Comment on "First-principles calculation of the superconducting transition in MgB_2 within the anisotropic Eliashberg formalism"

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Choi *et al.* [Phys. Rev. B **66**, 020513 (2002)] recently presented first-principles calculations of the electron-phonon coupling and superconductivity in MgB_2, emphasizing the importance of anisotropy and anharmonicity. We point out that (1) variation of the superconducting gap inside the σ or the π bands can hardly be observed in real samples, and (2) taking the anisotropy of the Coulomb repulsion into account influences the size of the small gap, Δ_π.

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In a recent paper,[1] as well as in a follow-up paper,[2] Choi *et al.* presented an *ab initio* calculation of the superconducting transition and superconducting properties of MgB_2. The important improvement over existing calculations was that they allowed the order parameter to vary freely over the Fermi surface, i.e., $\Delta=\Delta(\mathbf{k})$, and at the same time took the anharmonicity into account. As a consequence, they had to compute the fully anisotropic electron-phonon interaction, $\lambda(\mathbf{k},\mathbf{k}')$ and solve the corresponding Eliashberg equation. The Coulomb pseudopotential $\mu^*(\mathbf{k},\mathbf{k}')$ was assumed not to depend on $\mathbf{k}$ and $\mathbf{k}'$, and was treated as an adjustable parameter. First-generation *ab initio* calculations of the superconducting transition and superconducting properties of MgB_2 had assumed Δ to be constant and had therefore solved merely the isotropic Eliashberg equation.[3] Moreover, anharmonicity had been neglected. It was soon pointed out[4] that the calculated electron-phonon coupling suggests that the gap on the two π sheets of the Fermi surface is smaller than that on the two σ sheets, and that anharmonicity is important. This led to the so-called two-band model. *Ab initio* calculations of the second generation[4–6] allowed for two, and sometimes four gaps, Δ_n, and thus had to compute $\lambda_{nn'}$ to estimate the anisotropy of $\mu^*_{nn'}$, and to solve the corresponding Eliashberg equations.

Here we shall comment on (1) whether consequences of anisotropy beyond that of the two-band model may be observed and (2) whether at this level of detail the assumption of a uniform Coulomb repulsion made by Choi *et al.* is warranted.

(1) Reference 1 implies that there is a distribution of gaps *within* the σ and the π sheets, not only in the calculations for perfectly clean MgB_2, but also in the actual material; in other words, that the distribution of gaps shown in Fig. 2 of Ref. 2 is *observable.* However, in the theory of anisotropic superconductivity it is known that any intraband nonuniformity of the order parameter is suppressed by strong intraband impurity scattering. It is not immediately obvious, though, when scattering should be considered strong in this connection. Since excitation gaps are not equal to the order parameters anymore, one needs to compare individual densities of states (DOS), $N(E)$, for the two σ bands (or the two π bands) with each other for a given scattering strength, and check whether $|N_{\sigma 1}-N_{\sigma 2}|\ll|N_{\sigma 1}+N_{\sigma 2}|$. The relevant expression can be found in Ref. 7. In the limit of large scattering rates, γ, one can derive an analytical expression for this criterion, namely, $\gamma>\sqrt{\langle\Delta\rangle\delta\Delta}$ (Ref. 8), where $\langle\Delta\rangle$ is the average *order parameter*, and $\delta\Delta$ is the variation of the *order parameter* over the Fermi surface in question. With the data from Refs. 1 and 2 for $\langle\Delta\rangle$ and $\delta\Delta$, this gives characteristic scattering rates of, respectively, 2 and 1.5 meV for the σ and π bands. Therefore, to observe four distinct gaps in MgB_2 one needs samples with scattering rates smaller than 2 meV, that is, with mean free paths beyond 1500 Å. To observe gap variations beyond the four-band model, far cleaner samples are needed. This is the reason why at most two distinct gaps have been observed in experiments. It is even surprising that the difference of 5 meV between the gaps of the σ and the π bands is not smeared out. This seems to be due to the inability of common impurities to couple between the disparate σ and π band wave functions,[9] so that $\gamma_{\sigma\pi}\ll\gamma_{\sigma\sigma}\sim\gamma_{\pi\pi}$.

(2) For the Coulomb pseudopotential, Choi *et al.* used $\mu^*(\mathbf{k},\mathbf{k}')=\mu^*(\omega_c)=0.12$ (with the cutoff frequency $\omega_c\approx 5\omega_{ph}^{\max}$) and stated that the superconducting properties of MgB_2 were not very sensitive to the choice of $\mu^*(\omega_c)$. This at first seems plausible, because the Coulomb pseudopotential enters the Eliashberg equation only in the combination $\lambda(\mathbf{k},\mathbf{k}',\nu-\nu')-\mu^*(\mathbf{k},\mathbf{k}')$, and the λ distribution varies on the scale of ~ 1.8, ~ 0.3, and ~ 0.2 for $\sigma\sigma$, $\pi\pi$, and $\sigma\pi$ scattering, respectively [see Fig. 3 of Ref. 1]. Therefore, at most the $\sigma\pi$ scattering can be influenced by anisotropy of μ^*. We shall argue that the $\sigma\pi$ interband Coulomb matrix elements *are* considerably smaller than the intraband matrix elements due to the very small overlap of the σ- and π-band charge densities[5] and that this is sufficient to influence the superconducting properties, in particular the size of the small gap, Δ_π.

Choi *et al.* do not give the band-integrated values of their coupling constants, but by integrating Fig. 3 of Ref. 1 with the DOS ratio $N_\pi/N_\sigma=1.37$ according to

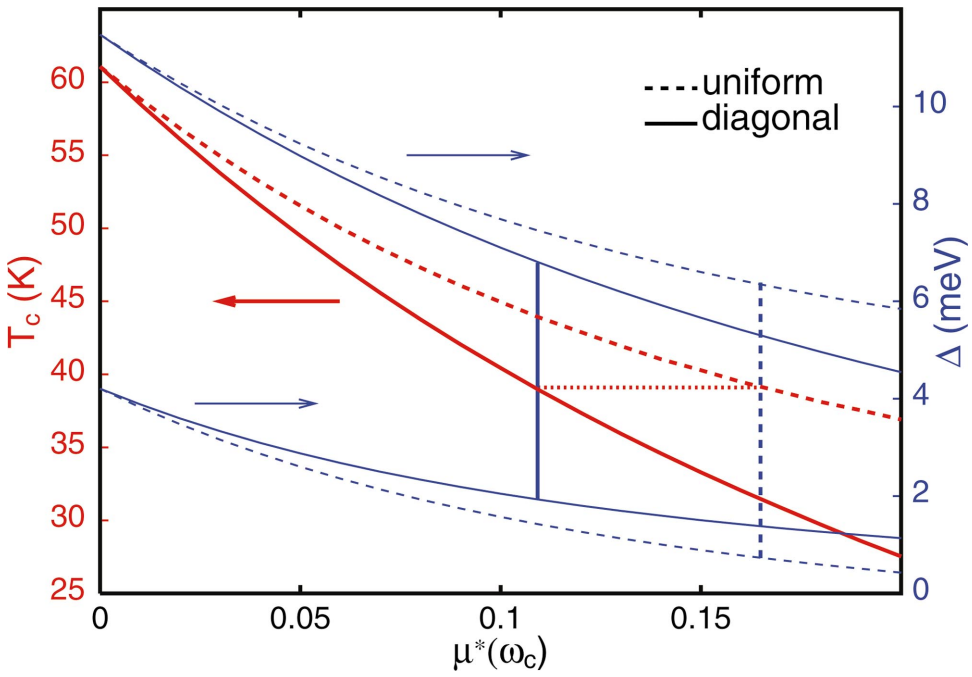

FIG. 1. (Color) Critical temperature and the values of the σ and π gaps at 1 K as functions of the renormalized Coulomb pseudopotential, $\mu^*(\omega_c)$, in two models: the uniform model where all matrix elements of the Coulomb repulsion are equal and the diagonal model where the interband matrix elements are zero. In both cases the normalization is chosen so as to produce given values of $\mu^*(\omega_c)$ after proper summation over all bands. The two gaps obtained from $\mu^*(\omega_c)$'s giving T_c's of 39 K in the two models are connected vertically.

$$\lambda_{nn'}(0) \equiv \frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'} \frac{\delta(\varepsilon_{n\mathbf{k}})}{N_n}\lambda(\mathbf{k},\mathbf{k}',0)\delta(\varepsilon_{n'\mathbf{k}'})$$

$$= \frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'} W_{n\mathbf{k}}\lambda(\mathbf{k},\mathbf{k}',0)W_{n'\mathbf{k}'}N_{n'} \qquad (1)$$

for the phonon-mediated coupling of an electron in band n to all electrons in band n', we can map the fully anisotropic model of Choi *et al.* onto a two-gap model with $\lambda_{\sigma\sigma}=0.78$, $\lambda_{\sigma\pi}=0.15$, $\lambda_{\pi\sigma}=0.11$, and $\lambda_{\pi\pi}=0.21$. These λ values yield the mass-renormalization parameters in Fig. 2 of Ref. 1: $m^*/m-1=\lambda_\sigma=\lambda_{\sigma\pi}+\lambda_{\sigma\sigma}\approx 0.94$ and $\lambda_\pi=\lambda_{\pi\sigma}+\lambda_{\pi\pi}\approx 0.32$. The total isotropic (thermodynamic) $\lambda=(N_\sigma\lambda_\sigma+N_\pi\lambda_\pi)/N=0.61$, which of course is the same as the one given by Choi *et al.* Here, and in Eq. (1), N is the DOS summed over all bands. With this two-gap model we have performed strong-coupling Eliashberg calculations in order to compare the results for T_c and the gaps with those resulting from the fully anisotropic treatment. For all four spectral functions we used the isotropic $\alpha^2F(\omega)$ from Fig. 1 of Ref. 1 scaled to produce the λ matrix given above. The $\mu^*(\omega_c)$ matrix is obtained from Eq. (1) with $\lambda(\mathbf{k},\mathbf{k}',0)$ substituted by $\mu^*(\omega_c)$ of Choi *et al.* The resulting T_c and the gaps are shown by dashed lines in Fig. 1 as functions of $\mu^*(\omega_c)$. At $\mu^*(\omega_c)=0.12$, as used by Choi *et al.*, we get $T_c=43$ K, $\Delta_\sigma=7.2$ meV, and $\Delta_\pi=1.3$ meV. The corresponding values quoted by Choi *et al.* are 39 K, 6.8 meV, and 1.8 meV. These differences are hardly due to intraband anisotropy, first of all because it can only increase T_c. Second, increasing the number of gaps from two to four in the Eliashberg equations, which should account for most of the anisotropy beyond the two-gap model, we found rather small changes.[10]

If, on the other extreme, we assume that there is no Coulomb repulsion between the σ and π electrons, then the corresponding two-gap treatment gives the full lines in Fig. 1 and, hence, $T_c=38$ K, $\Delta_\sigma=6.5$ meV, and $\Delta_\pi=1.8$ meV for $\mu^*(\omega_c)=0.12$, incidentally, rather close to the values quoted in Refs. 1 and 2. If the magnitude of μ^* in both calculations

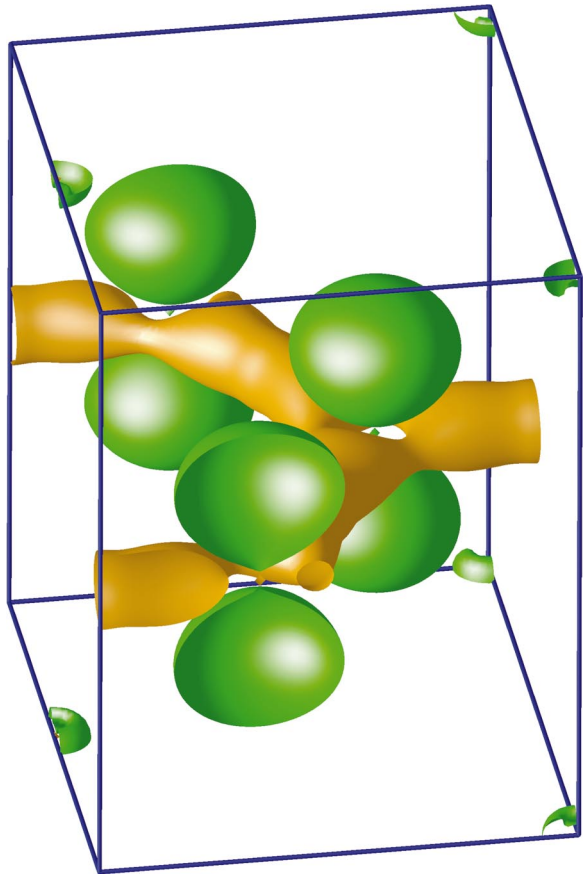

FIG. 2. (Color) Constant-density contour for the normalized σ (orange) and π (green) electron densities, $|\psi(\mathbf{r})|^2_\sigma$ and $|\psi(\mathbf{r})|^2_\pi$, at the Fermi level.

shown in Fig. 1 is adjusted to produce the same T_c of 39 K, the value of the lower gap changes from ≈ 2 meV (diagonal) to ≈ 0.4 meV (uniform).

That uniform and diagonal Coulomb pseudopotentials yield different results is not surprising. The same total Eliashberg μ^* in the uniform case is distributed over intra- and interband terms so that the $\sigma\sigma$ part of the pairing interaction suffers less than in the case of a diagonal μ^*. $\lambda_{\sigma\sigma}$ is more important for the critical temperature, and $\lambda_{\sigma\pi}$ for generating Δ_π. For uniform μ^*, therefore, the T_c and Δ_σ are larger, and Δ_π is much smaller.

Having demonstrated that the assumed structure of μ^* matters for the details of the superconducting properties of MgB_2,[11] the size of Δ_π in particular, let us finally estimate this structure from first principles. The unrenormalized μ is the matrix element $\langle n\mathbf{k}\uparrow, n-\mathbf{k}\downarrow|V_C|n'\mathbf{k}'\uparrow, n'-\mathbf{k}'\downarrow\rangle$ for scattering a Cooper pair from state $|n'\mathbf{k}'\rangle$ to state $|n\mathbf{k}\rangle$ via a phonon with wave vector $\mathbf{k}-\mathbf{k}'$. Inserting this matrix element in Eq. (1) instead of $\lambda(\mathbf{k},\mathbf{k}',0)$ yields $\mu_{nn'}$. Here $V_C(\mathbf{r},\mathbf{r}')$ is the screened Coulomb interaction between the electrons, and since it has short range in good metals, it makes sense to take it proportional to the delta function $\delta(\mathbf{r}-\mathbf{r}')$. This leads to the following estimate:

$$\mu \propto N \int |\psi_{n\mathbf{k}}(\mathbf{r})|^2 |\psi_{n'\mathbf{k}'}(\mathbf{r})|^2 d^3r,$$

$$\mu_{nn'} \propto N_{n'} \int |\psi(\mathbf{r})|_n^2 |\psi(\mathbf{r})|_{n'}^2 d^3r, \qquad (2)$$

where $|\psi(\mathbf{r})|_n^2 \equiv \Sigma_{\mathbf{k}} |\psi_{n\mathbf{k}}(\mathbf{r})|^2 \delta(\varepsilon_{n\mathbf{k}})/N_n$ is the shape, normalized to 1 in the cell or the crystal, of the electron density of band n at the Fermi level. These σ and π densities are shown in Fig. 2, and they yield for the ratios of the integrals in Eq. (2)

$$\langle|\psi|_\sigma^4\rangle : \langle|\psi|_\pi^4\rangle : \langle|\psi|_\sigma^2|\psi|_\pi^2\rangle \sim 3.0:1.8:1. \qquad (3)$$

These ratios reflect the facts that the σ density is more compact than the π density, and that the overlap of these two densities is small. Note that the exceptional smallness of the interband impurity scattering[9] in MgB_2 is due not only to this difference in charge density, but also to a disparity of the σ and π *wave functions*.

From Eqs. (2) and (3) we get $\mu_{\sigma\sigma}:\mu_{\pi\pi}:\mu_{\sigma\pi}:\mu_{\pi\sigma} = 3.1:2.6:1.4:1$. Now, any anisotropy in the *bare* pseudopotential is further *enhanced* in the renormalized μ^*. In the one-band case μ is renormalized as $\mu^*(\omega_c) = \mu/[1 + \mu \ln(W/\omega_c)]$, where W is a characteristic electronic energy of the order of the bandwidth or plasma frequency. For the multiband case, this is a matrix equation with W being a diagonal matrix with elements W_n. Assuming for simplicity that $\mu_{\sigma\sigma} = \mu_{\pi\pi} = A\mu_{\sigma\pi}$ with $A > 1$, and that $\mu_{\sigma\sigma}\log(W_\sigma/\omega_c) = \mu_{\pi\pi}\log(W_\pi/\omega_c) = L$, one obtains $A^* = A + (A - A^{-1})L$. For MgB_2, $L \sim 0.5$–1 and $A \sim 2.3$, so that $A^* \sim 3$–4, which is very different from the uniform μ.

In conclusion, any difference between the results of the fully anisotropic Eliashberg formalism and those of the two-gap formalism will hardly be observable in real MgB_2 samples. On the other hand, the anisotropy of the Coulomb pseudopotential is likely to have an observable effect on the size of the small gap, Δ_π.

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[10] This is not surprising, given that the variation of the calculated order parameter (Ref. 2), as well as of the mass renormalization (Ref. 1), within each band is at most 10%.

[11] The problem of proper structure of the μ^* matrix is specific for multiband superconductivity with large gap disparity. When the pairing interaction is fully uniform, the μ^* matrix can have any structure as long as it is normalized to the total μ^*. This has no effect on either T_c or gap ratios.