# Investigation of the Structure of Amorphous Substances by Means of Electron Diffraction

Jürgen Ankele, Joachim Mayer, Peter Lamparter, and Siegfried Steeb

Max-Planck-Institut für Metallforschung, Institut für Werkstoffwissenschaft, Seestraße 92, D-70174 Stuttgart

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The structure factor of amorphous germanium was determined using 120 kV electrons, an Ω-filter for the elimination of inelastically scattered electrons and a correction procedure for multiple scattering. The structure factor thus obtained is in good accordance to that obtained with X-rays and neutrons, respectively.

## 1. Introduction

The main problems in performing reliable quantitative electron diffraction experiments with amorphous specimens are the strong inelastic and the strong multiple scattering of the electrons. Compared to the experimental facility as used in a previous work [1] we describe in the present paper the use of a commercial electron microscope equipped with a so-called omega-filter. Useful energy filtering has earlier been applied to the study of amorphous materials by Cockayne et al. [2] and Reimer et al. [3]. In the present study for the first time an imaging omega filter and a slow scan CCD-camera as detector was applied. Concerning the correction for multiple scattering, the method applied in [1, 4] is improved.

Using the novel methods we will evaluate the structure factor of amorphous germanium and compare it with the structure factor as obtained using X-rays and neutrons.

## 2. Theoretical Background

### 2.1. Coherently Elastically Scattered Electrons

The structural information obtained by scattering of electrons with an amorphous element is given by the structure factor $S(Q)$, which is calculated from the normalized coherently scattered intensity per atom $I_{\mathrm{coh}}$ according to

$$S(Q) = I_{\mathrm{coh}}(Q)/f^2(Q) \tag{1}$$

with $Q = 4\pi(\sin\Theta)/\lambda$ = modulus of the scattering vector, $\Theta$ = half of the scattering angle, $\lambda$ = wavelength, $f(Q)$ = scattering factor.

The pair correlation function $G(R)$ for the case of spherical symmetry is obtained from $S(Q)$ by the equation

$$G(R) = 4\pi R[\varrho(R) - \varrho_0] = \frac{2}{\pi}\int_0^\infty Q[S(Q) - 1]\sin(QR)\,\mathrm{d}Q \tag{2}$$

with $R$ = coordinate in real space, $\varrho(R)$ = local atomic number density, $\varrho_0$ = average atomic number density.

### 2.2. Inelastically Scattered Electrons

Electrons may loose energy by the excitation of plasmons, by inner shell excitations, by thermal diffuse scattering, and due to Compton scattering. In the present experimental arrangement we use an omega-filter with its energy resolution of about 5 eV. Thus besides the elastically scattered electrons only the thermal diffuse scattered ones and part of those electrons which have undergone Compton scattering reach the electron detector.

Thermal diffuse scattering may be avoided by cooling the specimen with liquid nitrogen. Compton scattering can be neglected, since Compton scattering with $\Delta E \leq 5$ eV occurs at $Q$-values of maximum 15 $\mathrm{nm}^{-1}$, which is smaller than the $Q$-value of the first maximum of the structure factor $S(Q)$.

### 2.3. Multiple Elastically Scattered Electrons

The observed electron intensity has to be corrected for multiple scattering effects. This is accomplished

Reprint requests to Prof. Dr. S. Steeb.

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according to [5] in the slightly improved version of the one presented in [4]. The data needed to perform the correction for multiple scattering are the total number of the scattered electrons, the specimen thickness, and the mean free path for elastic scattering, which can be obtained experimentally. For the determination of the mean free path one has to determine the number of the unscattered and the scattered electrons. This is done by a separate set of experiments (see Chapter 4).

### *2.4. Evaluation and Normalization of the Experimental Intensity*

Since no correction for polarization and absorption is necessary, the following relationship can be used for the extraction of the coherently scattered intensity $I^{eu}_{coh}(Q)$ from the experimentally measured intensity $I_{exp}(Q)$:

$$I_{exp}(Q) = \beta(I^{eu}_{coh}(Q) + I^{eu}_{msc}(Q)) \tag{3}$$

with $I_{exp}(Q)$ = measured intensity,
$\beta$ = normalization constant (see below),
$I^{eu}_{coh}(Q)$ = coherently scattered intensity per atom in electron units (eu),
$I^{eu}_{msc}(Q)$ = multiple scattered intensity per atom in eu,
1 eu $= 7.9 \cdot 10^{-30}$ m$^2$/sterad.

The determination of the normalization constant $\beta$ was done according to [6].

## 3. Experimental

### *3.1. Sample Preparation*

Amorphous germanium-films with 20 nm thickness were prepared in a high vacuum evaporation apparatus using a directly heated tungsten boat as source and a freshly cleaved rocksalt substrate at room temperature. The thickness was controlled by an oscillating quartz sensor.

### *3.2. Diffraction Apparatus*

The electron diffraction experiments were performed with an energy filtering transmission electron microscope (Zeiss EM 912 Omega, Carl Zeiss, Oberkochen) using an accelerating voltage of 120 kV. Between the specimen and the detector, the diffracted electrons pass an imaging magnetostatic so-called omega-filter which corrects imaging errors up to the second order. Figure 1 schematically shows the design of the filter [7, 8]. The diffracted electrons are deflected by four magnetic prisms. By this an energy loss spectrum is projected in the final energy dispersive plane where the inelastically diffracted electrons are eliminated by a slit aperture. The width of the slit determines the width $\Delta E$ of the energy window, which for the present investigation was adjusted to $\pm 5$ eV.

As detector a slow scan CCD-camera (CCD = Charge-Coupled Device; manufacturer: Gatan, Pleasanton) is used, which is equipped with a YAG-scintillator (YAG = Yttrium-Aluminum-Garnet). The spatial resolution amounts to 1024 × 1024 pixel with a maximum of 4096 counts per pixel. The primary beam is eliminated by a beam stop.

## 4. Results and Discussion

Figure 2 shows the intensity curve of amorphous Ge as obtained with and without energy filtering. Obviously, the peak to background ratio is substantially improved by applying the $\Omega$-filter.

Figure 3 shows the structure factor of amorphous Ge as obtained with and without correction for multiple scattering. The structure factor as obtained without correction for multiple scattering is rather small at $Q$-values below about 50 to 60 nm, which according to [4] points to the necessity of performing a correction for multiple scattering. For the correction procedure we need the total scattered intensity, the thickness of the specimen, and the mean free path for elastic scattering of the electrons.

The thickness $D$ of the specimen was measured during the evaporation process and results in about 20 nm. The mean free path $L$ follows from the equation

$$I_{scatt} = (I_{unscatt} + I_{scatt})\,(1 - e^{-D/L}) \tag{4}$$

with $I_{scatt}$ = intensity of the scattered electrons,
$I_{unscatt}$ = intensity of the unscattered electrons.

The determination of $I_{scatt}$ and $I_{unscatt}$ is not possible from a single CCD-camera image because in the diffraction pattern all unscattered electrons are focused into a small primary spot. Since the dynamic range of the CCD-camera is limited to a maximum of 4096 counts per pixel, the intensities scattered into the amorphous rings in the diffraction pattern are very low even if the spot formed by the unscattered elec-

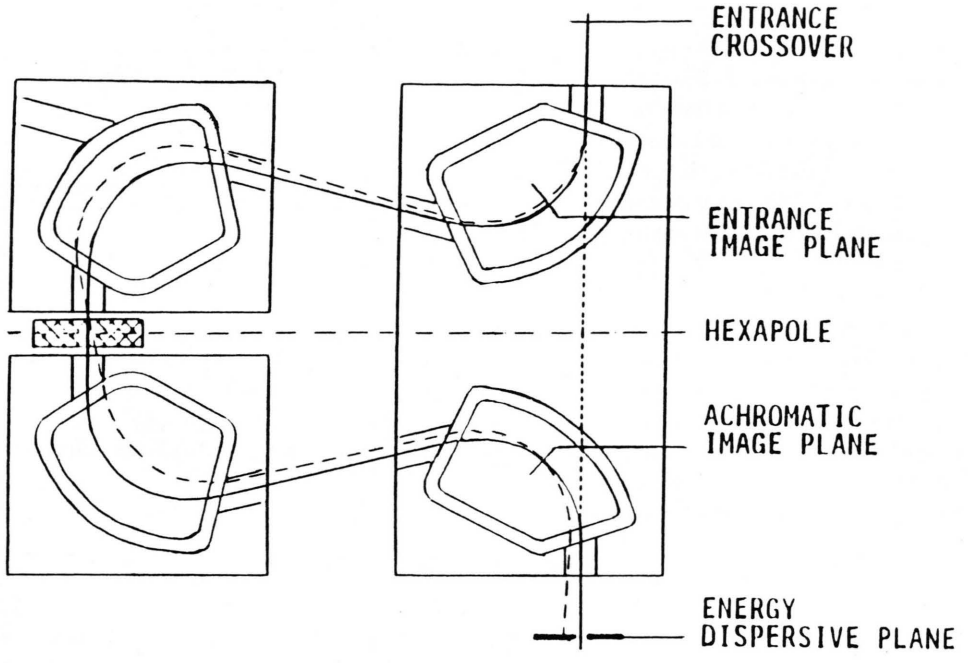


Fig. 1. Schematic design of the Ω-filter [8]; (——) path of elastically scattered electrons; (– – –) path of inelastically scattered electrons.

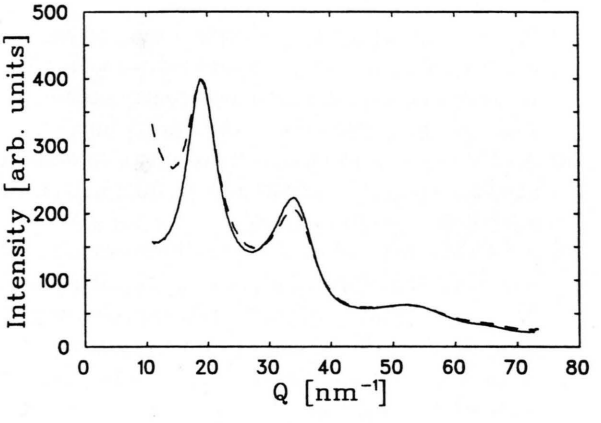


Fig. 2. Electron diffraction pattern of amorphous germanium at 120 kV; intensity vs. $Q$; intensities normalized at the first maximum; (——) with Ω-filter; (– – –) without Ω-filter.

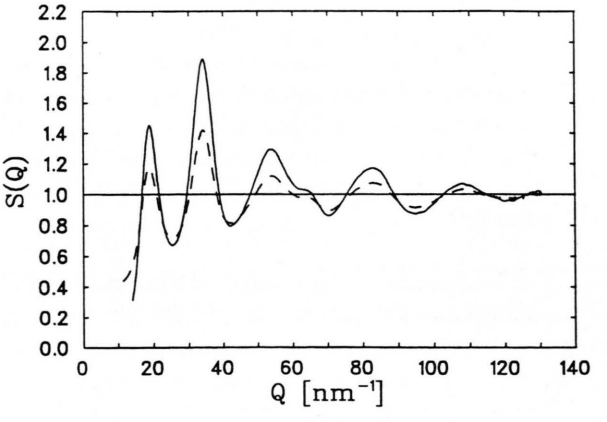


Fig. 3. Electron diffraction pattern of amorphous germanium at 120 kV; structure factor $S(Q)$; (——) corrected for multiple scattering; (– – –) without correction for multiple scattering.

trons is brought close to saturation. For the used specimen thickness of 20 nm, the ratio of the unscattered electrons per pixel in the primary spot to the scattered electrons per pixel at the first diffraction maximum is about 10 000, which already exceeds the dynamic range of the CCD-camera. Furthermore, the intensity is proportional to $(\sin \Theta)^{-4}$, which means that it drops by another factor of about 1000 from the first diffraction maximum to the largest scattering angles which were considered in the present experiments.

To overcome the difficulties produced by these enormous intensity variations, more than one measure-

ment is necessary for the evaluation of $L$:

i) Measurement of the flux of all scattered electrons: A diffraction pattern is recorded at the smallest camera length with high intensity of the incident beam. During this experiment the beam stop has to be applied in order to prevent the very intense primary spot from damaging the CCD-camera. The exposure time is chosen such that the first diffraction ring is close to saturation.

ii) Measurement of the flux of all unscattered electrons: A diffraction pattern is recorded at a camera length at which the primary spot and part of the first diffraction ring fit into the CCD-array. The intensity of the incident beam has to be substantially reduced and the exposure time short (1 sec) so that the primary spot is not saturated. In this pattern the first ring is not visible because of the low intensities.

iii) In an additional measurement, a normalization factor between the patterns obtained in i) and ii) is determined. This additional step is necessary because the modifications in incident beam intensity and camera length between i) and ii) are not calibrated with the required accuracy. The third pattern is recorded under identical microscope conditions as in ii), but the beam stop is applied and the exposure time is increased by a factor of 100. Since the first diffraction ring now appears with an intensity increase defined by the exposure time, its radius and total intensity can be used to normalize the camera length and intensity in i) and ii).

The evaluation of these three measurements with the amorphous germanium-film yielded the following data:

$$I_{\text{unscatt}} = 6.9 \cdot 10^8 \text{ electrons};$$
$$I_{\text{scatt}} = 1.8 \cdot 10^8 \text{ electrons}.$$

According to (4) we finally obtained $L = 86$ nm.

The single scattering intensity, which results from the correction procedure, can be well normalized and is presented as full line in Figure 3.

In Fig. 4 we present again this corrected curve (——) and compare it with the X-ray scattering result from [9] (– – –) and the neutron scattering result from [10] (· · · ·). The accordance between these three structure factors, which were obtained with three different radiations, is surprisingly good. Compared to [1] this good

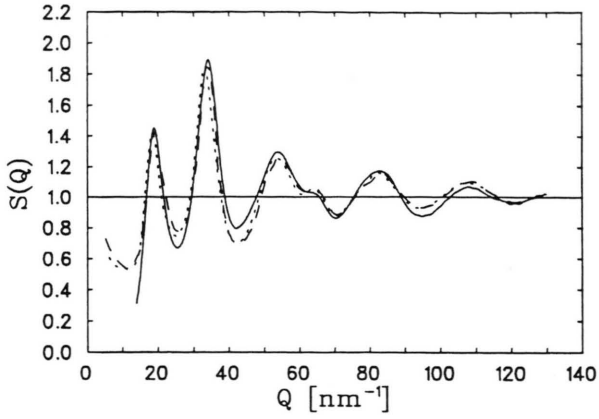


Fig. 4. Amorphous germanium; structure factor $S(Q)$; (——) electron diffraction (present paper); (– – –) X-ray diffraction [9]; (· · · ·) neutron diffraction [10].

accordance was mainly achieved by the following items:

i) An accelerating voltage of 120 kV was used instead of 50 kV as in [1]. Increasing the electron energy means to increase the mean free path for elastic scattering so that the ratio of multiple scattered electrons to single scattered electrons decreases.

ii) Rather monoenergetic electrons were used emerging from a $LaB_6$-source compared with the electrons from a cold gas discharge source used in [1] with its widely spread energy distribution.

iii) An $\Omega$-filter was applied with an energy resolution smaller than 5 eV compared to the filter in [1] with a resolution of 10 to 15 eV.

iv) A CCD-camera allows for two dimensional registration of the diffraction patterns with the possibility to eliminate partly crystalline specimens and specimens which show anisotropic scattering patterns. This is a big advantage compared to the one dimensional registration used in [1].

v) In the present work the parameters needed to perform the correction for multiple scattering were determined by electron diffraction experiments whereas in [1] these parameters were obtained by optimization.

## 5. Conclusion

Electron diffraction with amorphous germanium was performed using an electron microscope (EM 912

Omega) equipped with an energy filter. Thus inelastic scattering was eliminated. A correction for multiple scattering was performed. The structure factor as obtained by this procedure is in good accordance with that obtained by X-ray- and neutron-diffraction.

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