Comment on “Probing two- and three-dimensional electrons in MgB$_2$ with soft x-ray angle-resolved photoemission”

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A recent article by Sassa et al. [Phys. Rev. B 91, 045114 (2015)] reports on a soft-x-ray angle-resolved photoemission study of MgB$_2$. The analysis and/or presentation of the collected data and the corresponding calculations appear to be partially inconsistent. The aim of this comment is to provide a guide to these inconsistencies and to discuss their influence on the presented conclusions.

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INTRODUCTION

In a recent article Sassa et al. presented a soft-x-ray angle-resolved photoemission (SX-ARPES) study of MgB$_2$ [1], claiming to have established “the full 3D electronic structure” of the compound. The experimental data and the results of band-structure calculations are compared and certain deviations, e.g., for the band width and Fermi-surface area are reported. However, the illustrations used to circumstantiate the article’s quantitative claims seem to be mutually inconsistent. In the following, firstly these inconsistencies will be described in detail (but not necessarily in any hierarchical order). Secondly, their influence on the conclusions of Ref. 1 is discussed.

OBSERVATIONS

The main figure (Fig. 5 [2]) involved in the band-structure analysis contains a set of particular oddities. All the calculated bands depicted in Figs. 5(a) and 5(c) ought to be symmetric about $M$ and $L$—evidently, some are not: in Fig. 5(a) the $\pi$ band from $\Gamma$ to $M$ is farther away from $M$ than the $\pi$ band beyond $M$, leading to differences in the crossing points with the $\sigma$ bands (1.67 eV vs. 1.58 eV and 2.47 eV vs. 2.37 eV). In Fig. 5(c) the second $\pi$ band and the third $\sigma$ band (always counting at $L$ from low to high binding energy $E_B$) are shifted significantly toward $A$. The top of both bands is found at about $-1.13 \pi/a$ instead of $-2/\sqrt{3} \pi/a$ [3]. Hence, also the crossings of the second $\pi$ and second $\sigma$ bands are not at a constant $E_B$ (5.01 eV vs. 4.86 eV). Apparently, this is not an effect of inaccuracies of the calculations, otherwise, the second band shown in the zoom-in Fig. 5(g) would not be centered at $L$, either.

Staying with Fig. 5 and reviewing the energy-distribution-curve (EDC) analysis reveals several inconsistencies between the presentations in the various subfigures. According to the text in Ref. 1, the EDCs are analyzed using “resolution broadened Lorentzian functions”. Following the same route, here, EDCs around $M$ obtained from the grayscale map in Fig. 5(a) and the (assumed equally spaced) EDCs in Fig. 5(f) are modeled with a pair of Voigtians [Gaussian-broadened ($\sigma = 0.1$ eV) Lorentzians] residing on a polynomial background [either parametrized from Fig. 5(f) or fitted]. Two fits are performed for each set of data: using a $E_B$ fitting range between 5.5 eV and 10.5 eV firstly the curves are modeled with two Lorentzian widths that are shared by all EDCs and secondly the fit is repeated with two independent Lorentzian widths for each EDC. Both procedures yield fits that are comparable to those shown in Fig. 5(f). In the first case the obtained Lorentzian half-widths at half maximum (HWHM) are 0.6 eV and 0.9 eV, respectively. In the second case, the HWHM range from 0.1 eV (with small corresponding amplitudes) to 0.6 eV and from 1.1 eV to 1.2 eV, respectively. The obtained EDC peak positions are shown in Fig. 1 and compared to the peak positions marked in Fig. 5(h). If the presentation was consistent throughout Fig. 5, the curves sampled by the peak positions shown in the three upper panels of Fig. 1 would coincide. What is observed instead is that the presentation in Fig. 5(f) is shifted in energy by about 0.85 eV with respect to the data in Figs. 5(a) as well as 5(h) and that it features erroneous labeling; the top-most EDC in Fig. 5(f) appears to be rather at $k_x \approx -1.00 \pi/a$ or $k_x \approx -1.30 \pi/a$ [if the same decreasing $k_x$—not $k_y$—as in Fig. 5(e) is used for higher up EDCs]. In addition, while the band gap for the analysis of the data presented in Figs. 5(a) and 5(f) is consistently sized about 0.9 eV to 1.0 eV, the purported peak positions shown in Fig. 5(h) yield a gap of 0.4 eV only. Moreover, both gap labels in Fig. 5(h) seem to be inadequate: to be consistent with the points shown in this subfigure as well as the number given in the main text (ignoring for the moment the variance with the other subfigures) the “experimental gap” should equal 0.4 eV; the gap between the calculated curves is 0.6 eV also at the $M$ point. However, taking into account all available information, the experimental gap at $M$ seems more likely to have a size of about 0.9 eV to 1.0 eV—thus being larger than calculated and at major variance with the peak positions depicted in Fig. 5(h).

An analogous analysis for the data close to $L$ presented
in Figs. 5(c), 5(e), and 5(g) yields an overall much more consistent picture as shown in the lower panels of Fig. 1. Also here, a comparably small shift in energy of about 0.2 eV might be present between the data in Fig. 5(c) and the other two subfigures, yet, the overall dispersions and gap sizes match rather well as they should.

The calculated band structure along the $\Gamma$-$K$-$M$ high-symmetry line is shown twice in Ref. 1: once in Fig. 5(b) and as well in the left panel of Fig. 4. Naturally, both presentations are expected to show identical results. However, when plotted together in Fig. 2, it is evident that there are nonnegligible differences between the two presentations. The deviations in the depicted band positions partially reach up to 0.4 eV or more than 0.05 $\pi/a$.

The data in Fig. 5 is also used to compare variations in the band width between the experiment and the calculation. For instance, “(f)or the $\sigma$ bands, the calculation gives a width that is $\sim 8\%$ smaller in the $A$-$L$ than in the $\Gamma$-$M$ direction. The experiment, however, shows the same bandwidth for both of these direction(s) (...)” [1]. Again, using the data from Figs. 5(a) and 5(c) it is possible to estimate the bottom of the first two $\sigma$ bands and thus their band width. The bottom of the first $\sigma$ band is at 2.43 eV at $M$ and at 2.06 eV at $L$, the bottom of the second $\sigma$ band is at 8.21 eV at $M$ and at 7.38 eV at $L$. Thus, the experimental $\sigma$ band width is reduced by 10% to 15% for $A$-$L$ compared to $\Gamma$-$M$. Furthermore, using the results of the calculations shown in the same figures, one finds the bottom of the first $\sigma$ band at 2.31 eV at $M$ and at 1.91 eV at $L$. The bottom of the second $\sigma$ band is at 8.10 eV at $M$ and at 6.95 eV at $L$. Hence, the presented calculation actually suggests a reduction of the band width by 14% to 17% for $A$-$L$ compared to $\Gamma$-$M$. Therefore, the experiment seems to be in full qualitative agreement with the shown calculations. Yet, neither the supposed observed equal experimental band width nor the expected $\sim 8\%$ band-width reduction quoted above can be deduced from the figures.

Another contestable practice used in Figs. 5 and 3 is the disregard of $k_z$ variations for constant photon energies when $k_\parallel$ becomes comparably large, despite quoting Eq. (1) in Ref. 1. For instance, in the used photon-energy range in the vicinity of the Fermi energy $k_z$ changes by almost 0.1 $\pi/c$ when increasing $k_\parallel$ from 0 to 2/$\sqrt[3]{3} \pi/a$. This implies that some caution is in order when comparing constant-photon-energy measurements with constant-$k_z$ calculations, even more so if bands with strong $k_z$ dispersion like the $\pi/\pi^*$ bands of MgB$_2$ are studied. Despite the claim of Ref. 1 to have revealed the “complete 2D and 3D electronic band structure of MgB$_2$”, with the exception of parts of the Fermi surface and bands at two distinct $k_z$ points, no out-of-plane dispersion has been presented. Therefore, it is difficult to assess quantitatively the effects of the $k_z$ variations on the measured band structure within the calculations of Ref. 1. However, as evident from the calculated Fermi surface, expected qualitative effects include, e.g., the diminishing of the $L$ electron band close to the Fermi energy in Fig. 5(c) and the corresponding growth of the “$\pi$ Fermi hexagons” in Fig. 3(b) when $k_z$ is moving away from (11) $\pi/c$.

Focusing now on the $k_z$-dependent cuts through the Fermi surface shown in Figs. 2(a) through 2(e), also...
Comparison of the absolute values of the calculated in-plane components of the Fermi crystal momenta in units of $\pi/a$ along the different high-symmetry lines shown in Figs. 1, 2(d), 2(e), 3, 4, and 5 of Ref. 1. Using the plots in Fig. 3 as reference, values deviating by more than 0.02 $\pi/a$ (0.05 $\pi/a$) are marked in blue (red).

| Figure | Notes |
|--------|-------|
| $k_x = 0, k_y < 0$ | base plane, left |
| $k_z = 16 \pi/c, k_x < 0$ | Fermi-surface cylinders in $\Gamma-M$ |
| $k_z = 16 \pi/c, k_y < 2 \pi/a$ | Fermi-surface cylinders in $M-K$ |
| $k_z = 15 \pi/c, k_x < 0$ | central plane, left |
| $k_z = 0, k_y < 0$ | $k_z$ integer multiples of $\pi/c$. Maybe most prominently, this is seen at the orange curve off-centered from $k_z = 14 \pi/c$ in Fig. 2(e) as well as the cross section of the warped $\sigma$ Fermi-surface cylinders in Fig. 2(d) which are not “narrowest at $\Gamma$” as stated in the text of Ref. 1. |
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Finally, a few minor issues in the experimental description of Ref. 1 are identified. It has been mentioned that “by scanning the photon energy ($h\nu$) at fixed binding energy $E_B$ and emission angle” one could scan, e.g., along $k\parallel = 0$. Due to the finite momentum transfer from the incident photons this is not strictly the case: $k\parallel = 0$ does not correspond to electrons emitted normally to the sample surface and the emission angle changes continuously with the photon energy/momentum. Also, the correction term taking into account the photon momentum transfer for the calculation of $k_z$ in Eq. (1) lacks a division by $h$. Eventually, while the angle between the analyzer axis and the incident photon beam is 70° at the ADDRESS SX-ARPES end station, the angle used in Eq. (1) ought to be the one between the inclined analyzer axis and the laboratory normal (20°) [4].
Having identified a multitude of inconsistencies in the presentation of the results, an important question is how these impact the analysis and conclusions of Ref. 1. This question should be addressed in two parts: Firstly, Ref. 1 establishes experimentally the topology of the 3D Fermi-surface as well as of the in-plane dispersion of MgB$_2$. The observed features are qualitatively well reproduced by calculations in the literature (cf., e.g., references within Ref. 1) and the apparent flaws described above do not affect this result. The second part of the question concerns if the detailed quantitative comparisons between the ARPES data and the presented calculations still hold despite the inconsistencies within and across the different illustrations. To answer that a few concrete examples shall be discussed in the following.

Ref. 1 has a particular emphasis on deviations of the $\pi/\pi^*$ calculations from the measured bands. For instance, “(a) $L$, the measured electron pocket has a smaller FS cross section (by $\sim 10\%$) than what is estimated by the calculation [Fig. 5(c)]. (…)” To correct for this, the calculated antibonding $\pi^*$ band would have to be shifted by approximately $200 \pm 50$ meV toward $E_F$. This is at substantial variance with the Fermi surface shown in Fig. 2(d) (cf. $k_{F,3}^{\perp}$ and $k_{F,4}^{\parallel}$ in Table I). To reproduce the values of $k_{F,3}^{\perp}$ and $k_{F,4}^{\parallel}$ in Fig. 2(d), this very band would have to be shifted by $0.8$ eV toward $E_F$—multiple times more than the deviation discussed here. Although in fact this correction seems unlikely and would probably be at variance with the $k_z$ dispersion of the band, it nonetheless illustrates the order of magnitude of the observed inconsistencies in Ref. 1 and thus a substantial uncertainty introduced by them.

Next, “(f)or the $\pi$ bands, the measured width for the band dispersing from $L$ to $H$ to $A$ [Fig. 5(d)] is $\sim 10-15\%$ larger than the calculated one” [1]. Without taking into account the argument just made and assuming the illustration in Fig. 5(d) represents the valid result of the calculation for $k_z = \pi/c$, the consequences of the $k_z$ variation with changing $k_y$ in the data can be estimated. As mentioned above, Ref. 1 does not provide out-of-plane dispersions. Therefore, calculations of Ref. 5 for the $L$-$M$ direction are used for an estimate. According to Eq. (1) $k_z$ varies by about $0.24 \pi/c$ between $L$ ($k_y = \pm 2 \pi/a$) and $A$ ($k_y = 0$) for $h\nu = 370$ eV and energies close to the Fermi energy. Moving $0.24 \pi/c$ away from $L$ toward $M$, changes the topmost occupied energy level by about $0.4$ eV toward $E_F$ [5] which corresponds to about $6.5\%$ of the band width. Hence, the deviation reported in Ref. 1 is potentially overestimated by about a factor of 2.

Already in the previous section the comparison of the $\sigma$ band width between $A$-$L$ and $\Gamma$-$M$ as seen in Figs. 5(a) and 5(c) has been discussed in detail. It is emphasized once more that neither the supposed observed equal experimental band width along these directions nor the expected $\sim 8\%$ band-width reduction for $A$-$L$ compared to $\Gamma$-$M$ quoted in Ref. 1 can be deduced from the figures.

The detailed analysis presented around Fig. 1 suggests that contrary to the claim of Ref. 1 the band gap at $M$ at high binding energies is most likely not smaller than calculated, but rather has a similar size than the deduced band gap at $L$ and thus is somewhat larger than predicted. It is also worth noting here, that this band gap is expected to have a negligible $k_z$ dependence [5], which—in contrast to the findings of Ref. 1—is nicely confirmed in Fig. 1.

In connection with Figs. 5(g) and 5(h) it is also pointed out in Ref. 1 that “a shift of the calculated band(s) edges by approximately 0.6 eV is needed in order to fit the data (…).” Given the uncertainty of the calculations arising from the comparison of the calculated band structures depicted in Figs. 5(b) and 4, the relevant energy levels at $M$ might be shifted up to 0.4 eV to lower binding energies, thus increasing the postulated deviation by 2/3 to about 1.0 eV (keeping the originally marked peak positions as reference).

Statements like “the $\sigma$ band width ‘is 1–2 % wider than the calculated one along $\Gamma$-$M$’ [1]” depend on a very high degree of accuracy in the calculations. The uncertainty originating from Fig. 2—which easily amounts up to 5 % of the band width—renders such conclusions not reliable.

Eventually, the host of significant variations in the calculated Fermi crystal momenta summarized in Table I as well as the overall uncertainty in the presented band calculations discussed in the preceding examples most likely preclude a meaningful detailed quantitative comparison between the experimental data and the calculations. Therefore, also the comparison of the conclusions obtained in Ref. 1 with previous de Haas–van Alphen Fermi-surface measurements has to be seen overall critically.

Taking into account all the effects discussed above, it is concluded that the results of the quantitative analysis in Ref. 1 are indeed substantially influenced by the inconsistencies reported here. With the only exception of the band-gap determination at the $L$ point, the quantitative results are found to be questionable or at least subject to major uncertainty which could only be reduced by a fully consistent presentation.

**SUMMARY**

In summary, the article by Sassa *et al.* provides experimental data of high quality and any arguments regarding the topology of the in-plane electronic band structure and of the 3D Fermi surface of MgB$_2$ appear valid. Yet, a multitude of inconsistencies in the analysis and/or presentation of the results cast doubt on the reliability of the obtained quantitative conclusions. Moreover, the funda-
mental claim of the article to have established “the full 3D electronic structure” of MgB$_2$ is found to be somewhat exaggerated.

The following free software was particularly useful while preparing this comment: Debian GNU/Linux \[6\] including KDE \[7\], Inkscape \[8\], Engauge \[9\], GNU Octave \[10\], ROOT \[11\], gnuplot \[12\], and TeX Live \[13\] as well as musrfit \[14\].

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[1] Y. Sassa, M. Månsson, M. Kobayashi, O. Göteborg, V. N. Strocov, T. Schmitt, N. D. Zhigadlo, O. Tjernberg, and B. Batlogg, “Probing two- and three-dimensional electrons in MgB$_2$ with soft x-ray angle-resolved photoemission,” Phys. Rev. B 91, 045114 (2015).

[2] With the exception of Fig. 1 and Fig. 2 (without subfigure label), all references to figures and equations refer to Ref. 1.

[3] Throughout this work, the ambiguous labels 1.15 and 1.33 in Ref. 1 are interpreted as $2/\sqrt{3}$ and $4/3$, respectively.

[4] http://www.psi.ch/sls/adress/ManualsEN/Momentum_ARPES.pdf.

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