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High resolution electron energy loss spectroscopy of spin waves in ultra-thin film — The return of the adiabatic approximation?

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ABSTRACT

The paper reports on recent considerable improvements in electron energy loss spectroscopy (EELS) of spin waves in ultra-thin films. Spin wave spectra with 4 meV resolution are shown. The high energy resolution enables the observation of standing modes in ultra-thin films in the wave vector range of $0.15\ \text{Å}^{-1} < q_{||} < 0.3\ \text{Å}^{-1}$. In this range, Landau damping is comparatively small and standing spin wave modes are well-defined Lorentzians for which the adiabatic approximation is well suited, an approximation which was rightly dismissed by Mills and collaborators for spin waves near the Brillouin zone boundary. With the help of published exchange coupling constants, the Heisenberg model, and a simple model for the spectral response function, experimental spectra for Co-films on Cu(100) as well as for Co films capped with further copper layers are successfully simulated. It is shown that, depending on the wave vector and film thickness, the most prominent contribution to the spin wave spectrum may come from the first standing mode, not from the so-called surface mode. In general, the peak position of a low-resolution spin wave spectrum does not correspond to a single mode. A discussion of spin waves based on the "dispersion" of the peak positions in low resolution spectra is therefore subject to errors.

1. Introduction

Douglas L. Mills contributed to many fields in Solid State Physics. He has afforded inspiration to generations of students and collaborators. Of all his fields of interest there is one which stands out by the continuity of his efforts throughout his entire scientific career, the interaction of electrons with elementary excitations at surfaces. It was already in 1967 that he published a paper in which he considered the inelastic interaction of electrons from spin wave excitations [1], thereby anticipating electron energy loss spectroscopy (EELS) on spin waves by almost four decades. Because of the higher cross section, EELS of surface vibrations became possible much earlier, and Douglas Mills has contributed seminal studies to electron/phonon interactions at surfaces, firstly by considering the small-angle dipole scattering regime [2], later the impact scattering regime [3,4]. Ever since we have met in 1969 I had the privilege to work alongside with Douglas Mills on the experimental aspects of electron/solid interactions, through the development of electron spectrometers and by bringing the technique to bear in studies of the physics and chemistry of surfaces. It stresses the foresight of Douglas that our very last collaboration finally was on spin waves in thin magnetic film as studied by inelastic electron scattering [5].

In this contribution I firstly describe the challenges involved in the design of high-resolution energy loss spectrometers and in-how-far these challenges are met with the latest generation of instruments. Despite the small cross section for electron/spin wave interaction spin wave spectra can be obtained with energy resolution down to 4 meV (Section 2) because of high energy resolution spin waves of 3d-metals can be successfully studied in the wave vector regime of $0.15\ \text{Å}^{-1} < q_{||} < 0.3\ \text{Å}^{-1}$. In this range, Landau damping is much smaller than near the boundary of the Brillouin zone and spin waves are well defined Lorentzians. Thence, spin wave energies and Landau damping may be considered as separate entities. The easiest access to spin wave energies is provided by the adiabatic approximation in which the itinerant electron system is mapped onto an effective Heisenberg Hamiltonian [6]. Section 3 deals with the standing spin waves in thin films within that framework. The relative weight of modes in an EEL spectrum is determined by the layer-dependent spectral densities in combination with the finite penetration depth of the scattered electrons. In Section 4, a simple expression for the relative weight of various standing modes is obtained within the Heisenberg model. Using published exchange parameters of Bergqvist et al. [7] experimental spectra of cobalt layers of various thicknesses on Cu(100) substrates are simulated in Section 5. It is shown that the spin wave signal observed in EELS is composed of contributions from several modes. For small wave vectors the energy resolution is high enough to resolve these modes even without resorting to modeling. Section 6 deals with the spin wave signal observed in EELS after capping the cobalt layers with copper. The spectra of Cu-capped surfaces are well described by a simulation in which the exchange parameters near the free cobalt surface are

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replaced by the corresponding parameters known for the Co/Cu(100) interface. The final section considers the present experimental EELS results in the light of some earlier publications of Doug Mills and collaborators. Difference and similarities in the spin wave spectral response in experiments using EELS, scanning tunneling microscopy and light scattering are also discussed.

2. Challenges in the development of electron energy loss spectrometers

In superficial view, today's electron energy loss spectrometer employed for low-energy electron scattering appears to look much the same as they did when they came into use in the 60s of the last century for studies of gas-phase molecules (see e.g. [8]). The spectrometers feature a cathode, one or two electrostatic energy dispersive elements to cut out electrons in a small energy window, a lens system between monochromator and sample, a lens system between sample and analyzer and finally a single or a double-stage analyzer with an electron multiplier for detection (Fig. 1). The details however are grossly different. For example, the electrostatic analyzers are no longer cylindrical or spherical deflectors but rather involve a free-form design in which the inner and outer deflection plates feature cross sections of opposite curvature (see inset in the lower right corner of Fig. 1). The device was invented in 1992 [9]. It combines stigmatic focusing with low angular aberrations. Contrary to the spherical analyzer the stigmatic focusing is achieved by deflection in the dispersion plane as well as orthogonal to the dispersion plane. The active focusing ensures that the monochromator can carry a high current load without being too much affected by defocusing due to electron/electron repulsive forces ("space charge effects"). Spectrometers equipped with those deflectors have shown to produce vibration spectra of adsorbed species with energy resolutions of less than 1 meV [10] and spectra of high momentum phonons with resolution of 2 meV [11–14].

While spectrometers performed satisfactorily in investigations of phonons they did not meet the challenges involved in the detection of spin waves at surfaces, which is the topic of interest in this paper. The main reason for the failure of early attempts is the significant lower cross section for spin wave scattering. This is illustrated in Fig. 2. Fig. 2a shows the probability per solid angle for inelastic scattering of electrons from the so-called S_4-phonon of the Ni(100) surface at

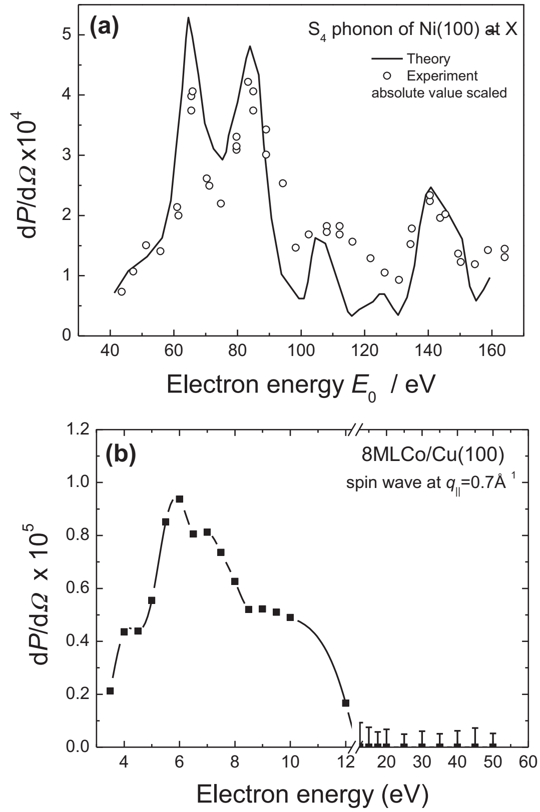

Fig. 2. (a) Probability per solid angle for inelastic scattering of electrons from the so-called S_4-phonon of the Ni(100) surface at the zone boundary. Solid line is a full dynamical scattering theory, the data points are measurements [15]. Their absolute values are scaled to match theory since at the time spectrometers were not calibrated with respect to the solid angle. (b) Measured probability per solid angle for inelastic scattering of electrons from spin waves in a cobalt film.

the $\overline{X}$-point of the surface Brillouin zone (SBZ) [15]. The solid line represents the result of the full dynamical scattering theory, the open circles are experimental data scaled in their absolute value to match the theory since at the time spectrometers were not calibrated with respect to the

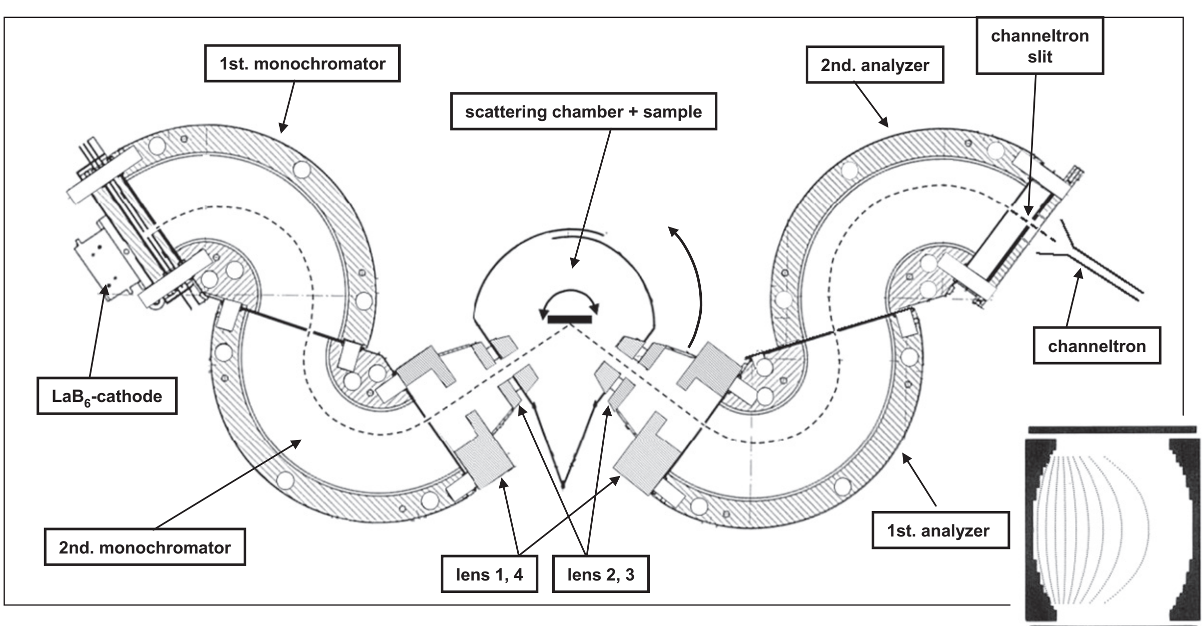

Fig. 1. Top view of the electron energy loss spectrometer as used today. The inset on the lower right side shows the cross section of the electrostatic deflectors.