Appendix A: Description of electron scattering

The electron scattering inside the bulk is determined by the total mean free path :

$$\frac{1}{\lambda_{tot}} = \frac{1}{\lambda_{el}} + \frac{1}{\lambda_{el}} \tag{1}$$

The probability p(s) that an electron scatters in the solid at a distance s from it's current position is given by:

$$p(s) = \frac{1}{\lambda_{tot}} e^{-\frac{s}{\lambda}} \tag{2}$$

Consequently using the inversion cumulative distribution function, s following the statistics given by the above equation can be computed from a uniform probability distribution P in the interval [0,1]:

$$s = -\lambda_{tot} \log(1 - P) \tag{3}$$

Using the velocity v of the electron this can also be converted to a scattering time $\tau = s/v$. Note that this formulation makes the assumption that the electron velocity is constant. For the electrons inside solids in an attosecond streaking experiment this is only approximately true. If the electron has crossed the surface before travelling the distance s, no bulk scattering occurs. Otherwise the probability of an elastic or inelastic scattering event, P_{el} and P_{inel} can be computed from the ratios of the inverse mean free paths:

$$P_{el} = \frac{\lambda_{inel}}{\lambda_{el} + \lambda_{inel}} \tag{4}$$

Using again a uniform random number distribution P, an elastic scattering event occurs if $P < P_{el}$ else an inelastic scattering event occurs.

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BestMasters, DOI 10.1007/978-3-658-13713-7

All the other distribution functions of elastic and inelastic scattering follow more or less complex curves and the inverse cumulative distribution function is not known analytically. Sampling the distributions function at closely spaced positions the cumulative distribution function can numerically be calculated. Inversion is then achieved by switching position vector and cumulative distribution function. Again using linear interpolation the inverse cumulative distribution function can now be calculated. For multidimensional distribution functions, the dimension is subsequently reduced by calculating conditional inverse cumulative distribution functions. For a given random variable the conditional inverse cumulative distribution function is then calculated by multidimensional linear interpolation. In this approach only a single random number has to be generated per dimension involved in the distribution function however at the price of increased memory requirements. However due to the smooth change of the distribution functions involved in the description of scattering in solids a database of a few 100 Mb yielded a good sampling of the distribution functions. For an inelastic scattering event using a uniform random number distribution first the energy loss Q is calculated and then for the given Q the scattering angle θ. Only the scattering angle θ has to be calculated in an elastic scattering event.

When the electron reaches the surface it has to cross the surface barrier and may undergo surface scattering. According to the description in chapter 3 the surface scattering is symmetrically separated into a part before and after the surface transmission. Due to the Poissonian statistics of the surface scattering the total SEP therefore has to be divided by a factor of 2. If the electron undergoes scattering, one scattering event is calculated. Then for the new angle and energy this procedure is maximally repeated 5 times until no scattering occurs. Then the electron is diffracted at at surface barrier and weighted by the transmission factor. Finally the other part of the surface scattering is calculated analogously to the first part.

The splitting of the surface scattering into two equal parts is questionable, but it was found to be more consistent when including the transmission. Also the possibility to switch off the surface scattering is implemented.

Bibliography

[1] Herb Sutter. The free lunch is over: A fundamental turn toward concurrency in software. *Dr. Dobb's Journal*, 30, 205. graph updated in 2009 taken from: "http://www.gotw.ca/publications/concurrency-ddj.htm"; last accessed 08.06.2014 Online.

[2] Ferenc Krausz and Mark I. Stockman. Attosecond metrology: from electron capture to future signal processing. *Nature Photonics*, 8:205–213, 2014.

[3] E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, and F. Krausz. Attosecond control and measurement: Lightwave electronics. *Science*, 317:769–775, 2007.

[4] Martin Schultze, Elisabeth M. Bothschafter, Annkatrin Sommer, Simon Holzner, Wolfgang Schweinberger, Markus Fiess, Michael Hofstetter, Reinhard Kienberger, Vadym Apalkov, Vladislav S. Yakovlev, Mark I. Stockman, and Ferenc Krausz. Controlling dielectrics with the electric field of light. *Nature*, 493:75–78, 2013.

[5] Agustin Schiffrin, Tim Paasch-Colberg, Nicholas Karpowicz, Vadym Apalkov, Daniel Gerster, Sascha Mühlbrandt, Michael Korbman, Joachim Reichert, Martin Schultze, Simon Holzner, Johannes V. Barth, Reinhard Kienberger, Ralph Ernstorfer, Vladislav S. Yakovlev, Mark I. Stockman, and Ferenc Krausz. Optical-field-induced current in dielectrics. *Nature*, 493:70–74, 2013.

J. Schötz, *Attosecond Experiments on Plasmonic Nanostructures*,
BestMasters, DOI 10.1007/978-3-658-13713-7

[6] Mark I. Stockman. Nanoplasmonics: past, present, and glimpse into future. *Optics Express*, 19:22029, 2011.

[7] Stefan A. Maier. *Plasmonics: Fundamentals and Applications*. Springer, New York, 2007.

[8] Mark I. Stockman. Nanoplasmonics: The physics behind the applications. *Phys. Today*, 64:3944, 2011.

[9] Markus Schenk, Michael Krüger, and Peter Hommelhoff. Strong-field above-threshold photoemission from sharp metal tips. *Review of Scientific Instruments*, 82:026101, 2011.

[10] C. Ropers, C. C. Neacsu, T. Elsaesser, M. Albrecht, M. B. Raschke, and C. Lienau. Grating coupling of surface plasmons onto metallic tips: a nano-confined light source. *Nano Lett.*, 7:2784–2789, 2007.

[11] S. Berweger, J. M. Atkin, X. G. Xu, R. L. Olmon, and M. B. Raschke. Femtosecond nanofocusing with full optical waveform control. *Nano Lett.*, 11:4309–4313, 2011.

[12] F. De Angelis, G. Das, P. Candeloro, M. Patrini M. Galli, A. Bek, M. Lazzarino, I. Maksymov, C. Liberale, and L. C. Andreani and. Di Fabrizio. Nanoscale chemical mapping using three-dimensional adiabatic compression of surface plasmon polaritons. *Nature Nanotechnology*, 5:67–72, 2010.

[13] Diyar Sadiq, Javid Shirdel, Jae Sung Lee, Elena Selishcheva, Namkyoo Park, , and Christoph Lienau. Adiabatic nanofocusing scattering-type optical nanoscopy of individual gold nanoparticles. *Nano Lett.*, 11:16091613, 2011.

[14] Catalin C. Neacsu, Samuel Berweger, Robert L. Olmon, Laxmikant V. Saraf, Claus Ropers, and Markus B. Raschke. Near-field localization in plasmonic superfocusing: A nanoemitter on a tip. *Nano Lett.*, 10:592–596, 2010.

[15] M. Krüger, M. Schenk, and P. Hommelhoff. Attosecond control of electrons emitted from a nanoscale metal tip. *Nature*, 475:78–81, 2011.

[16] G. Wachter, Ch. Lemell, J. Burgdörfer, M. Schenk, M. Krüger, and P. Hommelhoff. Electron rescattering at metal nanotips induced by ultrashort laser pulses. *Phys. Rev. B*, 86:035402, 2012.

[17] Björn Piglosiewicz, Slawa Schmidt, Doo Jae Park, Jan Vogelsang, PetraGroß, Cristian Manzoni, Paolo Farinello, Giulio Cerullo, and Christoph Lienau. Carrier-envelope phase effects on the strong-field photoemission of electrons from metallic nanostructures. *Nature Photonics*, 8:37–42, 2014.

[18] J. Itatani, F. Quere, G. L. Yudin, M. Yu. Ivanov, F. Krausz, and P. B. Corkum. Attosecond streak camera. *Physical Review Letters*, 88:173903, 2002.

[19] Markus Kitzler, Nenad Milosevic, Armin Scrinzi, Ferenc Krausz, and Thomas Brabec. Quantum theory of attosecond xuv pulse measurement by laser dressed photoionization. *Physical Review Letters*, 88:173904, 2002.

[20] R. Kienberger, E. Goulielmakis1, M. Uiberacker, A. Baltuska, V. Yakovlev1, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz. Atomic transient recorder. *Nature*, 427:817–821, 2004.

[21] M. Schultze, M. Fieß, N. Karpowicz, J. Gagnon, M. Korbman, M. Hofstetter, S. Neppl, A. L. Cavalieri, Y. Komninos, Th. Mercouris, C. A. Nicolaides, R. Pazourek, S. Nagele, J. Feist, J. Burgdrfer, A. M. Azzeer, R. Ernstorfer, R. Kienberger, U. Kleineberg, E. Goulielmakis, F. Krausz, and V. S. Yakovlev. Delay in photoemission. *Science*, 328:1658–1662, 2010.

[22] A. L. Cavalieri, N. M üller, T. Uphues, V. S. Yakovlev, A. Baltuska, B. Horvath, B. Schmidt, L. Blümel, R. Holzwarth, S. Hen-

del, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann. Attosecond spectroscopy in condensed matter. *Nature*, 449:1029–1032, 2007.

[23] A. Cavalieri, F. Krausz, R. Ernstorfer, R. Kienberger, P. Feulner, J. Barth, and D. Menzel. *Attosecond Time-Resolved Spectroscopy at Surfaces. In Dynamics at Solid State Surfaces and Interfaces.* 2010.

[24] S. Neppl, R. Ernstorfer, E. M. Bothschafter, A. L. Cavalieri, D. Menzel, J. Barth, J. V., F.Krausz, R. Kienberger, and P. Feulner. Attosecond time-resolved photoemission from core and valence states of magnesium. *Physical Review Letters*, 109:087401, 2012.

[25] Frederik Süßmann and Matthis F. Kling. Attosecond nanoplasmonic streaking of localized fields near metal nanospheres. *Phys. Rev. B*, 84:241–248, 2011.

[26] E. Skopalova, D. Y. Lei, T. Witting, C. Arrell, F. Frank, Y. Sonnefraud, S. A. Maier, J. W. G. Tisch, and J. P. Marangos. Numerical simulation of attosecond nanoplasmonic streaking. *New J. Phys.*, 13:083003, 2011.

[27] F. Kelkenberg, A. F. Koenderink, and M. J. J. Vrakking. Attosecond streaking in a nano-plasmonic field. *New J. Phys.*, 14:093034, 2012.

[28] A. G. Borisov, P. M. Echenique, and A. K. Kazansky. Attostreaking with metallic nano-objects. *New J. Phys.*, 14:23036, 2012.

[29] James S. Prell, Lauren J. Borja, Daniel M. Neumark, and Stephen R. Leone. Simulation of attosecond-resolved imaging of the plasmon electric field in metallic nanoparticles. *Ann. Phys.*, 535:151–161, 2013.

[30] Mark I. Stockman, Matthias F. Kling, Ulf Kleineberg, and Ferenc Krausz. Attosecond nanoplasmonic-field microscope. *Nature Photonics*, 1:539, 2007.

[31] Frederik Suessmann Michael Foerster Michael Krueger Byung-Nam Ahn Karen Wintersperger Sergey Zherebtsov Alexander Guggenmos Vladimir Pervak Alexander Kessel Sergei Trushin Abdallah Azzeer Mark Stockman Dong-Eon Kim Ferenc Krausz Peter Hommelhoff Benjamin Frg, Johannes Schoetz and Matthias Kling. Attosecond nanoscale near-field sampling. *arXiv:1508.05611v1 [physics.optics]*, 2015.

[32] Jean-Claude Diels and Wolfgang Rudolph. *Ultrashort Laser Pulse Phenomena.* Elsevier Inc., Burlington, 2006.

[33] Uwe Thumm, Qing Lao, Elisabeth Bothschafter, Frederik Süßmann, Matthias F. Kling, and Reinhard Kienberger. *Handbook of Photonics*, chapter Attosecond Physics: Attosecond Streaking Spectroscopy of Atoms and Solids. Wiley-Blackwell. to be published.

[34] Pierre Berini, Alexandre Bouhelier, Javier Garcia de Abajo, and Namkyoo Park. Focus issue on surface plasmon photonics introduction. *Optics Express*, 21:27286–27290, 2013.

[35] Oliver Benson. Assembly of hybrid photonic architectures from nanophotonic constituents. *Nature*, 480:193199, 2011.

[36] Craig F. Bohren and Donald R. Huffman. *Absorption and Scattering of Light by Small Particles.* John Wiley and Sons, New York, 1983.

[37] Michael Quinten. *Optical Properties of Nanoparticle Systems: Mie and Beyond.* Wiley-VCH Verlag, Weinheim, 2011.

[38] Zenghu Chang. *Fundamentals of Attosecond Optics.* CRC Press, Boca Raton, 2011.

[39] *Solid-State Photoemission and Related Methods: Theory and Experiment*, chapter Overview of core and valence photoemission. Wiley VCH, 2003.

[40] Friedrich Reinert and Stefan Hüfner. Photoemission spectroscopy - from early days to recent applications. *New J. Phys.*, 7:97, 2005.

[41] Stefan Neppl. *Attosecond Time-Resolved Photoemission from Surfaces and Interfaces*. PhD thesis, TU München, 2011.

[42] C. Lemell, B. Solleder, K. Tökesi, and J. Burgdörfer. Simulation of attosecond streaking of electrons emitted from a tungsten surface. *Physical Review A*, 79:062901, 2009.

[43] The electronic structure project, 2012. [Online; accessed 2-June-2014].

[44] C. Ortiz, O. Eriksson, and M. Klintenberg. Data mining and accelerated electronic structure theory as a tool in the search for new functional materials. *Comput. Mater. Sci.*, 44:1042–1049, 2009.

[45] Frederik Süßmann. *Attosecond dynamics of nano-localized light fields*. PhD thesis, LMU München, 2013.

[46] Andrius Baltuska, Matthias Uiberacker, Eleftherios Goulielmakis, Reinhard Kienberger, Vladislav S. Yakovlev, Thomas Udem, Theodor W. Hnsch, , and Ferenc Krausz. Phase-controlled amplification of few-cycle laser pulses. *IEEE JOURNAL OF SELECTED TOPICS IN QUANTUM ELECTRONICS*, 9:972–989, 2003.

[47] Albert Messiah. *Quantum Mechanics*. Interscience Publishers, New York, 1962.

[48] Aleksander Jablonski, Francesc Salvat, and Cedric J. Powell. Comparison of electron elastic-scattering cross sections calculated from two commonly used atomic potentials. *J. Phys. Chem. Ref. Data*, 33:409–451, 2004.

[49] Francesc Salvat, Aleksander Jablonski, and Cedric J. Powell. Elsepa dirac partial-wave calculation of elastic scattering of electrons and positrons by atoms, positive ions and molecules. *Computer Physics Communications*, 165:157–190, 2005.

[50] G. Wachter, C. Lemell, J. Burgdörfer, M. Schenk, M. Krüger, and P.Hommelhoff. Electron rescattering at metal nanotips induced by ultrashort laser pulses. *Phys. Rev. B*, 86:035402–035406, 2012.

[51] Francesc Salvat. Optical-model potential for electron and positron elastic scattering by atoms. *Physical Review A*, 68, 2003.

[52] H.-J. Fitting, E. Schreiber, J.-Chr., and A. von Czarnowski. Attenuation and escape depths of low-energy electron emission. *Journal of Electron Spectroscopy and Related Phenomena*, 119:3547, 2001.

[53] *Surface and Interface Science*, chapter An Introduction to the Theory of Crystalline Elemental Solids and their Surfaces. Wiley VCH, 2012.

[54] Wolfgang S. M. Werner and Francesc Salvat-Pujol. Surface excitations in electron spectroscopy. part i: dielectric formalism and monte carlo algorithm. *Surface Interface Analysis*, 45:873–894, 2012.

[55] D. Emfietzoglou, I. Kyriakou1, I. Abril, R. Garcia-Molina, and H. Nikjoo. Inelastic scattering of low-energy electrons in liquid water computed from optical-data models of the bethe surface. *International Journal of Radiation Biology*, 88:22–28, 2012.

[56] Wolfgang S. M. Werner, Kathrin Glantschnig, and Claudia Ambrosch-Draxl. Optical constants and inelastic electron scattering data for 17 elemental metals. *Journal of Physics: Chem. Ref. Data*, 38:1013–1092, 2009.

[57] N. D. Mermin. Lindhard dielectric function in the relaxation -time approximation. *Physical Rev. B*, 1:2362–2363, 1970.

[58] V. Cataudella, V. Marigliano Ramagua, and G.P. Zucchelli. On the analytical struture of the lindhard dielectric function. *Physics Letters*, 92A:359–362, 1982.

[59] N. D. Mermin. Lindhard dielectric function in the relaxation-time approximation. *Physical Review B*, 1:2362–2363, 1970.

[60] Z.-J. Ding and R. Shimizu. Inelastic collisions of kev electrons in solids. *Surface Science*, 222:313–331, 1989.

[61] D. Emfietzoglou, I. Abril, R. Garcia-Molina, I.D. Petsalakis, H. Nikjoo, I. Kyriakou, and A. Pathak. Semi-empirical dielectric descriptions of the bethe surface of the valence bands of condensed water. *Nuclear Instruments and Methods in Physics Research B*, 266:11541161, 2008.

[62] Edward D. Palik. *Handbook of Optical Constants of Solids.* Academic Press, Boston, 1985.

[63] C. J. Tung, Y. F. Chen, C. M. Kwei, and T. L. Chou. Differential cross sections for plasmon excitations and reflected electron-energy-loss spectra. *Physical Review B*, 49:16684–16693, 1994.

[64] J.-Ch. Kuhr and H.-J. Fitting. Monte-carlo simulation of low energy electron scattering in solids. *phys. stat. sol.*, 172:433–449, 1999.

[65] D. Emfietzoglou, I. Kyriakou, I. Abril, R. Garcia-Molina, I.D. Petsalakis, H. Nikjoo, and A. Pathak. Electron inelastic mean free paths in biological matter based on dielectric theory and local-field corrections. *Nuclear Instruments and Methods in Physics Research B*, 267:4552, 2009.

[66] J.-Ch. Kuhr and H.-J. Fitting. Monte carlo simulation of electron emission from solids. *J. Phys. Chem. Ref. Data*, 28:19–61, 1999.

[67] C. J. Powell and A. Jablonski. Nist electron inelastic-mean-free-path database. Technical report, National Institute of Standards and Technology NIST, 2010.

[68] Dimitris Emfietzoglou, Ioanna Kyriakou, Rafael Garcia-Molina, and Isabel Abril. The effect of static many-body local-field corrections to inelastic electron scattering in condensed media. *Journal of Applied Physics*, 114:144907, 2013.

[69] Michael Bosman et al. Surface plasmon damping quantified with an electron nanoprobe. *Scientific Reports*, 3:1312, 2013.

[70] F. J. Garcia de Abajo. Optical excitations in electron microscopy. *Rev. Mod. Phys.*, 82:209, 2010.

[71] Wolfgang S. M. Werner, Werner Smekal, and Francesc Salvat-Pujol. Angular dependence of electron induced surface plasmon excitation. *Applied Physics Letters*, 98, 2011.

[72] Michael Krüger. *Attosecond Physics in Strong-Field Photoemission from Metal Nanotips.* PhD thesis, LMU München, 2013.

[73] Lumerical solutions, inc.

[74] P. B. Johnson and R. W. Christy. Optical constants of the noble metals. *Phys. Rev. B*, 6:4370–4379, 1972.

[75] Sebastian Thomas, Michael Krüger, Michael Förster, Markus Schenk, and Peter Hommelhoff. Probing of optical near-fields by electron rescattering on the 1 nm scale. *Nano Lett.*, 13:47904794, 2013.

[76] Loup Verlet. "computer "experiments" on classical fluids. i. thermodynamical properties of lennardjones molecules". *Physical Review*, 159:98–103, 1967.

[77] J. Schäfer, S.-C. Lee, and A. Kienle. Calculation of the near fields for the scattering of electromagnetic waves by multiple infinite cylinders at perpendicular incidence. *J. Quant. Spectrosc. Radiat. Trans.*, 113, 2012.

[78] C. H. Zhang and U. Thumm. Probing dielectric-response effects with attosecond time-resolved streaked photoelectron spectroscopy of metal surfaces. *Physical Review A*, 84:063403, 2011.

[79] M. Moskovits. surface-enhanced spectroscopy. *Reviews of Modern Physics*, 57:783–826, 1985.

[80] Yuda Zhao, Xin Liu, Dang Yuan Lei, and Yang Chai. Effects of surface roughness of ag thin films on surface-enhanced raman spectroscopy of graphene: spatial nonlocality and physisorption strain. *Nanoscale*, 6:1311–1317, 2014.