Elastic and inelastic Scattering in Electron Momentum Spectroscopy of Amorphous Ge Films

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Abstract

Electron momentum spectroscopy (an *(e,2e)* experiment in the high-energy limit) is a scattering approach to the study of the electronic structure. The intensity observed in these experiment, is for an infinitely thin film, simply proportional to the spectral momentum density. For experiments using free-standing films of a more realistic thickness (100 Å or so) elastic and inelastic multiple scattering events are frequent and their influence on the observed intensity cannot be neglected. Here we study germanium films where the sp derived valence band can be measured simultaneously with the shallow, non-dispersing, 3d level. Somewhat surprisingly the intensity of the 3d level, relative to that of the valence band, increases with film thickness. This effect is attributed to elastic multiple scattering. Monte Carlo simulations reproduce the changes in the observed intensity distributions with increasing film thickness.

Key words: Germanium; electron momentum spectroscopy; multiple scattering;
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1. Introduction

There is a variety of techniques available for the study of the electronic structure of materials. In particular electron spectroscopy has been very successful in testing our understanding of the electronic structure of matter. However, the interpretation of the electron spectroscopy data is almost always complicated by additional structures (often referred to as extrinsic structures) that develop as a consequence of elastic and inelastic scattering occurring during transport of electrons to the surface. These extrinsic processes have to be understood in order to access fully the wealth of information that is contained in the intrinsic features of the spectra.

Electron momentum spectroscopy (EMS) is a relatively new technique for studying solids and has been successful for measuring the electronic properties of a number of materials. Until recently these measurements have been mostly performed on low Z targets such as carbon and aluminum. It was thought that for high Z targets elastic scattering would be too strong and would limit the use of this technique. Recently it became clear that,

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even for very heavy targets such as gold films, this technique can provide high-contrast data of the electronic structure [1]. This does not mean that elastic scattering is not important, it reduces for example the count rate for the high-Z materials. Here we will show that it also affects the intensity of different features in different ways.

In an EMS measurement high energy electrons impinge on a thin self-supporting film. Most of the incoming electrons are transmitted through the film, without any scattering event occurring, but some scatter from a target electron, transferring a large fraction of its energy and causing the target electron to be ejected. In these binary collision the energy and momentum of the target electron is completely described by the kinematics of the two outgoing electrons. Such that

$$\varepsilon = E_0 - E_1 - E_2 \tag{1}$$

$$\mathbf{q} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_0 \tag{2}$$

where ε and $\mathbf{q}$ is the binding energy and momentum of the target electron and $E_{0,1,2}, \mathbf{k}_{0,1,2}$ is the energy and momentum of the incoming and two outgoing electrons respectively. Thus by detecting both scattered and ejected electrons (momentum and energy resolved) in coincidence it is possible to infer the energy and momentum of the target electron by using the simple conservation laws Eqs. (1) and (2). By combining many of these events it is possible to determine the momentum and energy resolved electron density of the target.

As electrons are used as the probe in an EMS experiment it is affected with many of the same problems as more traditional methods for measuring electronic structure, however, the small thickness of the films used limits the range of possible trajectories and the task of unraveling intrinsic and extrinsic contributions is simplified significantly. Moreover we can test our understanding of the multiple scattering effects by changing the film thickness in a systematic way. Inelastic multiple scattering (e.g. by plasmon excitation by the incoming or outgoing electrons) causes a shift of the observed intensity to higher binding energy. Elastic deflections affect the momentum balance equation (eq. 2)

In our first attempt to understand these effects we have had considerable success using a simple Monte Carlo approach [2] to simulate the effects of elastic and inelastic scattering. These simulations, while providing insight into the mechanisms behind the measured spectra, did only qualitatively describe the measurements. For example, at the Fermi level, where inelastic scattering only reduces the intensity, but not affects the shape of the observed momentum distribution the simulations did not reproduce quantitatively the momentum distribution. It was thought that the neglect of diffraction was the root of this problem. Recently the Monte Carlo simulations was successfully extended to include diffraction in polycrystalline samples [3] by using cross sections derived from clusters of atoms. This dramatically improved the agreement between experiment and simulation at the Fermi level.

In this work we present measurements of the electron momentum density (EMD) for amorphous Ge films of varying thickness. Monte Carlo simulations using Linear-Muffin-Tin-Orbital (LMTO) calculations as input are presented for comparison and to aid in describing the features observed in the experimental results. The inelastic mean free path of Germanium (320 Å at 25 keV, 587 Å at 50 keV according to ref. [4] is larger than the elastic mean free path (110 Å at 25 keV, 190 Å at 50 keV, values derived from elastic scattering cross section calculations as described in [5]), as is usually the case for high Z elements. For germanium we can simultaneously measure the non-dispersive 'core-like' 3d level as well as the dispersive valence band as both features are separated by only about 20 eV.

2. Experimental Details

The Australian National University spectrometer was used for these studies (see Fig. (1)). Details of the spectrometer can be found in [6] and as such only specifics relating to these measurements will be discussed here.

A 30 Å thick amorphous carbon film (ACF metals) is mounted onto a mesh containing 0.3 mm

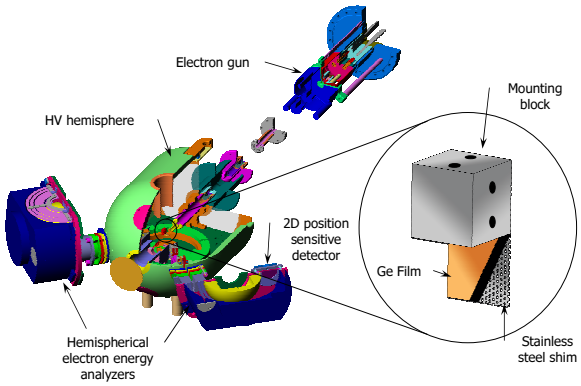


Fig. 1. Schematic diagram of the ANU spectrometer.

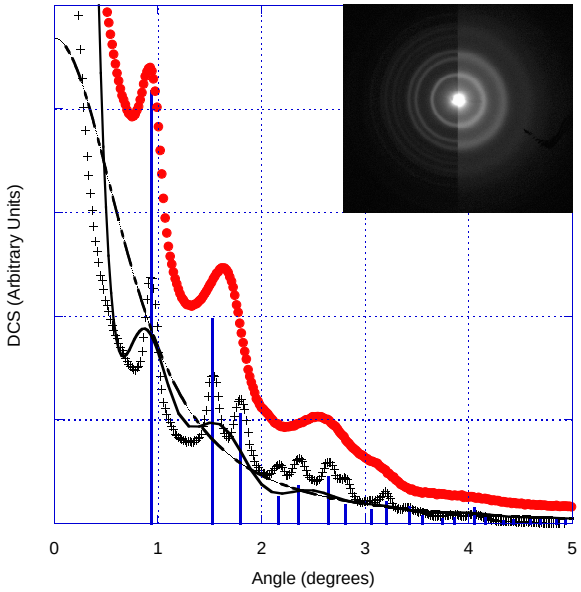


Fig. 2. Comparison of single atom and amorphous elastic cross-sections. Measured diffraction pattern (data points), 5 atom simulation (solid line), single atom cross section (dot-dashed line), solid bars are simulations of the diffraction rings for a polycrystalline sample, crosses are measured diffraction pattern for polycrystalline sample, obtained by annealing the evaporated Ge film. The insert shows the diffraction image as captured by the CMOS camera for polycrystalline (left) and amorphous (right).

diameter holes Fig. (1). Ge is evaporated onto the carbon film by resistive heating of a tantalum boat containing Ge chips, the thickness of the film is monitored during evaporation by a vibrating crystal thickness monitor parallel to the sample film. The vacuum during evaporation was in the 1×10^{-9}

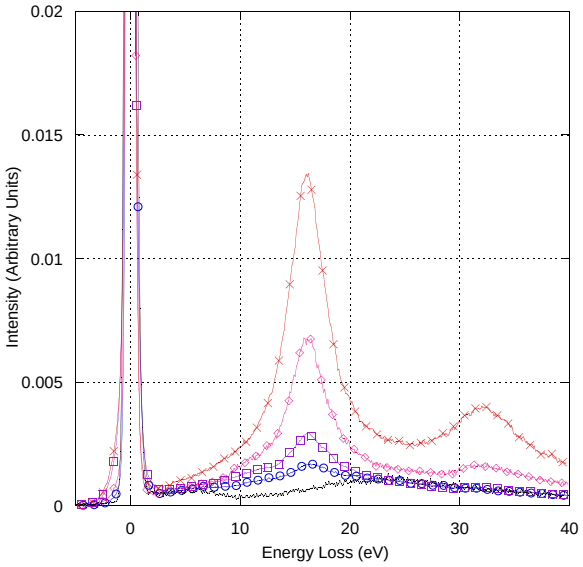


Fig. 3. EELS spectra for various thicknesses of Ge deposited on a 30 Å carbon film (30 Å Ge circles, 60 Å Ge squares, 150 Å Ge diamonds, 350 Å Ge crosses). The line without data points is the bare 30 Å carbon film. Spectra are scaled such that elastic peak area is equal to 1.

torr range. After preparation the sample is moved immediately under vacuum to the spectrometer proper with an operating pressure is better than 1×10^{-10} torr. After each measurement the original sample was returned to the evaporation chamber for additional Ge evaporation. In this way we obtained a series of EMS results for films with increasing thickness.

The kinematics of a measurement are as follows: 25 keV electrons are produced in an electron gun and directed towards the sample which is held at +25 kV, thus 50 keV electrons strike the sample. The majority of the incident electrons pass through the film and are collected in a Faraday cup. The Faraday cup can be moved out of the way and the intensity distribution of the transmitted electrons can then be observed on a phosphor screen. Diffraction patterns of the samples are taken during a measurement by imaging, with a CMOS camera. This makes it possible to determine *in situ* the quality and crystalline form of the sample. Diffraction patterns of these films are shown in Fig. (2)

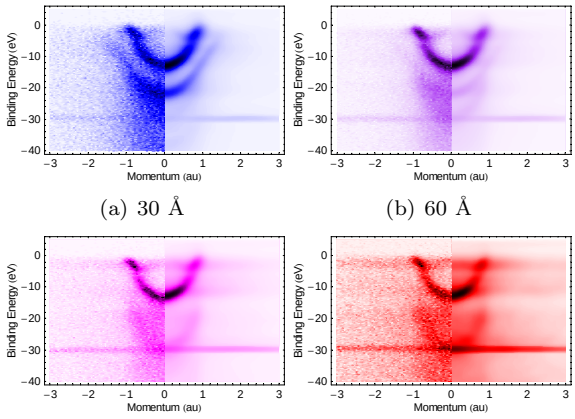
A few of the incident electrons are scattered in the field of a bound electron causing it to be

ejected. The scattered and ejected electrons are then detected by two electron energy spectrometers with slit lenses at $\simeq \pm 45^\circ$ relative to the incoming beam direction. The two analyzers are held close to ground allowing 25 keV electrons to be detected. This configuration simplifies the data acquisition as the electron detectors (channel plates plus resistive anode)are close to ground. The electrons pass through the analyzers and their energy and azimuthal angle (momentum) are determined using position sensitive detectors. In a typical measurement sufficient statistics is obtained in a 2-3 day period.

Energy loss spectra (see Fig. (3)) can also be measured in the apparatus by reducing the gun potential near to ground and holding the sample at 25 keV. Thus elastically scattered electrons are detected in either detector. By scanning the pass energy of the analyzers electrons that are scattered inelastically can also be measured. As the detector is at 45° all electrons that are detected have been scattered elastically over this angle. A fraction of the detected electrons have a reduced energy due to plasmon creation in the film. This fraction increases with increasing thickness. The EELS spectra are hence an independent monitor of the deposited Ge film thickness.

3. Results and Discussion

The measured intensity distribution as a function of momentum and energy are presented as a grey-scale plot in Fig. (4) for a number of different Ge thicknesses. For the smallest thickness (30 Å) two parabolas are visible, the main one, with a minimum around 12 eV binding energy, is followed by a slightly weaker one extending down to $\simeq$ 22 eV. The first one is due to the Germanium layer, the second one is caused by the carbon support film. For larger thicknesses the carbon contribution disappears. The carbon film faces the gun side. Both scattered and ejected electrons have to transverse the Ge layer on their way to the analyzer. The probability that both electrons reach the analyzers, without deflections or energy loss in the Ge layer becomes vanishing small for the thicker Ge films, and hence the carbon contribution is not visible for larger thicknesses . Near 29.5 eV there is a line of increased intensity, especially visible in the distributions obtained for larger Ge thicknesses. This is due to the Ge 3d level. These electrons are confined in coordinate space and thus, as a consequence of the uncertainty principle extend over a wide range in momentum space.



(a) 30 Å (b) 60 Å

(c) 150 Å (d) 350 Å

Fig. 4. Electron momentum density for a Ge film deposited on a 30 Å carbon film (left). Electron momentum density Monte Carlo simulation based on LMTO calculations (right).

For each thickness the left hand side of the plot represents the measured distribution, the right hand side is that obtained by Monte Carlo simulations, using the LMTO calculation of the spectral momentum density as in input(see eg. [7]). The LMTO calculation does not include many-body effects and as such lifetime broadening is included phenomenologically by convoluting the calculations with a 2 eV FWHM Lorentzian. In the calculation core electrons are treated as pseudo valence electrons which causes the d-band to appear at a lower binding energy. This was corrected before applying the Monte Carlo simulations by shifting the d band to higher binding energies as determined by recent 3d core level measurements [8]. The carbon backing is included into the simulations as an additional layer. The measured total band width of the carbon is found to be significantly larger than calculated. This discrepancy was recently reported for these type of calculations

by Kihlgren *et al* [9] who suggested that significant improvements could be made if the binding energy in the calculations are multiplied by 1.13. A large improvement in the total bandwidth of the LMTO calculation, when compared to the measurements, was found when modified in this way.

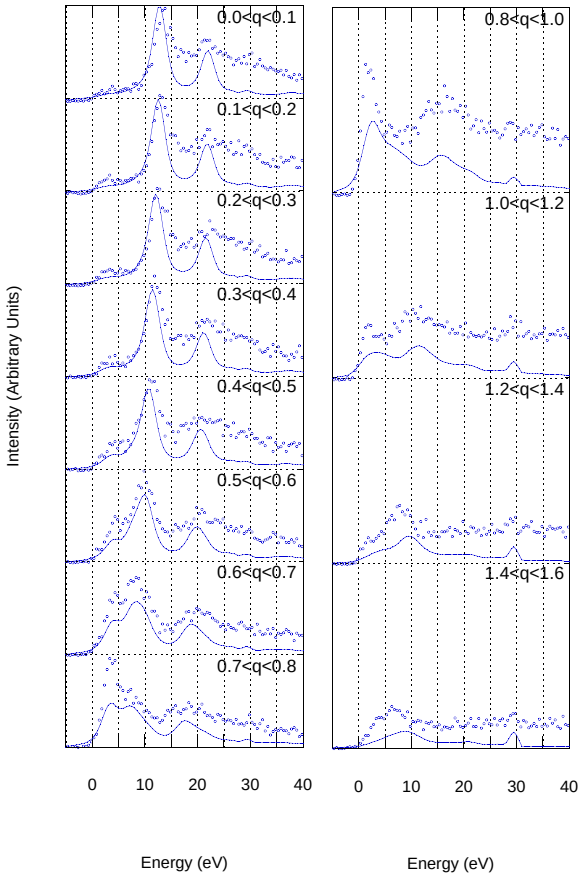


Fig. 5. Energy plots for various momentum slices for a 30 Å Ge film (data points). Monte Carlo computer simulations based on an LMTO calculation (solid line).

The Monte Carlo simulations have been described previously [2]. The simulations need as an input the spectral momentum density. Ideally the spectral momentum density is based on a calculation that takes both the lattice and electron-electron correlations into account. For elements as heavy as Ge this is a very time-consuming approach. Hence we used a simple angular-averaged spectral momentum density of Ge, based on an LMTO calculation as an input. Intrinsic satellites

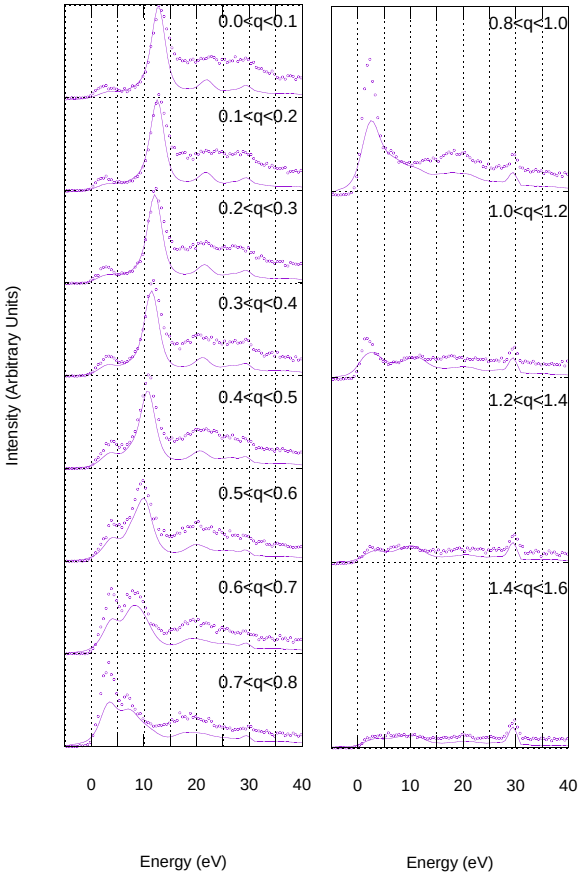


Fig. 6. Energy plots for various momentum slices for a 60 Å Ge film (data points). Monte Carlo computer simulations based on an LMTO calculation (solid line).

causing intensity at higher binding energy are thus not present in the input data.

In the Monte Carlo simulation we model the effect of changes in either energy or trajectory of any of the three electrons involved in an (e,2e) event will infer the incorrect momentum, energy or both for the bound electron. In order to determine the mean depth at which these events occur, the elastic and inelastic mean free paths are required. To simulate this energy loss spectra and elastic cross sections for incident, scattered and ejected electrons are also required. Of particular importance is the elastic cross section at small angles as it is these small angle scattering events which cause the incorrect momentum to be inferred. Electrons scattered at large angles are unlikely to cause a coin-

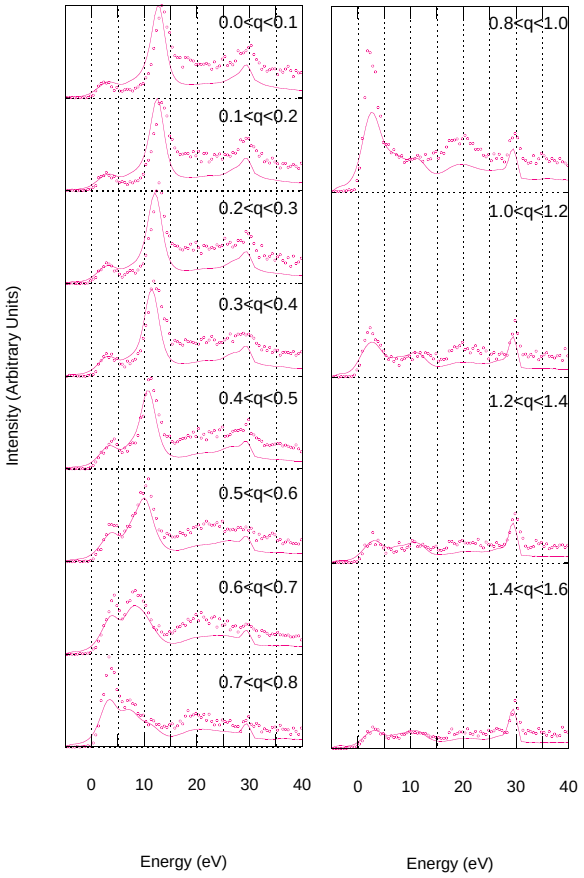


Fig. 7. Energy plots for various momentum slices for a 150 Å Ge film (data points). Monte Carlo computer simulations based on an LMTO calculation (solid line).

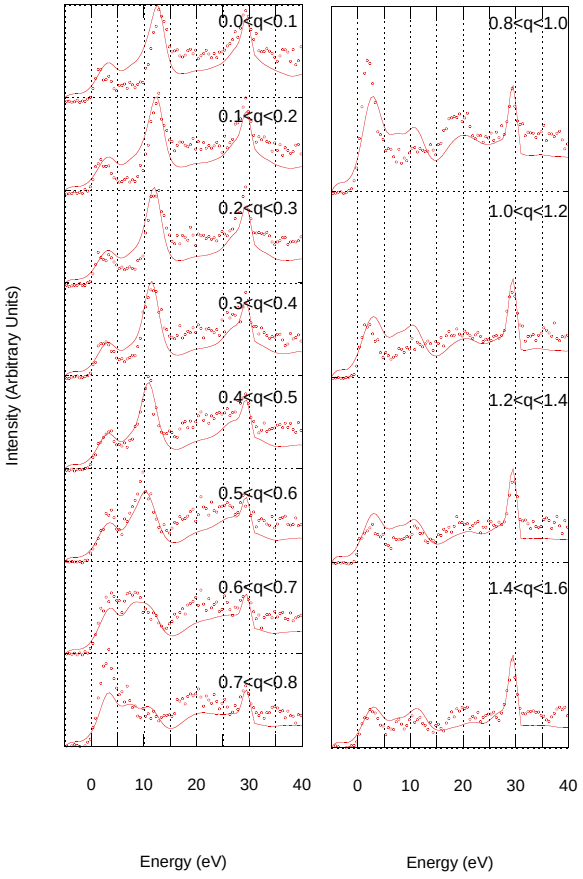


Fig. 8. Energy plots for various momentum slices for a 350 Å Ge film (data points). Monte Carlo computer simulations based on an LMTO calculation (solid line).

cidence in the detectors as the move outside the acceptance angle.

The differential elastic cross section is obtained from a calculation of electron scattering from an isolated atom [5]. The distribution of energy loss in an inelastic scattering event used in the Monte Carlo simulations was for Ge taken from [10], for carbon we used our data for a 30 Å thick film.

The EELS spectra presented in Fig. (3) highlights some of the features seen in the EMDs for the 30 Å film the majority of the inelastic scattering observed is due to the carbon film which can be seen by the similarity between the bare carbon and the 30 Å Ge EELS spectra. The feature at 16 eV energy loss is characteristic of Ge is barely visible in the 30 Å EELS. For the 60 Å film this feature has increased while the contribution from the carbon film has reduced. For the 150 and 350 Å the contribution from the carbon becomes negligible and that from Ge begins to dominate the spectra. This is evident in the strong plasmon peak at 16 eV and a second plasmon peak at 32 eV. For the thicker films we expect hence a ghost of the main features in the spectral momentum distribution to appear at 16 eV larger binding energies.

In order to more closely examine the spectra vertical slices have been taken through the EMDs for various momentum intervals. These are compared with the Monte Carlo simulations. For the 30 Å film Fig. (5) the presence of the carbon backing is evident and constitutes a significantly to the intensity at higher binding energies. For the thicker films

Fig. (6) – (8) this is not the case. For increasing film thickness the intensity of the d-band relative to the valence band increases [11]. Elastic scattering causes electrons that pass through the material to be deflected hence the incorrect momentum is inferred for these electrons. The effect of this can be seen readily in the measured spectra, the d-band intensity appears to increase as the thickness of the film increases. Electrons in the valence band that are elastically scattered are shifted to different momenta left and right of the band (Fig. (4)) thus intensity is shifted out of the band. However for the d-band which extends over all a large momentum range electrons that are elastically scattered remain in the band. The effect is essentially a convolution of the d-band momentum profile by the elastic cross section. As the film thickness increases the degree of elastic scattering increases smearing out the valence band while leaving the d-band intensity the same. This was demonstrated by taking the energy plots in the d-band region integrated over small momentum intervals and fitting a linear background and a gaussian peak, the area under the gaussian is then plotted as a function of momentum Fig. (9). The differences due to the different film thicknesses can be clearly seen when compared with the calculations of the atomic Ge 3d level Fig. (9). The thinnest film showing a typical d electron momentum profile while the 350 Å film has a greater intensity (relative to the valence band) and does not have the broad minimum at zero momentum evident in the calculation and the 30 Å film. The peak at zero momentum in the 350 Å film is due to additional intensity of the bottom of the valence band shifted by the Ge plasmon energy, which energy coincides with that of the Ge 3d-level.

At the top of the valence band in the $0.6 < |\mathbf{q}| < 1.2$ region there is a significant discrepancy in intensity between the measured and simulated spectra Fig. (5) – (8). The theory which is calculated for a single crystal and then averaged over all possible directions, should simulate a polycrystalline sample but will perform worse for an amorphous material where second neighbor distances are not well defined or where dangling bonds, which will contribute significantly to the electronic structure, are likely to occur [12]. In an effort to examine this

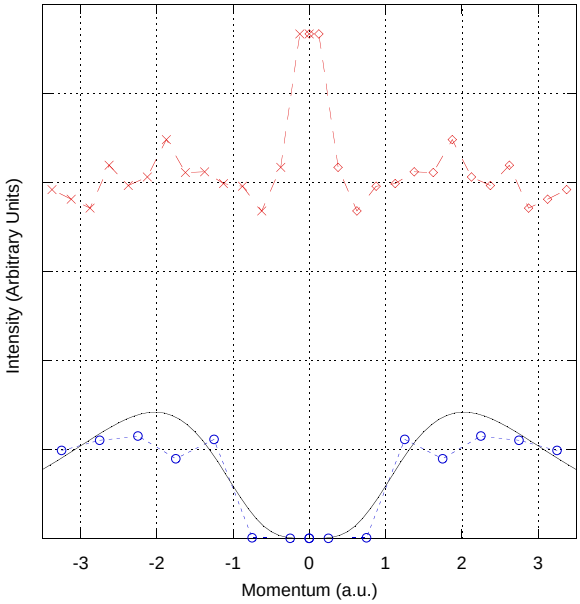


Fig. 9. Plot of the d-level for a 30 Å film (dotted line), a 350 Å (dashed line) and a Hartree-Fock calculation of the atomic Ge 3d momentum profile (solid line).

further the 350 Å amorphous Ge sample was annealed by electron bombardment into the polycrystalline form Fig. (2). In Fig. (10) we present the a comparison of the energy plots at the top and bottom of the band. While the polycrystalline sample exhibits less intensity at the top of the band it is hardly significant and not enough to explain the differences observed in the simulations.

In all of the samples the degree of inelastic scattering seems to be underestimated by the simulation though qualitative agreement is achieved. This is to be expected as intrinsic satellites are not present in the LMTO calculation, used as an input. This takes intensity away from the main quasi-particle peak and causes additional intensity at larger binding energy. Other possible causes that could contribute are errors in the experimental determination of the film thickness and/or uncertainties in the mean free path used. However, these combined errors are certainly less than 50%, and such a large value would still account only in part for the additional intensity seen at large binding energy. Thus the additional intensity at large binding energies are interpreted as mainly due to intrinsic satellites.

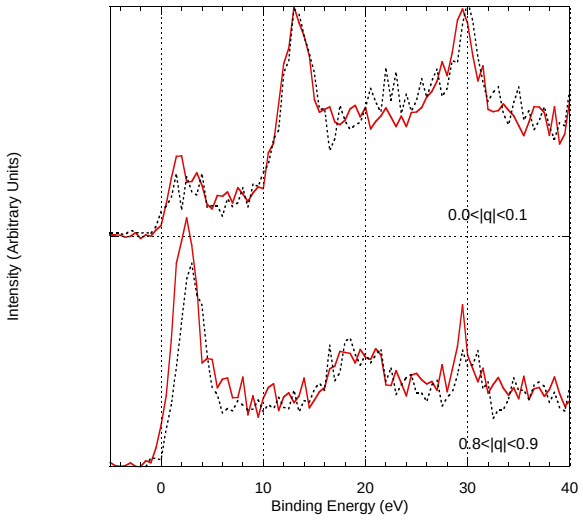


Fig. 10. Comparison of the bottom and top of the band for polycrystalline (dashed) and amorphous (solid) 350 Å germanium.

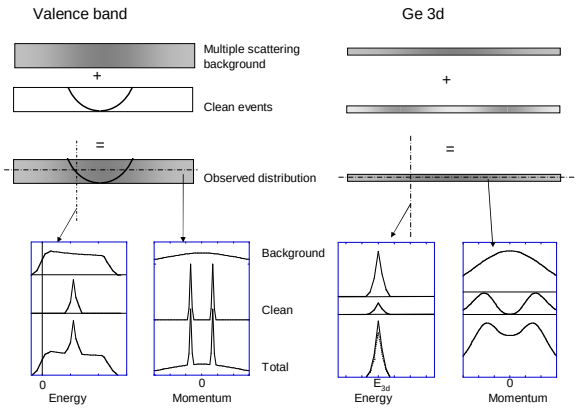


Fig. 11. The different effects of elastic scattering on the valence band and the core level. For the valence band the elastic scattering causes a smooth background under the sharp features due to the clean events, for both the energy spectrum and momentum profiles. For the Ge 3d level the elastic scattering processes do not cause a background in the energy spectrum, but influences the shape of the momentum profile. In these sketches the total intensity of the background due to elastic scattering is always larger than the area of the clean events, as is the case in the experiment, for all, but the thinnest film.

4. Summary and Conclusion

In this paper we have described how features in the electron momentum densities of Ge are affected by inelastic and elastic scattering. The elastic scattering appears to have a different effect on the dispersing valence band, compared to the non-dispersive core level, as summarized again in fig. Fig. (11). It is important to consider when analyzing these spectra how these contributions vary as the film thickness increases and how this can cause differences in the resulting spectra. It is also important that the films be thick enough such that contributions from the carbon backing are insignificant. Monte Carlo simulations allow an insight into the effects of these phenomena but further work is required particularly in the treatment of inelastic scattering.

5. Acknowledgement

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