A semi-classical theory of magnetic inelastic scattering in transmission electron energy loss spectroscopy

BG Mendis[1]

1. Dept. of Physics, Durham University, South Road, Durham, DH1 3LE, UK

Abstract

The feasibility of detecting magnetic excitations using monochromated electron energy loss spectroscopy in the transmission electron microscope is examined. Inelastic scattering cross-sections are derived using a semi-classical electrodynamic model, and applied to AC magnetic susceptibility measurements and magnon characterisation. Consideration is given to electron probes with a magnetic moment, such as vortex beams, where additional inelastic scattering can take place due to the change in magnetic potential energy of the incident electron in a non-uniform magnetic field. This so-called ‘Stern-Gerlach’ energy loss can be used to enhance the strength of the scattering by increasing the orbital angular momentum of the vortex beam, and enables separation of magnetic from non-magnetic (i.e. dielectric) energy losses, thus providing a promising experimental route for detecting magnons. AC magnetic susceptibility measurements are however not feasible using Stern-Gerlach energy losses for a vortex beam.

Keywords: magnons, AC magnetic susceptibility, vortex beams, electron energy loss spectroscopy

1. Introduction

Recent advances [1] in monochromation in the transmission electron microscope has enabled an energy resolution better than 10 meV in electron energy loss spectroscopy (EELS). Consequently, the scope of high spatial resolution EELS mapping has broadened to include phonon spectroscopy (e.g. [2-6]). This raises the intriguing possibility of similar analyses on the phonon counterpart in magnetic materials, namely magnons, which also have energies in the meV range [7-8]. These measurements are traditionally carried out using neutron diffraction, although electrons have also shown to reveal information on magnons, as well as Stoner excitations, using the technique of spin-polarised EELS (SPEELS), e.g. [9-14]. In SPEELS low energy (i.e. <10 eV) spin-polarised electrons are reflected off the specimen surface, so that the bulk properties of the material are not accessible, and furthermore the spatial resolution is poor compared to most other electron microscopy techniques. High spatial and energy resolution transmission electron microscopy could offer a suitable route to carry out these measurements within the ‘bulk’ of the specimen. Transmission EELS can also measure the material dielectric function over a frequency range much larger than optical techniques, by utilising the (broadband) electric field of the incident, high energy electron probe [15-17]. The equivalent technique for magnetic materials is AC magnetic susceptibility measurements, which cover a frequency range of up to 10^4 Hz and can therefore only examine slow processes, such as spin-lattice relaxation [18]. If the magnetic field of the incident electron is utilised in a similar manner to its electric field, then with 10 meV energy resolution the AC magnetic susceptibility can potentially be measured at 10^{12} Hz frequency using transmission EELS. This frequency range is also accessible by muon spin relaxation and neutron scattering techniques, but is still too fast to measure spin-spin relaxations, which have characteristic times of ~10^{-9}-10^{-10} s [18].

Thus far transmission EELS of magnetic materials has been limited to magnetic linear [19] and circular [20-22] dichroism measurements of core loss edges. In SPEELS magnons are detected via an exchange mechanism, where the incident and 'reflected' (actually exchanged) electrons have opposite spins, with a magnon excited in order to conserve angular momentum [14]. However, exchange mechanisms are suppressed at the high accelerating voltages used in transmission EELS (see, for example, Figure 3 in [9]). The standard energy loss mechanism in transmission EELS is due to the electric field of the incident electron interacting with the sample [23-25]. An electrodynamic calculation has shown that the total energy loss for high energy incident electrons due to magnetism is small compared to the (non-magnetic) dielectric losses [26]. Therefore, in order to detect magnetic excitations in transmission EELS it is required to look beyond the standard electric field-induced inelastic scattering mechanisms. Here the classical electrodynamic theory of energy loss for high energy charged particles is broadened to include the magnetic moment of the electron. The magnetic moment could be due to the intrinsic spin (for a spin polarised electron beam) or orbital angular momentum of electron vortex beams [27-31], and can interact with any surrounding magnetic field(s). Provided the magnetic field is spatially non-uniform along the electron trajectory there will be a force on the incident electron which results in an energy loss, or in some cases an energy gain depending on the orientation of the magnetic moment. The condition of non-uniform magnetic field is satisfied for the self-field of a moving charged particle [32] or gradient in the sample magnetisation (i.e. bound currents [7]). This new energy loss/gain mechanism is referred to as the 'Stern-Gerlach' energy loss/gain, since it is caused by the same force in the seminal Stern-Gerlach experiment used to detect electron spin [33].

In this paper the feasibility of detecting magnetic excitations via the Stern-Gerlach force in transmission EELS is theoretically evaluated. Emphasis is placed on magnon excitations and AC susceptibility measurements. Both these measurements involve a change in the specimen spin state and therefore an energy exchange with the incident electron probe. In AC magnetic susceptibility the specimen transitions into a magnetically more ordered state, while for magnon EELS there is a change in the magnon population. For completeness Section 2 covers magnetic energy losses due to the electric field of a standard, unpolarised electron beam. It is shown that such an electron probe is sensitive to AC magnetic susceptibility, but is difficult to implement in practice, since there is no convenient method for separating magnetic energy losses from the much larger dielectric losses. Magnon detection is also not possible, due to the incident electrons lacking the angular momentum required for magnon generation. In Section 3 the Stern-Gerlach inelastic scattering cross-sections due to the magnetic moment of the electron probe are derived. The discussion is limited to vortex beams, since at the time of writing it is difficult to produce spin polarised electron beams of sufficient brightness in transmission electron microscopy. Previous EELS theories of vortex beams have examined energy losses in chiral structures [34] and metal split ring resonators [35]. It is shown here that the Stern-Gerlach force for vortex beams cannot be used in AC magnetic susceptibility measurements, but nevertheless provide several advantages for detecting magnons, such as the ability to exploit the orbital angular momentum of the vortex beam to increase the strength of the magnetic scattering, as well as separating magnetic from dielectric energy losses. Vortex electron probes are proposed as the most suitable option for magnon EELS characterisation in magnetic materials.

2. Magnetic energy loss for an unpolarised electron beam

In this section magnetic energy losses for a high energy electron moving along the optic z-axis with speed v is calculated. The electron beam is not spin polarised, so that the energy loss is due only to the electric field of the incident electron. This scenario corresponds to conventional electron beams found in most transmission electron microscopes. Maxwell's first and fourth equations for an incident electron in a magnetic material are given by:

$$\vec{\nabla}\cdot\mathbf{D} = \varepsilon_0\varepsilon(\mathbf{r},t)\otimes\left(\vec{\nabla}\cdot\mathbf{E}\right) = \rho_f \quad \ldots (1a)$$

$$\vec{\nabla}\times\mathbf{B} = \mu_0\left(\mathbf{J}_f+\mathbf{J}_b\right)+\mu_0\frac{\partial\mathbf{D}}{\partial t} = \mu_0\left(\mathbf{J}_f+\mathbf{J}_b\right)+\frac{\varepsilon(\mathbf{r},t)}{c^2}\otimes\frac{\partial\mathbf{E}}{\partial t} \quad \ldots (1b)$$

where $\mathbf{D}$, $\mathbf{E}$ and $\mathbf{B}$ are the electric displacement, electric and magnetic fields respectively, ρ_f and $\mathbf{J}_f$ are the charge density and current density vector for the incident electron, ε_0 and μ_0 are the permittivity and permeability of free space, c is the speed of light and t is time. The electric displacement field is given by $\mathbf{D}(\mathbf{r},t) = \varepsilon_0\varepsilon(\mathbf{r},t)\otimes\mathbf{E}(\mathbf{r},t)$, where $\varepsilon(\mathbf{r},t)$ is the real space-time dielectric function and $\otimes$ represents a convolution operation [36]. The magnetic properties of the material are contained in the bound current term $\mathbf{J}_b = \vec{\nabla}\times\mathbf{M}$, where $\mathbf{M}$ is the magnetisation. The (retarded) $\mathbf{E}$ and $\mathbf{B}$-fields can be expressed in terms of the electric scalar (ϕ) and magnetic vector ($\mathbf{A}$) potentials, i.e. $\mathbf{E} = -\vec{\nabla}\phi-\frac{\partial\mathbf{A}}{\partial t}$ and $\mathbf{B} = \vec{\nabla}\times\mathbf{A}$. Using the vector identity $\vec{\nabla}\times\left(\vec{\nabla}\times\mathbf{A}\right) = \vec{\nabla}\left(\vec{\nabla}\cdot\mathbf{A}\right)-\nabla^2\mathbf{A}$ in Equation 1b gives:

$$\vec{\nabla}\left[\vec{\nabla}\cdot\mathbf{A}+\frac{\varepsilon(\mathbf{r},t)}{c^2}\otimes\frac{\partial\phi}{\partial t}\right]-\nabla^2\mathbf{A}+\frac{\varepsilon(\mathbf{r},t)}{c^2}\otimes\frac{\partial^2\mathbf{A}}{\partial t^2} = \mu_0\left(\mathbf{J}_f+\mathbf{J}_b\right) \quad \ldots (2)$$

The gauge for $\mathbf{A}$ is chosen so that the term within the square brackets in Equation 2 is zero. Therefore, from Equations 1a and 2 we obtain the following expressions for ϕ and $\mathbf{A}$:

$$\varepsilon\otimes\nabla^2\phi-\frac{\varepsilon}{c^2}\otimes\left(\varepsilon\otimes\frac{\partial^2\phi}{\partial t^2}\right) = -\frac{\rho_f}{\varepsilon_0} \quad \ldots (3a)$$

$$\nabla^2\mathbf{A}-\frac{\varepsilon}{c^2}\otimes\frac{\partial^2\mathbf{A}}{\partial t^2} = -\mu_0\left(\mathbf{J}_f+\mathbf{J}_b\right) \quad \ldots (3b)$$

It is convenient to take the Fourier transform of the above equations, i.e.:

$$\left[4\pi^2 q^2 - \frac{\omega^2 \varepsilon(\omega)}{c^2}\right] \tilde{\phi}(\mathbf{q}, \omega) = \frac{\tilde{\rho}_f(\mathbf{q}, \omega)}{\varepsilon_0 \varepsilon(\omega)}$$

… (4a)

$$\left[4\pi^2 q^2 - \frac{\omega^2 \varepsilon(\omega)}{c^2}\right] \tilde{\mathbf{A}}(\mathbf{q}, \omega) = \mu_0 [\tilde{\mathbf{J}}_f(\mathbf{q}, \omega) + \tilde{\mathbf{J}}_b(\mathbf{q}, \omega)]$$

… (4b)

where we adopt the convention that a function $f(\mathbf{r}, t)$ and its Fourier transform $\tilde{f}(\mathbf{q}, \omega)$ are related by:

$$\tilde{f}(\mathbf{q}, \omega) = \int f(\mathbf{r}, t) e^{-2\pi i \mathbf{q} \cdot \mathbf{r} + i\omega t} d\mathbf{r} dt$$

… (5a)

$$f(\mathbf{r}, t) = \frac{1}{2\pi} \int \tilde{f}(\mathbf{q}, \omega) e^{2\pi i \mathbf{q} \cdot \mathbf{r} - i\omega t} d\mathbf{q} d\omega$$

… (5b)

Fourier transformed variables are identified with a tilde sign. The local approximation is assumed for the dielectric function, so that its Fourier transform $\varepsilon(\omega)$ is a function of angular frequency ω only, and independent of the scattering vector $\mathbf{q}$ (the tilde sign is omitted from $\varepsilon(\omega)$ for convenience) [36]. Substituting Equations 4a and 4b in $\tilde{\mathbf{E}}(\mathbf{q}, \omega) = -2\pi i \mathbf{q} \tilde{\phi}(\mathbf{q}, \omega) + i\omega \tilde{\mathbf{A}}(\mathbf{q}, \omega)$ we get:

$$\tilde{\mathbf{E}}(\mathbf{q}, \omega) = \underbrace{i\left[-2\pi \mathbf{q} + \frac{\omega \varepsilon(\omega)}{c^2} \left(\frac{\tilde{\mathbf{J}}_f}{\tilde{\rho}_f}\right)\right] \tilde{\phi}}_{\text{dielectric}} + \underbrace{\frac{i\omega \tilde{\mathbf{J}}_b}{\varepsilon_0 c^2 \left[4\pi^2 q^2 - \frac{\omega^2 \varepsilon(\omega)}{c^2}\right]}}_{\text{magnetic}}$$

… (6)

The first and second terms in Equation 6 represent the sample dielectric and magnetic contributions to the electric field of the incident electron. The electron stopping power, *dW/dz*, is defined as being positive for energy loss events, so that:

$$\frac{dW}{dz} = eE_z(\mathbf{r}, t) = \frac{e}{2\pi} \int \tilde{E}_z(\mathbf{q}, \omega) e^{2\pi i \mathbf{q} \cdot \mathbf{r} - i\omega t} d\mathbf{q} d\omega$$

… (7)

where e is the magnitude of the electron charge and $E_z(\mathbf{r}, t)$ is the *z*-component of the electric field. From Equation 6 the magnetic contribution to the stopping power along the electron trajectory point $x, y = 0, z = vt$ is then:

$$\frac{dW_{\text{mag}}}{dz} = -\frac{e}{\varepsilon_0 c^2} \int \omega \left[\frac{q_x \tilde{M}_y(\mathbf{q}, \omega) - q_y \tilde{M}_x(\mathbf{q}, \omega)}{4\pi^2 q^2 - \frac{\omega^2 \varepsilon(\omega)}{c^2}}\right] e^{i(2\pi q_z v - \omega)t} d\mathbf{q} d\omega$$

… (8)

where we have used the result $\tilde{\mathbf{J}}_b(\mathbf{q},\omega) = 2\pi i\mathbf{q} \times \widetilde{\mathbf{M}}(\mathbf{q},\omega)$ and expressed the scattering vector in component form $\mathbf{q} = (q_x, q_y, q_z)$. In Equation 8 it is assumed that the energy loss is small, so that any changes to the speed v and electron trajectory are negligible. Consider the energy loss δW_{mag} over the time interval [-T,T]. We have from Equation 8:

$$\delta W_{\text{mag}} = \int \left(\frac{dW_{\text{mag}}}{dz}\right) dz = v \int_{-T}^{T} \frac{dW_{\text{mag}}}{dz} dt$$
$$= -\frac{2ev}{\varepsilon_0 c^2} \int \omega \left[\frac{q_x \widetilde{M}_y(\mathbf{q},\omega) - q_y \widetilde{M}_x(\mathbf{q},\omega)}{4\pi^2 q^2 - \frac{\omega^2 \varepsilon(\omega)}{c^2}}\right] \left\{\frac{\sin(2\pi q_z v - \omega)T}{(2\pi q_z v - \omega)}\right\} d\mathbf{q} d\omega \quad \ldots (9)$$

For large T the term $\left\{\frac{\sin(2\pi q_z v-\omega)T}{(2\pi q_z v-\omega)}\right\}$ approximates to a delta function centred at $q_z = \omega/2\pi v$. Therefore, only the scattering vector component $q_z = \omega/2\pi v$ contributes to the energy loss $\hbar\omega$, where $\hbar$ is Planck's reduced constant. In practice, it is not strictly correct to integrate Equation 8 over large distances of the electron trajectory, since it violates the assumption of a constant electron speed v. Nevertheless, for the small energy losses that are of interest here, the electron trajectory can be made sufficiently long so that the condition $q_z = \omega/2\pi v$ is approximately satisfied. In what follows however the condition $q_z = \omega/2\pi v$ is treated as being exact, i.e. it is assumed that the transmission EELS specimen is sufficiently thick for 'bulk'-like behaviour to be observed. For thinner specimens the condition $q_z = \omega/2\pi v$ is relaxed; in fact, Bloch wave analysis of core electron ionisation has also shown that wavevector conservation along the film thickness direction is relaxed for a thin specimen [37].

For $q_z = \omega/2\pi v$, we have $\left\{\frac{\sin(2\pi q_z v-\omega)T}{(2\pi q_z v-\omega)}\right\} = T$ and the energy loss increases linearly with distance travelled by the electron. The stopping power is therefore simply $\delta W_{\text{mag}}/(2vT)$, so that Equation 9 simplifies to:

$$\frac{dW_{\text{mag}}}{dz} = -\frac{e}{\varepsilon_0 c^2} \int \omega \left[\frac{q_x \widetilde{M}_y\left(\mathbf{q}_\perp, \frac{\omega}{2\pi v}, \omega\right) - q_y \widetilde{M}_x\left(\mathbf{q}_\perp, \frac{\omega}{2\pi v}, \omega\right)}{4\pi^2\left(q_\perp^2 + \left(\frac{\omega}{2\pi v}\right)^2\right) - \frac{\omega^2 \varepsilon(\omega)}{c^2}}\right] \delta\left(q_z - \frac{\omega}{2\pi v}\right) d\mathbf{q} d\omega \quad \ldots (10)$$

where we have expressed the scattering vector $\mathbf{q}$ as $\left(\mathbf{q}_\perp, q_z = \frac{\omega}{2\pi v}\right)$, with $\mathbf{q}_\perp$ being the component of $\mathbf{q}$ in the *xy*-plane of the specimen.

In Equation 10 the ω-integral has integration limits between -∞ to ∞, although in a typical EELS experiment only the positive frequencies are measured. Since the dielectric function and magnetisation are real, $\varepsilon(-\omega) = \varepsilon(\omega)^*$ and $\widetilde{\mathbf{M}}(-\mathbf{q},-\omega) = \widetilde{\mathbf{M}}(\mathbf{q},\omega)^*$, where the asterisk denotes the complex conjugate. This means that for any scattering vector, frequency pair $(\mathbf{q},\omega)$ and its negative $(-\mathbf{q},-\omega)$ the following relationships are valid:

$$(-\omega)(-q_x)\widetilde{M}_y\left(-\mathbf{q}_\perp,-\frac{\omega}{2\pi v},-\omega\right)=\omega q_x\widetilde{M}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)^*$$

… (11a)

$$(-\omega)(-q_y)\widetilde{M}_x\left(-\mathbf{q}_\perp,-\frac{\omega}{2\pi v},-\omega\right)=\omega q_y\widetilde{M}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)^*$$

… (11b)

Therefore, for a circular EELS aperture centred about the optic axis Equation 10 gives:

$$\frac{dW_{\mathrm{mag}}}{dz}=-\frac{e}{\varepsilon_0 c^2}\int\omega\mathrm{Re}\left[\frac{q_x\widetilde{M}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)-q_y\widetilde{M}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)}{4\pi^2\left(q_\perp^2+\left(\frac{\omega}{2\pi v}\right)^2\right)-\frac{\omega^2\varepsilon(\omega)}{c^2}}\right]\delta\left(q_z-\frac{\omega}{2\pi v}\right)d\mathbf{q}d\omega$$

… (12)

where 'Re' denotes the real part of a complex number and the limits of the ω-integral are now between 0 and ∞. Since $q_z=\omega/2\pi v$ we can express $d\mathbf{q}$ as $(d\mathbf{q}_\perp d\omega)/2\pi v$ in Equation 12:

$$\frac{dW_{\mathrm{mag}}}{dz}=-\frac{e}{2\pi\varepsilon_0 v c^2}\int\omega\mathrm{Re}\left[\frac{q_x\widetilde{M}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)-q_y\widetilde{M}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)}{4\pi^2\left(q_\perp^2+\left(\frac{\omega}{2\pi v}\right)^2\right)-\frac{\omega^2\varepsilon(\omega)}{c^2}}\right]d\mathbf{q}_\perp d^2\omega$$

… (13)

Define P_{mag} as the 'probability' of magnetic energy loss per unit path length, so that:

$$\frac{dW_{\mathrm{mag}}}{dz}=\int\hbar\omega\left(\frac{\partial^4P_{\mathrm{mag}}}{\partial\mathbf{q}_\perp\partial^2\omega}\right)d\mathbf{q}_\perp d^2\omega$$

… (14)

Comparing Equations 13 and 14 gives the differential cross-section for magnetic energy loss:

$$\frac{\partial^4P_{\mathrm{mag}}}{\partial\mathbf{q}_\perp\partial^2\omega}=-\frac{e}{2\pi\varepsilon_0\hbar v c^2}\mathrm{Re}\left[\frac{q_x\widetilde{M}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)-q_y\widetilde{M}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)}{4\pi^2\left(q_\perp^2+\left(\frac{\omega}{2\pi v}\right)^2\right)-\frac{\omega^2\varepsilon(\omega)}{c^2}}\right]$$

… (15)

Equation 15 indicates that only the curl of the magnetisation around the optic z-axis can contribute to the energy loss. Consider now AC magnetic susceptibility measurements, where the self-magnetic field of the incident electron induces a magnetisation within the sample. Assuming a linear, local response, we have $\widetilde{\mathbf{M}}(\mathbf{q},\omega)=\chi(\omega)\widetilde{\mathbf{H}}(\mathbf{q},\omega)$ [7], where $\chi(\omega)$ is the AC magnetic susceptibility and $\widetilde{\mathbf{H}}$ is the Fourier transform of the auxiliary magnetic $\mathbf{H}$-field of the incident electron. In real space $\mathbf{B}=\vec{\nabla}\times\mathbf{A}_f=\mu_0\mathbf{H}$, so that $\widetilde{\mathbf{H}}(\mathbf{q},\omega)=\frac{2\pi i}{\mu_0}\mathbf{q}\times\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$; the subscript '$f$' denotes the fact that $\mathbf{A}_f$ is the vector potential for the self-field. For an electron moving along the z-axis with velocity $\mathbf{v}$ the charge density is $\rho_f(\mathbf{r},t) = -e\delta(x)\delta(y)\delta(z - vt)$, and therefore the current density is $\mathbf{J}_f(\mathbf{r},t) = \mathbf{v}\rho_f(\mathbf{r},t)$. Substituting $\tilde{\mathbf{J}}_f(\mathbf{q},\omega) = -2\pi e\mathbf{v}\,\delta(2\pi q_z v - \omega)$ and $\tilde{\mathbf{J}}_b(\mathbf{q},\omega) = 0$ in Equation 4b we obtain $\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ for the self-field of the incident electron:

$$\widetilde{\mathbf{A}}_f(\mathbf{q},\omega) = -\frac{2\pi e\mu_0\mathbf{v}\,\delta(2\pi q_z v - \omega)}{\left[4\pi^2 q^2 - \frac{\omega^2\varepsilon(\omega)}{c^2}\right]} \qquad \ldots (16)$$

$\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ is non-zero for $q_z = \omega/2\pi v$, which is precisely the condition for magnetic energy loss in Equation 15. Furthermore, since the electron velocity $\mathbf{v}$ is along the z-axis, the x,y components of $\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ are zero. Therefore, Equation 15 for AC magnetic susceptibility becomes:

$$\frac{\partial^4 P_{\mathrm{mag}}}{\partial\mathbf{q}_\perp\partial^2\omega} = \frac{2\pi e^2 q_\perp^2}{\varepsilon_0\hbar c^2}\,\mathrm{Im}\left\{\frac{\chi(\omega)}{\left[4\pi^2\left(q_\perp^2 + \left(\frac{\omega}{2\pi v}\right)^2\right) - \frac{\omega^2\varepsilon(\omega)}{c^2}\right]^2}\right\} \qquad \ldots (17)$$

where 'Im' denotes the imaginary part of a complex number. Integrating over a circular EELS aperture with radius q_{EELS} gives:

$$\frac{\partial^2 P_{\mathrm{mag}}}{\partial\omega^2} = \frac{e^2}{8\pi^2\varepsilon_0\hbar c^2}\,\mathrm{Im}\left\{\chi(\omega)\left[\ln\left(1 + \frac{4\pi^2 q_{\mathrm{EELS}}^2}{\theta}\right) + \frac{\theta}{4\pi^2 q_{\mathrm{EELS}}^2 + \theta} - 1\right]\right\} \qquad \ldots (18a)$$

$$\theta = \omega^2\left(\frac{1}{v^2} - \frac{\varepsilon(\omega)}{c^2}\right) \qquad \ldots (18b)$$

Equation 18a must be integrated with respect to ω to obtain the magnetic energy loss spectrum $\frac{\partial P_{\mathrm{mag}}}{\partial\omega}$. For AC magnetic susceptibility measurements it is important to separate magnetic from dielectric energy losses in the EELS spectrum. Equation 18a indicates that the cross-section for the former is only weakly dependent on the electron speed v, while the latter varies approximately as v^{-2} (see Equation 23 below). This provides a means for identifying magnetic energy losses, although changing the accelerating voltage in a transmission electron microscope is inconvenient. Furthermore, magnetic energy losses are predicted to be small compared to dielectric losses [26], presumably because from Equation 6 they only appear as solutions to the retarded form of the electric field. Magnetic energy losses would therefore require far better signal-to-noise ratio for their detection.

Equation 15 can also be used to calculate magnon energy losses in a ferromagnetic material where the spontaneous magnetisation is along the z-axis. A magnon spin wave with wave vector $\mathbf{q}_m$ and angular frequency ω_m will induce in-plane magnetisation gradients that can lead to an energy loss of the incident electron. Following Ref [7] the in-plane magnetisation components due to the magnon is expressed as:

$$M_x(\mathbf{r},t) = A_m e^{2\pi i \mathbf{q}_m \cdot \mathbf{r} - i\omega_m t} \iff \widetilde{M}_x(\mathbf{q},\omega) = 2\pi A_m \delta(\mathbf{q}-\mathbf{q}_m)\delta(\omega-\omega_m)$$
… (19a)

$$M_y(\mathbf{r},t) = iA_m e^{2\pi i \mathbf{q}_m \cdot \mathbf{r} - i\omega_m t} \iff \widetilde{M}_y(\mathbf{q},\omega) = 2\pi i A_m \delta(\mathbf{q}-\mathbf{q}_m)\delta(\omega-\omega_m)$$
… (19b)

where A_m is the amplitude of the magnon wave. Substituting Equations 19a and 19b in Equation 15 and integrating over $\mathbf{q}_\perp$ and ω leads to the following expression for the magnon energy loss:

$$\frac{\partial P_{\mathrm{mag}}}{\partial \omega} = \frac{eA_m}{\varepsilon_0 \hbar v c^2} \mathrm{Re}\left\{\frac{q_{my} - iq_{mx}}{4\pi^2\left(q_{m\perp}^2 + \left(\frac{\omega_m}{2\pi v}\right)^2\right) - \frac{\omega_m^2 \varepsilon(\omega_m)}{c^2}}\right\}$$
… (20)

where q_{my} and $q_{m\perp}$ are the *y*- and *xy*-components of $\mathbf{q}_m$ respectively. In deriving Equation 20 it is assumed that $q_{mz} = \omega_m/2\pi v$, which ensures conservation of linear momentum during scattering. Consider a magnon energy $\hbar\omega_m$ equal to 10 meV, which is within the energy resolution of modern monochromated electron microscopes [1]. Taking ferromagnetic iron as an example the magnon dispersion relationship, as measured by neutron scattering, is given by $\hbar\omega_m = Dq_m^2$, where the constant D is equal to 9.08 eV·$Å^2$ [8] (the value of D is adjusted for the fact that in Ref [8] the definition of the magnon wavevector includes the factor 2π; cf. Equations 19a and 19b). Therefore, for a 10 meV magnon in iron and 60 kV electron beam q_m >> $q_{mz} = \omega_m/2\pi v$. Assume that the in-plane component of the magnon wavevector is along the *y*-direction, so that $q_{my} \approx q_m$ in Equation 20. The magnon amplitude A_m is assumed to be $10^{-3}M_s$, where the room temperature spontaneous magnetisation M_s for iron is 1.7×10^6 $\mathrm{Am^{-1}}$ [38]. For simplicity the dielectric function $\varepsilon(\omega)$ is modelled on a free electron metal [39]:

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\tau)}$$
… (21)

where ω_p is the plasmon frequency and τ is the damping constant. The EELS spectrum from a ferritic stainless steel specimen was used to estimate the plasmon peak energy $\hbar\omega_p = 22$ eV, and from the plasmon full-width-at-half-maximum the damping energy was estimated to be $\hbar\tau = 16$ eV [15]. The relatively large damping energy may partly be due to the fact that a strong interband transition is observed close to the plasmon peak maximum. Nevertheless, with these numerical estimates Equation 20 gives $\frac{\partial P_{\mathrm{mag}}}{\partial \omega} = 1.9\times10^{-6}$ $\mathrm{m^{-1}s}$ (the result does not strongly depend on the precise value of τ). This can be compared with dielectric energy losses, which can be derived starting from the first term in Equation 6 using a procedure similar to that outlined for magnetic energy losses. The result is:

$$\frac{\partial^3 P_{\text{diel}}}{\partial \mathbf{q}_\perp \partial\omega} = \frac{e^2}{4\pi^3\varepsilon_0\hbar v^2}\text{Im}\left\{-\frac{(1-\varepsilon(\omega)\beta^2)}{\varepsilon(\omega)\left[q_\perp^2+\left(\frac{\omega}{2\pi v}\right)^2(1-\varepsilon(\omega)\beta^2)\right]}\right\}$$

… (22)

Integrating over a circular EELS aperture with radius q_{EELS} gives:

$$\frac{\partial P_{\text{diel}}}{\partial\omega} = \frac{e^2}{4\pi^2\varepsilon_0\hbar v^2}\text{Im}\left\{-\frac{(1-\varepsilon(\omega)\beta^2)}{\varepsilon(\omega)}\ln\left[1+\frac{q_{\text{EELS}}^2}{\left(\frac{\omega}{2\pi v}\right)^2(1-\varepsilon(\omega)\beta^2)}\right]\right\}$$

… (23)

For a 20 mrad EELS aperture and 60 kV electron beam the largest value of $\frac{\partial P_{\text{diel}}}{\partial\omega}$ is 5.2×10^{-10} m^{-1}s at the plasmon peak, which is considerably less than $\frac{\partial P_{\text{mag}}}{\partial\omega}$. Hence our simple estimate suggests that magnons should be easily detected using standard electron beams. However, there is an important subtlety that has been overlooked. From the Einstein-de Haas effect [7] magnetisation is linked to angular momentum, so that generation of magnons by the electron beam requires a transfer of $1\hbar$ unit of angular momentum from the incident electron to the specimen. Standard, unpolarised electron beams do not however possess angular momentum, so that magnon excitation is prohibited. For this reason, it is useful to explore energy losses in vortex electron beams, which contain orbital angular momentum.

3. Magnetic energy loss for a vortex electron beam with orbital angular momentum

The three-dimensional electron wavefunction (ψ) for a vortex beam is a radial function in the xy-plane of the specimen and has an azimuthal phase dependence $\exp(il\phi)$, where l is the winding number and ϕ the azimuthal angle. The orbital angular momentum (OAM) of the vortex beam in free space is therefore $l\hbar$ [31]. The areal electron density is $n_v(\mathbf{R},z) = \alpha|\psi|^2\delta(z-vt)$ and the current density is $\mathbf{J}_f(\mathbf{R},z) = -e\frac{\hbar}{m}\text{Im}(\psi^*\vec{\nabla}\psi)\delta(z-vt)$, where m is the electron mass, $\mathbf{R}$ is the position vector in the xy-plane and α is a normalisation constant for the electron density in the specimen plane at depth $z = vt$ [33]; the term $\delta(z-vt)$ signifies the fact that in our semi-classical electrodynamic model the electron is treated as a particle moving with speed v along the z-axis. The current density has an azimuthal component as well as a component along the z-direction [40]. Vortex beams in free space include Bessel and Laguerre-Gaussian (LG) beams [40]. For a uniform magnetic field along the z-axis the wavefunction has solutions of Landau states, which are modified LG beams, where the OAM is enhanced provided the magnetic vector potential $\mathbf{A}$ and azimuthal current flow in the same sense [40-41].

The energy loss for a vortex beam will now be calculated. First consider the magnetic energy loss due to the electric field (Equation 6). The stopping power in Equation 7 is modified to:

$$\frac{dW}{dz} = \int eE_z(\mathbf{r},t)n_v(\mathbf{R},z)d\mathbf{R} = \frac{e}{2\pi}\int \tilde{E}_z(\mathbf{q},\omega)n_v(\mathbf{R},z)e^{2\pi i\mathbf{q}\cdot\mathbf{r}-i\omega t}d\mathbf{q}d\omega d\mathbf{R}$$

… (24)

Equation 24 can be simplified as:

$$\frac{dW}{dz} = \frac{e}{2\pi}\int \tilde{E}_z\,(\mathbf{q},\omega)\left[\iint n_v(\mathbf{R},\mathrm{z})e^{2\pi i\mathbf{q}_\perp\cdot\mathbf{R}}\,d\mathbf{R}\right]e^{2\pi i q_z z - i\omega t}d\mathbf{q}d\omega$$
$$= \frac{e}{2\pi}\int \tilde{E}_z\,(\mathbf{q},\omega)\tilde{n}_v(\mathbf{q}_\perp,z)e^{2\pi i q_z z - i\omega t}d\mathbf{q}d\omega$$
… (25)

where we have used the relationship $\tilde{n}_v(-\mathbf{q}_\perp,z) = \tilde{n}_v(\mathbf{q}_\perp,z)$, since for a vortex beam the electron density is a real quantity with even parity in the *xy*-plane, i.e. $n_v(-\mathbf{R},\mathrm{z}) = n_v(\mathbf{R},z)$; even parity of the electron density also implies that $\tilde{n}_v(\mathbf{q}_\perp,z)$ is a real quantity. For a vortex beam in a crystalline specimen however this simple relationship breaks down due to dynamic diffraction [42-44].

The differential cross-section for magnetic energy loss can be calculated from Equation 25 using a procedure similar to that outlined in Section 2. First consider the integration over the electron trajectory $z = vt$ to calculate the energy change δW (see Equation 9). For simplicity, it is assumed that the vortex beam is non-diffracting, so that $\tilde{n}_v(\mathbf{q}_\perp,z)$ is independent of *z*. This is exactly true for modified LG (i.e. Landau) beams in a uniform magnetic field in the *z*-direction, as well as Bessel beams, but not LG beams, which are approximate solutions to the paraxial Schrödinger equation [40]. These results however assume a uniform electrostatic potential, and do not always hold in a crystal [42-44], so that our analysis is only applicable for weakly diffracting specimens. Integration along the electron trajectory with the assumption of a depth independent electron density leads to the quantisation condition $q_z = \omega/2\pi v$. Making use of the relationships in Equations 11a and 11b, we obtain:

$$\frac{\partial^4 P_{\mathrm{mag1}}}{\partial\mathbf{q}_\perp\partial^2\omega} = -\frac{e}{2\pi\varepsilon_0\hbar v c^2}\,\tilde{n}_v(\mathbf{q}_\perp)\mathrm{Re}\left[\frac{q_x\widetilde{M}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right) - q_y\widetilde{M}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)}{4\pi^2\left(q_\perp^2+\left(\frac{\omega}{2\pi v}\right)^2\right) - \frac{\omega^2\varepsilon(\omega)}{c^2}}\right]$$
… (26)

where P_{mag1} is the 'probability' for magnetic energy loss per unit path length due to the vortex beam electric field. Apart from the $\tilde{n}_v(\mathbf{q}_\perp)$ term, Equation 26 is otherwise similar to Equation 15, and hence there is no obvious benefit of vortex beams over standard, unpolarised electron beams. Instead, we look for energy loss mechanisms that depend on the magnetic moment (magnitude μ_v) of the vortex beam, since vortex beams with arbitrarily large OAM can in principle be generated using the holographic aperture method [28], thus amplifying any magnetic energy losses in the sample. The energy loss mechanism of interest here is the 'Stern-Gerlach' force (Section 1), which is due to the change in magnetic potential energy in a non-uniform magnetic field:

$$\frac{dW_{\mathrm{mag2}}}{dz} = \mu_v\int\frac{\partial B_z}{\partial z}n_v(\mathbf{R},z)d\mathbf{R} = i\mu_v\int q_z\tilde{B}_z\,(\mathbf{q},\omega)\tilde{n}_v(\mathbf{q}_\perp)e^{2\pi i q_z z - i\omega t}d\mathbf{q}d\omega$$
… (27)

where we have assumed that the vortex beam is non-diffracting and used the methods outlined previously to simplify the above expression. The stopping power is defined as being positive for energy loss events. Furthermore, Equation 27 includes only the *z*-component of the magnetic field B_z, since μ_v is parallel to the optic *z*-axis. Using the fact that $\mathbf{B} = \vec{\nabla} \times \mathbf{A}$ Equation 27 reduces to:

$$\frac{dW_{\mathrm{mag2}}}{dz} = -2\pi\mu_v \int q_z\left[q_x\tilde{A}_y(\mathbf{q},\omega) - q_y\tilde{A}_x(\mathbf{q},\omega)\right]\tilde{n}_v(\mathbf{q}_\perp)e^{2\pi i q_z z - i\omega t}d\mathbf{q}d\omega \qquad \dots (28)$$

Integrating over the electron trajectory $z = vt$ to calculate the energy change δW (see Equation 9) leads to the quantisation condition $q_z = \omega/2\pi v$, and therefore:

$$\frac{dW_{\mathrm{mag2}}}{dz} = -\frac{\mu_v}{v}\int \omega\left[q_x\tilde{A}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right) - q_y\tilde{A}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)\right]\tilde{n}_v(\mathbf{q}_\perp)d\mathbf{q}d\omega \qquad \dots (29)$$

Since the magnetic vector potential is a real quantity $\widetilde{\mathbf{A}}(-\mathbf{q},-\omega) = \widetilde{\mathbf{A}}(\mathbf{q},\omega)^*$, so that for any scattering vector, frequency pair $(\mathbf{q},\omega)$ and its negative $(-\mathbf{q},-\omega)$ we have:

$$(-\omega)(-q_x)\tilde{A}_y\left(-\mathbf{q}_\perp,-\frac{\omega}{2\pi v},-\omega\right)\tilde{n}_v(-\mathbf{q}_\perp) = \omega q_x\tilde{A}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)^*\tilde{n}_v(\mathbf{q}_\perp) \qquad \dots (30a)$$

$$(-\omega)(-q_y)\tilde{A}_x\left(-\mathbf{q}_\perp,-\frac{\omega}{2\pi v},-\omega\right)\tilde{n}_v(-\mathbf{q}_\perp) = \omega q_y\tilde{A}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)^*\tilde{n}_v(\mathbf{q}_\perp) \qquad \dots (30b)$$

Therefore, for a circular EELS aperture centred about the optic axis Equation 29 gives:

$$\frac{dW_{\mathrm{mag2}}}{dz} = -\frac{\mu_v}{v}\int \omega\tilde{n}_v(\mathbf{q}_\perp)\mathrm{Re}\left[q_x\tilde{A}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right) - q_y\tilde{A}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)\right]d\mathbf{q}d\omega \qquad \dots (31)$$

From this a differential scattering cross-section can be derived (see Equation 14):

$$\frac{\partial^4 P_{\mathrm{mag2}}}{\partial\mathbf{q}_\perp\partial^2\omega} = -\frac{\mu_v}{2\pi\hbar v^2}\tilde{n}_v(\mathbf{q}_\perp)\mathrm{Re}\left[q_x\tilde{A}_y\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right) - q_y\tilde{A}_x\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)\right] \qquad \dots (32)$$

where P_{mag2} is the 'probability' for magnetic energy loss per unit path length due to the Stern-Gerlach force. From Equation 4b, $\widetilde{\mathbf{A}}(\mathbf{q},\omega)$ contains contributions from both the free current density of the incident electron and bound current density due to sample magnetisation. Consider first the free current density, which for a vortex beam has azimuthal symmetry in the *xy*-plane. This means that when integrating Equation 32 over a circular EELS aperture to obtain the total energy loss, the contribution at wavevector (q_x, q_y) will exactly cancel that at (q_y, q_x).

The symmetry of the vortex beam therefore prohibits any Stern-Gerlach energy loss due to the self-field of the electron. This result has a simple physical explanation. For the non-diffracting vortex beams in Reference [40] the relationship $\mathbf{J}_{fz}(\mathbf{R}) = -e\mathbf{v}n_v(\mathbf{R})$ was obtained, i.e. apart from the azimuthal current flow in the *xy*-plane, the electrons also move with constant velocity **v** along the optic *z*-axis. The electron trajectory is therefore helical, and its magnetic field can be shown to be uniform along the optic axis [45]. From Equation 27 the Stern-Gerlach force must therefore be zero. It naturally follows that vortex beams are not suitable for AC susceptibility measurements, since the magnetisation $\mathbf{M}(\mathbf{R}, z, \omega) = \chi(\omega)\mathbf{H}(\mathbf{R}, z, \omega)$ will also be uniform along the *z*-axis, thereby resulting in zero Stern-Gerlach energy loss. A derivation of this result is given in the Appendix.

Since only bound currents can contribute to the Stern-Gerlach energy loss, using $\tilde{\mathbf{J}}_b(\mathbf{q}, \omega) = 2\pi i\mathbf{q} \times \widetilde{\mathbf{M}}(\mathbf{q}, \omega)$ and Equation 4b, Equation 32 becomes:

$$\frac{\partial^4 P_{\mathrm{mag2}}}{\partial \mathbf{q}_\perp \partial^2 \omega} = \frac{\mu_v \mu_0}{\hbar v^2} \tilde{n}_v(\mathbf{q}_\perp) \mathrm{Im} \left\{ \frac{\left(\frac{\omega}{2\pi v}\right)\left[q_x \widetilde{M}_x\left(\mathbf{q}_\perp, \frac{\omega}{2\pi v}, \omega\right) + q_y \widetilde{M}_y\left(\mathbf{q}_\perp, \frac{\omega}{2\pi v}, \omega\right)\right] - q_\perp^2 \widetilde{M}_z\left(\mathbf{q}_\perp, \frac{\omega}{2\pi v}, \omega\right)}{4\pi^2\left(q_\perp^2 + \left(\frac{\omega}{2\pi v}\right)^2\right) - \frac{\omega^2 \varepsilon(\omega)}{c^2}} \right\} \quad \ldots (33)$$

For a given magnetisation reversing the sign of μ_v will convert energy losses to energy gains and vice-versa. Equation 33 can be used to calculate the Stern-Gerlach energy loss due to magnons. The in-plane magnetisation due to magnons are given by Equations 19a and 19b. The out-of-plane magnetisation M_z is spatially uniform and time-independent, so that $\widetilde{M}_z(\mathbf{q}, \omega) = 2\pi M_s \delta(\mathbf{q})\delta(\omega)$, where M_s is the spontaneous magnetisation. Substituting in Equation 33 and integrating over $\mathbf{q}_\perp$ and ω we obtain:

$$\frac{\partial P_{\mathrm{mag2}}}{\partial \omega} = \tilde{n}_v(q_{m\perp})\mu_v \left(\frac{\mu_0 \omega_m A_m}{\hbar v^3}\right) \mathrm{Im} \left\{ \frac{q_{mx} + i q_{my}}{4\pi^2\left(q_{m\perp}^2 + \left(\frac{\omega_m}{2\pi v}\right)^2\right) - \frac{\omega_m^2 \varepsilon(\omega_m)}{c^2}} \right\} \quad \ldots (34)$$

where it is assumed that $q_{mz} = \omega_m / 2\pi v$ (conservation of linear momentum). Equation 34 is evaluated for a 10 meV magnon in iron using the numerical values given in Section 2. For a vortex beam $\mu_v = l\mu_B$, where μ_B is the Bohr magneton. Since $\int n_v(\mathbf{R}) d\mathbf{R} = 1$, it follows that $\tilde{n}_v(\mathbf{q}_\perp) \leq 1$, and here we assume $\tilde{n}_v(\mathbf{q}_\perp) = 1$ for convenience.

For a $1\hbar$ OAM vortex beam Equation 34 gives a cross-section 9.2×10^{-14} $\mathrm{m^{-1}s}$. This is three orders of magnitude smaller than the plasmon scattering cross-section in iron for unpolarised electron beams (Section 2). Magnon detection is therefore challenging, although vortex beams have several benefits over unpolarised beams that, in theory, should make the task easier. First the OAM of a vortex beam can provide the angular momentum required for magnon generation. Here the sign of the OAM is important. For example, magnons can only be generated if the vortex beam magnetic moment μ_v is anti-parallel to the sample magnetisation. For the opposite case only thermally generated magnons can be annihilated; this is less probable, since by the principle of detailed balance energy gain and loss processes are linked through a Boltzmann factor [5, 46]. Second the scattering cross-section can be enhanced by using vortex beams with larger OAM and therefore larger magnetic moment (Equation 34). Vortex beams with OAM up to $1000\hbar$ have been produced in the electron microscope [29]. Variable OAM also provides a method for separating magnetic energy losses from dielectric losses. By subtracting EELS spectra acquired for vortex beams with different μ_v, the electric field induced magnetic and dielectric energy losses cancel provided $\tilde{n}_v(\mathbf{q}_\perp)$ for the two vortex beams are identical, leaving only the difference in Stern-Gerlach magnetic energy losses (Equations 26 and 33). $\tilde{n}_v(\mathbf{q}_\perp)$ is identical for free space vortex beams of opposite sign. Note that the cross-sections, e.g. Equations 26 and 32, represent single inelastic scattering of the vortex beam. Following scattering there can be a change in the OAM as a result of angular momentum transfer during the magnetic excitation (e.g. magnons). Therefore, if multiple scattering is involved the cross-section must be modified to reflect the changes in magnetic moment μ_v and electron density $\tilde{n}_v(\mathbf{q}_\perp)$ of the new vortex state. The $\tilde{n}_v(q_{m\perp})$ term in Equation 34 has further implications for magnon characterisation. For example, a 10 meV magnon in iron has wavevector $q_m = 3.3\times10^{-2}$ Å^{-1}, so that for $\tilde{n}_v(q_{m\perp})$ and the Stern-Gerlach cross-section (Equation 34) to have large values the vortex beam must have an intensity maximum at $\sim q_m^{-1}$ or 30 Å. This limits the spatial resolution that can be achieved in practice. Furthermore, dynamic scattering within the specimen means that the incident electrons do not have well defined OAM [42-44], and the magnon lifetime will also be limited by decay via single particle Stoner excitation [8] (note that Equations 19a and 19b do not take into account magnon damping). These two factors will also negatively impact the measurement.

4. Conclusions

Transmission EELS scattering cross-sections for magnetic excitations are derived for standard, unpolarised and vortex electron beams. The latter contains a magnetic moment due to its orbital angular momentum, which results in a 'Stern-Gerlach' energy loss mechanism in addition to the energy losses caused by the electric field of the incident electron. The Stern-Gerlach energy loss is due to the change in magnetic potential energy of the incident electron in a non-uniform magnetic field. The prospect of AC magnetic susceptibility measurements and magnon characterisation in a transmission electron microscope are explored. It is shown that the former can be carried out using unpolarised electron beams, but the weak scattering cross-section and difficulty in separating magnetic energy losses from dielectric losses makes this technique difficult to implement. There are no advantages in using vortex beams for measuring AC magnetic susceptibility, since the Stern-Gerlach force is zero due to the self-magnetic field being spatially uniform along the optic axis. Vortex beams are however highly suited for detecting magnons. They can provide the angular momentum required for magnon excitation and the strength of the Stern-Gerlach interaction can be enhanced by using vortex beams of larger orbital angular momentum. Furthermore, magnetic energy losses can be separated from dielectric losses by subtracting EELS spectra acquired with vortex beams of opposite sign. Vortex beams are therefore a potential route for experimental observation of magnons at much higher spatial resolution compared to traditional neutron scattering measurements.

Finally, although the emphasis has been on magnons and AC magnetic susceptibility, the semi-classical results derived in this paper are general and can be applied to other large scale magnetic features where a continuum description is appropriate, such as, for example, demagnetising fields, domain walls and skyrmions. The $\mathbf{q}$-dependence in $\widetilde{\mathbf{M}}(\mathbf{q},\omega)$ is determined by the magnetic feature of interest. The ω (or equivalently time) dependence is due to the precessional motion of the spontaneous magnetisation vector under the influence of the incident electron self-magnetic field, which can be modelled using the Landau-Lifshitz-Gilbert equation. For localised magnetic signals, such as Stoner excitations or short wavelength magnons, a continuum description is no longer appropriate, and a full quantum mechanical treatment is required to calculate the energy loss.

5. Acknowledgments

I would like to thank the anonymous referee for his/her helpful comments.

6. Appendix

Here it will be shown that the Stern-Gerlach energy loss due to magnetisation of a medium by a vortex beam is zero, which prohibits AC magnetic susceptibility measurements. Substituting $\widetilde{\mathbf{M}}(\mathbf{q},\omega) = \chi(\omega)\widetilde{\mathbf{H}}(\mathbf{q},\omega)$ and $\widetilde{\mathbf{H}}(\mathbf{q},\omega) = \frac{2\pi i}{\mu_0}\mathbf{q} \times \widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ in Equation 33 we obtain,

$$\frac{\partial^4 P_{\mathrm{mag2}}}{\partial \mathbf{q}_\perp \partial^2 \omega} = -\frac{2\pi\mu_v}{\hbar v^2}\tilde{n}_v(\mathbf{q}_\perp)\mathrm{Re}\left\{\chi(\omega)\frac{\left(q_\perp^2 + \left(\frac{\omega}{2\pi v}\right)^2\right)\left[q_x\tilde{A}_{fy}\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right) - q_y\tilde{A}_{fx}\left(\mathbf{q}_\perp,\frac{\omega}{2\pi v},\omega\right)\right]}{4\pi^2\left(q_\perp^2 + \left(\frac{\omega}{2\pi v}\right)^2\right) - \frac{\omega^2\varepsilon(\omega)}{c^2}}\right\} \quad \ldots \text{(A1)}$$

The dependence on $\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ is similar to the expression in Equation 32. Due to azimuthal symmetry of the vortex beam and $\widetilde{\mathbf{A}}_f(\mathbf{q},\omega)$ the cross-section at wavevector (q_x, q_y) is equal and opposite to that at (q_y, q_x). Integrating over a circular EELS aperture the net energy loss is therefore zero.

References

[1] O.L. Krivanek, J.P. Ursin, N.J. Bacon, G.J. Corbin, N. Dellby, P. Hrncirik, M.F. Murfitt, C.S. Own and Z.S. Szilagyi, *Phil. Trans. R. Soc. A* **367** (2009) 3683.
[2] F.S. Hage, R.J. Nicholls, J.R. Yates, D.G. McCulloch, T.C. Lovejoy, N. Dellby, O.J. Krivanek, K. Refson and Q.M. Ramasse, *Sci. Adv.* **4** (2018) eaar7495.
[3] F.S. Hage, D.M. Kepaptsoglou, Q.M. Ramasse and L.J. Allen, *Phys. Rev. Lett.* **122** (2019) 016103.
[4] M.J. Lagos, A. Trügler, U. Hohenester and P.E. Batson, *Nature* **543** (2017) 529.
[5] M.J. Lagos and P.E. Batson, *Nano Lett.* **18** (2018) 4556.

[6] J.A. Hachtel, J. Huang, I. Popovs, S. J-Popova, J.K. Keum, J. Jakowski, T.C. Lovejoy, N. Dellby, O.L. Krivanek and J.C. Idrobo, *Science* **363** (2019) 525.
[7] S. Blundell, *Magnetism in Condensed Matter*, Oxford University Press, Oxford (2014).
[8] J.W. Lynn, *Phys. Rev. B* **11** (1975) 2624.
[9] H. Hopster, R. Raue and R. Clauberg, *Phys. Rev. Lett.* **53** (1984) 695.
[10] J. Kirschner, D. Rebenstorff and H. Ibach, *Phys. Rev. Lett.* **53** (1984) 698.
[11] D. Venus and J. Kirschner, *Phys. Rev. B.* **37** (1988) 2199.
[12] K.-P. Kämper, D.L. Abraham and H. Hopster, *Phys. Rev. B.* **45** (1992) 14335.
[13] R. Vollmer, M. Etzkorn, P.S.A. Kumar, H. Ibach and J. Kirschner, *J. Magn. Magn. Mater.* **272-276** (2004) 2126.
[14] Kh. Zakeri, Y. Zhang and J. Kirschner, *J. Electron. Spectros Relat. Phenomena.* **189** (2013) 157.
[15] R.F. Egerton, *Electron Energy-Loss Spectroscopy in the Electron Microscope*, Plenum Press, New York (1996).
[16] J.A. Alexander, F.J. Scheltens, L.F. Drummy, M.F. Durstock, F.S. Hage, Q.M. Ramasse and D.W. McComb, *Ultramicroscopy* **180** (2017) 125.
[17] B.G. Mendis, Q.M. Ramasse, T.P. Shalvey, J.D. Major and K. Durose, *ACS Appl. Energy Mater.* **2** (2019) 1419.
[18] C.V. Topping and S.J. Blundell, *J. Phys.*: Condens. Matter **31** (2019) 013001.
[19] J. Yuan and N.K. Menon, *J. Appl. Phys.* **81** (1997) 5087.
[20] P. Schattschneider, S. Rubino, C. Hébert, J. Rusz, J. Kuneš, P. Novak, M. Fabrizioli, G. Panaccione and G. Rossi, *Nature* **441** (2006) 486.
[21] C. Hébert, P. Schattschneider, S. Rubino, P. Novak, J. Rusz and M. Stöger-Pollach, *Ultramicroscopy* **108** (2008) 277.
[22] J. Rusz, J.C. Idrobo and S. Bhowmick, *Phys. Rev. Lett.* **113** (2014) 145501.
[23] R.H. Ritchie, *Phys. Rev.* **106**, 874 (1957).
[24] F.J. Garcia de Abajo, *Rev. Mod. Phys.* **82** (2010) 209.
[25] U. Hohenester, *Comput. Phys. Commun.* **185** (2014) 1177.
[26] D. Ivanenko and V.N. Tsytovich, *Soviet Physics JETP* **1** (1955) 135.
[27] M. Uchida and A. Tonomura, *Nature* **464** (2010) 737.
[28] J. Verbeeck, H. Tian and P. Schattschneider, *Nature* **467** (2010) 301.
[29] E. Mafakheri, A.H. Tavabi, P.-H. Lu, R. Balboni, F. Venturi, C. Menozzi, G.C. Gazzadi, S. Frabboni, A. Sit, R.E. Dunin-Borkowski, E. Karimi, V. Grillo, *Appl. Phys. Lett.* **110** (2017) 093113.
[30] J. Verbeeck, H. Tian and A. Béché, *Ultramicroscopy* **113** (2012) 83.
[31] P. Schattschneider and J. Verbeeck, *Ultramicroscopy* **111** (2011) 14612.
[32] J.D. Jackson, *Classical Electrodynamics*, John Wiley & Sons, New York (1998).
[33] A.I.M. Rae, *Quantum Mechanics*, Fifth Edition, Taylor & Francis, Florida (2008).
[34] A. Asenjo-Garcia, F.J. Garcia de Abajo, *Phys. Rev. Lett.* **113** (2014) 066102.
[35] Z. Mohammadi, C.P. Van Vlack, S. Hughes, J. Bornemann, R. Gordon, *Opt. Express* **20** (2012) 15024.
[36] P. Schattschneider, *Fundamentals of Inelastic Electron Scattering*, Springer-Verlag, New York (1986).
[37] A.P. Young and P. Rez, *J. Phys. C* **8** (1975) L1.
[38] J. Crangle and G.M. Goodman, *Proc. Roy. Soc. Lond. A* **321** (1971) 477.
[39] E Hecht, *Optics*, 4th Edition, Addison Wesley, San Francisco, USA (2002).

[40] K.Y. Bliokh, P. Schattschneider, J. Verbeeck and F. Nori, *Phys. Rev. X* **2** (2012) 041011.
[41] P. Schattschneider, Th. Schachinger, M. Stöger-Pollach, S. Löffler, A. Steiger-Thrisfeld, K.Y. Bliokh, F. Nori, *Nature Commun.* **5** (2014) 4586.
[42] S. Löffler and P. Schattschneider, *Acta Cryst. A* **68** (2012) 443.
[43] B.G.Mendis, *Ultramicroscopy* **149** (2015) 74.
[44] B.G.Mendis, *Ultramicroscopy* **157** (2015) 1.
[45] S.M. Lloyd, M. Babiker, J. Yuan, C. Kerr-Edwards, *Phys. Rev. Lett.* **109** (2012) 254801.
[46] T. Guénault, *Statistical Physics*, Second Edition, Springer, Netherlands (2007).