The manuscript entitled “The asymmetric Fermi surface of (Pby,Bi1−y)2Sr2−xLaxCuO6+δ” investigates the asymmetric Fermi surface of the cuprate Bi2201 using angle-resolved photoemission spectroscopy (ARPES). It reveals a significant asymmetry in Fermi momenta along orthogonal nodal directions, exceeding the orthorhombic distortion observed via X-ray crystallography. Using a tight-binding model and first-principle calculations, the authors propose a mechanism rooted in crystal-field splitting. The study presents a new, linear relationship between ARPES-derived and transport-derived doping estimates, providing insights into the electronic structure of high-temperature superconductors.

This manuscript is scientifically rigorous, novel, and well-aligned with SciPost Physics's standards for high-quality research. It makes a significant contribution to the understanding of electronic structure in high-temperature superconductors and fulfills the journal’s publication criteria. With that said, I have some points the authors should clarify:

1. Precision of Manipulator: To perform the cuts along ΓX and ΓY directions, it is necessary to rotate the sample and ensure that the cut is done precisely along the nodal direction. In the method section, it is mentioned that the experiments were conducted at I05 at the Diamond Light Source. How precise is the manipulator? It is worth mentioning this to ensure the robustness of the data.

2. MDC Analysis Presentation: In the paper, only the resultant fit of the MDCs is shown. However, the MDCs and the fitting are not displayed. The authors subtracted the full width at half maximum (FWHM) at E_F from the energy-dependent Lorentzian width of the Voigt function fitted to the MDCs. As I understood, this subtraction technique was a key methodological step to ensure the anisotropy measured was intrinsic rather than influenced by extrinsic effects like experimental resolution or sample quality. Can the authors show the MDCs before and after subtraction as a function of energy and temperature, and conduct the same analysis to see if the results obtained are similar? Showing such comparative analysis will strengthen the paper and convince the readers about the robustness of their results. Such comparative analysis can be added as supplementary information.

3. Significance of Linear Relationship: The manuscript introduces a new linear relationship between ARPES-derived and transport-derived doping estimates. Could the authors elaborate on its significance and limitations more explicitly?

4. Quantification of Uncertainties: Systematic uncertainties in ARPES measurements are addressed, but an explicit quantification of experimental and theoretical errors would strengthen the conclusions.

5. Applicability to Other Cuprates: The paper focuses on Bi2201 and does not fully explore the potential applicability of the findings to other single layer cuprates. If possible, it would be interesting to put these findings in perspective with other cuprates.

6. Nematicity and Charge Order Discussion: The discussion of nematicity and charge order as alternative explanations for the asymmetry could be expanded to connect with ongoing debates in condensed matter physics. For example, resonant inelastic X-ray scattering (RIXS) studies on Bi2201 have provided significant insights into its electronic properties, particularly concerning charge order and spin excitations. A notable study by Y.Y. Peng et al. (https://doi.org/10.1103/PhysRevB.94.184511) utilized Cu L_3-edge RIXS to investigate charge density modulations in underdoped and optimally doped Bi2201. They observed short-range charge order with a momentum transfer of approximately 0.23 reciprocal lattice units, persisting up to optimal doping levels. This charge order was found to modulate along the Cu-O bond directions, with no evidence of modulation along the nodal (diagonal) direction. Additionally, the out-of-plane measurements indicated a lack of phase correlation, suggesting that the charge order in single-layer Bi2201 is primarily two-dimensional.

In the context of this manuscript, the authors reveal a 6-10% difference in the nodal vectors along the ΓY and ΓX directions. This asymmetry was attributed to crystal-field splitting and orthorhombic distortions in the CuO_2 plane. Connecting these findings, the RIXS-detected charge order modulating along the Cu-O bond directions aligns with the ARPES-observed Fermi surface asymmetry. Both studies highlight the significance of anisotropic electronic properties in Bi2201. The charge order observed in RIXS could influence the electronic structure probed by ARPES, potentially contributing to the detected Fermi surface asymmetries. Furthermore, the absence of charge modulation along the nodal direction, as reported in the RIXS study, complements the ARPES findings by indicating that the observed anisotropies are more pronounced along specific crystallographic directions.

7. In the sentence starting by Berben et al., a subscript is missing for R(T).