Emergent electronic phases in cuprate strange metals

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Publication date
2022

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LIST OF PUBLICATIONS

This thesis contains work based on the following manuscripts:

- *Chapter 3:* M. Berben*, **S. Smit***, C. Duffy, Y.-T. Hsu, F. Heringa, F. Gerritsen, S. Cassanelli, X. Feng, S. Bron, E. van Heumen, Y. Huang, F. Bertran, T. K. Kim, C. Cacho, A. Carrington, M. S. Golden and N. E. Hussey, *'Superconducting dome and pseudogap endpoint in Bi2201'* (*Equal contribution), Physical Review Materials 6, 044804 (2022) (DOI: 10.1103/PhysRevMaterials.6.044804)

  - Contributions: S.S., F.H, F.G., S.C., X.F., S.B., E.v.H. and M.S.G. performed ARPES experiments with help from F.B., T.K.K. and C.C.. S.S. performed the ARPES data-analysis. M.B., C.D., Y.T.H, A.C., N.E.H. performed and analysed the transport measurements. Y.H. made the samples. M.B., S.S., M.S.G. and N.E.H. wrote the manuscript.

- *Chapter 3:* W. O. Tromp, T. Benschop, J.-F. Ge, I Battisti, K. M. Bastiaans, D. Chatzopoulos, A. Vervloet, **S. Smit**, E. van Heumen, M. S. Golden, Y. Huang, T. Kondo, Y. Yin, J. E. Hoffman, M. A. Sulangi, J. Zaanen, M. P. Allan, *'Puddle formation, persistent gaps, and non-mean-field breakdown of superconductivity in overdoped* $(Pb,Bi)_2Sr_2CuO_{6+\delta}$*'*, (under review)

  - Contributions: W.O.T, T.B., J-F.G., K.M.B, D.C., and Y.Y performed STM experiments. S.S. and M.S.G performed the ARPES experiments and analysis. W.O.T, I.B., and A.V. did data analysis. J.Z and M.A.S. performed the theoretical simulations. W.O.T., T.B. J-F.G., J.Z., and M.P.A. wrote the manuscript. Y.H., T.K., and E.v.H. provided the samples.

- *Chapter 4:* **S. Smit**, E. Mauri, L. Bawden, F. Heringa, F. Gerritsen, E. van Heumen, Y.K. Huang, T. Kondo, T. Takeuchi, N. E. Hussey, T.K. Kim, C. Cacho, A. Krikun, K. Schalm, H.T.C. Stoof, and M.S. Golden, *'Momentum-dependent scaling exponents of nodal self-energies measured in strange metal cuprates and modelled using semi-holography'*, (under review) `https://arxiv.org/pdf/2112.06576.pdf`

  - Contributions: S.S., L.B, F.H., F.G., E.v.H. and M.S.G. performed the ARPES experiments, with support from T.K.K and C.C. Data-analysis was done by S.S. with input from A.K., K.S., E.v.H., E.M., H.T.C.S. and M.S.G.. Y.K.H., T.K., T.T. and N.E.H. provided the samples. The theoretical calculations were performed by E.M. and H.T.C.S. The manuscript was written by S.S., E.M., H.T.C.S., M.S.G., with input from K.S. and A.K.

- *Chapter 5:* **S. Smit**, L. Bawden, Y. Huang, J. Henke, J. van Wezel, F. Heringa, F. Gerritsen, E. van Heumen, M. Roslova, L. Folkers, A. Isaeva, J. I. Facio, T. Kondo, and M.S. Golden, *'Fast and slow nodes: the asymmetric Fermi surface of Bi2201'* (in preparation)

  - Contributions: S.S., L.B, F.H., F.G., E.v.H. and M.S.G. performed the ARPES experiments. ARPES data-analysis was done by S.S., with help from J.H. and J.v.W.. Y.K.H. and T.K., provided the samples. The XRD was performed and analysed by M.R., L.F. and A.I.. The theoretical calculations were performed by J.I.F. The manuscript was written by S.S., M.R., A.I. and M.S.G.,

Other publications involving the author

- L. Mulder, C. Castenmiller, F. J. Witmans, **S. Smit**, M. S. Golden, H. J. W. Zandvliet, P. L. de Boeij, and A. Brinkman, *'Spectroscopic signature of surface states and bunching of bulk subbands in topological insulator $(Bi_{0.4}Sb_{0.6})_2Te_3$ thin films'*, Physical Review B 105, 035122 (2022) (DOI: 10.1103/PhysRevB.105.035122)

- G. Araizi-Kanoutas, J. Geessinck, N. Gauquelin, **S. Smit**, X. H. Verbeek, S. K. Mishra, P. Bencok, C. Schlueter, T. Lee, D. Krishnan, J. Fatermans, J. Verbeeck, G. Rijnders, G. Koster, and M. S. Golden, *'Co valence transformation in isopolar $LaCoO_3/LaTiO_3$ perovskite heterostructures via interfacial engineering'*, Physical Review Materials 4, 026001 (2020) (DOI: 10.1103/PhysRevMaterials.4.026001)

- P.T. Phong Le, K. Hofhuis, A. Rana, M. Huijben, H. Hilgenkamp, G. A. J. H. M. Rijnders, J. E. ten Elshof, G. Koster, N. Gauquelin, G. Lumbeeck, C. Schüßler-Langeheine, H. Popescu, F. Fortuna, **S. Smit**, X. H. Verbeek, G. Araizi-Kanoutas, S. Mishra, I. Vaskivskyi, H. A. Dürr, and M. S. Golden, *'Tailoring Vanadium Dioxide Film Orientation Using Nanosheets: a Combined Microscopy, Diffraction, Transport, and Soft X-Ray in Transmission Study'*, Advanced Functional Materials 30, 1900028 (2020) (DOI: 10.1002/adfm.201900028)

- N. de Jong, R. Heimbuch, S. Elins, **S. Smit**, E. Frantzeskakis, J.-S. Caux, H.J.W. Zandvliet and M. S. Golden, *'Gold-induced nanowires on the Ge(100) surface yield a 2D and not a 1D electronic structure'*, Physical Review B 93, 235444 (2016) (DOI: 10.1103/PhysRevB.93.235444)

SUMMARY

Emergent electronic phases in cuprate strange metals

In this thesis, the electronic structure of $(Pb,Bi)_2Sr_{2-x}La_xCuO_{6+\delta}$ (or Bi2201), is investigated using angle resolved photoemission spectroscopy (ARPES). Bi2201 and other cuprates, while being famous for being unconventional, high temperature superconductors, are actually more mysterious due to their anomalous normal state. In this 'strange metallic' phase, they show properties that cannot be reconciled with any conventional theory of metals, and understanding this phase is proposed to hold the key for understanding the high T_c superconducting ground state.

Following over 30 years of intense investigation, cuprates have a vast and complicated literature, with sometimes contradicting information. For example, the phase diagram of Bi2201 as function of hole doping and temperature is disputed, as there are competing claims regarding how many doped holes are needed to enable the superconductivity and the pseudogap phases. In **Chapter 3**, using a study combining ARPES and electrical transport, the phase diagram of Bi2201 is brought in line with that of other cuprates. By comparing both in-plane resistivity and results from Luttinger counting between LSCO and Bi2212, we propose a similar dependence of the superconducting dome on hole-doping for these compounds. The location of p^*, the endpoint of the pseudogap regime, is found at a doping level of $p^* \approx 0.2$, both by showing a sharp decrease in gap magnitude upon hole doping above $p > p^*$, and a sudden change from incoherent-to-coherent spectral lineshapes, similar to what has been found in the bilayer compound Bi2212. This observation is reinforced by the disappearance of the characteristic 's-shape' in the resistivity data around the same doping level. These experimental facts strengthen the evidence for universality within the cuprate families, implying that most of the important electronic properties can be derived simply by the theoretical modelling of doped CuO_2 planes. Having established that Bi2201 is not that different from other cuprate compounds, **Chapter 4** turns the attention to a specific region in momentum space, namely the node, where the lack of gaps and the steep dispersion allows for precise quantitative analysis of the spectral function. Here an extensive survey as function of hole doping and temperature was carried out, yielding a large data set of very high quality.

By careful fitting, we find that the electron-electron interactions are excellently described by a single power law, with a momentum dependent scaling exponent. This momentum dependence reveals itself in the lineshape of the momentum distribution curves, which deviate from being Lorentzians. A mathematical description for this follows naturally from the holographic duality, where strongly interacting systems such as the cuprates can be modelled by a dual, gravitational problem in the framework of general relativity. This work constitutes the first time that real ARPES data is compared to holographic calculations.

In the last two chapters of this thesis the Fermi surface of Bi2201 is more closely investigated. First in **Chapter 5**, an asymmetry of the Fermi surface is uncovered that is deeply rooted in the underlying crystal structure. The CuO_2 planes that form the core of the electronic properties are not four-fold rotationally symmetric, and this is reflected in the electronic behavior. The bands in the two nodal k-space directions have different Fermi wave vectors, velocities and widths in their momentum distribution. First-principle Density Functional Theory (DFT) calculations, using the lattice parameters attained from X-ray diffraction experiments, confirm the crystallographic origin of the asymmetry. Using these DFT calculations as a proxy for the bare band structure, we support a notion of differing lifetime between the carriers in the two nodal directions. Finally in **Chapter 6**, we use the unique strength of ARPES in measuring the k-resolved spectral function to connect to an anomalous feature visible in transport experiments, namely that of the gradual loss of active carriers in Hall measurements (n_H) with underdoping. Over a broad range in doping, unconnected to the opening of the pseudogap or a topological transformation of the Fermi surface, the transport-active carriers change from the total number of carriers per copper site $1 + p$ at high overdoping, to only the extra doped holes, p, as p_{opt} is approached. The fact that this correlates with an increase in both the linear component of the resistivity and the superfluid density, has brought forward the idea that this drop in n_H is caused by an increasing number of incoherent carriers that do not contribute to the Hall number. By investigating the width of the spectral peaks around the Fermi surface, a trend is uncovered in the very same doping region disconnected to the pseudogap. In this doping range the states around the antinode get increasingly incoherent with underdoping, something expressed in the width of the spectral function for $k = k_F$. This could possibly account for the missing carriers in the transport experiments.

Emergente elektronische fases in vreemde metalen van koper-oxides

In deze thesis wordt de elektronische structuur van $(Pb,Bi)_2Sr_{2-x}La_xCuO_{6+\delta}$, (ofwel Bi2201) onderzocht met behulp van hoekopgeloste foto-emissie spectroscopie (ARPES). Bi2201 en vergelijkbare koper-oxides zijn bekend vanwege hun onconventionele hoge temperatuur supergeleiding, maar hun abnormale metallische toestand is eigenlijk nog veel mysterieuzer. In deze 'vreemde metaal'-toestand laten ze eigenschappen zien die niet verklaard kunnen worden door enige conventionele theorieen, en men denkt dat het begrijpen van deze toestand de sleutel is tot het begrijpen van de hoge temperatuur supergeleiding.

Na meer dan 30 jaar onderzoek, is er een enorme, gecompliceerde berg aan resultaten te vinden in de literatuur, met soms tegenstrijdige bevindingen. Bijvoorbeeld het fase-diagram van Bi2201 als functie van temperatuur en doping is onderwerp van discussie, omdat er tegengestelde resultaten zijn over hoeveel ladingsdragers er nodig zijn in het materiaal om de supergeleidende en pseudogap toestand te verkrijgen. In **Hoofdstuk 3**, wordt een combinatie van ARPES en elektrische weerstands metingen gecombineerd om het fase diagram van Bi2201 in lijn te brengen met dat van andere koper-oxides. Door zowel de geleiding in het vlak en Luttingers theorema te vergelijken tussen LSCO en Bi2201, stellen we een vergelijkbare afhankelijkheid van de supergeleiding voor op de doping tussen deze materialen. De positie van p^*, het einde van de pseudogap, wordt gevonden bij een doping van $p^* \approx 0.2$, zowel door het ineens krimpen van de supergeleidende energie-kloof, en een net zo plotselinge overgang van incoherente-naar-coherente spectra, net zoals eerder in het tweelaagse materiaal Bi2212 is gevonden. Deze observatie wordt ondersteund door het verdwijnen van de karakteristieke 's-vorm' in de elektrische weerstand bij eenzelfde doping. Deze experimentele bevindingen versterken het idee dat de koper-oxides universeel gedrag vertonen, wat impliceert dat belangrijke elektronische eigenschappen simpelweg gemodelleerd kunnen worden door het dopen van CuO_2 vlakken. Nu vastgesteld is dat Bi2201 niet zo anders is als andere koper-oxides, wordt de aandacht in **hoofdstuk 4** gevestigt op een specifieke richting in impuls-ruimte, die de 'node' wordt genoemd. Hier zijn geen energie-kloven, en is de

dispersie erg steil. Dit maakt zeer precieze kwantitatieve analyse van de spectraalfunctie mogelijk. In deze nodal richting is een uitgebreide studie als functie van doping en temperatuur uitgevoerd, wat een grote dataset heeft opgeleverd van hoge kwaliteit. Door zorgvuldig te fitten, hebben we gevonden dat de elektron-elektron interacties in dit materiaal zeer goed beschreven kunnen worden door een enkele machtsfunctie, met een exponent die van de impuls afhangt. Deze afhankelijkheid uit zich in de lijnvorm van de spectra als functie van impuls, die geen symmetrische Lorentzians zijn. Een wiskundige beschrijving hiervoor volgt vanuit de holografische dualiteit, waar een sterk interactief systeem, zoals de koper-oxides, gemodelleerd kan worden door een duaal zwaartekrachts systeem in de algemene relativiteits theorie. Dit resultaat is de eerste keer dat echte ARPES data vergeleken wordt met holografische berekeningen. In de laatste twee hoofdstukken van deze thesis wordt het Fermi oppervlak van Bi2201 zorgvuldig onderzocht. Eerst in **Hoofdstuk 5**, leggen we een asymmetrie van het het Fermi oppervlak bloot, die zijn oorsprong heeft in de onderliggende kristal structuur. De CuO_2 vlakken die aan de basis liggen van de elektronische eigenschappen zijn niet viervoudig symmetrisch, en dit is terug te zien in de elektronische eigenschappen. De banden in de twee 'knoop'-richtingen hebben verschillende impulsen aan het Fermi niveau, en ook verschillende steilheden en breedtes van de pieken. Berekeningen vanuit Dichtheids-Functionaal-Theorie (DFT), waarin de rooster parameters gebruikt worden die met X-ray diffractie experimenten zijn bepaald, ondersteunen een scenario waarin de elektronische interacties verschillend zijn in de twee richtingen. Uiteindelijk in **Hoofstuk 6**, gebruiken we de unieke kracht van ARPES in het meten van impuls-opgeloste spectraalfuncties om te verbinden met een abnormale observatie in weerstandsmetingen, namelijk die van een gradueel verlies van transport-actieve ladingsdragers in Hall-metingen (n_H) met verminderde doping. Over een breed bereik van doping, niet verbonden aan de pseudogap of een topologische transformatie van het Fermi oppervlak, verandert het aantal ladingsdragers van het totaal aantal dragers per koper-ion, $1 + p$ bij overdoping, naar alleen de extra gedoseerde dragers, p, als p_{opt} wordt benaderd. Het feit dat dit correleert met een verhoogde lineare component van de weerstand en de 'super-vloeistof dichtheid', leidt tot het idee dat de daling in n_H wordt veroorzaakt door een groeiend aantal incoherente ladingsdragers. Door de breedte van de spectraalfunctie langs het Fermi oppervlak te bestuderen, zien we een trend ontstaan in precies dit doping gebied. Hier worden de toestanden langs de antinode steeds meer incoherent met minder doping, wat geuit wordt in de breedte van de spectraalfunctie bij $k = k_F$. Dit zou mogelijk de 'missende' ladingsdragers in de weerstandsmetingen kunnen verklaren.

ACKNOWLEDGEMENTS

I'm very grateful for having had the opportunity to do my PhD with you. Mark. You have been a great supervisor, and I have learned a lot from you during our time working together. From hands on labwork, to deep scientific discussions and (workplace) politics, I really appreciate your never ending energy and enthusiasm, and the trust and responsibility you give to your students. Nigel, thanks a lot for stepping in to be my second advisor. Working together with you and your group has always been a great and very pleasant experience.

Many people have been instrumental to the success of my PhD. Yingkai, thanks for always working hard to grow amazing crystals. Hans and Johan, and the rest of the TC, many thanks for all your hard work on building and maintaining AMSTEL, this work would not have been possible without you. Astrid, Klaartje and Jirina, thanks for all your administrative help over the years. Thanks to Anne for all your help with my PPMS measurements.

The start of my PhD was greatly helped by the people already in the lab. Lewis, Maarten, Georgios, I appreciate you teaching me the ways of ARPES, data-analysis and the importance of Friday afternoon drinks. Now its time to hand the lab over to the new guard: Sergio, Saumya, Arnaud and Kourosh, I'm happy you joined the lab, you have made my last few months here a lot more enjoyable.

On the scientific front, I have enjoyed the collaboration with the Utrecht crew immensely. Henk and Enea, thanks for all your patient explanations and insightful discussions, I always looked forward to our weekly meetings and I am very proud of the results we achieved! Nigel and Maarten, our work together has been very rewarding, I learned a lot from both of you. It keeps amazing me how much information you can extract from your 1D curves. Maarten, I'm very impressed by ourselves for actually sticking to the one goal of the consortium: Publishing a combined transport-ARPES paper!

During my stay at the IoP I have had the pleasure of meeting and working together with many people: Florian, Floris, Sil, Vera and Mardou, thank you for all the energy and excitement you brought to the lab during your projects. Xuanbo, Stephan, Silvia, thanks for your help with the beamtimes, and our enjoyable physics discussions. Paul, I like that no matter how busy you are, you always have time for a chat and a coffee. Veldhoven 2020 won't be forgotten! Heleen, thanks for the

bikerides and swimming lessons, it seems I still have some catching up to do...

Jans, as my PhD colleague and friend I'm very happy that we took similar paths starting from AMEP, and proud that we managed to get this far together, even ending up being each others paranymphs! Noor and Alona, thanks for always taking the time to hang out and inviting me to your parties, and I really enjoyed the trip to Sweden!

Mannen (en dame) van Zwaar 16, ik ben erg blij dat we destijds bij elkaar in de boot terecht zijn gekomen. Ik kan altijd op jullie rekenen, zowel in de boot (ZRBlik!) als op de fiets en daarbuiten. De vakanties - Kyrgystan, Ardennen, Duitsland - waren fantastisch, en ik kan niet wachten op de volgende.

Bijna 10 jaar na ons afstuderen is het diner met de jongens die Ooit in de Bergerie waren nog altijd een van de hoogtepunten van mijn kerstvakantie. Luuk, Timpa en Max, ik vind het erg leuk dat we elkaar nog zo vaak zien, van Kos tot de Baltische staten, het is overal feest! Quincy, 15 jaar geleden had ik nooit gedacht dat wij vrienden zouden worden, maar ik vind het leuk dat het toch gelukt is.

Dorus, bedankt voor de introductie tot de prachtige fietssport, het heeft me veel geholpen tijdens de PhD. Van toertochten over de Veluwe tot klimmen in de Franse alpen, niks zo goed om je gedachten te verzetten als uren pijnlijden op de fiets. Lieve pap en mam, ontzettend bedankt voor al jullie onvoorwaardelijke steun met alles dat ik doe, en dat er altijd een bord eten en bed klaarstaat als ik thuiskom.