2738
doi:10.1017/S1431927622010303
Microsc. Microanal. 28 (Suppl 1), 2022

The Beauty and Clarity of a Well Designed Experiment

P.E. Batson

Department of Physics and Astronomy, Rutgers University, NJ, USA

During my graduate work at Cornell in the 1970's, it became very clear to me that Electron Energy Loss Spectroscopy provided a fascinating look into the detailed behavior of nanoscale materials. At the time, however, there were very few instruments that were capable of obtaining reproducible and detailed spectroscopic information, because of the lack of adequate spatial and energy resolution, as well as methods for recording data with sufficient statistical accuracy. My thesis research was, therefore, heavily involved with implementation of computer control for the Wien Filter electron spectrometer recently built there by Curtis and Silcox.[1] As I completed that work and began to take data, I became aware of the work of John Spence on the energy loss spectroscopy of aluminum during his graduate years in Melbourne, particularly regarding his discovery of excess multiple scattering at the two plasmon energy loss.[2] I also noticed this in my data, [3, 4] and so I was very interested to meet him at the second Cornell EELS meeting in 1978. I was impressed with his clarity, excitement, and vision of what might be obtained using the new STEM imaging he was exploring with John Cowley. He did not disappoint me throughout his career.

In this presentation, I will discuss some results that I have obtained over the years using EELS, beginning with my only joint publication with John. After spending several years building a new spectrometer design for the VG HB-501 STEM at IBM,[5] I was having trouble reproducing results, even with the new Wien Filter design, because small variations in the microscope and field emission tip conditions created shifts and shape changes in the Zero Loss that were hard to control at the level needed for measurement of electronic structure at the Si L2,3 edge. John pulled an incomplete manuscript from his desk discussing D.W. Johnson's work with deconvolution using an asymmetric zero loss. It still required hard work, but pretty soon my system routinely produced 0.2 eV resolution data with 20meV accuracy, using a 0.3-0.4 eV wide Zero Loss.[6]

An important understanding about EELS using a relativistic probe is the fact that the keV probe electron is accompanied by a hydrodynamic wake consisting of specimen charge moving in response to the passage of the relativistic electron.[7, 8, 9] This hydrodynamic wake response reflects the local charge disturbance that we call phonons, interband transitions, plasmons, single electron scattering, and core losses, among many others at higher and lower energies. It is anisotropic, extended in the longitudinal direction by the various excitations, and in the perpendicular directions by the Thomas-Fermi screening length, and it therefore modifies the applied coulomb potential in an anisotropic way. This behavior controls details of Drude damping and quantum confinement in nanoscale structures, [10, 11, 12] and significantly modifies the core hole exciton distortion at core edges. [13, 14]

The close correlation of charge density fluctuations with the relativistic electron motion leads inexorably to the question of coherency in EELS. In other words, does a swift electron scattered wavefunction share a coherent relationship with the electron wavefunction before scattering. Fermi's Golden Rule seems to beg this question, because both the swift electron and the specimen excitations are described by unconstrained sums of plane waves. High resolution microscopy has a similar problem. During both processes, momentum is transferred, and the question becomes, how random is the momentum transfer.

I will describe a couple experiments that argue for preservation of phase information during inelastic scattering, with limitations similar to high resolution imaging. [15, 16]

In 1999, the Nion aberration corrector came to town, improving the STEM spatial resolution from about 2.2˚A to 0.8˚A. [17] This small change was a huge step in a field where the silicon column spacings can range from 0.78˚A to 1.35˚A. It also revealed a wealth of kinetic behavior of atoms and nanoscale structures under the electron beam. This behavior included plasmonic forces driving coalescence in particle dimers, [18] and a completely unexpected force reversal from attractive to repulsive for very close encounters between the probe and a nanoparticle, apparently caused by a diamagnetic-like interaction with the magnetic field accompanying the swift electron. [19] This activity is fascinating and touches ongoing disagreements in the physics community concerning the sharing of momentum between an electromagnetic field and a materials structure during the scattering process.

With the introduction of the Nion Co. Hermes high resolution monochromator, sub-10 meV resolution EELS has become a reality for electron microscopy.[20] This has enabled an IR-like spectroscopy in aloof scattering,[21] and experiment and theory of bulk and surface phonon polariton modes in a single MgO nanocube.[22, 23] However, below about 50 meV, where the phonon system must be treated with a scattering theory at finite temperature, our understanding is still developing.[24] There, we expect boson occupation statistics to affect the observed EELS results, giving both energy gain, and modification of energy loss results. There turn out to be three kinds of scattering events: 1) spontaneous emission of a collective phonon by the probe, an energy loss process, 2) an induced emission by occupied phonon modes, also an energy loss process, and 3) an induced absorption, or electron energy gain. I will sketch a procedure for obtaining the total phonon density of states, appropriate for electron scattering.

I chose these observations for this discussion, because they remind me of the depth that is needed to get to the elegance of underlying physical principles. It is this depth of treatment that I admired in John Spence, that and always his sense of humor and clarity.[25]

References:

[1] G.H. Curtis and J. Silcox, Rev. Sci. Inst., **42** (1969), p. 630.
[2] J. Spence and A. Spargo, Phys. Rev. Lett., **26** (1971), p. 895.
[3] P.E. Batson, C.H. Chen and J. Silcox, Phys. Rev. Lett., **37** (1976), p. 937.
[4] P.E. Batson and J. Silcox, Phys. Rev. B, **27** (1983), p. 5224.
[5] P.E. Batson, Rev. Sci. Inst., **57** (1986), p. 43.
[6] P.E. Batson, D.W. Johnson and J.C.H. Spence, Ultram., **41** (1992), p. 137.
[7] P. Echenique, R. Ritchie and W. Brandt, Phys. Rev. B, **20** (1979), p. 2567.
[8] F.J. Abajo and P.M. Echenique, Phys. Rev. B, **46** (1992), p. 2663.
[9] P.E. Batson, Ultram., **47** (1992), p. 133.
[10] P.E. Batson, Sol. St. Comm., **34** (1980), p. 477.
[11] P.E. Batson, Ultram, **50** (1993), p. 1.
[12] P.E. Batson and J.R. Heath, Phys. Rev. Lett., **71** (1993), p. 911.
[13] P.E. Batson and J. Bruley, Phys. Rev. Lett., **67** (1991), p. 350.
[14] P.E. Batson, Phys. Rev. B, **47** (1993), p. 6898.
[15] P.E. Batson, Phys. Rev. Lett., **70** (1993), p. 1822.

https://doi.org/10.1017/S1431927622010303 Published online by Cambridge University Press

[16] P.E. Batson, Micron, **39** (2008), p. 648.
[17] P.E. Batson, N. Dellby and O.L. Krivanek, Nature, **418** (2002), p. 617.
[18] P.E. Batson, A. Reyes-Coronado, R. Barrera, A. Rivacoba, P. Echenique and J. Aizpurua, NanoLett., **11** (2011), p. 3388.
[19] M.J. Lagos, A. Reyes-Coronado, A. Konečná, P.M. Echenique, J. Aizpurua and P.E. Batson, Phys. Rev. B, **93** (2016), p. 205440.
[20] O.L. Krivanek, T.C. Lovejoy, N. Dellby, T. Aoki, R.W. Carpenter, P. Rez, E. Soignard, J. Zhu, P.E. Batson, M.J. Lagos, R.F. Egerton and P.A. Crozier, Nature, **514** (2014), p. 209.
[21] P. Rez, T. Aoki, K. March, D. Gur, O.L. Krivanek, N. Dellby, T.C. Lovejoy, S.G. Wolf and H. Cohen, Nature Comm., **7** (2016), p. 10945.
[22] M.J. Lagos, A. Trügler, U. Hohenester and P.E. Batson, Nature, **543** (2017), p. 529.
[23] U. Hohenester, A. Trügler, P.E. Batson and M.J. Lagos, Phys. Rev. B, **97** (2018), p. 165418.
[24] D. Pines and P. Nozieres, W. A. Benjamin (1966).
[25] Work supported by the Basic Energy Sciences Division of the Department of Energy, Award #DE-SC0005132.