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Estimation of the electron beam-induced specimen heating and the emitted X-rays spatial resolution by Kossel microdiffraction in a scanning electron microscope

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A B S T R A C T

A Kossel microdiffraction experimental setup has been developed inside a Scanning Electron Microscope for crystallographic orientation, strain and stress determination at a micrometer scale. This paper reports an estimation of copper and germanium specimens heating due to the electron beam bombardment. The temperature rise is calculated from precise lattice parameters measurement considering different currents induced in the specimens. The spatial resolution of the technique is then deduced.

1. Introduction

When an electron beam is focused on a material, various elastic and inelastic interactions can occur between electrons and the material. During inelastic scattering, most of the energy created is dissipated as heat within the specimen, causing a local temperature rise. When considering scanning electron microscopy, a conductive bulk specimen is normally used and the radial heat flow profile is three-dimensional, leading to a relatively small temperature rise in a stationary probe and even lower in a scanning probe [1]. However, materials with low thermal conductivity or thin films are more sensitive to heating [2]. The temperature rise in the zone directly impacted by the beam can be reduced by decreasing the accelerating voltage and the probe current or by applying a highly thermal conductive coating [3]. Kossel microdiffraction is a tool that has been adapted for use in the Scanning Electron Microscopes (SEM), which enables to determine not only the crystallographic orientation, but also the inter- and intragranular strain and stress state while observing the microstructure [4]. Since high probe currents are necessary for the technique, like Wavelength-Dispersive X-Ray Spectroscopy microanalysis, the temperature rise and the spatial resolution need to be estimated. Two main approaches have been used to measure the temperature rise in a material which is irradiated by an electron beam in a scanning electron microscope. The temperature profile can be first directly measured by a thermocouple but the spatial resolution of the device has to be less than one micrometer. A good candidate is a thin film thermocouple because of its high resolution [5]. Another method is to follow the melting of low fusion point samples using different electron beam illuminations [6]. The purpose of this paper is to propose a third solution to correctly estimate the temperature rise in materials bombarded by an electron beam, by taking advantage of the precise lattice parameters determined using the Kossel microdiffraction technique. Similar analyses have been carried out by Harris [7] in 1974 to obtain lattice parameters on Swedish steel, taking into account the possible specimen heating. With the development of powerful CCD camera detectors, the quality of the Kossel patterns is considerably improved, leading to a 10^{-5} precision in lattice parameters determination. This paper will first present how to obtain crystal lattice parameters by Kossel microdiffraction. Next, lattice values obtained at different electron beam currents on copper and germanium specimens, chosen for their differences in thermal conductivity, will be shown. The specimens' heating and the spatial resolution of Kossel microdiffraction will then be deduced according to theoretical formulations governing electron-beam-induced heating.

2. Principle of the technique

Kossel microdiffraction is a local analysis tool – the analysis volume is a few cubic micrometers – based on X-ray diffraction inside a scanning electron microscope. The crystallographic

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orientation, the lattice parameters and the full elastic strain tensor with a strain resolution of about 3×10^{-4} [8] can be deduced from experimental patterns using a semi-automatic program, KSLStrain, developed by Adam Morawiec [8].

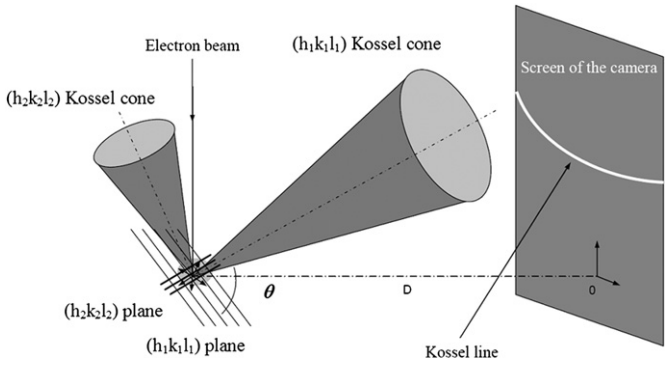

Fig. 1. Kossel microdiffraction: generation of Kossel line patterns.

When an electron beam is focused on a crystalline material, the latter is excited and emits fluorescent X-rays. A part of these X-rays is diffracted by the crystallographic planes forming Kossel cones, in accordance with the Bragg's law; the wavelength of Kossel interferences is that of the sample X-ray characteristic emission. The sample being a multidirectional X-ray source, several reflections occur simultaneously, each cone corresponding to one (h k l) diffracting plane (Fig. 1).

Kossel patterns are obtained in a JEOL 5800LV scanning electron microscope with a tungsten filament used as a source for thermionic emission. A secondary electron (SE) detector type Everhart–Thornley allows to see the sample microstructure and to choose precisely the area to probe with the electron beam. A chemical analysis of the sample can be performed using Energy-Dispersive X-ray Spectroscopy (EDS). The Kossel lines are observed using a very high-resolution X-ray digital camera from Photonics Science Limited featuring: direct straight coupled fiber-optic input, water cooled, on chip-up to 8×8 pixel binning, sub-area readout up to 4008×2672 pixel resolution, 9 μm^2 pixel size and motorized mechanical interface to the microscope.

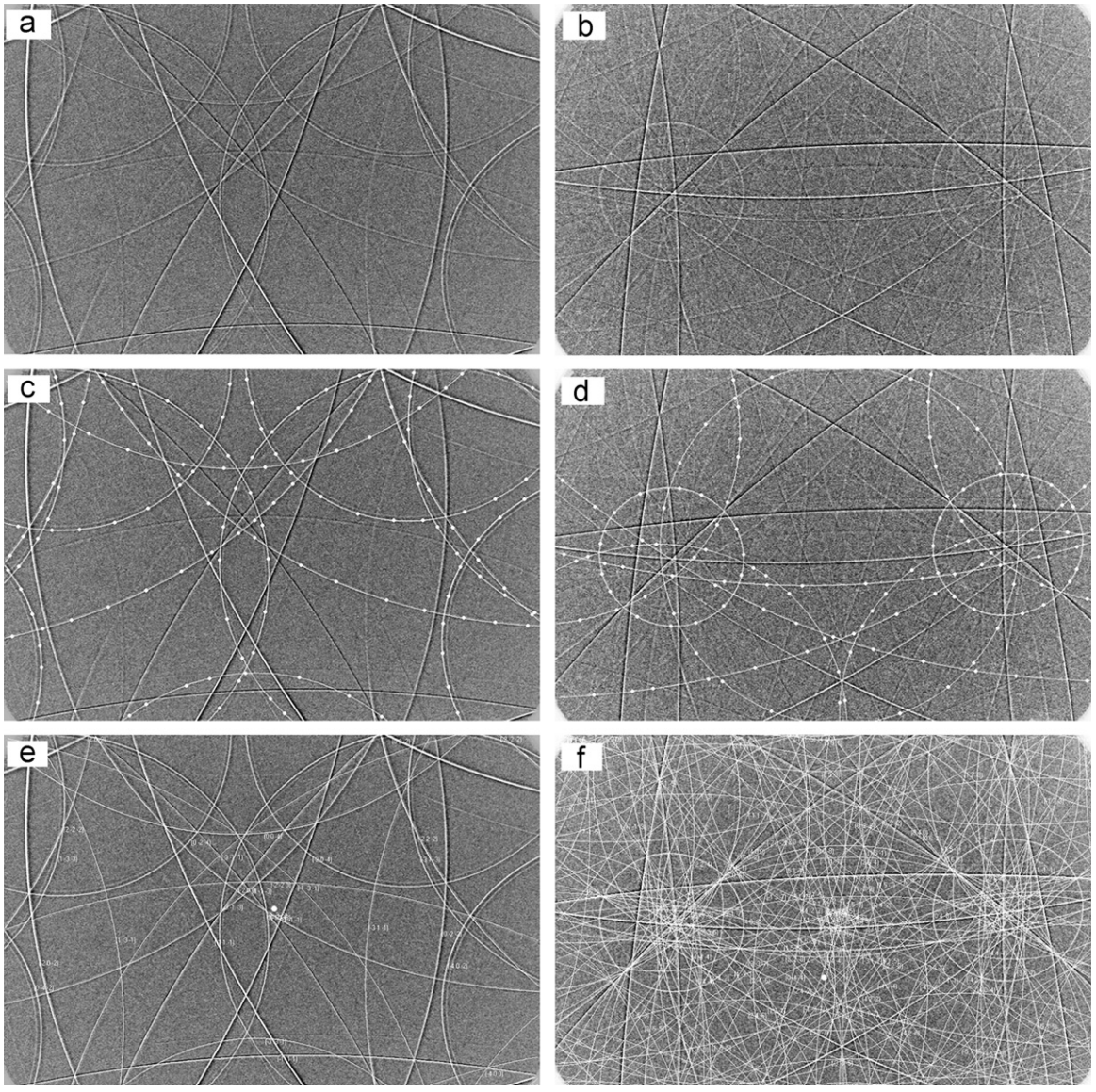

Fig. 2. Experimental Kossel line pattern for copper (a) and germanium (b) – conics manually marked on patterns for copper (c) and germanium (d) – simulated Kossel line pattern after the refinement for copper (e) and germanium (f).

Our approach to estimate the heating due to the electron-beam bombardment was to take patterns at different electron beam powers. Lattice parameters were then found for each beam power. Knowing the linear thermal coefficient of the material, the temperature rise could be deduced from the lattice dilatation. Two stress-free single crystals were chosen: one in germanium and one in copper. The germanium sample was part of a micro-electronic wafer provided by the CEA (French Atomic Energy Research Centre) and the copper sample was previously electro-polished. These two materials were chosen because their thermal conductivities are very different.

Kossel patterns were taken by focusing the electron beam on the same location on the samples surface, randomly chosen, to avoid lattice parameter fluctuations that can result from small changes in chemical composition or in surface preparation. Two sets of Kossel patterns were taken for the two materials to optimize the reliability of the technique. For germanium, it was decided to analyze two different samples. For copper, the evolution of the lattice parameter was followed at the same location with increasing and with decreasing electron beam power.

An accelerating voltage E_0 of 30 kV was taken and the absorbed current I_{abs} was measured by means of a picoammeter. As samples are conductive bulks, a linear relationship was found between I_{abs} and the probe current, I_0, by measuring the latter with a Faraday cage:

$$I_0 = 1.56 \times I_{abs} \tag{1}$$

The electron beam power W considered is

$$W = E_0 \times I_0 \tag{2}$$

The electron beam currents were taken from about 100 nA to 1.5 μA.

Examples of experimental Kossel line patterns are shown in Fig. 2a and b. Several parameters need to be fulfilled in the program to index the obtained patterns: the radiation wavelength of the emitted X-rays that is known with relatively high accuracy, the unit cell of the material, the pixel size of the camera and approximate values of the camera geometry parameters (sample-to-detector distance and location of the pattern center). Moreover, conics on the pattern need to be marked manually selecting a number of points per Kossel line. A number of ten and fourteen Kossel lines were marked for germanium and copper, respectively (Fig. 2c and d), and those corresponding to high diffracting planes were preferred because they are more sensitive to the lattice values.

Starting with approximate reference values of lattice parameters, a specific procedure is used to find the geometry of the pattern. Once the crystal orientation is determined and the camera geometry parameters are tuned, the program can proceed with the refinement of the lattice parameter. The full strain tensor values in the crystal coordinate system were also refined to check if samples were really stress-free and if values were similar between the different electron beam powers. The program uses the so-called "K-line equation based scheme" [8]. Kossel conics manually marked by the operator are matched to corresponding conics in simulated patterns (Fig. 2e and f).

Lattice parameter uncertainties, given by KSLStrain, are 6×10^{-5} for copper and 2×10^{-5} for germanium. Four additional areas were analyzed for a given electron beam power, on the germanium single crystal. They were chosen fifty micrometers apart (in four different directions) from the location of the primary spot. A standard deviation of about 2×10^{-5} was found.

3. Temperature rise and spatial resolution

The local temperature rise $\Delta\vartheta$ induced by the electron beam on a bulk specimen can be calculated from Eq. (3) [9]:

$$\Delta\vartheta = \frac{W}{2\pi JC}\left(\frac{1}{r} - \frac{1}{R}\right) \tag{3}$$

where W is the power supplied by the electron beam (W), J is the Joule conversion coefficient (4.18 J cal^{-1}), C is the thermal conductivity of the specimen (cal s^{-1} K^{-1} cm^{-1}), R is the dimension of the specimen and r is the distance from the probed zone on the material corresponding to the X-ray source (cm). Since the size of the specimen used in a SEM is typically of the order of 1 cm^3 and r is in the range of micrometers, the $1/R$ term can be neglected:

$$\Delta\vartheta = \frac{W}{2\pi JCr} \quad \text{or} \quad \Delta\vartheta = \frac{W}{2\pi\lambda r} \tag{4}$$

where λ is the thermal conductivity of the target (W m^{-1} K^{-1}).

Considering a_{true} the true lattice parameter without heating and a_{meas} the measured lattice parameter with beam heating, one has

$$a_{meas} = a_{true}(1 + \alpha \times \Delta\vartheta) \tag{5}$$

where α is the linear thermal expansion coefficient of the crystal (K^{-1}).

A linear relationship between a_{meas} and Wcan be then expressed as

$$a_{meas} = a_{true}\left(1 + \frac{\alpha W}{2\pi\lambda r}\right) \tag{6}$$

According to Eq. (6), there is a linear relationship between the lattice parameter and the electron beam power.

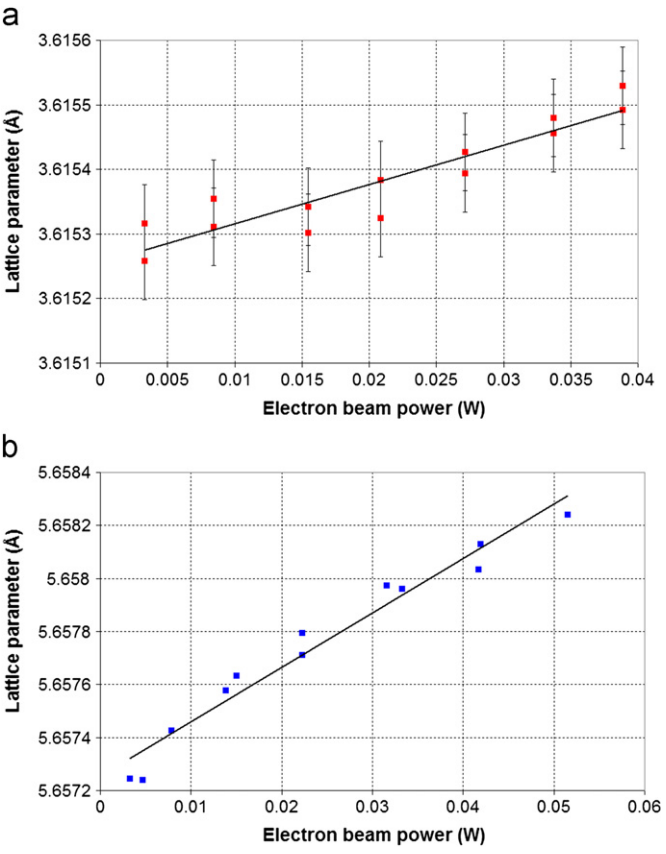

Fig. 3. Variation of the lattice parameter with the electron beam power considering both copper (a) and germanium (b) single crystals.

For each material, lattice parameter measurements and their uncertainties, given by KSLStrain, are plotted in Fig. 3. From these values, linear regressions were determined for both materials. Their equations (expressed in Angströms) are the following:

$$a_{meas}(\mathrm{Cu}) = 3.61526(\pm 3\times10^{-5}) + 0.00608(\pm 0.00133)\times W \quad (7)$$

$$a_{meas}(\mathrm{Ge}) = 5.65725(\pm 1\times10^{-5}) + 0.02057(\pm 0.00038)\times W \quad (8)$$

From Eqs. (7) and (8), assuming the coefficient of linear expansion of copper and germanium to be, respectively, 16.5×10^{-6} and $6\times10^{-6}\,\mathrm{K}^{-1}$, the local temperature rise can be expressed as:

$$\Delta\vartheta(\mathrm{Cu}) = 102(\pm 22)\times W \quad (9)$$

$$\Delta\vartheta(\mathrm{Ge}) = 606(\pm 11)\times W \quad (10)$$

The local temperature rise $\Delta\vartheta$ is the average temperature rise in the volume in which diffraction of X-rays excited by the electron beam has occurred. The maximum temperature rise ϑ_m at the center can be calculated from Eq. (11) in the case of a bulk specimen [9]

$$\vartheta_m = 1.5\times\Delta\vartheta \quad (11)$$

Several values of the temperature rise were calculated for three different electron beam powers (Table 1).

Considering these results, the local temperature rise appears to be insignificant for conductive bulk materials, even if a high probe current is applied. For example, the local temperature of a piece of copper impacted by a high energy electron beam (an accelerating voltage of 30 kV and a probe current of 1 μA) can reach 4.5 K more than the temperature without the beam. For a piece of germanium, considering the same beam conditions, a difference up to 27 K can be reached.

From these experiments and according to Eq. (4), the size of the volume required for the generation of Kossel patterns can be obtained. It was found 3.9 ± 0.9 μm for copper and 4.4 ± 0.1 μm for germanium, assuming the thermal conductivity of copper and germanium to be, respectively, 400 and $60\,\mathrm{W\,m^{-1}\,K^{-1}}$. The spatial resolution found for copper is less precise than for germanium because the copper thermal conductivity is higher so there is less lattice dilatation and because of the differences in crystallography. In fact, germanium has a diamond cubic unit cell with a high lattice parameter. So many high diffracting planes Kossel lines can be seen on patterns like small circles (Fig. 2f) which contribute to a better accuracy of the lattice parameter determination.

Interaction volume simulations based on the Monte-Carlo principle were performed in parallel using Electron Flight Simulator software, to estimate the shape and size of the emitted X-ray volume below the surface of the material. Emitted X-rays are generated X-rays which have sufficient energy to escape the sample and reach the detector. 32,000 electrons trajectories

Table 1
Specimens' heating at different electron beam powers.

Electron beam power(W)		0.003	0.015	0.03
Electron probe current(μA)		0,1	0,5	1
Cu	$\Delta\vartheta$(K)	0.3	1.5	3
	ϑ_m(K)	0.45	2.3	4.5
Ge	$\Delta\vartheta$(K)	1.8	9	18
	ϑ_m(K)	2.7	13.5	27

$\Delta\vartheta$: Local temperature rise in the volume probed and ϑ_m: Maximum temperature rise.

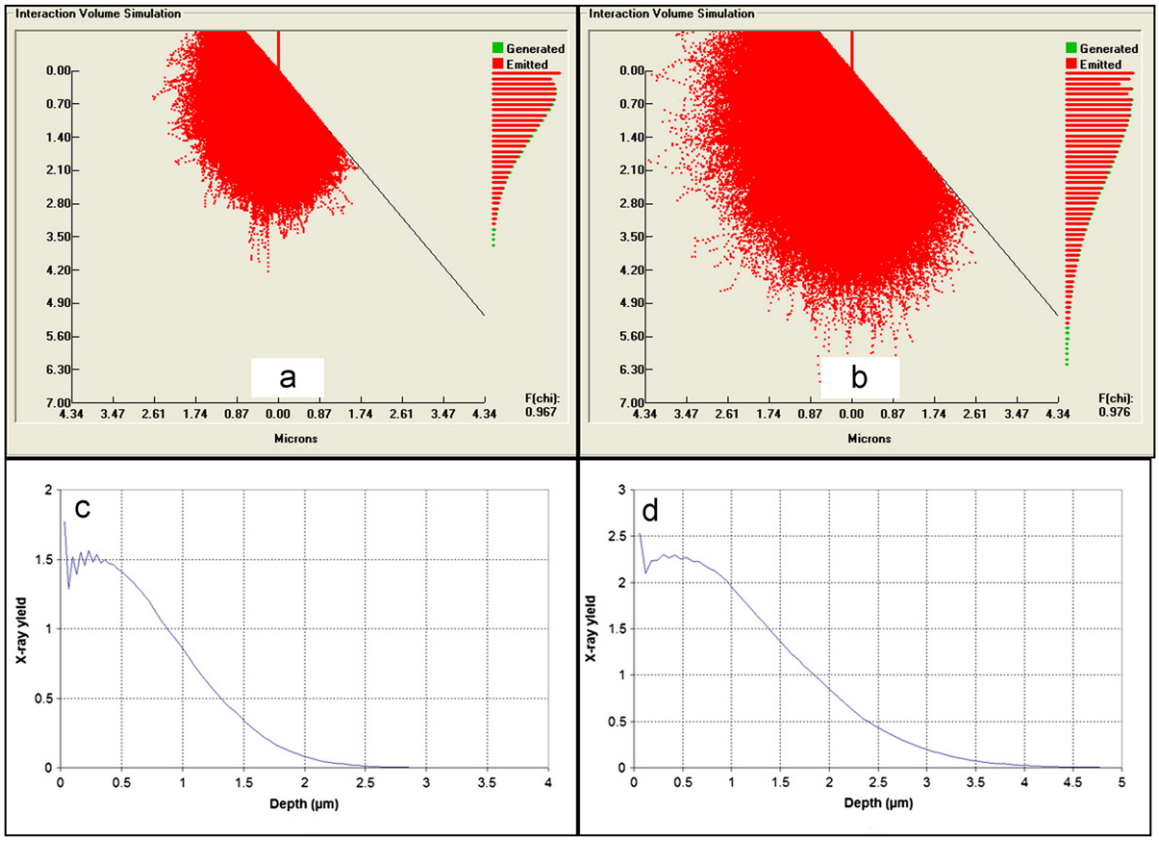

Fig. 4. Simulations of the emitted X-ray volume below the surface for copper (a) and germanium (b) and X-ray yields as a function of depth for copper (c) and germanium (d).

for a 30 kV electron beam voltage and a tilt angle of 50° were simulated, for both materials. The shape of the emitted X-ray volumes, for energies corresponding to the K-shell X-ray emission lines, were plotted on Fig. 4a and b for copper and germanium, respectively. The X-ray yields as a function of the depth below the material surface were also plotted on Fig. 4c and d for copper and germanium, respectively.

The dimensions of the emitted X-ray volume obtained for copper and germanium by Kossel microdiffraction are a little larger than the depth resolutions calculated with Monte-Carlo simulations. This small discrepancy can be explained because the Monte-Carlo software does not take into account the interaction volume widening due to an electron probe spot size that is not negligible compared to the electron diffusion volume. Moreover, a little drift of the sample during the Kossel line pattern acquisitions could lead to an underestimation of the temperature rise and an overestimation of the diffracted volume. It should be interesting to perform the same study using a field emission gun electron microscope. In fact, interaction volumes would be smaller and closer to those estimated by Monte-Carlo simulations.

4. Conclusion

Kossel microdiffraction is a characterization tool that needs high probe currents like Energy or Wavelength-Dispersive X-ray Spectroscopy. This study has shown that the temperature rise calculated from the lattice dilatation does not exceed about 5 K and 30 K for copper and germanium, respectively, when very energetic electrons (30 kV, 1 μA) collide with the sample. In addition, the spatial resolution of the technique with an accelerating voltage of 30 kV has been determined for both the materials. Values of 3.9 ± 0.9 μm and 4.4 ± 0.1 μm were found and are consistent with Monte-Carlo simulations. These results confirm that even if very energetic electrons are used, the temperature rise for a conductive material is insignificant and consequently Kossel microdiffraction can be used as a precise tool to get lattice parameter values or the full local elastic strain and stress tensors.

Acknowledgment

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