

Disentangling the role of bond lengths and orbital symmetries in controlling T_c in optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_7$

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Optimally doped YBCO ($\text{YBa}_2\text{Cu}_3\text{O}_7$) has a high critical temperature, at 92 K. It is largely believed that Cooper pairs form in YBCO and other cuprates because of spin fluctuations, the issue and the detailed mechanism is far from settled. In the present work, we employ a state-of-the-art *ab initio* ability to compute both the low and high energy spin fluctuations in optimally doped YBCO. We benchmark our results against recent inelastic neutron scattering and resonant inelastic X-ray scattering measurements. Further, we use strain as an external parameter to modulate the spin fluctuations and superconductivity. We disentangle the roles of Barium-apical Oxygen hybridization, the interlayer coupling and orbital symmetries by applying an idealized strain, and also a strain with a fully relaxed structure. We show that shortening the distance between Cu layers is conducive for enhanced Fermi surface nesting, that increases spin fluctuations and drives up T_c . However, when the structure is fully relaxed electrons flow to the d_{z^2} orbital as a consequence of a shortened Ba-O bond which is detrimental for superconductivity.

Even while more than three decades have passed since their discovery, the origin of superconductivity in cuprates remain highly debated. It is largely believed that at least for several variants of cuprates it is primarily spin fluctuations that drive superconductivity[1, 2]. However, the issue is far from settled. It is difficult to resolve because the phase diagram is very dense: small excursions in parameter space drives the material from one phase to a new phase. Multiple low energy scales are present and sometimes intertwined. Theoretically, the challenge has been to build a material specific ability that incorporates all such interactions in the right proportions which can also predict ways to disentangle their roles. Optimally doped YBCO ($\text{YBa}_2\text{Cu}_3\text{O}_7$) is one of the higher T_c superconductors in the cuprate family. In this work we present a high-fidelity *ab initio* theory that is designed to realize this objective, and we use it to explore how spin fluctuations and superconductivity can be modified through strain. In a recent ultrafast experiment [3] on $\text{YBa}_2\text{Cu}_3\text{O}_7$ large amplitude distortions on apical Oxygens were induced to modulate the inter layer coupling. Here we apply a uniaxial strain along the c -axis. We perform two distinct excursions under strain; (a) an ideal strain where all atoms displace in proportion to their height along the c axis, and (b) allowing the internal coordinates to relax under the strain. We show that strain modifies superconductivity in both cases, but in different ways, thus highlighting how a detailed understanding of the mechanism is essential in order to predict and control unconventional superconductivity.

High resolution inelastic neutron scattering (INS) data for spin fluctuations exists [4, 5], but the data is available only up to 60 meV. State-of-the-art recent resonant inelastic X-ray scattering (RIXS) picks up the signatures of bosonic fluctuations of different kinds [6], including those whose mechanisms are intertwined. Thus to de-

cipher what constitutes the primary component of the observed RIXS spectra is a challenge for both theorists and experimentalists alike. RIXS data has been taken from the Cu- L_3 edge for $\text{YBa}_2\text{Cu}_3\text{O}_7$ along the $(0,\pi)$ line [6] with excitations observed up to 300 meV. However, measurements could not be performed for the important momentum region around (π, π) , which likely drives superconductivity.

In the present letter, we calculate the magnetic susceptibilities and superconducting instability for optimally doped YBCO using a new high fidelity, *ab initio* approach [7, 8]. For the one-particle Green's function it combines the quasiparticle self consistent GW (QSGW) approximation [9] with CTQMC solver [10] based dynamical mean field theory (DMFT). This framework [11, 12] is extended by computing the local vertex from the two-particle Green's function by DMFT [13, 14], which is combined with nonlocal bubble diagrams to construct a Bethe-Salpeter equation [15, 16]. The latter is solved to yield the essential two-particle spin and charge susceptibilities χ^d and χ^m — physical observables which provide an important benchmark. Moreover they supply ingredients needed for the Eliashberg equation, which yields eigenvalues and eigenfunctions that describe instabilities to superconductivity. We will denote QSGW⁺⁺ as a shorthand for the four-tier QSGW+DMFT+BSE+Eliashberg theory. The numerical implementation is discussed in Pashov et al. [8] and codes are available on the open source electron structure suite Questaal [17]. Some details are also given in the supplemental material.

QSGW⁺⁺ has high fidelity because QSGW captures non-local dynamic correlation particularly well in the charge channel [8, 18], but cannot adequately capture effects of spin fluctuations. DMFT does an excellent job at the latter, which are strong but mostly controlled by a

local effective interaction given by U and J . In this letter, we have used $U = 8\text{eV}$ and $J = 0.7\text{eV}$ which is similar to what is generally used for cuprates [19]. That it can well describe superconductivity has now been established in several materials [15, 16].

In YBCO, we explore the full potential of our ability by performing rigorous bench-marking of our computed magnetic susceptibilities (resolved in momentum and energy) against the low and high energy spectral data from INS and RIXS respectively. We show that we reproduce all intricate structures in momentum and energy spaces observed in INS and RIXS from our theory in a parameter free manner. That it is possible to reproduce the RIXS spectra from the spin susceptibility alone indicates that RIXS is measuring an excitation primarily magnetic in nature in this compound.

The superconducting glue the present theory can characterize originates from some combination of spin and charge susceptibility [13, 15]. That we are able to reliably recover experimental neutron and RIXS data provides some confirmation that we have an adequate foundation to describe superconductivity of this kind. We can use the same method to probe how spin fluctuations and superconductivity are affected when the system is perturbed, in particular how T_c evolves with tensile strain. In a prior work[15], this machinery was used to explain in detail how T_c evolves with tensile strain in Sr_2RuO_4 where it could be benchmarked against experiments.

Fig. 1 benchmarks the dynamical structure factor $S(q, \omega) = (1 - e^{-\beta\omega})^{-1} \text{Im} \chi^{\text{mag}}(q, \omega)$ computed by QSGW^{++} (Fig. 1b) against direct INS measurements of S (Fig. 1a) in the vicinity of the antiferromagnetic point $Q_{AF} = (\pi, \pi)$; and also against RIXS which measures any bosonic excitation, including the magnetic structure factor (Fig. 1c). For the former, we can compare only up to maximum value reported, 60 meV. At least in this range of energy and momentum QSGW^{++} is in good qualitative and quantitative agreement. Since the RIXS measurement [21] did not sample the region close to Q_{AF} , we compare to RIXS along the line Q connecting Γ and $(\pi, 0)$. A second branch appears at high energy. For both low-energy and high-energy excitations the maxima of the peaks along Q line shown by the dashed line are in a very good agreement with the experimental data (blue dots). It confirms the magnetic nature of the excitation measured in this RIXS response, a subject of some controversy [22–24].

Next we use the Eliashberg theory derived from spin and charge susceptibilities to estimate T_c , and investigate how it is affected by strain. We use QSGW^{++} to simulate an uniaxial strain on the c -direction and study its effect on the superconducting order. The uniaxial strain is carried out by reducing the c -axis up to 8% with a concomitant expansion the plane. The volume change $\frac{\Delta V}{V}$ and the reduction of c are related by $\frac{\Delta V}{V} = (1 - 2\nu)\frac{\Delta c}{c}$ where the Poisson ratio used is $\nu = 0.25$, which is close

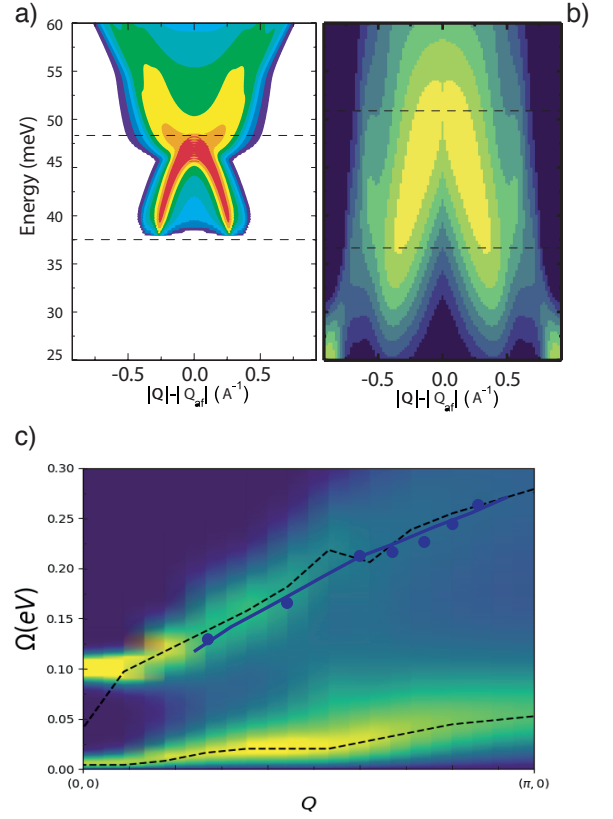


FIG. 1. (a) Structure factor obtained by Inelastic Neutron Scattering along the nodal line, reproduced from [4]. (b) Structure factor for the same energy window obtained by QSGW^{++} . (c): Heat map representing dynamical structure factor along the path Γ -X in the Brillouin Zone computed with QSGW^{++} . Dashed line indicates the position of the maxima. Circle dot :RIXS experimental data shown for comparison, reproduced from [20].

to experiment [25]. For each strain, we consider two scenarios : an ideal strain where all internal displacements are fixed to their projection along the c axis, and another case where atoms are relaxed to the zero-force condition. Forces are computed within density functional theory. We will denote these scenarios as ‘SS’ (for single shot) and ‘SO’ (for structure optimized). SO corresponds to the actual mechanical response of YBCO subject to an ϵ_{33} tensile strain. For convenience of presentation we define a strain with the opposite sign of the usual definition: $\epsilon_z = -\epsilon_{33} = \frac{c_0 - c}{c_0}$.

For both scenarios, we compute the variation of the critical temperature T_c by comparing the superconducting instability computed by solving the linearized Eliashberg equation (see Appendix). Comparing these two scenarios distinguishes two competing effects: on the one hand, the ideal strain changes the topology of the Fermi surface in a way that favors superconducting order. On the other hand, subsequently allowing the internal coordinates to fully relax empties the d_{z^2} orbital, which is

ϵ_z [%]	Cu ¹ -Cu ² [Å]		Cu-AO [Å]		Ba-AO [Å]	
	SO	SS	SO	SS	SO	SS
0.0	3.39	3.39	2.30	2.30	0.30	0.30
2.0	3.38	3.33	2.21	2.26	0.26	0.29
4.0	3.32	3.28	2.15	2.22	0.22	0.29

TABLE I. Interlayer Cu-Cu spacing (first column), copper to apical oxygen distance (second column), and vertical component of AO to Ba distance (third column). We report distances for the pristine material (first row), and under uniaxial strain (ϵ_z). Parameters for ideal (SS) and fully relaxed (SO) structures are shown (see text).

unfavorable for the superconducting order.

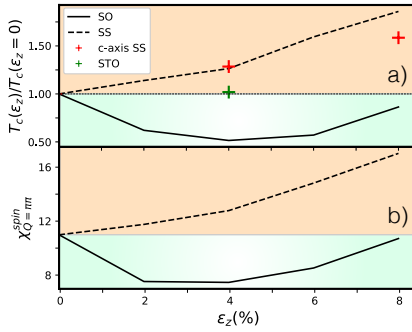


FIG. 2. (Top panel) T_c relative to the unstrained condition for the SS case (dashed line) and SO case (solid line). T_c increases in the former case, but decreases in the latter (see text). The red crosses show the SS case assuming a Poisson ratio of 0 (basal plane is kept frozen). This shows that the dominant effect is the change in coupling along the c axis. The green cross corresponds to a compression of 2% in the basal plane in SS case. Panel (b) shows the corresponding evolution of the static magnetic susceptibility at $Q = (\pi, \pi)$. It close correspondence with T_c indicates that spin fluctuations are the dominant contributor to the superconducting instability. Panel (c): evolution of the Stoner factor $s = 1/(1 - \alpha)$ where α is the highest eigenvalue of $\chi_0\Gamma$ in $\chi = \chi_0/(1 - \chi_0\Gamma)$.

Fig. 2 shows these results in more detail. T_c is greatly increased in the SS scenario, and the middle and bottom panels show how T_c is correlative to magnetic susceptibility at (π, π) . Both the spin fluctuation and the superconductivity instability show a similar trend which confirms that the spin fluctuation are an essential contributor of the superconductivity. We can also separate the contributions from stretching the c axis from the contributions by reductions in the basal plane by varying the Poisson ratio. In one case we used $\nu=0$ which freezes the lattice vector in the basal plane (red symbols in Fig. 2). T_c changes only marginally with ν , which indicate that the main effect are coming from the reduction of the c -axis. In another scenario, we expand a and b axes about 2% to match YBa₂Cu₃O₇ epitaxially on an STO substrate

(green symbol).

On the $(0, 0) - (\pi, \pi)$ line, the Cu $d_{x^2-y^2}$ in the two planes of YBa₂Cu₃O_{7- δ} couple weakly through the apical O, splitting these otherwise degenerate states into a bond-antibond pair. The Fermi surface connected with these orbitals splits into two sheets.

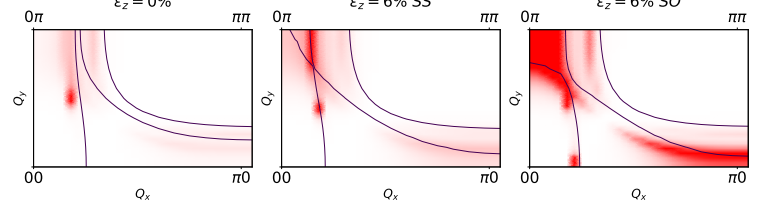


FIG. 3. Evolution of the Fermi surface against applied strain, as computed from QSGW⁺⁺. The Fermi surface (full lines) is composed of a vertical line originating from the CuO chain, and other lines from the bilayer coupling as explained in the text. Strain increases the separation between antibonding and bonding Fermi sheets for both (a) ideal strain (SS) and (b) fully relaxed strain (SO). SS and SO differ in the degree that $d_{x^2-y^2}$ and d_{z^2} are coupled, as discussed in the text. This is manifest by the colorbar, which shows the off-diagonal component $|G(Q, \omega=0)_{z^2, x^2-y^2}|$, an indicator of the hybridization between d_{z^2} and $d_{x^2-y^2}$ on the Fermi surface.

As shown in Fig. 3, the Fermi surface is formed of three bands. The two curved lines correspond to the bonding and antibonding $d_{x^2-y^2}$ bands noted above. The interlayer hybridization is strongest at the two antinodal points where either $Q_x=\pi$ or $Q_y=\pi$. As strain is applied, the interlayer distance decreases (Table I). This increases the interlayer hybridization, which further splits the bonding and antibonding $d_{x^2-y^2}$ Fermi surfaces. The antibonding surface becomes flatter and thus more square. Making the arc more square improves the nesting of momentum transfer $Q=(\pi, \pi)$ for electrons living at the antinodal point. In d-wave superconductivity, it increases the attractive interaction [26, 27], i.e works constructively for d -wave superconductivity. As shown in Fig. 4, not only the magnetic susceptibility increases but the nesting vector moves closer to Q_{AF} where the magnetic susceptibility is maximum. These two effects cumulatively explain the large enhancement of T_c observed in the SS scenario.

In SO scenario, we observe a similar splitting of the bonding and antibonding $d_{x^2-y^2}$ Fermi surfaces, but with an important difference. In SO case, the $d_{x^2-y^2}$ hybridize with d_{z^2} . This hybridization increases because relaxation reduces the Ba-AO vertical distance, e.g. by 25% when the c axis is reduced by 4% (Table I). As a consequence, the AO environment is changed which affects the Cu d_{z^2} orbital. In the unstrained case, d_{z^2} sits at -1.48 eV below the Fermi level, and it is marginally changed in the SS case (for $\epsilon_z=4\%$ it resides at -1.41 eV) while in the SO case, d_{z^2} is pushed closer to the Fermi

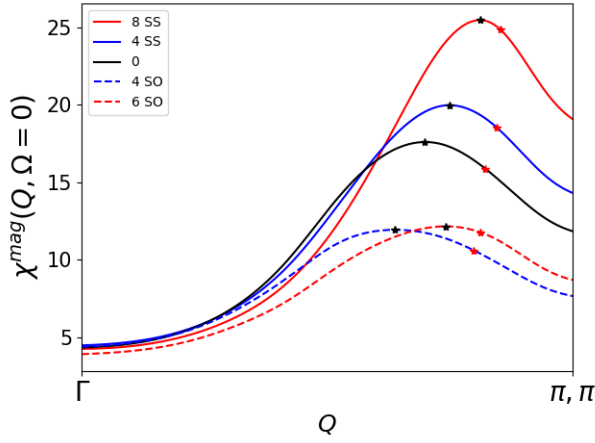


FIG. 4. Static magnetic susceptibility $\chi(q, \omega=0)$ on the $(0,0)-(\pi, \pi)$ line, for pristine YBCO (black line), and YBCO subject to SS and SO kinds of strain. SS and SO induce opposite effects on both the peak position and amplitude $\chi(q, \omega=0)$. In the SS case T_c increase both because $\chi(q, \omega=0)$ increases and the peak shifts closer to Q_{AF} .

level (-1.18 eV). The Fermi surface mainly composed of $d_{x^2-y^2}$ state becomes strongly hybridized with d_{z^2} . This is apparent from colorbar in Fig. 3. The two orbitals hybridize close the antinodal point which is known to be unfavorable to d -wave superconductivity [28–30]. Indeed, not only almost filled band, as d_{z^2} is unfavorable for T_c but opposite spin nearest-neighbor coupling to form Cooper pair is less favorable when two orbitals are active at the Fermi level. When d_{z^2} orbital are not included in correlated subspace and in the Eliashberg equation, this destructive effect disappears.

To recap, in the idealized scenario compression of the planes increases the interlayer hybridization which enhances Fermi surface nesting and, hence, T_c . However, with a proper relaxation (SO), the Ba-AO bond length decreases dramatically to force out-of-plane contributions to the planar physics and changes the orbital components of the Fermi surface. This contribution works destructively for T_c .

To conclude, we have first shown that QSGW++ is able to predict magnetic fluctuations in $\text{YBa}_2\text{Cu}_3\text{O}_7$ with high fidelity. Using this technique to explore the parameter space when $\text{YBa}_2\text{Cu}_3\text{O}_7$ is subject to strain, we find that T_c can be dramatically altered, but how it is altered depends on the details of the displacements. Subject to an ideal strain with no internal relaxation, T_c is increased owing to enhanced interlayer hybridization, which changes the shape of the Fermi surface and makes nesting more favorable. However, in a more realistic scenario, the Fermi surface also suffers from a competing d_{z^2} hybridization which is detrimental to T_c .

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SUPPLEMENTARY MATERIAL

The calculation in the unstrained case was performed using crystal structure reported in [31]. Paramagnetic DMFT is combined with non magnetic QSGW via local projection on Cu $3d$ on the Cu augmentation spheres to form the correlated subspace. DMFT loop was performed using CTQMC impurity solver [10] and [32]. The two particle susceptibility needed in BSE for the magnetic susceptibility was computed using Exact Diagonalisation impurity solver with 6 bath sites on a mesh of 50 bosonic frequencies and 500 fermionic frequencies. A benchmark was done with CTQMC solver to check the accuracy of the hybridization fit. The Eliashberg equation was solved using an impurity susceptibility computed with CTQMC impurity solver[33].

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- [1] T. Dahm, V. Hinkov, S. V. Borisenko, A. A. Kordyuk, V. B. Zabolotnyy, J. Fink, B. Büchner, D. J. Scalapino, W. Hanke, and B. Keimer, *Nature Physics* **5**, 217–221 (2009).
- [2] D. J. Scalapino, *Rev. Mod. Phys.* **84**, 1383 (2012).
- [3] W. Hu, S. Kaiser, D. Nicoletti, C. R. Hunt, I. Gierz, M. C. Hoffmann, M. Le Tacon, T. Loew, B. Keimer, and A. Cavalleri, *Nature Materials* **13**, 705–711 (2014).
- [4] D. Reznik, J.-P. Ismer, I. Eremin, L. Pintschovius, T. Wolf, M. Arai, Y. Endoh, T. Masui, and S. Tajima, *Physical Review B* **78** (2008), 10.1103/physrevb.78.132503.
- [5] H. Woo, P. Dai, S. M. Hayden, H. A. Mook, T. Dahm, D. J. Scalapino, T. Perring, and F. Doğan, *Nature Physics* **2**, 600–604 (2006).

- [6] L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, *Rev. Mod. Phys.* **83**, 705 (2011).
- [7] L. Sponza and et al, *Phys. Rev. B* **95**, 041112 (2017).
- [8] D. Pashov, S. Acharya, R. L. W. Lambrecht, J. Jackson, K. Belashchenko, D. A. Chantis, F. Jamet, and M. van Schilfgaarde, *Computer Physics Communications* (2019).
- [9] T. Kotani, M. van Schilfgaarde, and S. V. Faleev, *Phys. Rev. B* **76**, 165106 (2007).
- [10] K. Haule, *Physical Review B* **75**, 155113 (2007).
- [11] S. Acharya, C. Weber, E. Plekhanov, D. Pashov, A. Taraphder, and M. van Schilfgaarde, *Phys. Rev. X* **8**, 021038 (2018).
- [12] E. Baldini and et. al, *Proceedings of the National Academy of Sciences* **117**, 6409 (2020).
- [13] H. Park, *The study of two-particle response functions in strongly correlated electron systems within the dynamical mean field theory*, Ph.D. thesis, Rutgers University-Graduate School-New Brunswick (2011).
- [14] Z. Yin, K. Haule, and G. Kotliar, *Nature Physics* **10**, 845 (2014).
- [15] S. Acharya, D. Pashov, C. Weber, H. Park, L. Sponza, and M. van Schilfgaarde, *Communications Physics* **2** (2019), 10.1038/s42005-019-0254-1.
- [16] S. Acharya, D. Pashov, F. Jamet, and M. van Schilfgaarde, *Phys. Rev. Lett.* **124**, 237001 (2020).
- [17] “Questaal website,” <https://www.questaal.org>.
- [18] J. Tomczak, P. Liu, A. Toschi, G. Kresse, and K. Held, *The European Physical Journal Special Topics* **226**, 2565 (2017).
- [19] S. Choi, A. Kutepov, K. Haule, M. van Schilfgaarde, and G. Kotliar, *npj Quantum Materials* **1**, 16001 (2016).
- [20] M. Le Tacon, M. Minola, D. C. Peets, M. Moretti Sala, S. Blanco-Canosa, V. Hinkov, R. Liang, D. A. Bonn, W. N. Hardy, C. T. Lin, and et al., *Physical Review B* **88**, 020501(R) (2013).
- [21] M. Le Tacon, G. Ghiringhelli, J. Chaloupka, M. M. Sala, V. Hinkov, M. W. Haverkort, M. Minola, M. Bakr, K. J. Zhou, S. Blanco-Canosa, and et al., *Nature Physics* **7**, 725–730 (2011).
- [22] D. Benjamin, I. Klich, and E. Demler, *Physical Review Letters* **112** (2014), 10.1103/physrevlett.112.247002.
- [23] M. Minola, G. Dellea, H. Gretarsson, Y. Peng, Y. Lu, J. Porras, T. Loew, F. Yakhov, N. Brookes, Y. Huang, and et al., *Physical Review Letters* **114** (2015), 10.1103/physrevlett.114.217003.
- [24] M. Kanász-Nagy, Y. Shi, I. Klich, and E. A. Demler, *Physical Review B* **94** (2016), 10.1103/physrevb.94.165127.
- [25] C. Chen, L. Liu, Y. Wen, Y. Jiang, and L. Chen, *Crystals* **9**, 497 (2019).
- [26] N. Bickers, D. Scalapino, and R. Scalettar, *International Journal of Modern Physics B* **01**, 687–695 (1987).
- [27] C. Weber, C. Yee, K. Haule, and G. Kotliar, *EPL (Europhysics Letters)* **100**, 37001 (2012).
- [28] H. Sakakibara, H. Usui, K. Kuroki, R. Arita, and H. Aoki, *Phys. Rev. B* **85**, 064501 (2012).
- [29] C. E. Matt, D. Sutter, A. M. Cook, Y. Sassa, M. Månsson, O. Tjernberg, L. Das, M. Horio, D. De-straz, C. G. Fatuzzo, and et al., *Nature Communications* **9** (2018), 10.1038/s41467-018-03266-0.
- [30] H. Sakakibara, H. Usui, K. Kuroki, R. Arita, and H. Aoki, *Phys. Rev. Lett.* **105**, 057003 (2010).
- [31] J. D. Jorgensen, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus, and W. K. Kwok, *Phys. Rev. B* **41**, 1863 (1990).
- [32] P. Seth, I. Krivenko, M. Ferrero, and O. Parcollet, *Computer Physics Communications* **200**, 274 (2016).
- [33] S. Acharya, D. Pashov, C. Weber, H. Park, L. Sponza, and M. V. Schilfgaarde, *Communications Physics* **2** (2019), 10.1038/s42005-019-0254-1.