

# Fluctuation electron microscopy of a bulk metallic glass

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Bulk metallic glasses (BMG) are currently the focus of intense research in the materials community due to their potential for structural applications [1]. The attractive properties as very high strength and high elastic limits are closely related to their atomic arrangement. Therefore, a quantitative description of the structure of BMG is necessary to understand their properties. Unlike crystalline materials, the structure of BMG is characterized by the lack of long-range periodicity as in an amorphous structure; still it is assumed that short-range order (SRO) and medium-range order (MRO) are present in BMG. Conventional diffraction methods, e. g. by X-rays or electrons are only sensitive to SRO involving atoms of neighbouring shells. In order to study atomic correlations beyond SRO, new methods are required that are sensitive to detect correlations of MRO (0.5 to 3 nm). Here, we use fluctuation electron microscopy (FEM) [2] to study atomic correlations of a BMG on the MRO scale. FEM is a spatially resolving diffraction technique that is sensitive to structural fluctuations on nanometer scale.

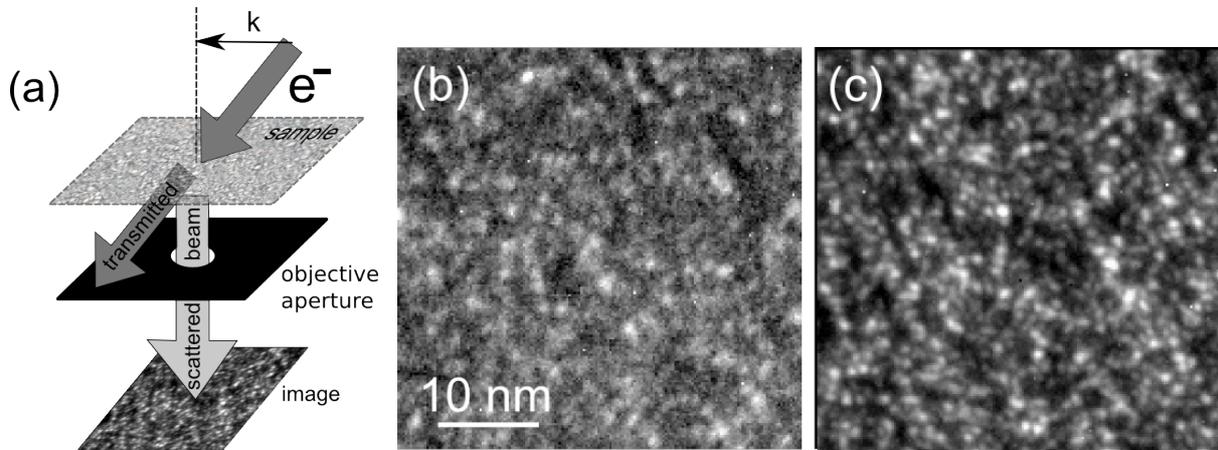
Samples of CuZrAlAg, a BMG produced by copper mould suction casting, were prepared by electropolishing to achieve TEM foils. FEM experiments by taking tilted dark-field images (cf. Fig. 1(a)) were carried out in a TEM operating at 120 and 200kV. The dark-field images show intensity variations due to local structural correlations on the MRO scale (cf. Fig. 1(b,c)). In order to measure MRO, dark-field images taken at different scattering vectors  $k$  were analyzed statistically by calculating the normalized variance  $V(k)$  of the image intensity  $I(k,r)$  [2]:

$$V(k) = \frac{\langle I(k,r)^2 \rangle - \langle I(k,r) \rangle^2}{\langle I(k,r) \rangle^2} - 1, \text{ where } \langle \rangle \text{ means averaging over sample position } r.$$

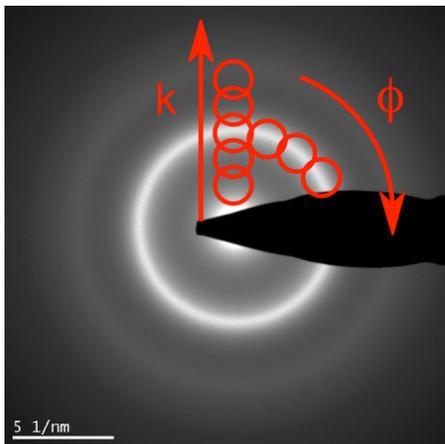
Since  $V(k)$  depends on the imaging condition, the exact focus found by maximizing  $V(k)$  in the image as well as constant condenser settings and no specimen drift are mandatory. Before calculating  $V(k)$  the dark-field images were corrected for instrumental effects (e.g. modular transfer function, read-out noise) due to the acquisition using a CCD camera. In order to increase the reliability of  $V(k)$  a set of dark-field images was taken by varying the scattering vector  $k$  and the angle  $\varphi$  (cf. Fig. 2). The acquisition of dark-field images probing the reciprocal space between given values of  $k$  and  $\varphi$  was automated by a script running under "Digital Micrograph". Averaging over  $\varphi$  was done in two alternative ways: (i)  $V(k)$  was calculated from images summed over  $\varphi$  or (ii)  $V(k)$  calculated for all images was averaged over  $\varphi$ . Both averaging methods lead to curves of the normalized variance as a function of  $k$  with similar characteristics but the absolute values are different (cf. Fig. 3). A peak in the curve indicates that intensity speckles in the images measured by the normalized variance are especially pronounced at a given  $k$ -value ( $4.2 \text{ nm}^{-1}$ ). The scattering vector of the maximum contains information of the MRO structure. It is interesting to note that in crystalline B2 ordered CuZr the strongest reflection corresponding to (110) has a very similar  $k$ -value indicating some similarity in the atomic correlations as in the MRO of the BMG (cf. Fig. 3).

In order to get information on the correlation length sets of dark-field images were taken with different objective apertures (2, 5, 10, 20  $\mu\text{m}$ ). Both the size of the objective aperture used to form the image and the wavelength of the electrons determine the resolution of the FEM images. By varying the resolution the sample is probed with respect to MRO on different length scales. The corresponding calculated  $V(k)$  curves show the same characteristics with a peak at  $4.2 \text{ nm}^{-1}$ ; as an example the curves obtained from images taken with 5 and 10  $\mu\text{m}$  objective apertures are shown in Figure 4. Plotting the peak values of  $V(k=4.2 \text{ nm}^{-1})$  as a function of resolution yields a curve containing two maxima (cf. Fig. 5). Two maxima in  $V$  as a function of resolution indicate the presence of two MRO correlation lengths of about 0.7 and 2.3 nm in the amorphous structure. The observed MRO correlation lengths can be linked to the size of individual clusters and to the correlation between similarly oriented clusters.

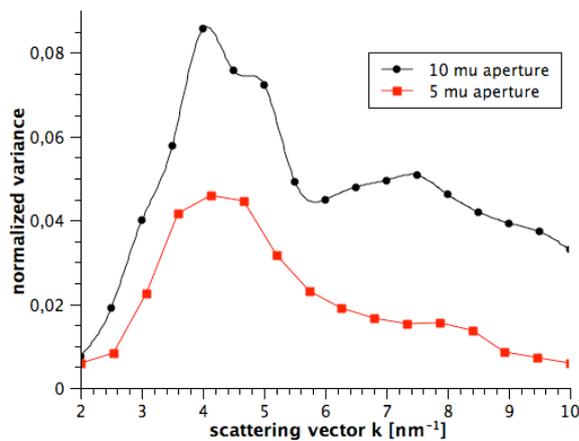
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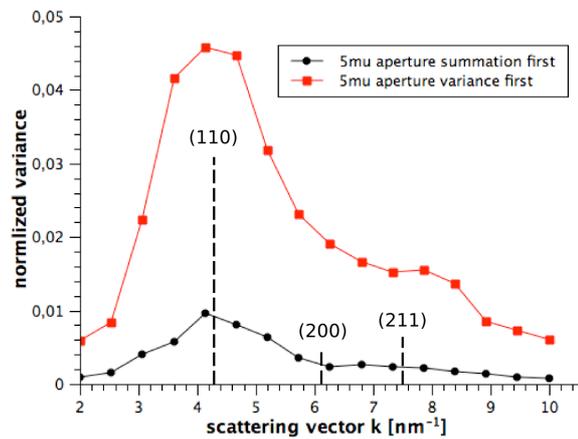
**Figure 1.** (a) Schematic drawing of the FEM technique using tilted illumination. Examples of tilted dark-field images showing low (b) and high values (c) of the normalized variance.



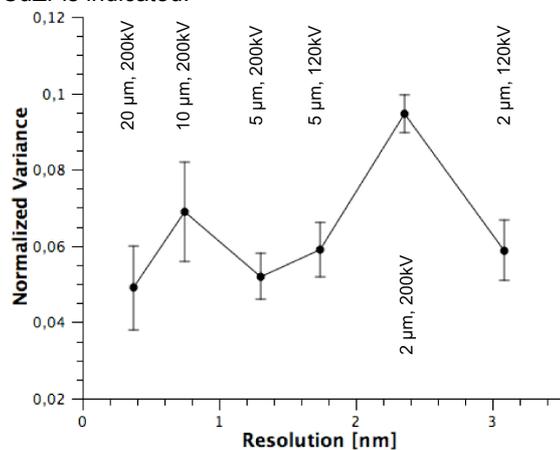
**Figure 2.** Diffraction pattern of CuZrAlAg bulk metallic glass is characterized by diffuse rings. For the FEM analysis a set of dark-field images is taken by varying the scattering vector  $k$  and the angle  $\phi$ . The rings illustrate the different positions of the objective apertures used to form the dark-field images.



**Figure 4.** Plot of  $V(k)$  calculated from sets of dark-field images taken with different objective apertures (5 and 10  $\mu\text{m}$  in diameter).



**Figure 3.** Plot of the normalized variance  $V(k)$  of the intensity of images taken with a 5  $\mu\text{m}$  objective aperture. The two curves differ by the method of averaging over the angle  $\phi$ . Lower values of  $V(k)$  are obtained by calculating  $V$  of images summed first over  $\phi$ . The position of structure factors  $(hkl)$  of B2 ordered CuZr is indicated.



**Figure 5.** Plot of the normalized variance as a function of the resolution determined by the size of the objective aperture and the acceleration voltage.