

Angle-resolved photoemission spectroscopy (ARPES): probing electronic structure and many-body interactions

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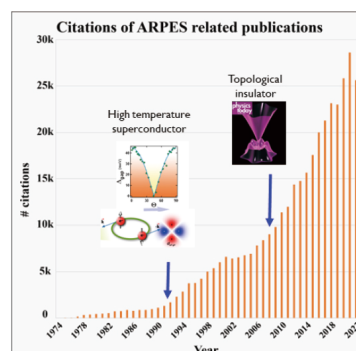
<https://doi.org/10.61109/cs.202405.130>

Citation: Z.-X. Shen, Angle-resolved photoemission spectroscopy (ARPES): probing electronic structure and many-body interactions, *Coshare Science* **02**, 04 (2024)

Author introduction

The significant enhancements in resolution, coupled with the expansion of modalities and meticulous alignment of problems, have propelled Angle-Resolved Photoemission Spectroscopy (ARPES) into the forefront as a primary tool for probing electronic structure and many-body effects in solids. The author stands as a pioneering figure in this advancement, marked by numerous groundbreaking discoveries and influential mentorship of key figures within the field.

Dr. Shen is the Paul Pigott Professor in Physical Sciences at Stanford University. His work has been recognized by many important awards, including the Centennial Lecture of the American Physical Society, the Kamerlingh Onnes Prize, the E.O. Lawrence Award, the Oliver E. Buckley Prize, Einstein Professorship Award, William and Flora Hewlett Foundation Fellow and Tage Erlander guest professor. In 2015, Dr. Shen was elected as a member of the National Academy of Sciences. In 2017, he was elected as a fellow of the American Academy of Arts and Sciences, and a foreign member of the Chinese Academy of Sciences.





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Abstract Complex phenomenon in quantum materials is a major theme of physics today. As better controlled model systems, a sophisticated understanding of the universality and diversity of these solids may lead to revelations well beyond themselves. Angle-resolved photoemission spectroscopy (ARPES), formulated after Einstein's photoelectric effect, has been a key tool to uncover the microscopic processes of the electrons that give rise to the rich physics in these solids. Over the last three decades, the improved resolution and carefully matched experiments have been the keys to turn this technique into a leading experimental probe of electronic structures and many-body effects.

Drawing upon examples spanning from novel superconductors and topological materials to magnetic and one-dimensional materials, we illustrate ARPES's pivotal role in testing ideas, benchmarking theoretical frameworks, uncovering unexpected phenomena, and elucidating the fingerprints of many-body interactions. Moreover, we demonstrate how the integration of modern ultrafast UV lasers and spin polarimetry has empowered photoemission spectroscopy to capture essential microscopic quantities of electrons—energy, momentum, spin, and temporal dynamics—yielding invaluable insights from a wealth of rich and precise information.

Keywords band structure, electron correlation, unconventional superconductors, topological insulators

Acknowledgements The works are supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under Contract No. DE-AC02-76SF00515.

References

1. J.R. Chelikowsky and M.L. Cohen, *Electronic structure of silicon*, *Phys. Rev. B* **10**, 5095 (1974).
2. A. Damascelli, Z. Hussain, and Z.-X. Shen, *Angle-resolved photoemission studies of the cuprate superconductors*, *Rev. Mod. Phys.* **75**, 473 (2003).
3. J.A. Sobota, Y. He, and Z.-X. Shen, *Angle-resolved photoemission studies of quantum materials*, *Rev. Mod. Phys.* **93**, 025006 (2021).
4. Z.-X. Shen, D.S. Dessau, B.O. Wells, D.M. King, W.E. Spicer, A.J. Arko, D. Marshall, L.W. Lombardo, A. Kapitulnik, P. Dickinson, S. Doniach, J. DiCarlo, T. Loeser, and C.H. Park, *Anomalous large gap anisotropy in the a - b plane of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$* , *Phys. Rev. Lett.* **70**, 1553 (1993).
5. Y.L. Chen, J.G. Analytis, J.H. Chu, Z.K. Liu, S.K. Mo, X.L. Qi, H.J. Zhang, D.H. Lu, X. Dai, Z. Fang, S.C. Zhang, I.R. Fisher, Z. Hussain, and Z.-X. Shen, *Experimental realization of a three-dimensional topological insulator, Bi_2Te_3* , *Science* **325**, 178 (2009).
6. B.O. Wells, Z.-X. Shen, A. Matsuura, D.M. King, M.A. Kastner, M. Greven, and R.J. Birgeneau, *E versus k relations and many body effects in the model insulating copper oxide $\text{Sr}_2\text{CuO}_2\text{Cl}_2$* , *Phys. Rev. Lett.* **74**, 964 (1995).
7. Z.Y. Chen, Y. Wang, S.N. Rebec, T. Jia, M. Hashimoto, D.H. Lu, B. Moritz, R.G. Moore, T.P. Devereaux, and Z.-X. Shen, *Anomalous strong near-neighbor attraction in doped 1D cuprate chains*, *Science* **373**, 1235 (2021).
8. D.M. King, D.S. Dessau, A.G. Loeser, Z.-X. Shen, and B.O. Wells, *Electronic structure evolution from mott insulator to superconductor — an angle-resolved photoemission investigation*, *J. Phys. Chem. Solids* **56**, 1865 (1995).
9. A.G. Loeser, Z.-X. Shen, D.S. Dessau, D.S. Marshall, C.H. Park, P. Fournier, and A. Kapitulnik, *Excitation gap in the normal state of underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$* , *Science* **273**, 325 (1996).
10. D.S. Marshall, D.S. Dessau, A.G. Loeser, C.H. Park, A.Y. Matsuura, J.N. Eckstein, I. Bozovic, P. Fournier, A. Kapitulnik, W.E. Spicer, and Z.-X. Shen, *Unconventional electronic structure evolution with hole doping in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$: angle-resolved photoemission results*, *Phys. Rev. Lett.* **76**, 4841 (1996).
11. K. Tanaka, W.S. Lee, D.H. Lu, A. Fujimori, T. Fujii, Risdiana, I. Terasaki, D.J. Scalapino, T.P. Devereaux, Z. Hussain, and Z.-X. Shen, *Distinct Fermi-momentum-dependent energy gaps in deeply underdoped Bi_2Te_3* , *Science* **314**, 1910 (2006).
12. S.D. Chen, M. Hashimoto, Y. He, D.J. Song, K.J. Xu, J.F. He, T.P. Devereaux, H. Eisaki, D.H. Lu, J. Zaanen, and Z.-X. Shen, *Incoherent*

strange metal sharply bounded by a critical doping in Bi2212, *Science* **366**, 1099 (2019).

13. S.D. Chen, M. Hashimoto, Y. He, D.J. Song, J.F. He, Y.F. Li, S. Ishida, H. Eisaki, J. Zaanen, T.P. Devereaux, D.H. Lee, D.H. Lu, and Z.-X. Shen, *Unconventional spectral signature of T_c in a pure d -wave superconductor*, *Nature* **601**, 562 (2022).
14. A. Lanzara, P.V. Bogdanov, X.J. Zhou, S.A. Kellar, D.L. Feng, E.D. Lu, T. Yoshida, H. Eisaki, A. Fujimori, K. Kishio, J.I. Shimoyama, T. Noda, S. Uchida, Z. Hussain, and Z.-X. Shen, *Evidence for ubiquitous strong electron-phonon coupling in high-temperature superconductors*, *Nature* **412**, 510 (2001).
15. T. Cuk, F. Baumberger, D.H. Lu, N. Ingle, X.J. Zhou, H. Eisaki, N. Kaneko, Z. Hussain, T.P. Devereaux, N. Nagaosa, and Z.-X. Shen, *Coupling of the B_{1g} phonon to the antinodal electronic states of $\text{Bi}_2\text{Sr}_2\text{Ca}_{0.92}\text{Y}_{0.08}\text{Cu}_2\text{O}_{8+\delta}$* , *Phys. Rev. Lett.* **93**, 117003 (2004).
16. J.J. Lee, F.T. Schmitt, R.G. Moore, S. Johnston, Y.T. Cui, W. Li, M. Yi, Z.K. Liu, M. Hashimoto, Y. Zhang, D.H. Lu, T.P. Devereaux, D.H. Lee, and Z.-X. Shen, *Interfacial mode coupling as the origin of the enhancement of T_c in FeSe films on SrTiO_3* , *Nature* **515**, 245 (2014).
17. S. Gerber, S.L. Yang, D. Zhu, H. Soifer, J.A. Sobota, S. Rebec, J.J. Lee, T. Jia, B. Moritz, C. Jia, A. Gauthier, Y. Li, D. Leuenberger, Y. Zhang, L. Chaix, W. Li, H. Jang, J.S. Lee, M. Yi, G.L. Dakovski, S. Song, J.M. Glowia, S. Nelson, K.W. Kim, Y.D. Chuang, Z. Hussain, R.G. Moore, T.P. Devereaux, W.S. Lee, P.S. Kirchmann, and Z.-X. Shen, *Femtosecond electron-phonon lock-in by photoemission and x-ray free-electron laser*, *Science* **357**, 71 (2017).
18. P.V. Bogdanov, *Study of high temperature superconductors with angle-resolved photoemission spectroscopy* (Doctoral dissertation, Stanford University, 2002).
19. M. Hashimoto, I.M. Vishik, R.H. He, T.P. Devereaux, and Z.-X. Shen, *Energy gaps in high-transition-temperature cuprate superconductors*, *Nat. Phys.* **10**, 483 (2014).
20. I.M. Vishik, M. Hashimoto, R.H. He, W.S. Lee, F. Schmitt, D.H. Lu, R.G. Moore, C. Zhang, W. Meevasana, T. Sasagawa, S. Uchida, K. Fujita, S. Ishida, M. Ishikado, Y. Yoshida, H. Eisaki, Z. Hussain, T.P. Devereaux, and Z.-X. Shen, *Phase competition in trisected superconducting dome*, *Proc. Natl. Acad. Sci.* **109**, 18332 (2012).
21. K.M. Shen, F. Ronning, D.H. Lu, F. Baumberger, N.J.C. Ingle, W.S. Lee, W. Meevasana, Y. Kohsaka, M. Azuma, M. Takano, H. Takagi, and Z.-X. Shen, *Nodal quasiparticles and antinodal charge ordering in $\text{Ca}_{2-x}\text{Na}_x\text{CuO}_2\text{Cl}_2$* , *Science* **307**, 901 (2005).
22. D.S. Dessau, B.O. Wells, Z.-X. Shen, W.E. Spicer, A.J. Arko, R.S. List, D.B. Mitzi, and A. Kapitulnik, *Anomalous spectral weight transfer at the superconducting transition of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$* , *Phys. Rev. Lett.* **66**, 2160 (1991).
23. Z.-X. Shen, W.E. Spicer, D.M. King, D.S. Dessau, and B.O. Wells, *Photoemission studies of high- T_c superconductors: the superconducting gap*, *Science* **267**, 343 (1995).
24. N.P. Armitage, D.H. Lu, C. Kim, A. Damascelli, K.M. Shen, F. Ronning, D.L. Feng, P.V. Bogdanov, Z.-X. Shen, Y. Onose, Y. Taguchi, Y. Tokura, P.K. Mang, N. Kaneko, and M. Greven, *Anomalous electronic structure and pseudogap effects in $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$* , *Phys. Rev. Lett.* **87**, 147003 (2001).
25. W.S. Lee, I.M. Vishik, K. Tanaka, D.H. Lu, T. Sasagawa, N. Nagaosa, T.P. Devereaux, Z. Hussain, and Z.-X. Shen, *Abrupt onset of a second energy gap at the superconducting transition of underdoped Bi2212*, *Nature* **450**, 81 (2007).
26. Z.K. Liu, B. Zhou, Y. Zhang, Z.J. Wang, H.M. Weng, D. Prabhakaran, S.K. Mo, Z.-X. Shen, Z. Fang, X. Dai, Z. Hussain, and Y.L. Chen, *Discovery of a three-dimensional topological Dirac semimetal, Na_3Bi* , *Science* **343**, 864 (2014).
27. Y. Zhang, T.R. Chang, B. Zhou, Y.T. Cui, H. Yan, Z.K. Liu, F. Schmitt, J. Lee, R. Moore, Y.L. Chen, H. Lin, H.T. Jeng, S.K. Mo, Z. Hussain, A. Bansil, and Z.-X. Shen, *Direct observation of the transition from indirect to direct bandgap in atomically thin epitaxial MoSe_2* , *Nat. Nanotechnol.* **9**, 111 (2014).
28. S.J. Tang, C.F. Zhang, D. Wong, Z. Pedramrazi, H.Z. Tsai, C.J. Jia, B. Moritz, M. Claassen, H. Ryu, S. Kahn, J. Jiang, H. Yan, M. Hashimoto, D.H. Lu, R.G. Moore, C.C. Hwang, C. Hwang, Z. Hussain, Y.L. Chen, M.M. Ugeda, Z. Liu, X.M. Xie, T.P. Devereaux, M.F. Crommie, S.K. Mo, and Z.-X. Shen, *Quantum spin Hall state in monolayer $1\text{T}'\text{-WTe}_2$* , *Nat. Phys.* **13**, 683 (2017).
29. Y. He, M. Hashimoto, D. Song, S.D. Chen, J. He, I.M. Vishik, B. Moritz, D.H. Lee, N. Nagaosa, J. Zaanen, T.P. Devereaux, Y. Yoshida, H. Eisaki, D.H. Lu, and Z.-X. Shen, *Rapid change of superconductivity and electron-phonon coupling through critical doping in Bi-2212*, *Science* **362**, 62 (2018).
30. H.J. Zhang, C.X. Liu, X.L. Qi, X. Dai, Z. Fang, and S.C. Zhang, *Topological insulators in Bi_2Se_3 , Bi_2Te_3 and Sb_2Te_3 with a single Dirac cone on the surface*, *Nat. Phys.* **5**, 438 (2009).
31. C.L. Kane and E.J. Mele, *Z_2 topological order and the quantum spin Hall effect*, *Phys. Rev. Lett.* **95**, 146802 (2005).
32. B.A. Bernevig, T.L. Hughes, and S.C. Zhang, *Quantum spin Hall effect and topological phase transition in HgTe quantum wells*, *Science* **314**, 1757 (2006).
33. L. Fu and C.L. Kane, *Topological insulators with inversion symmetry*, *Phys. Rev. B* **76**, 045302 (2007).
34. J.E. Moore and L. Balents, *Topological invariants of time-reversal-invariant band structures*, *Phys. Rev. B* **75**, 121306(R) (2007).
35. X.L. Qi and S.C. Zhang, *Spin-charge separation in the quantum spin Hall state*, *Phys. Rev. Lett.* **101**, 086802 (2008).
36. M.Z. Hasan and C. L. Kane, *Colloquium: topological insulators*, *Rev. Mod. Phys.* **82**, 3045 (2010).
37. D. Hsieh, D. Qian, L. Wray, Y. Xia, Y.S. Hor, R.J. Cava, and M.Z. Hasan, *A topological Dirac insulator in a quantum spin Hall phase*, *Nature* **452**, 970 (2008).
38. Y. Xia, D. Qian, D. Hsieh, L. Wray, A. Pal, H. Lin, A. Bansil, D. Grauer, Y.S. Hor, R.J. Cava, and M.Z. Hasan, *Observation of a large-gap topological-insulator class with a single Dirac cone on the surface*, *Nat. Phys.* **5**, 398 (2009).
39. N. Nagaosa, *A new state of quantum matter*, *Science* **318**, 758 (2007).
40. C. Day, *Quantum spin Hall effect shows up in a quantum well insulator, just as predicted*, *Phys. Today* **61(1)**, 19 (2008).
41. Y.M. Shi, J. Kahn, B. Niu, Z.Y. Fei, B.S. Sun, X.H. Cai, B.A. Francisco, D. Wu, Z.-X. Shen, X.D. Xu, D.H. Cobden, and Y.T. Cui, *Imaging quantum spin Hall edges in monolayer WTe_2* , *Sci. Adv.* **5**, eaat8799 (2019).
42. X.F. Qian, J.W. Liu, L. Fu, and J. Li, *Quantum spin Hall effect in two-dimensional transition metal dichalcogenides*, *Science* **346**, 1344 (2014).
43. M.M. Ugeda, A. Pulkin, S.J. Tang, H. Ryu, Q.S. Wu, Y. Zhang, D. Wong, Z. Pedramrazi, A.M. Recio, Y. Chen, F. Wang, Z.-X. Shen, S.K. Mo, O.V. Yazyev, and M.F. Crommie, *Observation of topologically protected states at crystalline phase boundaries in single-layer WSe_2* , *Nat. Commun.* **9**, 3401 (2018).
44. P. Zhang, K. Yaji, T. Hashimoto, Y. Ota, T. Kondo, K. Okazaki, Z.J. Wang, J.S. Wen, G.D. Gu, H. Ding, and S. Shin, *Observation of topological superconductivity on the surface of an iron-based superconductor*, *Science* **360**, 182 (2018).
45. Y.F. Li, S.D. Chen, M. Garcia-Diez, M.I. Iraola, H. Pfau, Y.L. Zhu, Z.Q. Mao, T. Chen, M. Yi, P.C. Dai, J.A. Sobota, M. Hashimoto, M.G. Vergniory, D.H. Lu, and Z.-X. Shen, *Spectroscopic evidence for topological band structure in $\text{FeTe}_{0.55}\text{Se}_{0.45}$* , arXiv:2307.03861v2 (2023).
46. C. Kim, A.Y. Matsuura, Z.-X. Shen, N. Motoyama, H. Eisaki, S. Uchida, T. Tohyama, and S. Maekawa, *Observation of spin-charge separation in one-dimensional SrCuO_2* , *Phys. Rev. Lett.* **77**, 4054 (1996).
47. B.J. Kim, H. Koh, E. Rotenberg, S.J. Oh, H. Eisaki, N. Motoyama, S. Uchida, T. Tohyama, S. Maekawa, Z.-X. Shen, and C. Kim, *Distinct spin and holon dispersions in photoemission spectral functions from one-dimensional SrCuO_2* , *Nat. Phys.* **2**, 397 (2006).
48. H.M. Hill, *Augmented model captures behavior in one-dimensional cuprates*, *Phys. Today* (2021).
49. H. Ding, T. Yokoya, J.C. Campuzano, T. Takahashi, M. Randeria, M.R. Norman, T. Mochiku, K. Kadowaki, and J. Giapintzakis, *Spectroscopic evidence for a pseudogap in the normal state of underdoped high- T_c superconductors*, *Nature* **382**, 51 (1996).
50. M. Hashimoto, R.H. He, K. Tanaka, J.P. Testaud, W. Meevasana, R.G.



- Moore, D.H. Lu, H. Yao, Y. Yoshida, H. Eisaki, T.P. Devereaux, Z. Hussain, and Z.-X. Shen, Particle-hole symmetry breaking in the pseudogap state of Bi2201, *Nat. Phys.* **6**, 414 (2010).
51. R.H. He, M. Hashimoto, H. Karapetyan, J.D. Koralek, J.P. Hinton, J.P. Testaud, V. Nathan, Y. Yoshida, H. Yao, K. Tanaka, W. Meevasana, R.G. Moore, D.H. Lu, S.K. Mo, M. Ishikado, H. Eisaki, Z. Hussain, T.P. Devereaux, S.A. Kivelson, J. Orenstein, A. Kapitulnik, and Z.-X. Shen, From a single-band metal to a high-temperature superconductor via two thermal phase transitions, *Science* **331**, 1579 (2011).
 52. M. Hashimoto, E.A. Nowadnick, R.H. He, I.M. Vishik, B. Moritz, Y. He, K. Tanaka, R.G. Moore, D.H. Lu, Y. Yoshida, M. Ishikado, T. Sasagawa, K. Fujita, S. Ishida, S. Uchida, H. Eisaki, Z. Hussain, T.P. Devereaux, and Z.-X. Shen, Direct spectroscopic evidence for phase competition between the pseudogap and superconductivity in Bi₂Sr₂CaCu₂O_{8+δ}, *Nat. Mater.* **14**, 37 (2015).
 53. K.J. Xu, Q.D. Guo, M. Hashimoto, Z.X. Li, S.D. Chen, J.F. He, Y. He, C. Li, M.H. Berntsen, C.R. Rotundu, Y.S. Lee, T.P. Devereaux, A. Rydh, D.H. Lu, D.H. Lee, O. Tjernberg, and Z.-X. Shen, Bogoliubov quasiparticle on the gossamer Fermi surface in electron-doped cuprates, *Nat. Phys.* **19**, 1834 (2023).
 54. K.J. Xu et al., *Science*, accepted (2024)
 55. Q.Y. Wang, Z. Li, W.H. Zhang, Z.C. Zhang, J.S. Zhang, W. Li, H. Ding, Y.B. Ou, P. Deng, K. Chang, J. Wen, C.L. Song, K. He, J.F. Jia, S.H. Ji, Y.Y. Wang, L.L. Wang, X. Chen, X.C. Ma, and Q.K. Xue, Interface-induced high-temperature superconductivity in single unit-cell FeSe films on SrTiO₃, *Chin. Phys. Lett.* **29**, 037402 (2012).
 56. N. Choudhury, E.J. Walter, A.I. Kolesnikov, and C.K. Loong, Large phonon band gap in SrTiO₃ and the vibrational signatures of ferroelectricity in ATiO₃ perovskites: first-principles lattice dynamics and inelastic neutron scattering, *Phys. Rev. B* **77**, 134111 (2008).
 57. J. Qu, X. Han, S. Sakamoto, C.J. Jia, J. Liu, H. Li, D. Guan, Y.J. Zeng, M. Schüler, P.S. Kirchmann, B. Moritz, Z. Hussain, T.P. Devereaux, Z.-X. Shen, and J.A. Sobota, Reversal of spin-polarization near the Fermi level of the Rashba semiconductor BiTeCl, *npj Quantum Mater.* **8**, 13 (2023).
 58. X. Han, J. Qu, S. Sakamoto, D.Y. Liu, D.D. Guan, J. Liu, H. Li, C.R. Rotundu, N. Andresen, C. Jozwiak, Z. Hussain, Z.-X. Shen, and J.A. Sobota, Development of deflector mode for spin-resolved time-of-flight photoemission spectroscopy, *Rev. Sci. Instrum.* **94**, 103906 (2023).
 59. J.A. Sobota, S. Yang, J.G. Analytis, Y.L. Chen, I.R. Fisher, P.S. Kirchmann, and Z.-X. Shen, Ultrafast optical excitation of a persistent surface-state population in the topological insulator Bi₂Se₃, *Phys. Rev. Lett.* **108**, 117403 (2012).
 60. Y. Mizuguchi, Y. Hara, K. Deguchi, S. Tsuda, T. Yamaguchi, K. Takeda, H. Kotegawa, H. Tou, and Y. Takano, Anion height dependence of T_c for the Fe-based superconductor, *Supercond. Sci. Technol.* **23**, 054013 (2010).
 61. S. Medvedev, T.M. McQueen, I.A. Troyan, T. Palasyuk, M.I. Erements, R.J. Cava, S. Naghavi, F. Casper, V. Ksenofontov, G. Wortmann, and C. Felser, Electronic and magnetic phase diagram of β -Fe_{1.01}Se with superconductivity at 36.7 K under pressure, *Nat. Mater.* **8**, 630 (2009).
 62. S. Mandal, R.E. Cohen, and K. Haule, Strong pressure-dependent electron-phonon coupling in FeSe, *Phys. Rev. B* **89**, 220502(R) (2014).
 63. P.W. Anderson, More is different: broken symmetry and the nature of the hierarchical structure of science, *Science* **177**, 393 (1972).

