Ultramicroscopy 176 (2017) 161–169

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Local temperature measurement in TEM by parallel beam electron diffraction

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ARTICLE INFO

Keywords:
In situ microscopy
Temperature measurement
Beam parallelity
Microscope alignment
Metallic nanoparticles
Transmission electron microscopy

ABSTRACT

With the recent advances in instrumentation pushing the limits of *in situ* transmission electron microscopy, the question of local sample temperature comes into focus again. In this work the applicability of parallel beam electron diffraction to locally measure and monitor the sample temperature in TEM is assessed, with applications for *in situ* heating experiments in mind. With Au nanoparticles applied to the sample surface, temperature is measured in the range from RT to 890 °C by evaluating the change in scattering angle upon thermal expansion. Repeated measurements at constant temperature reveal a statistical precision of the method as good as 2.8 K. The applicability to locally measure the temperature is demonstrated mapping the temperature gradient across a heating chip. Owing to instantaneous response of thermal expansion to temperature changes, the method is well suited for monitoring even quick temperature changes, as demonstrated by quenching experiments. In order to enable extensive *in situ* studies, an evaluation method capable of processing large datasets with high precision is developed. Beam parallelity is identified as crucial experimental prerequisite and a routine is established, optimizing the microscope alignment in terms of beam parallelity. Apart from establishing a procedure for local temperature measurement, the present work demonstrates the unique capabilities of MEMS-based *in situ* heating equipment.

1. Introduction

From the early days of transmission electron microscopy (TEM), the questions of sample temperature and how to measure it have ever been intensively and controversially discussed. Without a heating (or cooling) device in operation, the sample temperature is expected to be close to room temperature. However the use of cold traps at liquid nitrogen temperature could lower the temperature of the sample. Moreover the sample is heated locally by the electron beam as part of the fundamental electron–matter interaction. Depending on the properties and geometry of the electron beam and the sample determining heat generation and dissipation, the deviation in temperature can be significant [1]. However, a precise knowledge of the sample temperature is important to correctly interpret the observations and measurements, *e.g.* for the determination of lattice constants presumably at room temperature.

With the rising interest in *in situ* TEM fuelled by the advances in instrumentation, the question of sample temperature regains growing importance. *In situ* heating equipment based on microelectromechanical systems (MEMS) to miniaturize the dimensions of the heater has revolutionized this field [2–4]. The small heat capacities allow rapid temperature changes at high temperature stability and vanishing drift rates, making the systems applicable for high resolution TEM (HRTEM) [5,6]. Despite the excellent performance of these devices allowing for quantitative *in situ* TEM observations [7,8], their temperature measurement relies on an external calibration. As the conditions inside the transmission electron microscope are special and probably differ from microscope to microscope, a method to precisely measure temperature *in operando* in the microscope is of high interest.

Over the past decades several methods for measuring local temperature in TEM have been developed and tested. The first attempts were based on thin film thermocouples deposited onto TEM samples [9,10] or electron diffraction to measure thermal expansion [11]. Characteristic temperatures were measured based on phase transitions, for example order transitions in AuCu [12] and the melting of In [13]. More recent works adapted this approach to measure temperature gradients from the melting and/or evaporation of Au [14] and In [15]. The reaction front corresponding to the isotherm of the phase transition temperature was monitored and its progression was used together with modelling to resolve the spatial temperature distribution [14,15].

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http://dx.doi.org/10.1016/j.ultramic.2016.11.028
Received 12 August 2016; Received in revised form 6 November 2016; Accepted 25 November 2016
Available online 07 December 2016

Mecklenburg et al. [16] developed a technique based on electron energy-loss spectroscopy (EELS) in scanning transmission electron microscopy (STEM). The minuscule shift in plasmon energy of Al structures was correlated to temperature using thermal expansion and a free electron model. Vendelbo et al. [17] employed a comparable approach to measure the temperature in a gas cell via its density determined from EELS. Winterstein et al. [18] employed selected area electron diffraction (SAED) of Ag nanoparticles to measure temperature from thermal expansion. They obtained an accuracy of ± 30 K and measured the pressure dependent influence of a gas atmosphere in environmental (E)TEM. He et al. [19] used a different approach based on the temperature dependency of thermally diffuse scattered (TDS) intensity from Ge and Si samples. Resembling the expansion thermometers known from everyday life, Gao et al. [20] and Gong et al. [21] fabricated Ga filled carbon nanotube and Au(Si) filed $\beta - Ga_2O_3$ nanotube thermometers. Temperature was determined from the movement of the meniscus due to thermal expansion of the filling.

In this work we reassess the applicability of parallel beam electron diffraction (PBED) to precisely measure sample temperature in TEM. Instrumentation has seen great advances since the seminal work by Reimer et al. [11], who has been the first to apply this approach for measuring temperature from the diffraction of polycrystalline Pb and Ni thin films. For example the development of three condenser lens systems improved the flexibility in forming the beam. Together with the increased stability of the microscope and the availability of cameras capable of recording the diffraction patterns with high resolution and high dynamics, a high accuracy achievable by electron diffraction in TEM can be expected. It is therefore highly promising to revisit electron diffraction as an alternative approach to the temperature measurement methods reviewed above. The basic principle is shown in Fig. 1a in form of a simplified beam path. An array of nanoparticles is applied to the sample surface, each acting as a single thermometer. Their temperature is read out by illumination with a parallel electron beam by acquiring a SAED pattern. Temperature is measured from the diameters of the rings observed in the diffraction pattern, correlated to thermal expansion of the crystal lattice of the nanoparticles. The observed ring pattern averages over a large number of particles in the selected area aperture (SAA) to increase the accuracy of the measurement. To resolve a temperature change of 10 K from a change in diffraction ring diameters, these have to be determined to a precision of 0.01 % assuming a thermal expansion coefficient in the order of $10 \cdot 10^{-6}$ K^{-1} (typical value for metals). This imposes high requirements on the stability of the experimental conditions in the microscope as well as the employed evaluation method.

The parallelity of the electron beam used for PBED is of crucial importance, as will be seen from the results shown in this work. Beam parallelity in this work is taken as the variation in incident beam angle over the illuminated area, as defined by Christenson et al. [22], and should not be mixed with the local beam convergence as seen by each object point describing the spatial coherence of the illumination. Beam

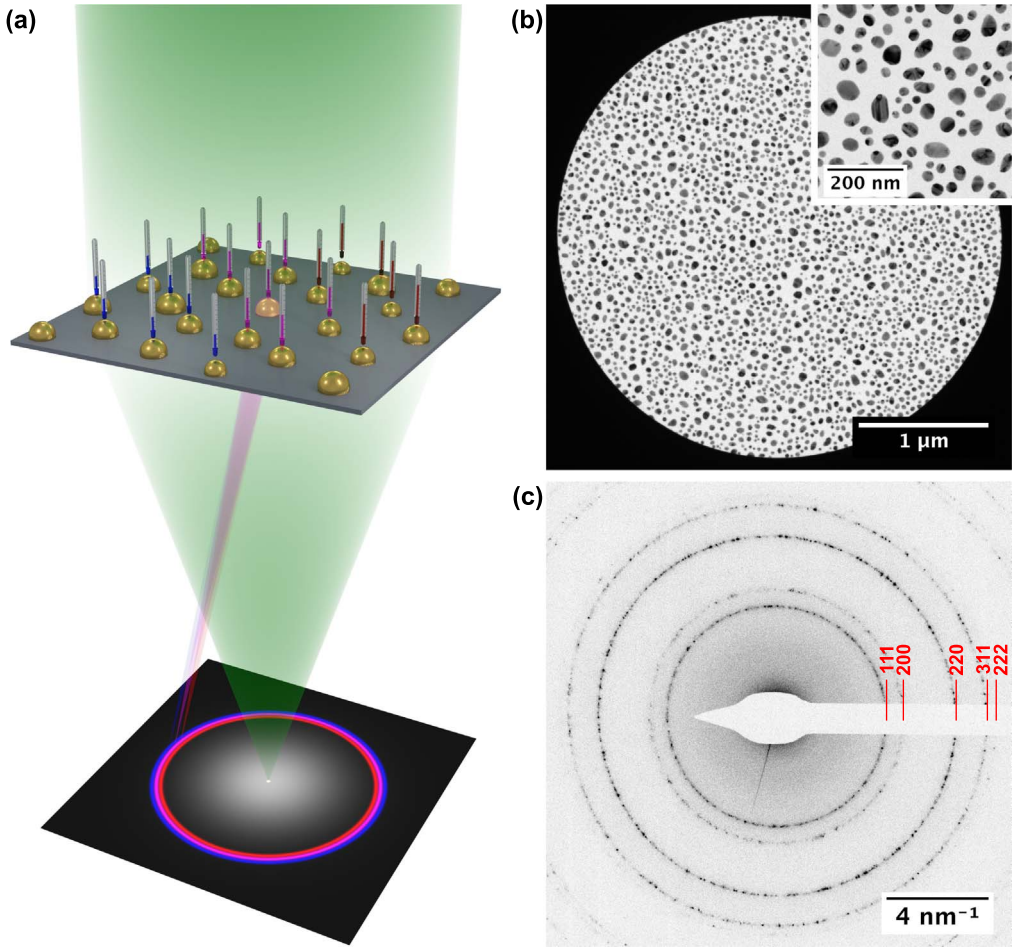

Fig. 1. (a) Schematic illustration of the approach employed to measure sample temperature in TEM. An array of nanoparticles is applied on the sample surface acting as thermometers, read out by PBED based on thermal expansion leading to changes in scattering angle. A characterization of the Au nanoparticle sample used in this work is shown in (b) the BF TEM images and (c) the corresponding SAED pattern. The size of the SAA is seen from the shadow in (b).