} - \phi^{1/3}\right]^2\right] \quad (19)$$

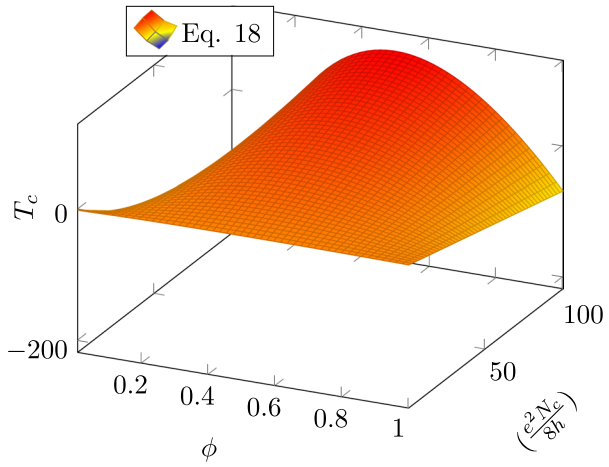
Equations 18 and 19 show the relationship between T_c and the doped hole/electron concentration ϕ for 2D systems. Both Eqs. 18 and 19 are very similar to the empirical equation Eqs. 1 and 2, especially the Eq. 19. They render the physical meanings of several parameters/constants in Eq. 1: (1) T_{max}^c is equal to $\frac{e^2N_c}{8h}$; (2) the number "82.6" actually is $\frac{16V_m\phi_h^{1/3}}{\pi N_d}$ in Eq. 18 and $\frac{3V_m}{4\pi N_d})^{2/3}$ in Eq. 19; (3) the value "0.16" actually is $\phi_m^{1/3}$), depending on the maximum packing fraction; (4) it is the doped hole/electron concentration to the power of 1/3 instead of 1 that correlates with T_c; (5) ϕ by definition should be less than ϕ_m, as ϕ_m is the maximum packing fraction of holes/electrons.

Results

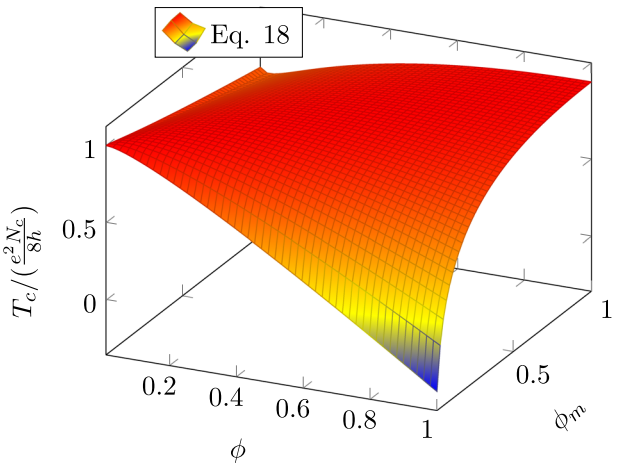
First, we will plot Eqs. 18 and 19 with various parameters to gain an idea of whether these equations can predict a dome-shape relationship, which is shown in Figs. 1 and 2, respectively. The regular and normalized superconductivity transition temperature, T_c and $T_c/\left(\frac{e^2N_c}{8h}\right)$, do show a maximum against the hole concentration. The heights of the maximum point increase with both $\left(\frac{e^2N_c}{8h}\right)$ and $\frac{16V_m}{\pi N_d}$, implying that both parameters control the maximum superconductivity transition temperature. The maximum packing fraction ϕ_m, on the other hand, does not create a dome-like relationship between T_c and ϕ. The normalized superconductivity transition temperature only monotonically decreases with the doped hole/



(a) Normalized superconductivity transition temperature vs. hole concentration and the parameter $\frac{16V_m}{\pi N_d}$ in Eq. 18



(b) Superconductivity transition temperature vs. hole concentration and the parameter $(\frac{e^2N_c}{8h})$ in Eq. 18

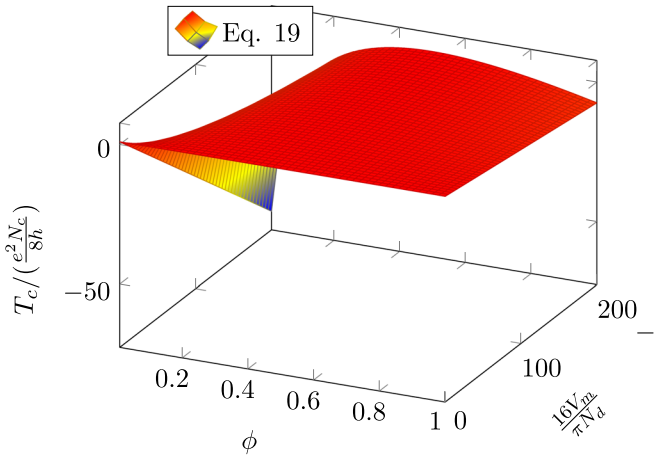


(c) Normalized superconductivity transition temperature vs. hole concentration and the maximum packing fraction in Eq. 18

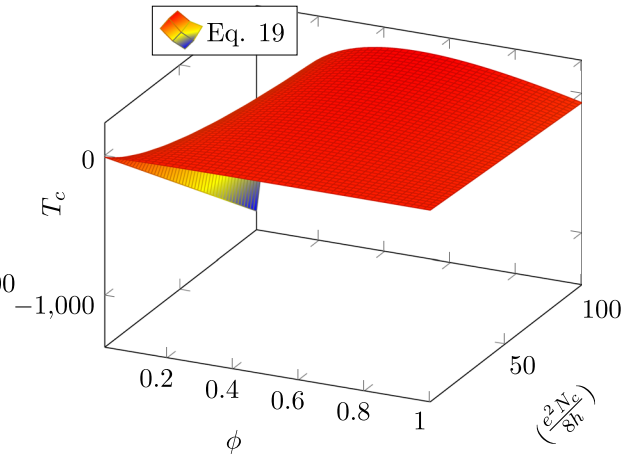
Fig. 1 Illustration of superconductivity transition temperature against the hole concentration and other three key parameters in Eq. 18

electron concentration at low ϕ_m but slightly increases at high ϕ_m. Figures 1 and 2 demonstrate that Eqs. 18 and 19 seem working for the doping induced process. The dome-shape is most pronounced when both $\left(\frac{e^2N_c}{8h}\right)$ and $\frac{16V_m\phi^{1/3}}{\pi N_d}$ are higher and diminishes when these two parameters are low. Note that T_c and $T_c/\left(\frac{e^2N_c}{8h}\right)$ should be positive, and the negative axis is shown for easy viewing purpose. Comparing Figs. 1 and 2, we may easily tell that the former gives a taller dome than the latter.

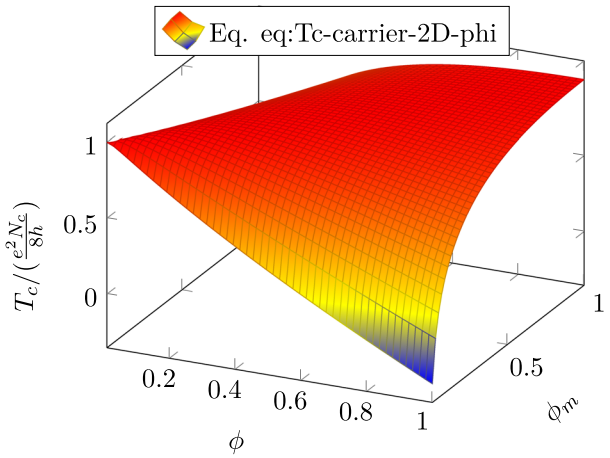
Let us compare empirical equations Eqs. 1 and 2 with newly derived ones, Eqs. 18 and 19. Figure 3 shows the normalized superconductivity transition temperature vs. the hole concentrations for these three equations. For Eq. 18, $\frac{16V_m}{\pi N_d}$ is assumed to be 82.6, same as the one in Eq. 1, and ϕ_m is assumed to be 0.125. In Eq. 2, the term $p_{3D} \times 10^{-22}$ is assumed to be ϕ. Equations 1 and 2 are overlapping together, and they peak at the same place. However, Eq. 18 peaks at different places and is unable to overlap with Eqs. 1 and 2, no matter how I change the parameters. A different shape will be obtained if they are forced to peak at the same location. Eq. 19 is very similar to the empirical equations but shows a narrower dome



(a) Normalized superconductivity transition temperature vs. hole concentration and the parameter $\frac{16V_m}{\pi N_d}$ in Eq. 19



(b) Superconductivity transition temperature vs. hole concentration and the parameter $(\frac{e^2N_c}{8h})$ in Eq. 19



(c) Normalized superconductivity transition temperature vs. hole concentration and the maximum packing fraction in Eq. 19

Fig. 2 Illustration of superconductivity transition temperature against the hole concentration and other three key parameters in Eq. 19

shape with the same parameters, $\frac{3V_m}{4\pi N_d}^{2/3}$ is assumed to be 82.6, same as the one in Eq. 1 and ϕ_m is assumed to be 0.125. At least all four equations predict a dome-like relationship.

The best approach to test the equations would be how they can fit the experimental data. Figure 4 shows the data points extracted from the literature and fitted with Eq. 1 in Fig. 4a, with Eq. 18 in Fig. 4b, and with Eq. 19 in Fig. 4c. The fitting parameters and quality R^2 are shown in Table 1. Note that we keep using *"p"* to represent the doped concentration for consistency with the literature, but it is replaced with ϕ during the fitting process. All three equations can give a good fitting for SrD-La214 superconductor, and the fitting quality indicated with R^2 is 0.91 for Eq. 1, 0.94 for Eq. 18, and 0.91 for Eq. 19. This superconductor has a perfect dome shape. However, both CD-Bi2201 and OD-Hg1201 superconductors show skewed dome-shapes, and Eq. 1 shows a very poor fitting quality with negative R^2. However, Eqs. 18 and 19 can still fit these two materials with very good

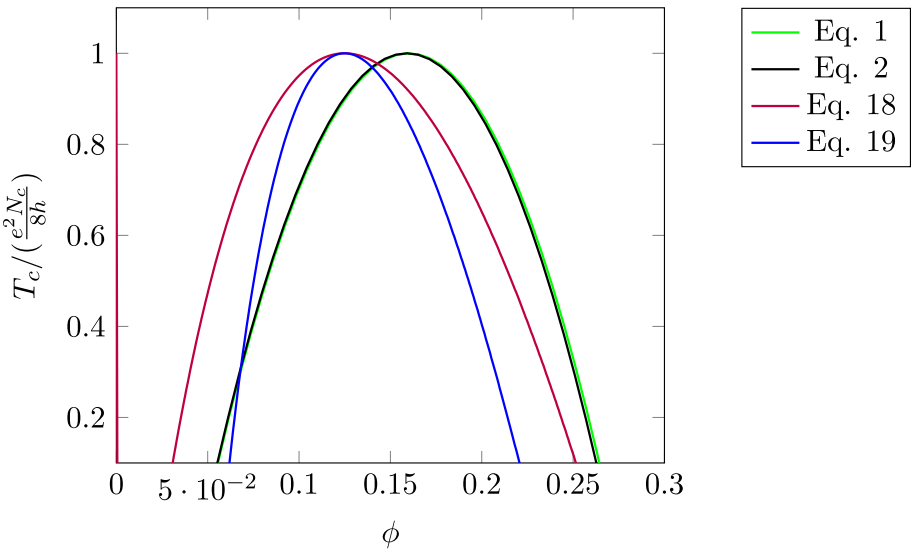


Fig. 3 The normalized superconductivity transition temperature $T_c/\left(\frac{e^2N_c}{8h}\right)$ vs. the doped hole/electron concentration $p(p = \phi)$ with Eqs. 1, 2, 18 and 19.

fitting quality; R^2 is 0.97 and 0.97 for OD-Hg1201 and 0.95 and 0.94 for CD-Bi2201 with Eqs. 18 and 19, respectively. Excellent fitting quality with Eqs. 18 and 19 indicates that these two equations work for dome-shaped superconductivity phenomena observed for many superconductors, though Eq. 18 shows a slight better fit than Eq. 19 does. The numeric values before the quadratic term obtained with Eq. 18 are close to an empirical constant 82.6. The average of these three values is 78, which is not far away from 82.6 and may explain why the empirical Eq. 1 sometimes works for a lot of superconductors. On the other hand, the numeric values before the quadratic term obtained with Eq. 19 are far away from 82.6.

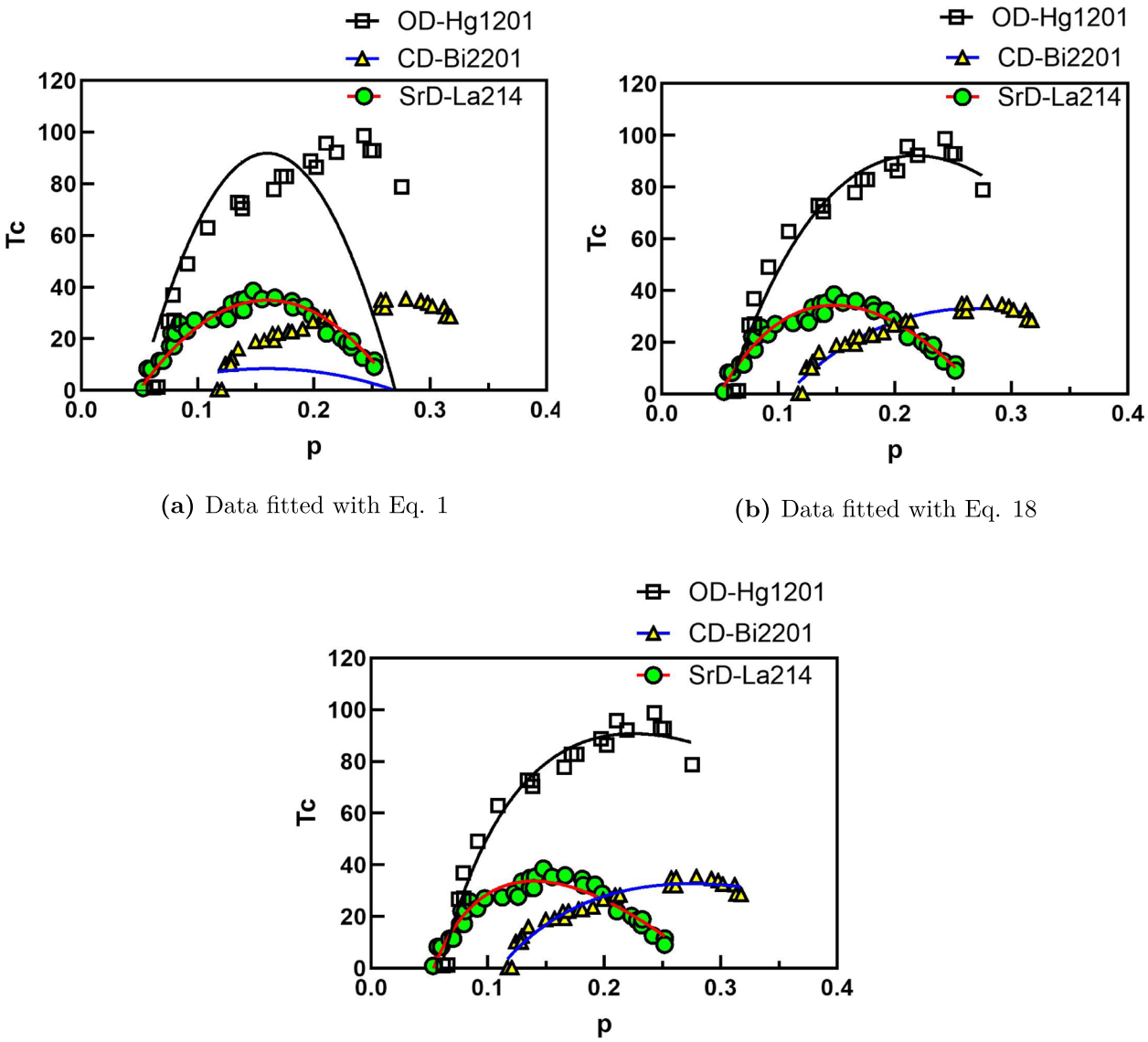
Theoretically, ϕ should be less than ϕ_m based on the original concept that there is a maximum packing volume fraction. However, as shown in Table 1, the obtained ϕ_m from the best fitting process is less than the doping concentration. This seems to be contradictory to our original assumptions. However, please note that the ϕ_m corresponds to the peak of T_c, implying that ϕ_m is the maximum doping level; beyond this value, T_c is going to decrease. Over-doping is harmful but of experimental significance.

There are many examples in literature that the dome-shape is skewed rather than the perfect (Lin et al. 2021; Li et al. 2019). Equations 18 and 19 should be more powerful to describe this universal phenomenon, as it is derived from the generic conductivity equation without being bound to any specific materials. The empirical equation can only work for perfect dome-shaped superconductors.

A "dip" in the dome has been identified recently in $Nd_{1-x}Sr_xNiO_2$ type (Chow and Ariando 2022; Li et al. 2020). Such a dip happens when the Hall coefficient transfers from the negative to the positive, implying that the system experiences from the electron-doped to the hole-doped process. As we may imagine, the maximum packing fraction of holes should be larger than that of electrons, i.e., ϕ_m should increase slightly. Figure 5 demonstrates that a dip happens if we increase ϕ_m from 0.125 to 0.216. The curves are predicted with Eq. 18. The lowest point corresponds to the average of these two ϕ_m values divided by 2, i.e., $\phi_{dip} = (\phi_{m1} + \phi_{m2})/2$. How to predict ϕ_m? It is dependent on the material and its electronic structure, which is out of the scope of this article. The dome is formed due to the suppression of the free volume from the doped holes or electrons. So it is not strange to see that the dome-shape superconductivity can be induced by an external pressure (Chen et al. 2021; Zhang et al. 2021). External pressure can suppress the free volume of electrons as well, as detailed in my recent article (Hao 2022). A transition from hole to electron-doped (Zeng et al. 2020), from charge density wave to superconductivity (Chen et al. 2021), or any changes that may vary the maximum packing fraction, will induce double domes.

Discussion

The limitations of Eq. 18 come from the approximations of Eq. 3. (1) N_c and λ that contains N_c are independent of temperature. Generally, this assumption is valid, except in extremely low temperatures; (2)



(a) Data fitted with Eq. 1

(b) Data fitted with Eq. 18

(c) Data fitted with Eq. 19

Fig. 4 Experimental data points extracted from the literature (Honma and Hor 2006) and therein for SrD-La214 ($La_{2-x}Sr_xCUO_4$), CD-Bi2201($Bi_2Sr_{2-x}La_xCuO_{6+\delta}$), and OD-Hg1201 ($HgBa_2CuO_{4+\delta}$) are regressed with Eqs. 1, 18, and 19. The solid lines are best fitted with the equations

$\frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{k_B} \approx \frac{\Delta G}{R} - \alpha\frac{eE\lambda}{k_B} + \frac{eE\lambda}{2k_B}$, which is not exact; (3) we use $e^{-x} \approx 1 - x + \frac{x^2}{2}$ to simplify the exponential function. They should be valid unless the temperature is extremely low close to zero Kelvin. These choices are unfortunately needed to find the relationship between T_c and the free volume of doped holes or electrons. Equations 18 and 19 are derived for both hole and electron-doped systems. No matter which particle is, the free volume of the doped holes or electrons should be excluded from the free volume that the conduction electrons can have.

I would like to emphasize that "conduction electrons" means the electrons that participate conduction process. The term originated from the conduction equation developed for the electron conduction process. It should include any conduction carriers including both electrons and holes. Since we are dealing with doped electrons/holes in this article, the doped electrons/holes can only be distinguished from the free volume confounding effect as expressed with Eq. (16).

The free volume argument is valid for all superconductors. The impact of doped electrons and holes on T_c

Table 1 The fitting parameters and quality R^2 with Eqs. 1, 18, and 19

Equation	Parameter	SrD-La214	OD-Hg-1201	CD-Bi2201
Eq. 1	T_c^{max}	34.95	91.84	8.43
	ϕ_m		0.16	
	Coefficient before the quadratic term		82.6	
	Regression R^2	0.91	-0.236	-5.304
Eq. 18	T_c^{max}	34.34	92.19	33.14
	ϕ_m	0.148	0.218	0.269
	Coefficient before the quadratic term	105.0	54.83	72.52
	Regression R^2	0.94	0.97	0.95
Eq. 19	T_c^{max}	33.72	90.78	32.69
	ϕ_m	0.140	0.225	0.275
	Coefficient before the quadratic term	49.53	21.32	34.14
	Regression R^2	0.91	0.97	0.94

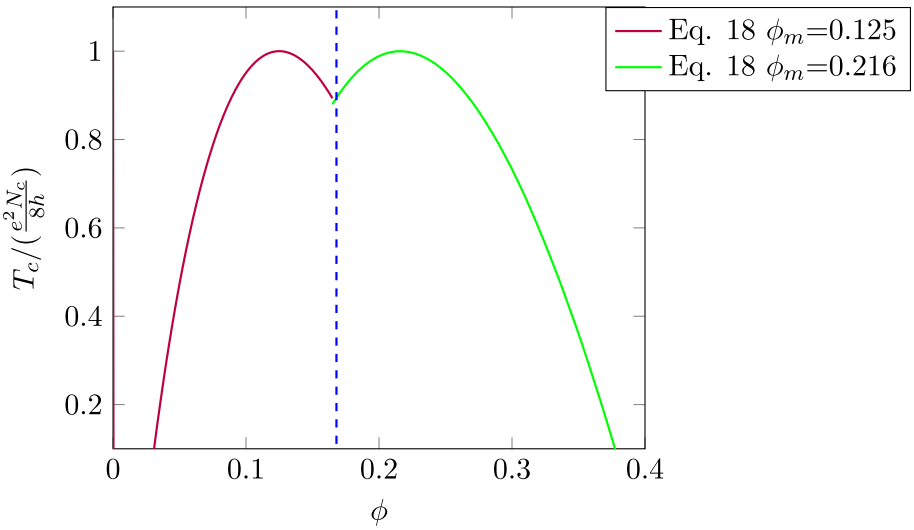


Fig. 5 The normalized superconductivity transition temperature $T_c/\left(\frac{e^2N_c}{8h}\right)$ vs. the doped hole/electron concentration with Eq. 18 at different maximum packaging fractions, which forms double domes and creates a dip

may be very pronounced for low-T_c superconductors but not too much for high-T_c superconductors, which is of course material-dependent. Please take a look at Eq. (5), T_c is related to the parameter α that is small for the low-T_c superconductors and large for high-T_c superconductors (Hao 2019). When α is large, the change of α may make T_c lower rather than higher. The enhancement is limited for high-T_c superconductors via the doping method. In overdosed regions, the doped electrons/holes would not further impact the free volume of the conduction electrons. In such a situation, we deal with a mixture of two materials rather than one compound and do not expect further suppression effect, which is the reason that there is a dome-shaped relationship.

Free volumes of electrons and holes should be quite different due to their different maximum packing fractions and packing structures of these two, which may explain the dips for any materials, including infinite-layer nickelates, during the doping process. The concepts are demonstrated in Figs. 3 and 5. Anything different in terms of the packing structures and the ultimate ϕ_m will shift the dome from one location to another, creating a dip.

We further demonstrate that the free volume of conduction electrons is critical. The superconductivity transition temperature is tied to the electron travel distance that in turn associated with free volume. Doped holes or electrons cannot share the free volume with

the conduction electrons, which leads to dome-like behaviors. The external pressure of course can induce dome-shape correlation, as it can change the free volume of conduction electrons, the same as doped holes or electrons.

The number of conduction electrons and the maximum doping level to achieve the highest transition temperature can be obtained from Eq. 18. These parameters may be useful when we design, synthesize, and optimize superconductivity materials. Compared with empirical equations, our equation renders physical meanings for every constant.

Conclusion

Utilizing the generic conductivity equation developed in our previous article, we have correlated the superconductivity transition temperature with the doped hole or electron concentration. The basic idea is simple: the doped species, holes or electrons, will suppress the free volume of the conduction electrons, resulting in a different travel distance of electrons. The obtained equations do reproduce a dome-like relationship, show an identical prediction as the empirical equations when the dome-shape is perfect, and can accurately fit the experimental data when the dome-shape is skewed, much better than the empirical equations. Since our equation is not bound to any specific materials, it should be capable of describing various superconductors. The dome-shaped behaviors of doped systems should be a common feature across a broad of materials. Double domes with a dip can happen when the maximum packing fraction change happens, either due to the doping type or doping structure changes. Anything that can change the free volume of conduction electrons, such as external pressure, can generate a dome-shaped behavior, which will be covered in our future article. Our equations can work for both holes and electrons-doped systems.

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Declarations

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