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#### Optical spectroscopy of quantum spin systems

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## Acknowledgements

Meeting Dick van der Marel was just a coincidence. At that time I was working in the field of nonlinear optics and I had never heard of him. Then again, he had never heard of me either! Now, after the four years that I spent working with him, I can indeed say that this coincidence was a very lucky one. I really appreciate his way of both doing research and guiding the students in their projects. As an experimental physicist, his aim is not only to obtain relevant 'experimental facts', but also to understand them on a deep theoretical and intellectual level. As a supervisor, he is constantly inspiring and confronting his students, but, at the same time, he grants them all the freedom they wish. In other words, thanks Dick for what I learned from you, for your contagious enthusiasm, and for the great time we had together. I hope that these four years will be the basis of a long friendship and fruitful future collaborations.

Working in Groningen has been a special experience also because of the high level scientific environment. I had the opportunity to interact with scientists who have vast knowledge and experience, like George Sawatzky, Daniel Khomskii, and Thom Palstra. I would like to thank George Sawatzky for the interest he always showed in my work and, in particular, for the series of evening lectures he gave for Ph.D. students. It was extremely useful and pleasant being there: I never saw anybody else able to talk about solid state physics in such a fascinating way. I am very grateful to Daniel Khomskii for the careful reading of my manuscript but, above all, for the 'daily discussions' we had, which were extremely important for me when entering the realm of low dimensional spin systems. He is indeed a theoretician who knows how to talk to experimentalists. I am particularly grateful to Thom Palstra for the intense collaboration we had on the project of  $\alpha'$ -NaV<sub>2</sub>O<sub>5</sub>. His alternative experimental approach and the occasional 'divergence of opinions' have been extremely precious and stimulating.

Obviously, not only the 'big shots' made Groningen such a pleasant place for me. The number of people is much larger and, among them, I would like to thank, first of all, Cor Bos, the godfather of our 'Cor-related experimental systems'. His technical and much more than technical assistance was irreplaceable. Secondly, I would like to express my gratitude to Auke Meetsma and Jan de Boer, who tried hard to turn me into a crystallographer; Henk Bron, who polished and 'smoked' most of my samples; and Frans van der Horst, who was never tired of having to enlarge my quota on our work-station.

Special thanks to Maxim Mostovoy, a brilliant theoretician and an even more brilliant (and patient) teacher. I really enjoyed our frequent discussions.

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And now, the people of my group, who I could write a book about. Markus Grüninger, the German paranimf: We shared everything, life inside and outside the lab, the experience of having a son (I mean, you had yours and I had mine!)...not to mention our tough discussions (the one we had at the bottom of the Grand Canyon was great!). Thanks Markus, I'm sure our friendship will not be just a four year experience. Johan Feenstra: thanks Johan, you also are a tremendous friend, even though we did not really share everything: I'm sorry, but I was not brave enough to eat a kroket together with you. However, I was very pleased to see how much you did appreciate my risotto and, remember, you will always be welcome. Anna-Maria Janner: What can I say? We already told each other everything, and in Italian! One remark: Life has indeed changed in our office since you left-it's just so quiet! Thanks Anna-Maria, and when Johan comes to our place for a risotto, don't be shy and join him! Karina Schulte: I really enjoyed working with you when you were my student and later when you became my k-space teacher. Most important of all, we had a great time: You, me and the Bomem. Dank je, Karina, for the translation of the Samenvatting, along with all the 'Dutch mail' that, strangely enough, I was receiving every day. One last thing, toti toti! Let me now continue by thanking people in a more concise way: Jeroen van der Eb (see you in Stanford?), Diana Dulic (see you and Walter in Sardinia?), Artem Tsvetkov (spasibo, tovaristch: We had a short but good time together), Cristian Presura (who is taking good care of my samples), Hajo Molegraaf (who took care of the layout of the front cover), Ronald Hesper (and his brilliant car), Mark James (who took care of considerably improving the English here and there), Jae H. Kim and Jürgen Schützmann (my personal FTIR trainers, together with Clay Paays from Bomem), and the 'Italian Mafia': Salvatore Altieri and Alberto Morpurgo (se vedemo, testine di silicio). Although he doesn't belong to the Solid State Physics Lab of the University of Groningen, I would also like to thank Fabio Fiorani, the *Italian paranimf*: thanks Fabio, for being such a great *contrappeso* for so many years!

During my Ph.D research I had the opportunity to interact with many people outside the University of Groningen. First of all, a big thank you goes to the people who grew the beautiful and large single crystals used in this experimental investigation: A.A. Menovsky for the transition-metal mono-silicide samples, and J. Jegoudez, G. Dhalenne, and A. Revcolevschi for the  $CuGeO_3$  and  $\alpha'$ - $NaV_2O_5$  samples.

I am indebted to M. Fäth, J. Aarts, and J.A. Mydosh for their intense cooperation on the FeSi project (with a continuous exchange of 'unpublished' data), and to V. Vescoli and L. Degiorgi for numerous discussions and for providing me with their optical data on RuSi. I also enjoyed fruitful discussions with P.H.M. van Loosdrecht, D. Smirnov, J. Leotin, B. Büchner, T. Mizokawa, J.E. Lorenzo, P. Lemmens, G.S. Uhrig, and M. Braden.

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The last thank you goes to Fulvio Parmigiani, who supervised my undergraduate thesis project and suggested to me to come to Groningen for my Ph.D. research. Since then, he has never stopped giving me good advice, and unlimited scientific and moral support.

At this point some people may wonder:

Isn't this guy going to thank all those friends not scientifically involved in his thesis work?

And what about his parents, who made everything possible in the first place??

And, even more surprisingly, isn't he going to thank his beloved wife Barbara, who let him go to The Netherlands for his Ph.D. after only two weeks of marriage, who one year later managed to find an occupation in Groningen and moved over there so that they could live together, who gave birth to their son Matteo, who was waiting for him to come home from the lab all those nights and weekends, who always encouraged him, and who even accepted to be put on the front cover of his thesis???

Don't worry, I will do that, but in real life!!!

### List of Publications

- A. Damascelli, G. Gabetta, A. Lumachi, L. Fini, and F. Parmigiani. Multiphoton Electron Emission from Cu and W: an Angle-Resolved Study. Physical Review B **54**, 6031 (1996).
- A. Damascelli, K. Schulte, D. van der Marel, M. Fäth, and A.A. Menovsky. *Optical Phonons in the Reflectivity Spectrum of FeSi*. Physica B **230-232**, 787 (1997).
- A. Damascelli, K. Schulte, D. van der Marel, and A.A. Menovsky.

  Infrared Spectroscopic Study of Phonons Coupled to Charge Excitations in FeSi.

  Physical Review B (Rapid Communications) 55, R4863 (1997).
- A. Damascelli, D. van der Marel, F. Parmigiani, G. Dhalenne, and A. Revcolevschi. Infrared Signatures of the Spin-Peierls Transition in CuGeO<sub>3</sub>. Physical Review B (Rapid Communications) **56**, R11373 (1997).
- A. Damascelli, D. van der Marel, F. Parmigiani, G. Dhalenne, and A. Revcolevschi. *Infrared Reflectivity of Pure and Doped CuGeO*<sub>3</sub>. Physica B **244**, 114 (1998).
- D. van der Marel, A. Damascelli, K. Schulte, and A.A. Menovsky. Spin, Charge and Bonding in Transition Metal Mono-Silicides. Physica B **244**, 138 (1998).
- A. Damascelli, D. van der Marel, M. Grüninger, C. Presura, T.T.M. Palstra, J. Jegoudez, and A. Revcolevschi.
   Direct Two-Magnon Optical Absorption in α'-NaV<sub>2</sub>O<sub>5</sub>: "Charged" Magnons. Physical Review Letters 81, 918 (1998).
- A.A. Tsvetkov, D. van der Marel, K.A. Moler, J.R. Kirtley, J.L. de Boer, A. Meetsma, Z.F. Ren, N. Koleshnikov, D. Dulic, A. Damascelli, M. Grüninger, J. Schützmann, J.W. van der Eb, H.S. Somal, and J.H. Wang.

  Global and Local Measures of the Intrinsic Josephson Coupling in Tl<sub>2</sub>Ba<sub>2</sub>CuO<sub>6</sub> as a Test of the Interlayer Tunneling Model.

  Nature 395, 360 (1998).

- A. Meetsma, J.L. de Boer, A. Damascelli, J. Jegoudez, A. Revcolevschi, and T.T.M. Palstra.
   Inversion Symmetry in the Spin-Peierls Compound α'-NaV<sub>2</sub>O<sub>5</sub>.
   Acta Crystallographica C 54, 1558 (1998).
- A. Damascelli, D. van der Marel, J. Jegoudez, G. Dhalenne, and A. Revcolevschi. The Symmetry Problem in  $\alpha'$ -NaV<sub>2</sub>O<sub>5</sub>. Physica B, in press (1999).
- M. Grüninger, D. van der Marel, A. Damascelli, A. Zibold, H.P. Geserich, A. Erb, M. Kläser, Th. Wolf, T. Nunner, and T. Kopp.
   Charged Magnons and Magneto-Elastic Polarons in the MIR Spectrum of YBa<sub>2</sub> Cu<sub>3</sub> O<sub>6</sub>. Physica C, in press (1999).
- A.A. Tsvetkov, D. Dulic, D. van der Marel, A. Damascelli, G.A. Kaljushnaia, J.I. Gorina, N.N. Senturina, B. Willemsen, N.N. Kolesnikov, Z.F. Ren, J.H. Wang, A.A. Menovsky, and T.T.M. Palstra.

  Systematics of c-axis Phonons in the Tl and Bi Based Cuprate Superconductors.

Submitted to Physical Review B (1999).

## Happy End

During the four years I spent in Groningen working on my Ph.D., I tried many and many times to explain to people, not belonging to the 'Physics Flying Circus', what my work was all about. In particular, we usually went through a rather reproducible series of questions and answers (to be completely honest, I was often questioning myself in a similar way). Below, I would like to go through these questions once more:

#### What are you working on?

Once this question has been formulated, I take a deep breath, I see all my life passing before my eyes like in a movie, and then I start talking. In the simplest possible way. After a little while we all feel we are dealing with a major communication problem here. With hindsight, my interrogators realize this was not such a clever question, in the first place. After all, what I am exactly doing is not so important to them. The essential point is:

#### Is it useful?

With this question people usually mean whether the things I am doing, independently of how am I doing them, will eventually result in any practical application that could improve the quality of life. Unfortunately, I have to answer that I do not think so. Not even in the very far future. But I do not miss the opportunity to stress how interesting these experimental studies are from the point of view of 'fundamental research'. However, the disappointment at this point is usually so deep that people reply:

#### That's what we are paying you for?!

You can imagine that this could easily be the end of our conversation and, probably, of my job: People could completely disagree with the use the government is making of their tax contributions and decide not to finance academic activities any longer. Fortunately, this is usually not the case, at least not in The Netherlands. About Italy, I am not that sure.

#### Stellingen

# behorende bij het proefschrift Optical Spectroscopy of Quantum Spin Systems van Andrea Damascelli

- 1. In the transition-metal mono-silicides vibrational degrees of freedom are coupled to low-energy electron-hole excitations: "Charged Phonons".

  Chapter 2 of this thesis
- 2. Spin-flip excitations can carry a finite electric dipole moment: "Charged Magnons". Chapter 4 and 5 of this thesis
- 3. In reflectivity spectra an optical phonon does not always have to look like a peak.
- 4. The nonexistence of phonon softening at  $T_{\rm SP}$  in CuGeO<sub>3</sub> is an experimental fact. M. Braden et al., Phys. Rev. Lett. 80, 3634 (1998)
- 5. The expression 'this is an experimental fact' means a lot of work for theoreticians.
- 6. Fluid dynamics can be successfully applied to the study of plant organ growth.
- 7. Leonardo da Vinci said: 'It is not enough to believe what you see, you must also understand what you see'. For completeness, I would personally like to add: '...But you will never understand what you see, if you do not believe it first'.
- 8.  $\tau \delta \delta \rho \tilde{\alpha} \nu \pi \alpha \theta \tilde{\epsilon} \iota \nu$ : To act implies suffering.  $\pi \tilde{\alpha} \theta \epsilon \iota \mu \dot{\alpha} \theta o \varsigma$ : Suffering leads to knowledge. Aeschylus
- 9. The water cycle is well known to everybody: Water falls on the earth in form of rain, snow or hail. It collects in streams, lakes and rivers, finally reaching the ocean. This water then evaporates forming clouds and, eventually, falls again and the cycle repeats. What is not known to the majority of the people is that 'most' of this water falls on The Netherlands.
- 10. The tradition of Sinterklaas is wonderful for kids. But what an exhausting job for the Zwarte Pieten!