



Leibniz Institute for Solid State and Materials Research Dresden

Electron Holography

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Converting phase shifts to contrasts: Fresnel imaging







Fresnel imaging: Pros & Cons

Pro:

- simple
- fast
- sensitivity adjustable

Con:

- (partially) non-linear contrast
- defocus \rightarrow unsharp images
- quantification difficult (but possible)
- sensitiv to dynamical scattering

Can be overcome by Holography! (now)

Recommended reading:

 Völkl, Edgar, Allard, Lawrence F., Joy, David C. (Eds.), Introduction to Electron Holography, Springer (1999).





- 1. Fundamentals of electron scattering
 - a. Axial scattering
 - b. Magnetic and electric Ehrenberg–Siday–Aharonov–Bohm effect

2. Fundamentals of Electron Holography and Tomography

- a. Holographic Principle (interference, reconstruction)
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How do fields act on electrons waves? *



paraxial approximation

no backscattering

small-angle scattering

 $\Psi = e^{ik_z z} \psi$

$$E\Psi = \left[\frac{\hat{p}^2}{2m} - eV\right]\Psi$$

kinetic momentum operator

 $\hat{p} = -i\hbar \nabla + eA$

$$\begin{aligned} -2k_{z}\hbar\hat{p}_{z}\psi &= \left[\hat{p}_{\perp}^{2} - 2meV\right]\psi\\ \partial_{z}\psi &\approx i\left[\frac{-\hat{p}_{\perp}^{2}}{2\hbar^{2}k_{z}} + \sigma V - \frac{e}{\hbar}A_{z}\right]\psi\end{aligned}$$

 \cong 2D time-dependent Schrödinger equation

axial approximation (wavelength << object details)

very small angle scattering

$$\partial_z \psi \approx i \left[\sigma V - \frac{e}{\hbar} A_z \right] \psi$$
$$\psi = e^{i\varphi} \psi_0 \rightarrow \varphi = \int_{\text{object}} \left(\frac{e}{\hbar v} V - \frac{e}{\hbar} A_z \right) dz$$



* It is a good exercise to do derivation by yourself.



Phase shift by electric potential



$$\varphi = k \left[\int_{s_2 - s_1} n ds \right] = \frac{e}{\hbar v} \int_{\text{object}} V dz$$

refractive index





Detectable phase shift *

$$\Delta \varphi = \sigma \int_{s_2 - s_1} V \, ds$$

electric
$$\int \phi$$

$$\Delta \varphi = \sigma \left(V_{p,1} - V_{p,2} \right)$$





* Why can we only detect phase differences?



Detectable phase shift















Ehrenberg - Siday – Aharonov - Bohm Effect Proposal: Ehrenberg & Siday 1949 Aharonov & Bohm 1958



Experiment: Möllenstedt & Bayh 1962







Magnetic phase shift







Summary: object exit wave







Summary: object exit wave

phase modulation $\varphi(x, y)$:

micro-/nanofields

- electric
- magnetic

amplitude modulation a(x, y):

- scattering into large angles
- interference effects
- inelastic scattering





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Dennis Gabor



Easter 1947, on the tennis court:

... and all of sudden it came to me, without any effort on my side.

Interference and diffraction are mutually inverse

1902-1979 Nobel Prize 1971

Electron Holography measures phases





Dennis Gabor

Holography

Object wave







Common Forms of Electron Holography





J.M. Cowley, 20 forms of holography, Ultramicroscopy 41 (1992), 335-348



Holography - Dennis Gabor's idea







Holography - Dennis Gabor's idea



Holography - reconstruction of wave



Holography: basic scheme







Holography: recording hologram



$$hol = (\psi + r)(\psi + r)^*$$
$$= \psi\psi^* + rr^* + \psi r^* + \psi^* r$$





Holography: reconstruction of wave







Holography: reconstruction of wave







Plane reference wave r





Where to take the hologram ?

- **Object plane**
- Fresnel region
- Fraunhofer region
- Fourier plane

In principle: "where" is not essential, but with electrons we are "coherency-limited"





Where to take the hologram ?

Inline Holography

Scattering Regimes

Illumination
$$k = 2\pi / \lambda$$

Fresnel (near field)

Fraunhofer (far field)



Reconstruction Schemes

Differential Defocus / Transport of Intensity Reconstruction

Defocus Series Reconstruction

Fraunhofer Holography



Figure from Lee, Optics Express Vol. 15, Issue 26, pp. 18275-18282 (2007)



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Transport of Intensity Reconstruction

Paraxial Eq.

$$\frac{\partial \Psi(\mathbf{r}_{\perp}, z)}{\partial z} = \frac{i}{2k} \Delta_{\perp} \Psi(\mathbf{r}_{\perp}, z)$$

Continuity Eq. / Transport of Intensity Eq.

density / intensity

$$\rho \equiv \left| \Psi \right|^2$$

experimental data from 2 slightly defocussed images

$$\frac{\overline{\rho(z+\delta z), \rho(z-\delta z)}}{\rho(z+\delta z), \rho(z-\delta z)} + O(\delta z^{2})$$

$$\rho(z) = \frac{\rho(z+\delta z) + \rho(z-\delta z)}{2} + O(\delta z^{2})$$



Transport of Intensity Reconstruction



TIE: Pros & Cons

Pro:

- linear signal
- simple reconstruction
- simple experiment
- no external reference / vacuum required
- works at moderate coherency

Con:

- not so fast (2 recordings)
- not sensitiv to small spatial frequencies (large scale variations)
- ambiguous result (because of unknown boundary conditions)











iterate over initial waves to find Φ with min R_{Φ}

 $\tilde{\psi}$

 $R^{(n+1)} - R^{(n)}$

Focal Series Reconstruction

Experimental focus series Reconstruction of B-Field







Focal Series: Pros & Cons

Pro:

- sensitiv to smaller (but still not very small) spatial frequencies
- works at every TEM
- no external reference / vacuum required

Con:

- very slow
- ambiguous result (depending on starting guess)
- complicated reconstruction




Biprism-Holder







Biprism-Holder









Off-axis electron holography

Hologram







Magnetic phase shift in Cobalt stripe domains

Amplitude image





$$\frac{\partial \varphi_{\text{mag}}}{\partial x} = -\frac{e}{\hbar} \int B_y(x, y, z) \, \mathrm{d}z$$

Projected B-field







Electric and magnetic phase shift







Electric and magnetic phase shift





Fernandez-Pacheco, A. et al., Nat Commun 2017, 8, 15756.

Sample provided by Denys Makarov, Helmholtz-Zentrum Dresden-Rossendorf.



Liquid Helium Cryostage



FIG. 4. Cross section of electron microscope with cooling apparatus. A, field emission gun; B, liquid-He reservoir; C, cooling stage; D, ion pumps; E, biprism; F, condenser lens; G, objective lens; H, intermediate lenses; I, projector lenses.



FIG. 5. Cross section of cooling stage. A, second shield; B, first shield; C, specimen holder; D, conducting rods; E, heater; F, objective pole-piece; G, superconducting coil; H, heater; I, Ge resistor; J, insulating supports; K, specimen.





Superconductivity: Vortex lattice



Nb-film T=4.5K < Tc=9.2K B=15 mT (150 Gauss) Phase amplification 16*



J.E. Bonevich, K. Harada, T. Matsuda, H. Kasai, T. Yoshida, G. Pozzi and A. Tonomura, Phys.Rev.Letters, 70 (1993), 2952



Off-axis: Pros & Cons

Pro:

- linear signal
- simple reconstruction
- unambiguous result
- sensitiv to the whole spatial frequency range

Con:

- (multiple) biprisms required
- reference (vacuum) required
- large coherency requirements





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3. Magnetic fields and textures in solids

- a. Magnetization, Magnetic induction, Magnetic field
- b. Magnetostatics
- c. Micromagnetics





Separation of magnetic and electric phase shift *



 $\varphi_1(x, y) = \varphi_{el}(x, y) + \varphi_{mag}(x, y)$

$$\varphi_2(x,y) = \varphi_{el}(x,y) - \varphi_{mag}(x,y)$$

$$\varphi_{mag}(x, y) = (\varphi_1(x, y) - \varphi_2(x, y))/2$$

$$\varphi_{el}(x, y) = (\varphi_1(x, y) + \varphi_2(x, y))/2$$





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Separation of magnetic and electric phase shift

$\varphi_{mag} = (\varphi_1 - \varphi_2)/2$



$$\varphi_{el} = (\varphi_1 + \varphi_2)/2$$



 $\begin{pmatrix} \partial_x \varphi_{mag} \\ \partial_v \varphi_{mag} \end{pmatrix} = -\frac{e}{\hbar} \int \begin{pmatrix} B_y(x, y, z) \\ -B_x(x, y, z) \end{pmatrix} dz \qquad \varphi_{el}(x, y) = C_E \int V(x, y, z) dz$

 $+\infty$





Towards 3D nanomagnetism







Towards 3D nanomagnetism



Towards 3D nanomagnetism

+ structural, chemical data

Reyes et al., Nano Lett. 16 (2016) 1230





DW in <u>50 nm</u>

Biziere et al., Nano Lett. 13 (2013) 2053

Comparison





3D modelling of M,B (eg. micromagnetic simulation)

Single tilt axis holographic tomography of nanomagnets





Wolf et al., *Chem. Mater.* **27** (2015) 6771 Simon, Wolf et al., *Nano Lett.* **16** (2016) 114

Electron holographic tomography of magnetic samples

3D reconstruction of V(x, y, z) and **B**(x, y, z)

- Tilt series acquisition of off-axis electron holograms: Two 360° tilt series around *x*- and *y*-axis (gaps due to experimental limitations)
- 2. Phase shift retrieval from electron holograms
- Separation of electric and magnetic phase shift and alignment ⁴
- 4. Tomographic reconstruction of

 $V(x, y, z) \text{ from } \varphi_{el},$ $B_x(x, y, z) \text{ from } \frac{\partial \varphi_{mag}}{\partial y},$ $B_y(x, y, z) \text{ from } \frac{\partial \varphi_{mag}}{\partial x}$

5. Computation of $B_z(x, y, z)$ from $\nabla \cdot \mathbf{B}(x, y, z) = 0$







Dual tilt axis holographic tomography of nanomagnets

Electron Holographic Tomography



Implementation

THOMAS				0 0
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Projection Compustage Eu Read Compustage Reset Compustage	sentricity Tracking Tit Seri	es Holography e Shift Claimed Precision (µm Number of Tries	01 10	Stored Positions
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Correct A-Tilt Induced Disp	lacement Calculated	Read A-Tilt Displacements	it [pm] 0	I* Blank Beam

Fine Alignment Images Till series Zn0_ts_ppot Sino from ROI Average Threshold Displacements Manual Volume Threshold Analyze sinogram Analyze x Images Apply x Reference slice Lower slice 305 Down Shift table Lower slice Round Shift table Lower slice Genut Apply x All shift tables Create Get Apply All shift tables

Reconstruct 3D image: sinogram size of backprojection: x-size 440 z-size 200 t Sino 195 Extract Tilts pos. of backprojection: x-pos 0 z-pos 0 SIRT reconstruction /alues of projection dit select nath 1.0197 -36 534394 90 12421 12.635874 edge in px 10 7 900128 38.376365 1.738393 31.823999 0.8293 shape both 💌 28 297830 -4.390504 Smooth sine olina 18

Automated tomographic tilt series acquisition

- Installed at NCEM Berkeley, U Antwerp, TU Berlin
- Adapted for different TEMs

Wolf et al. Ultramic. 110 (2010) 390

Alignment

- Displacement correction
- Tilt axis finding

Wolf et al., *Chem. Mater.* **27** (2015) 6771

"Reconstruct 3D" software package

 Documentation at <u>www.triebenberg.de/wolf</u>

Wolf et al. Ultramic. 136 (2014) 15

Challenges	Problems
Magnetic sample	 beam damage diffraction contrast stray fields magnetization by Lorentz lens





Permalloy disks provided by J. Zweck, Regensburg



E. Dunin-Borkowski and T. Kasama, Microscopy and Microanalysis 10 (2004) 10



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$\int B_y(x(\alpha), y, z(\alpha)) dz = -\frac{\hbar}{e} \frac{\partial \varphi}{\partial x(\alpha)}$ y in the direction of tilt axis	 derivation enhances noise only B_y, i.e., parallel to tilt axis



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Two ultra-high-tilt series $(\pm 90^{\circ})$ about orthogonal axes to get B_x and B_y	sample geometryholder designstability



E. Dunin-Borkowski and T. Kasama, Microscopy and Microanalysis 10 (2004) 10





Tsuneta et al., Microscopy 63 (2014) p. 469

Solution

 preparation of free-standing samples combined with special holder designs





Dual-Axis Tomography Holder Model 2040, Fischione Instruments



Multiple-axis rotation holder



IFW Forschungstechnik: Steffen Ziller, Nico Richter, Rolf Morgner





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Separation electric (MIP contribution) magnetic phase shift	 acquisition of additional two tilt series with reversed magnetization precise alignment (2D Affine transformation)





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B_z from $\nabla \cdot B = 0$ with B_x and B_y inserted	 second derivation enhances noise further unknown boundary conditions
Liibriz & E. Dunin-Borkowski and T. Kasama	a. Microscopy and Microanalysis 10 (2004) 1010

E. Dunin-Borkowski and T. Kasama, Microscopy and Microanalysis 10 (2004) 1

Vectorfield tomography of Cu/Co multi-stacked NWs: Phase diagram for a single Co-disk from micromag simulation







Vectorfield tomography of Cu/Co multi-stacked NWs: Hologram tilt series

Tilt range -69° to +72°



Rotated 90° in-plane: Tilt range -69° to +72°



+ tilt series flipped upside-down

Vectorfield tomography of Cu/Co multi-stacked NWs: Hologram tilt series

Tilt range -69° to +72°



Rotated 90° in-plane: Tilt range -69° to +72°



Vectorfield tomography of Cu/Co multi-stacked NWs: Phase tilt series

Electric phase shift



Magnetic phase shift (smoothed)



Electrostatic 3D potential of Cu/Co multi-stacked NW



 3D reconstruction from electrostatic phase shift (Average of two tilt series)







Electrostatic 3D potential of Cu/Co multi-stacked NW: Quantification







MIPs reduced due to low purity (voids); 15% Cu amount in Co





Reconstructed magnetic configurations in Cu-Co NW



Nanoscale mapping for better understanding of 3D nanomagnetism



$$M_{S} = 1200 \times 10^{3} \ A/m$$
$$A = 22 \times 10^{-12} \ J/m$$
$$H_{k} = 100 \times 10^{3} \ J/m^{3}$$





Nanoscale mapping for better understanding of 3D nanomagnetism

+ structural, chemical data

Reyes et al., Nano Lett. 16 (2016) 1230

3D reconstruction by electron holographic vector field tomography of B-fields



DW in <u>50 nm</u> <u>50 nm</u> <u>50 nm</u> CO

Biziere et al., Nano Lett. 13 (2013) 2053

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