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Referee Report on the Thesis by Izabela Biało entitled*: Role of the Charge Correlations in the mechanism of high temperature superconductivity,* submitted to the Faculty of Physics and Applied Computer Science of the AGH University of Science and Technology in Kraków and to the Institute of Solid State Physics of the Technische Universität Wien, for fulfilment of PhD degree in physics

The PhD Thesis has been carried out under the supervision of Professors Andrzej Kozłowski from AGH Kraków and Neven Barišić from TU Wien. It contains original scientific material, partly published in renowned international scientific journals (8 publications), not included as an explicit part of the Thesis. In my report, I shall briefly discuss the material chapter by chapter and at the end, summarize my main conclusions.

The introductory **Chapter 1** contains a nice but demanding brief overview of the experimental situation with 130 references attached to 20 pages of text. Hard job for a theorist who likes to test his preconceptions when analyzing experimental results. I regard it as an overview of what follows in the next chapters. I have two questions though. Do I understand correctly that the pseudogap and T^* temperatures are indeed close to each other? Also, I presume that the model discussed there is that the electrons located on $3d_{x^2-y^2}$ orbitals (which are subsequently thermally activated) due to Cu^{2+} ions and are localized throughout the superconducting phase below the optimal doping, whereas the itinerant holes are located in itinerant 2p states due to $p_{x/y}$ states of oxygen. Why then the AF-SC coexistence phase is absent in the cuprates, even though the localized $d_{x^2-y^2}$ unpaired electrons are strongly antiferromagnetically coupled in the CuO_2 planes?

Chapter 2 contains discussion of the resonant X-ray scattering data carried out on single-layer $HgBa_2CuO_{4+\delta}$ (Hg 1201) and observed charge-density-wave (CDW) order, which turned out to be effectively short range. The CDW peaks are fitted to the Gaussians. It would be desirable to have an explicit form of the Gaussian, as well as a brief explanation of the relation between the CDW coherence length and of the full width at half maximum (FWHM). At this point, I do not think that the correlation length ξ is just inverse of CDW-peak half-width. In general, ξ should be determined from the corresponding charge-charge correlation function or static susceptibility, which has the Ornstein-Zernike form. The reference paper for this simplified analysis is that of Tabiś et al., Nat Comm **5** 5875 (2014). Here the results extend in detail those in the earlier paper (PRB 2017) by studying CDW evolution as a function of both doping and temperature. Due to the fact that the coherence length $\xi/a \sim 8{,}0(7)$ at best (for $p \cong 0.08$) and from the circumstance that the elastic RXS intensity is smeared out as a function of reciprocal vector h and/or k, it is difficult to claim that the CDW order is well established. This is probably the reason why the author speaks about CDW order and CDW correlations interchangeably. However, from the facts that relative FWHM values are much smaller than the peak amplitude, which seems to disappear at well-defined temperature (cf. Tabiś et al., Fig. 1e), one can claim that CDW ordering temperature is well defined. Is that so? Also, perhaps a relation to our theoretical paper (cf. PRB **98**, 155144 (2018)) should be noted, as we show that the CDW long-range order disappears exactly at the optimal doping, as observed. The nature of CDW states is discussed there as well. I should also emphasize that the results concerning the phase diagram on the plane temperature-hole doping (cf. Fig 2.9) are very interesting as they incorporate explicitly the detailed CDW ordering. The subject of this chapter is connected to Ref. [36], in which I. Biało has contributed in an important way.

In **Chapter 3**, the author analyzes the applied-magnetic-field dependence of CDW correlations/ordering in YBCO. The main effect observed is the transition from 2D $\rightarrow$ 3D character of a CDW as a function of the increasing pulsed field from zero to above 16.5 T. The implicit question which appears there is again whether the CDW has a long-range character. Its 2D component seems to be of short range, whereas its 3D correspondent is rather of longer-range nature. The most interesting question is to what extent the 2D CDW with q_{CDW}= (1/2, 0, 0) or q_{CDW}= (1/2, 1/2, 0) can be regarded as LRO, what comes out naturally from theory? The second half of this Section is devoted to X-ray observation spectroscopy (XAS, XANES) in the applied field. While I appreciate the experimental effort devoted to obtaining the corresponding data, I find the conclusions as qualitative only. It would be easier to understand those results for (Hg 1201) samples, as they are single-layer systems with no CuO-chain contributions, which obscure clarity of the results. This remark is caused by the fact that the author contributed to the paper on the latter (cf. PRX **10,** 021059 (2020)) and the applied-field studies would coherently match the material presented in Chapter 2. Nonetheless, the presented systematic study of CDW evolution with varying hole concentration should be emphasized in any case.

Chapter 4 contains yet another aspect of high-temperature superconductors, i.e., phonons, carried out on electron-doped system (NCCO). The measurements have been carried with the help of inelastic X-ray scattering technique (IXS). The main results concerning the phonon dispersion relation of acoustic and optic branches are displayed in Figs. 4.7 and 4.8 for doped and undoped samples, respectively. The measured spectra are generally in good agreement with DFT calculations. The optical mode exhibits softening around the wave vector $q = q_{CDW}$, but this is ascribed it to the anticrossing of the corresponding neighboring modes. In effect, the conclusion is that the CDW correlations and phonon modes are essentially decoupled from each other. This, in turn, may be caused by the circumstance that the charge and phonon modes operate on different energy scales. In other words, the group of velocities of the acoustic plasmon is much larger than the corresponding value for acoustic phonons (perhaps also of that of optical phonons).

In **Chapter 5**, the author discussed one of the principal features of the CDW ordered state - its symmetry. But strictly speaking, the author addresses only the effect of uniaxial pressure on the checkboard or stripe CDW orders. The Chapter starts with construction details of pressure cell, with a possibility of exerting uniaxial pressure on the mounted samples of NCCO. Such experimental arrangement allows for studying the evolution of CDW under application of the uniaxial stress and hence, reduced orthorhombicity. The results in the pressure range 0-500 MPa turned out to be inconclusive in the sense that the CDW characteristic, such as amplitude of RXS, q_{CDW}, and FWHM which are constant within the data accuracy. The measurements have been performed with a dedicated effort though, as in other instances in this Thesis.

The final **Chapter 6** contains preliminary soft X-ray absorption measurements due to the oxygen K-edge and Cu L-edge in LSCO under uniaxial pressure, applied in Cu-O plane along [0,1,1] direction. The aim of that study was to change the degree of orthorhombicity and, in effect, observe the change of electronic properties induced by the uniaxial stress applied in the CuO_2 plane. The most interesting feature was the interpretation of the two-peak structure as the Hubbard subbands MHP (mobile hole peak) and UHP (upper Hubbard peak), respectively. What is interesting, this split-band picture was detected near the oxygen K-edge, not in the CuL_3-edge. However, a moment of reflection leads us to concluding that this is originating from the charge-transfer character of this Mott system. Under those circumstances, the holes originate from 2p states, which subsequently hybridize strongly with Cu 3d states. The high-energy excitation from the 2p filled states are to the upper Hubbard subband, originating predominantly from $3d^{10}$ states of Cu^{1+}. Is it then the main contribution to the peak in Fig. 6.1b due to excitation of 3d electrons directly across the Hubbard gap?

In summary, the Thesis results from a solid work of a dedicated researcher. The original results are numerous and properly summarized in Chapter 7. Although the text is dense and not easy to follow, particularly for a theorist not accustomed to such a detailed description of experiment, the results such as a those presented in Fig. 2.9, 3.2, 4.7, 4.8, and 4.12, are of the outstanding character. Parenthetically, the results of Fig. 4.12 may represent a starting point to further studies of dynamic collective excitations - plasmons and paramagnons, which are conspicuously absent in this work. A clear distinction between the static and dynamic correlations may lead to a resolution of a longstanding question of determining the possible long-range-order nature of the CDW-state correlations. Whether this resolution will throw a new light on pairing mechanism in high-temperature superconductors based on strong inter-electronic correlations, remains to be seen. However, I underline again, the present results already speak for themselves.

In conclusion, the PhD Thesis of Izabella Biało fulfils all the respective formal regulations for obtaining the degree of *doctor in physical science* in Poland. Additionally, I rate the Thesis as outstanding (cum laude, with the highest honors), as I document it in a separate formal letter. I recommend this Thesis to the appropriate PhD Committees at AGH Kraków and TU Wien for the final approval.

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