# Micro-scale and nano-scale strain mapping techniques applied to creep of rocks

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**Abstract.** Usually several deformation mechanisms interact to accommodate plastic deformation. Quantifying the contribution of each to the total strain is necessary to bridge from observations of

- 10 microstructures to geomechanical descriptions and, thus, is a critical component in the extrapolation from laboratory data to field observations. Here, we describe experimental and computational techniques involved in micro-scale strain mapping (MSSM), which allows strain produced during high-pressure, hightemperature deformation experiments to be tracked with high resolution. MSSM relies on the analysis of relative displacement of initially regularly spaced markers after deformation. We present two lithography
- 15 techniques used to pattern rock substrates at different scales: photolithography and electron-beam lithography. Further, we discuss the challenges of applying the MSSM technique to samples used in high-temperature and pressure experiments. We applied the MSSM technique to a study of strain partitioning during creep of Carrara marble and grain boundary sliding in San Carlos olivine, synthetic forsterite, and Solnhofen limestone at a confining pressure, *Pc*, of 300 MPa and homologous temperatures, *T/Tm*, of 0.3 to
- 20 0.6. The MSSM technique works very well up to temperatures of 700°C. Experimental developments described here show promising results for higher temperature applications.

#### **1** Introduction

During plastic deformation of a crystalline material, strain is accommodated by a variety of deformation mechanisms. These deformation mechanisms can be described as combinations of several physical processes, which might include atomic diffusion; dislocation glide, climb, and cross slip; mechanical twinning; and grain-boundary sliding. Deformation mechanisms maps are conceptual tools that divide stress-temperature-strain rate space into different fields with a particular deformation mechanism prevalent in each. Simplified flow laws are established by assuming specific micromechanical models corresponding to the prevalent mechanisms and using experimental data to fit the material constants in the theoretical flow

30 laws [Ashby, 1972; Frost and Ashby, 1982]. Such simplified flow laws are a useful first step for the extrapolation of laboratory results to larger-scale geomechanical problems, but it is also possible that the relative activity of the various deformation processes might change significantly when extended to natural conditions of rate, mean pressure, chemical environment, and temperature. Thus, quantification of the

strain due to each process and determination of the interactions amongst the processes are crucial for establishing robust microphysical models that can be used to interpret observations of natural micro- and nano-structures and that can describe natural deformation at larger spatial and temporal scales.

Quantification of the strain accommodated by each mechanism is not an easy task. Ideally, the microstructural evolution of a polycrystalline material would be monitored continuously as a sample is strained. However, high pressures and temperatures are required when deforming rocks by plastic or viscous processes in the laboratory, making microscopic observations of deformation in-situ a significant

experimental challenge. Fortunately, even step-wise observations of the amount of strain accommodated by individual intra-crystalline mechanisms could significantly enhance our understanding of high-temperature creep.

In this paper, we describe a technique for micro-scale strain measurement (MSSM), which permits strain mapping at micrometer and sub-micrometer scales and discuss several observations of strain partitioning in rocks deformed at high pressures and temperatures. To quantify the deformation, the coordinates of the material points (at the micro-scale) must be identified before and after deformation. Then, the strain tensor can be computed from their relative displacements. We used two lithography techniques, described in Section 2, to print a grid of points onto the polished surface of a rock. After deformation, we computed strain following the formulation described in Section 3. We applied the MSSM technique to four different high-temperature experiments described in Section 4.

### 2 Lithographic techniques

- The literal meaning of lithography, writing on stones, is very appropriate for our application. More
  generally, lithography refers to different printing techniques that allow transferring a pattern onto a substrate. Micro- and nano-lithography are widely used in the semi-conductor industry [Razegui, 2006; Mack, 2006] to pattern micro- and nano-metric circuits onto silicon wafers. Applying these techniques to non-conductive, heterogeneous materials is challenging, but we developed two protocols, using photolithography and electron-beam (e-beam) lithography, to produce patterns on surfaces of Carrara
- 60 marble, Solnhofen limestone, San Carlos olivine, and synthetic forsterite. The patterns had features with spatial dimensions on the order of one µm down to tens of nm. In this section, we will first describe both lithographic processes and then provide the specific protocols used for our applications. The communities developing and using micro- and nano-fabrication techniques are extremely active. Undoubtedly, new processes and improvements will allow for other innovative applications to geomaterials.

#### 2.1 Photolithography and e-beam lithography

Figure 1 summarizes the steps needed in the lithographic process. Both photolithography and electron-beam lithography require special equipment, usually present in standard clean laboratories used to fabricate solid-state devices. The process described here is commonly referred as "lift-off" in the microfabrication
community. It uses a sacrificial layer of polymer, a film that can be spread uniformly on the sample, to create a pattern on the sample surface [Razeghi, 2006]. Note that the microfabrication processes used in solid-state engineering require extra steps for ensuring the functionalities of the devices (conductivity and reflectivity among others). We present here the steps required to deposit micrometric and nanometric patterns on a rock surface: our main objective being to print a particular grid designed to track strain at a granular level. Quintanilla-Terminel [2014] describes in more detail different exploratory steps that were taken before developing the following protocols.

- I- Sample preparation: the surface to be patterned needs to be polished to a roughness of less than  $0.5-\mu m$ . The details of the polishing technique will depend on the composition, porosity, and grain size of the rock. For e-beam lithography, the sample must be flat with less than a 1- $\mu m$  variation in height for each 1-mm<sup>2</sup> region.
- II- Coating with a polymer: the surface needs then to be uniformly coated with a polymer sensitive to the energy used to transfer the pattern. A photoresist is sensitive to UV light (photolithography) and Poly(methyl methacrylate) or PMMA is sensitive to an electron beam (e-beam lithography). The coating method will depend on the sample geometry. Spin coating is the standard method for flat samples: the sample is held on a rotating device chuck by vacuum and the polymer is spread onto the surface to uniform thickness by spinning the sample at a controlled speed, typically around 3000 rpm. For more challenging geometries, such as half-cylinders or curved surfaces, a paintbrush or an aerosol spray can be used, but care must be given to ensure that the polymer thickness is relatively uniform from sample to sample.
- 90 III- Exposure with UV rays for photolithography (IIIa) or with an electron beam for e-beam lithography (IIIb). Although both techniques allow the transfer of a custom designed pattern, the process by which this is achieved is different. Photolithography necessitates the creation of a shadow mask, a glass mask imprinted with the wanted design, analogous to a negative in the photographic process. UV-light is then shined through the mask onto the sample coated with the photoresist. E-beam lithography does not necessitate a mask, instead the pattern is directly rastered with an electron beam at a specific dose (typically set around 500 to 1000 μC per cm<sup>2</sup>) onto the PMMA covering the sample. The correct exposure time and the dose of the e-beam depend on the nature of the polymer used, its thickness, the chemistry of the substrate, and the resolution of the pattern. For both photolithography and e-beam lithography, the pattern can be

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over or under exposed. Overexposed features or patterns will appear larger than designed, while those underexposed will be smaller or not appear at all.

- IV- Development of the coating: the structure of the polymer exposed to the UV light or to the electron beam changes and a chemical bath is used to dissolve this sacrificial layer. The pattern is now exposed on the sample's surface. The development time depends on the exposure time and on the resolution of the pattern. Some play exists between the exposure time and the development time; ultimately, the lithographer must determine the optimum match for the intended application.
- V- Etching: the exposed surface is slightly etched before proceeding to the metal deposition. This step was found to be important in geomaterials to allow for a good adhesion of the metal layer. The etching step can be dry (via reactive or non reactive gas) or wet (typically in a diluted HCl or HF bath). If the sample cannot be in contact with water, a dry etch would be preferable.
- VI- Metal deposition: a physical vapor deposition is used to coat the sample's surface with a thin film of a chosen metal. There is latitude in the choice of the metal sputtered. Our applications require a metal that can be seen before and after deformation and that has a minimum interaction with the rock. Therefore, parameters that are important in the choice of metal include adhesion, melting temperature, visibility, and interactions with the rock.
- VII- Dissolution of the remaining polymer: the remaining polymer is dissolved, leaving only the metal deposited onto the sample's surface.
- VIII- Patterned sample: the patterned sample is now ready to be deformed.
- Both photolithography and e-beam lithography can be used for decorating rocks; a combination of both would be also possible. The main factors to take into account are the size of the area where the pattern will be deposited, the design of the pattern (shape and resolution), and the sample geometry. Photolithography is a good choice when many samples are required, when large areas are to be patterned, and when the sample geometry is more complicated. For applications requiring a pattern with submicron features, e-beam lithography will be more appropriate. Furthermore, because e-beam lithography does not necessitate a mask, it would be faster to implement for single applications. In the following sections we provide a detailed
- protocol for each technique. With some adaptation, particularly regarding the exposure and developing steps, and the choice of the metal to be deposited, these protocols might be used on other geomaterials.

# 2.2 Photolithography applied to Carrara marble

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Photolithography was carried out at the MIT Microsystems Technology Laboratory (MTL), where half
 cylinders of Carrara marble were patterned. First, each half cylinder was polished with a succession of aluminum oxide suspensions down to a 0.5-µm particle size to produce a flat, mirror-like surface. After polishing, the pattern was produced using the following steps:

I. The polished half cylinders were dried in a convection oven at 130°C for 20 minutes.

 II. The sample surfaces were manually coated with a light sensitive photoresist, Fujifilm OCG 825 g 135 line, using a paintbrush; the thickness of the polymer was determined to be ~400 nm using a profilometer. Each half cylinder was then baked for 20 minutes in a convection oven at 90°C.

III. The pattern inscribed in a photomask was transferred onto the polymer coating by exposing the coating to ultraviolet light in a mask aligner for 60 s. The exposure time depends on the substrate, the photoresist, and photoresist thickness. Our photomask was created using computer-aided design (CAD) software to create the pattern and a laser to draw the pattern onto a glass plate covered with chrome and photoresist, followed by a typical etch-back technique [Mack, 2006]. Photomasks are also available from various commercial sources in the microfabrication industry.

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IV. The non-exposed polymer was dissolved using a developer, Fujifilm OCG 934, for 30 to 60 s.

V. The samples were then very lightly etched with an acidic solution (a 1 mass% dilute solution of HCl for 1 s).

VI. The surface of the samples was sputtered with a double layer composed of 20-nm of chromium and 30-nm of gold using a plasma sputterer.

VII. Finally, the protective layer of photoresist was removed with 2-(2-Aminoethoxy)ethanol, Nmethyl-2-pyrrolidine at 100°C for 30 min.

- 150 Photolithography is a very flexible patterning technique. Patterns can be deposited on large surfaces with a customized design, little limitation exists regarding the geometry of the sample, and a wide selection of metals can be used. For example, the 10-mm by 20-mm patterned area in Figure 2a contains roughly 1.5 million markers spaced every 10-µm and is composed of a double layer of chromium and gold. The polished surface of the outside of a Carrara marble cylinder in Figure 2b is patterned with markers spaced
- 155 every 50-µm and composed of a double layer of titanium and gold. The main disadvantages of photolithography are related to pattern resolution. Although the resolution of photolithography has been pushed to sub-micrometric scales [Dong et al., 2014], the resolution is limited to a couple of micrometers for non-conventional substrates like rocks and to hundreds of micrometers for patterns on curved surfaces. Furthermore, a photomask is needed and its fabrication adds an additional step in the protocol. More details
- 160 of the photolithography process and the development of the described protocol are given by Quintanilla-Terminel [2014] and Quintanilla-Terminel and Evans [2016].

## 2.3 Electron-beam lithography applied to olivine and Solnhofen limestone

The e-beam lithography process was carried out at the Minnesota Nano Center (MNC). The forsterite and San Carlos olivine samples were polished using diamond-lapping film down to a 0.5-µm particle size. The

165 Solnhofen limestone samples were polished with aluminum oxide powder following the same procedure used on the Carrara marble samples. The electron-beam lithography steps are described below:

I. A wafer of San Carlos olivine, Solnhofen limestone, or synthetic forsterite was initially left in a vacuum oven at 130°C for 20 min to dry.

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II. The sample was first coated with PMMA diluted in 4 vol% chlorobenzene. The coating was done with a precision spin coater spinning at 3000 rpm for 60 s to produce a thickness of the PMMA film of 350-nm (evaluated with ellipsometry). The sample was then baked on a hot plate at 180°C for 10 min. Afterward, the sample was coated with 15-nm of gold in a plasma sputterer to avoid charging.

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III. The patterning was achieved using a Vistec EBPG5000+ e-beam lithography system. First, the design was created using CAD software. In e-beam lithography, a dose array is typically used in order to find the optimum exposure dose for each material and each resolution. To find the optimum exposure for San Carlos olivine, forsterite, and Solnhofen limestone, patterns of different resolution were exposed with

- 180 doses ranging from 300 to 1200  $\mu$ C/cm<sup>2</sup> in increments of 50  $\mu$ C/cm<sup>2</sup>. The optical microscope image in Figure 3a illustrates the results of a dose array performed on a sample of San Carlos olivine. The visible radial lines forming the spokes of a wheel are 10- $\mu$ m thick and were exposed to 900  $\mu$ C/ cm<sup>2</sup>. Inside each rectangle along the radial lines, different doses were tested for grids of lines 50-, 100- and 200-nm wide. Examples of good exposure and over-exposure are shown in the secondary electron micrographs, Figures
- 185 3a,b, respectively. Based on these results, we selected optimal doses of 900  $\mu$ C/cm<sup>2</sup> for sub-micron features, 750  $\mu$ C/cm<sup>2</sup> for micrometer features on San Carlos olivine and synthetic forsterite, and 800  $\mu$ C/cm<sup>2</sup> for the sub-micron features on Solnhofen limestone. A variety of layouts were designed to meet the needs of the type of application; for example, the wheel design (Figure 3a) was developed to evaluate the inelastic strain distribution produced during a torsion experiment.

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IV. The first step in the developing process required dissolution of the gold layer using a solution of KI, routinely called gold etcher. The sample was first submerged in the KI solution for 30 s, then rinsed in deionized (DI) water for 10 s, and finally rinsed in isopropanol for 10 s. The sample was then developed. Two developing processes were explored. The typical developing solution for PMMA is a 3:1 mixture of isopropanol and methyl isobutyl ketone. The coated sample was submerged in this solution for 60 s. If a

195 isopropanol and methyl isobutyl ketone. The coated sample was submerged in this solution for 60 s. If a higher resolution is required, during developing, a mixture of 3:1 isopropanol and DI water can be used. This second mixture was used with samples that were exposed to about half of the dose previously described in order to avoid over-developing of the features [Yasin et al., 2002]. In any case, the choice of the particular developer, exposure, and developing time should be adapted to the patterned substrate as well as the resolution needed.

V. Two etching processes were explored, a wet etch using HF diluted to 1 vol% applied for 1 s and a chlorine-gas etch using an Oxford plasma etcher for 1 min. Although the two processes gave similar results, the wet etch was simpler to implement.

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VI. A 110-nm thick layer of Cr was deposited in a plasma evaporator.

VII. To dissolve the remaining PMMA, the sample was immersed in 2-(2-aminoethoxy)ethanol, Nmethyl-2-pyrrolidine at 120°C for at least 4 h. Gentle, 1-s bursts of sonication were sometimes necessary to completely remove the PMMA.

The resulting patterned surfaces of San Carlos olivine and Solnhofen limestone are illustrated in Figure 4. Notice the difference in scale compared to the patterned marble surface in Figure 2; the images of the patterned Carrara marble in Figure 2 are optical light micrographs, whereas the images of the San Carlos

- 215 and Solnhofen limestone in Figure 4 are scanning secondary-electron micrographs. The high resolution, the sharpness of lines, and the pattern profile can be better appreciated in images of patterns deposited on San Carlos olivine obtained by atomic-force microscopy (AFM) (Figure 5). It is apparent that electronbeam lithography permits much higher resolution than photolithography; in theory, nanometer scales can be achieved [Vieu et al., 2000; Manfrinato et al., 2013]. For geomaterials, a resolution of 20-nm is easily
- 220 attained. The pattern is rastered directly onto the sample without the need for a mask, which facilitates tests of different designs. However, because the rastering process has to be repeated for each sample, e-beam lithography is much slower and more costly than photolithography. Furthermore, samples have to fit into the electron-beam lithography system, limiting the geometry of the sample that can be patterned. In the Vistec EBPG5000+ used at the MNC at the University of Minnesota, the sample has to be as thin and flat
- as a silicon wafer (i.e., no more than 1-mm thick), a geometric constraint that has to be considered when designing deformation experiments.

## **3** Strain analysis

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The regular grid that is printed onto the sample is used to track the strain accommodated by the sample. Indeed, the ultimate goal of using the MSSM technique is to understand which micromechanical processes accommodate strain in a deforming rock and to relate the observed microstructure to the imposed macroscopic deformation conditions. For this purpose, it is necessary to calculate the strain at different scales and, particularly, at a microscale.

## 3. 1 Need for a regular grid

Different techniques allow the computation of strain at a local scale, most are image-based particle tracking techniques and therefore use a Lagrangian description in which strain is calculated by following a material point before and after deformation [Reddy, 2013; Malvern, 1969]. The more widely used strain analysis

technique, digital image correlation (DIC), has been successfully applied in a variety of rock mechanics applications. For instance, DIC was used to map the localization of damage in a heterogeneous carbonate [Dautriat et al., 2011], to quantify the role of crystal slip and grain-boundary sliding during creep of synthetic halite [Bourcier et al., 2013], and to better understand creep of ice [Chauve et al., 2015; Grennerat et al., 2012]. DIC is an excellent tool for extracting displacement and strain measurements at a microscopic scale [Bruck et al., 1987; Sutton et al., 2009] by comparing digital micrographs of the sample at different stages of deformation. The strain is computed using various algorithms allowing the observer to track blocks of pixels and cross correlate them between deformation steps [Bornert et al., 2008]. Each block of

pixels needs therefore to be unique and recognizable between deformation steps.

- High-temperature and high-pressure deformation often changes the reflectivity of the material to a point
  where blocks of pixels are no longer recognizable between deformation steps. Furthermore, because our experimental set-up makes it difficult to analyze incrementally strained samples, we require a technique that allows us to the identify a material point independently of the amount deformation the material has experienced. We therefore rely on the analysis of an initially regular grid characterized after deformation [Allais et al., 1994; Ghadbeigi et al., 2012; Karimi, 1984; Moulart et al., 2007; Sharpe, 2008; Wu et al., 2006, Martin et al., 2014]. Because our experiments are performed at high temperature and pressure with a
- metal jacket surrounding the sample, the grid is introduced in the middle of the sample, following different variations of a split cylinder assembly [Raleigh, 1965]. The initial reference grid is formed by deposition using either photolithography or e-beam lithography. Other deposition methods have been used, such as sputtering through a commercial screen [Xu and Evans, 2010] or introducing a metal mesh in between the
- 260 sample halves [Spiers, 1979]. Recently, Hiraga [2015] used a focused ion beam to groove lines on samples composed of synthetic forsterite and diopside in order to track grain displacement and rotation during diffusion creep. In all the techniques, the main challenge is to accurately identify each individual marker or line before and after deformation. Various complications can arise, for instance the markers can be unstable at high temperature, or the split-cylinder surfaces can weld together at high pressures making the recovery
- 265 of the patterned surface challenging or impossible. Nevertheless, if the identification of individual markers after deformation is accomplished, then spatial variations in relative displacement can be used to compute the strain field across the reference surface. Lithography has the advantages that higher resolutions can be achieved, that the patterns are more robust, that they survive high-temperature, high-pressure deformation, and that specific patterns can be custom designed to account for the grain size of the material or the research questions to be answered. Furthermore, because lithographic techniques allow the experimentalist
- to create extremely regular patterns with known dimensions, the strain can be computed by locating the

markers only after deformation. It is however, advisable to image the sample before deformation as well, as this step could provide additional information (particularly if the grain boundaries are visible) and eliminate the potential error introduced by irregularities in the undeformed grid.

# 275 **3.2 Strain computation**

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#### 3.2.1 Marker location

Different image processing techniques can be used for locating the markers before and after deformation: for instance, a convolution [Biery et al., 2003] or a Hough transform algorithm [Quintanilla-Terminel and Evans, 2016]. Depending on the application, the user could rely on the regularity of the grid and not identify the markers before deformation. Often, some manual input is required to ensure that the marker is correctly identified after deformation.

#### 3.2.2 The *n*-point analysis

estimate.

The *n*-point analysis is an inversion technique that allows the user to probe the strain at different scales. The positions of n material points are registered before and after deformation, and the best fit of the 285 deformation gradient tensor transforming the undeformed to the deformed material lines related to these *n* material points is computed. This inversion assumes that the deformation is homogenous over the area spanned by the *n*-point ensemble. In practice, the technique requires the following steps. (1) Imaging of the region of interest (ROI) before deformation: Based on the size and spacing of the markers, this step can be performed with light or electron beam microscopy. Note that the strain can also be computed without 290 imaging the area before deformation, relying on the regularity of the grid. (2) Registration of the coordinates of all markers in the ROI before deformation. (3) Deformation experiments under pressure and temperature. (4) Imaging of the same ROI after deformation. (4) Registration of the coordinates of all markers in the ROI after deformation. (5) Determination of deformation gradient tensor F using the *n*-point inversion technique. To eliminate rigid-body translations, the material displacements are calculated relative 295 to a moving centroid for every set of n points. That is, the coordinates of each point i before and after deformation,  $X_i$  and  $x_i$  respectively, are referenced to the centroid of the set of n points before and after deformation,  $C_n$  and  $c_n$ . The 2-D deformation gradient tensor F is determined from the relation between material displacement vectors before,  $dX_i = X_i - C_n$ , and after deformation,  $dx_i = x_i - c_n$  using a least-square

300 The deformation gradient tensor,  $\mathbf{F}$ , is determined by calculating the least-square fit for the ensemble of *n* material vectors, i.e., minimizing the sum of the squares of the difference between the modeled and the measured material lines,  $d\mathbf{x_i}^* = \mathbf{F} \cdot d\mathbf{X}$  and  $d\mathbf{x_i}$ , respectively. Consequently, the tensor,  $\mathbf{F}$ , describes the homogeneous deformation that best fits the displacement field of *n* material points. Four different configurations of *n* points are illustrated in Figure 6a.

# 305 3.2.3 Computation of the strain tensor

The deformation gradient tensor  $\mathbf{F}$  can be decomposed into a product of a rotation tensor  $\mathbf{R}$  and either  $\mathbf{U}$ , a right-stretch tensor, or  $\mathbf{V}$ , a left-stretch tensor.  $\mathbf{U}$  can be diagonalized following Eq. (1):

$$\mathbf{U} = \mathbf{Q}^{\mathrm{T}} \mathbf{D} \, \mathbf{Q}. \tag{1}$$

The Hencky strain tensor,  $\varepsilon$ , is defined in Eq. (2):

$$310 \qquad \mathbf{\varepsilon} = \mathbf{Q}^1 \ln \mathbf{D} \, \mathbf{Q}. \tag{2}$$

An optical or electron micrograph allows for a 2-D strain inversion. The strain tensor,  $\varepsilon$ , is a 2 by 2 tensor with three independent components that can be computed in the sample reference as seen in the insert in Figure 9, one along the 1 axis, a second along the 2 axis, and a third shear component. In Figure 9, the three components for an area of Carrara marble deformed to 11% compressive strain at  $T = 600^{\circ}$ C,  $P_c = 300$  MPa and a strain rate of  $3\times10^{-5}$ s<sup>-1</sup> are contoured over a map of the grain boundaries of the analyzed area. The geological convention is used in which positive strains correspond to shortening strains and negative strains correspond to lengthening strains. Topographic information would be required to compute the full 3-D strain tensor. However, in the split cylinder set-up, the out-of plane component of the strain tensor can be inferred from the 2-D tensor if the deformation is isochoric and axisymmetric as explained in Quintanilla-

320 Terminel and Evans [2016].

## 3.3 Resolution of the technique

The resolution of the strain analysis technique depends on two main factors that should be assessed for each application before interpreting the strain results. The main sources of error are related to the location of the markers and to the strain induced by the sample preparation. Any error of markers location will propagate into the strain calculation. This error is a function of the resolution of the images and of the image analysis technique used to locate the markers. The second source of error comes from actual local strains produced by preparation of the sample. This error has to be estimated with a "zero strain" experiment, by measuring the local strain in a sample left under the pressure and temperature conditions for a time length typical of the experiment.

- The n-point inversion technique assumes homogeneous deformation for all *n* points, it therefore introduces a characteristic spatial dimension for the local inelastic strain. In practice, the inversion is realized point by point; for each point, its *n-1* neighbors are located. Thus, when *n* is larger, the computation error is reduced, providing that strain is actually homogenous within the ROI. However, when n is larger the characteristic length scale of the strain heterogeneity is also larger. Thus, a compromise exists between resolution of spatial variations in local strain and errors involved in its calculation [Ouintanilla-Terminel and Evans,
- 335 spatial variations in local strain and errors involved in its calculation [Quintanilla-Terminel and Evans, 2016; Xu and Evans, 2010; Bourcier et al., 2013]

Figure 6 illustrates the different *n* configurations and their corresponding strain map and strain distribution along the 1 axis (in the sample reference) for a "zero-strain" experiment. This experiment was performed on a half cylinder of Carrara marble at  $T = 700^{\circ}$ C and  $P_c = 300$  MPa in order to evaluate the resolution of the experimental series in Quintanilla-Terminel & Evans [2016]. The mean strain is zero for all *n* point

340 the experimental series in Quintanilla-Terminel & Evans [2016]. The mean strain is zero for all *n* point configurations, but the standard deviation is larger for smaller *n*. Both the "real" strain coming from experimental artifacts and the computational error coming from the marker location are evaluated. In the Carrara marble series we chose 9n point as a compromise, with a strain gauge length of 20-µm and an experimentally estimated uncertainty of strain of ±0.001 [Quintanilla-Terminel & Evans, 2016].

# 345 4 Application to creep of rocks

The choice of grid design and patterning technique should reflect the test geometry, physical characteristics of the target rocks, and research goals, including specific hypotheses regarding constitutive behavior. The design choices made for tests on Carrara marble and on San Carlos olivine, synthetic forsterite, and Solnhofen limestone can be used for illustration. In the first case, Carrara marble deforming by power-law

350 creep (cf. Renner and Evans, 2002; Renner et al., 2002), we wished to investigate the partitioning of strain between different deformation mechanisms, to determine which were dominant, and to identify internal state variables needed for a more accurate flow law [Evans, 2005]. Patterns in this study were made using photolithography. In the second case, we wished to observe microstructures produced during dislocation creep of San Carlos olivine [Hansen et al., 2011; Hirth and Kohlstedt, 2003] and during diffusion creep in synthetic forsterite [Dillman, 2016] and Solnhofen limestone [Schmid et al., 1977]. Here, our goals included determining the strain contribution of grain-boundary sliding in different creep regimes and testing current constitutive models. For this work, we produced patterns with e-beam lithography.

## 4.1 Grid design and sample geometry

- The average grain size of our Carrara marble samples was 130-µm [Xu and Evans, 2010; Quintanilla Terminel, 2014]; therefore, patterns with micrometer resolution, as formed by photolithography, were optimal. Our designs included circular markers, 2-µm in diameter, with centers spaced at 10-µm intervals, complemented by a printed numbering system. Thus, even though there were about 1.5 million markers, each could be assigned a unique address. Strain was mapped at a scale of 20-µm across the sample. For MSSM in samples of San Carlos olivine, synthetic forsterite, and Solnhofen limestone, all of which have
- 365 grain sizes ≤10 µm, higher resolution patterns were necessary, and so we used electron-beam lithography to print two intersecting grid patterns. In the first, lines were 500-nm thick, spaced by 4.5-µm; in the second, 200-nm thick, spaced by 2-µm. We chose lines rather than circular markers to emphasize localized offsets at grain boundaries.

Photolithography is a flexible technique that can be used to mark surfaces with varying geometry, including 370 split-cylinders or cylindrical surfaces (Figures 2a,b, 8a). E-beam lithography has better spatial resolution

but limits the surface geometry. As seen in Section 2, the sample has to be flat and not more than 1-mm thick. Two different composite cylinders sample configurations were used to introduce a 1-mm thick patterned sample. In the first, a 1-mm thick disk of San Carlos olivine disk was placed between two short olivine cylinders (Figure 8b). In the second, a rectangular slab, 1-mm thick, of patterned Solnhofen limestone was inserted between two half cylinders (Figure 8c).

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## 4.2 Application of photolithography: Creep of Carrara marble

We used a split cylinder set-up following Raleigh [1965] and Spiers [1979]. Half cylinders of "Lorano Bianco" Carrara marble, a standard material for deformation experiments [Molli and Heilbronner, 1999; Heege et. al., 2002], were polished down to 0.5-µm grit size using aluminum oxide and patterned by 380 photolithography. Optical micrographs of the surfaces were made before and after deformation. Matlab<sup>™</sup> was used for the strain computation. The circular markers were identified using a Hough transform and registered with the embedded coordinate system. The 2-D strain tensor was computed using the n-point algorithm. The MSSM technique allowed for an inversion of strain at different scales, up to shortening strains of 36% at a temperature of 700°C. Notice that strains measured over a ROI at sample-, macro-, and 385 micro- scales have the same average values, but larger standard deviations as the spatial scale of measurement decreases (Figure 9). The markers belonging to each grain are identified and classified; maps of the different strain tensor components can be used to determine strain accommodation owing to slip along grain boundaries, to twinning, and to intra-granular deformation [Quintanilla-Terminel, 2014]. Figure 10 shows samples shortened to 11% at T = 400, 500, 600 and 700°C and  $P_c = 300$  MPa; Figure 11 390 illustrates samples shortened to 11, 22 and 36% at 600°C. In all experiments, the means of the local strains agreed remarkably well with those measured during the mechanical tests. Importantly, the spatial heterogeneity varied with strain and temperature. For more detail, see Quintanilla-Terminel and Evans

[2016].

## 4.3 Application of electron-beam lithography: Creep of San Carlos olivine and synthetic forsterite

395 The greatest challenge in studying strain distributions during creep in olivine rocks is the separation of the surfaces after deformation. Because creep tests must be carried out at T > 1000 °C and at  $P_c = 300$  MPa, the metal of the grid and the rock surface strongly adhere. Nonetheless, the results described here are promising and suggest that, with further work, strain distributions during grain boundary sliding might be successfully measured (Figures 12-13).

# 400 4.3.1 Compression experiments on San Carlos olivine at 300 MPa and 1150°C

We used electron-beam lithography to pattern mono-phase, polycrystalline samples prepared from powders of San Carlos olivine, Fo<sub>90</sub>. First, samples were fabricated by cold pressing powders into a nickel can, followed by hot-pressing at  $P_c = 300$  MPa and T = 1250°C for 3 h [Hansen et al., 2011]. A 1-mm thick slice

was cut from the hot-pressed cylinder and one side was ground and polished with a final step using lapping films with 0.5- $\mu$ m diamond grit. The olivine wafers were then patterned following the protocol described in Section 2. The rest of the cylinder was cut in half, one end polished, and the three pieces assembled as shown in Figure 8b. This composite sample was deformed at  $P_c = 300$  MPa, T = 1150 °C, and a constant strain rate of  $1 \times 10^{-5}$  s<sup>-1</sup> to 15% shortening strain in a gas-medium high-pressure apparatus [Hansen et al., 2011]. The strength at steady state of the composite, 245 MPa, is in excellent agreement with that determined from earlier tests of intact cylinders [Hansen et al., 2011].

Recovering the grid after deformation was more challenging because of the high temperatures and normal

loads on the marked surface. We experimented with different metals and separation techniques. Sputtering a window shape of a high melting temperature metal such as tungsten on the facing sample (a technique similar to the one used for Carrara marble) was unsuccessful, and the two faces still could not be separated.

- 415 Inserting a thin (0.01-mm), ring-shaped, nickel foil between the gridded surface and the facing surface was still not sufficient, but the gridded surface was successfully recovered if a full nickel foil was placed between both surfaces. Secondary electron micrographs of grids with lines 200-nm and 1- $\mu$ m thick show offsets of 0.5 to 1- $\mu$ m along the grain boundaries (Figures 12a, b). On a recovered area (Figure 13a), the strain was inverted using the *n*-point algorithm described above; the component  $\varepsilon_{22}$  is mapped in Figure 13b.
- 420 Note that the gridded surface is perpendicular to the shortening axis, compared to the Carrara marble tests in which the gridded surface was parallel to the shortening axis (Figures 10-11). Because the size of the recovered area is small, we cannot draw meaningful conclusions regarding the partitioning between intragranular strain and grain boundary sliding, but the technique offers further opportunities for success.

## 4.3.2 Compression experiments on forsterite at 1 atm and 1100°C

- Samples of synthetic forsterite were synthetized following Koizumi et al. [2010]. After gridding, they were deformed at T = 1250°C and P<sub>c</sub> = 1 atm. [Dillman, 2016] with the grid on an outer surface of a sample with a square-cross section. The tests used a grid 500-nm thick and were designed to compare with previous measurements of creep [Dillman, 2016] using a line offset caused by grain-boundary sliding [Langdon, 2006]. Grid recovery for these samples was much easier because deformation occurred under atmospheric
  pressure. We used an AFM to measure the vertical displacement of grains and to map the deformed grid
- (Figure 14). The state of the initially regular grid after deformation highlights the deformation occurring along grain boundaries. 2-D offsets of ~500 nm are evident. More details on the 1 atm. compression experiments on synthetic forsterite can be found in Dillman [2016].

# 4.3.3 Creep of fine-grained Solnhofen limestone

Three-piece samples of Solnhofen limestone were prepared (Figure 8c), gridded using e-beam lithography, and deformed in compression in gas-medium apparatus [Paterson, 1990] at T = 700°C and  $P_c = 300$  MPa to a shortening strain of 9% at a strain rate of  $3 \times 10^{-4}$  s<sup>-1</sup>. The patterned surfaces could be separated without the addition of a metal foil, and the deformed pattern was easily found using electron microscopy. In Figure 15 with two secondary electron micrographs of the same area before and after deformation, the deformed lines

440 and grid are clearly visible, however the grain structure is not. Additional characterization permitting identification of grain boundaries (possibly imaging with electron backscatter diffraction), will be necessary to evaluate the contribution of grain boundary sliding to the total strain. Nonetheless, this application demonstrates that the deformation of the lines and grid is visible and that the electron-beam lithography provides the required resolution to quantify the strain at the granular level.

# 445 5. Conclusion

Patterning using photolithography and e-beam lithography can provide maps of strain calculated over spatial scales of 10-0.5  $\mu$ m, respectively. The experimental preparation is technologically demanding and labor intensive, but MSSM is a unique characterization tool that yields a detailed description of strain accommodation within intra- and inter-granular regions. The experimental protocols described here can be

450 adapted to many other rocks, and the pattern can be designed for specific purposes. The MSSM technique has the potential to improve the interpretation of microstructural observations of rocks deformed in nature and in the lab, to constrain the partitioning of strain amongst several deformation mechanisms, and to improve constitutive modeling of creep mechanisms in the Earth.

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Figure 1: Lithographic process: 1- rock, 2- photoresist or PMMA, 3-metal layer (sputtered or evaporated). Each step is described in more detail in the text: I- Sample preparation, II- Coating with a polymer (photoresist for photolithography, PMMA for e-beam lithography), III- Exposure with UV rays for photolithography (IIIa) or mith an electron for a beam lithography (IIIb). IV, Development of the section V. Etablica, VI. Metal

565 with an electron beam for e-beam lithography (IIIb), IV- Development of the coating, V- Etching, VI- Metal deposition, VII- Dissolution of the remaining polymer, VIII- Patterned sample.



Figure 2: Patterned Carrara marble using photolithography. a) The optical light micrograph on the left shows the patterned half-cylinder with more than 1.5 million markers, while the micrograph on the right presents a zoomed-in view of an area revealing the 2-µm wide markers with centers spaced every 10-µm as well as the embedded numbering system that allow us to locate each marker before and after deformation. b) The photograph on top illustrates the structure of a patterned cylinder, while the optical light micrograph on the bottom show a closer view of the patterned curved surface revealing 20-µm wide markers with centers spaced every 50-µm.



Figure 3: a) Light micrograph of a patterned San Carlos olivine crystal. Electron beam lithography was used
 with varying doses along the radial lines. All radial lines (10-μm thick and exposed with a 900μC/cm<sup>2</sup> dose are visible), inside each rectangle different doses were tested: an example of a well exposed and an over-exposed area are seen in (b) and (c) respectively. In the over-exposed area, the pattern appears blurred, and causes the rounding of the corners in the grid. An under-exposed area will typically show no signs of the pattern. The dose test allowed identification of the optimum dose for achieving the desired resolution: 700 μC/cm<sup>2</sup> for 200-nm wide

600 lines.



Figure 4: Secondary electron images of patterned San Carlos olivine (a and b) and Solnhofen limestone (c and d). The grid in the San Carlos olivine sample is composed of 500-µm lines spaced every 4.5-µm (a), a zoom in is seen in (b). For Solnhofen limestone, we combined 200-nm straight lines with a regular grid of 200-µm lines spaced every 2-µm (c), a zoom into both areas is seen in (d).

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Figure 5: Atomic force micrographs of patterned San Carlos olivine (a) and height profile (b) of the surface
 along the profile 1. The 500-nm wide chromium lines are spaced every 4.5 μm and are 110 nm high. The sharpness of the lines can be appreciated on the AFM data.



Figure 6: a) *n* point configurations for n=3, 5, 9 and 25. b) Corresponding strain maps and strain distributions for strain along the axis 1 in the sample reference (see insert for reference frame) for a Carrara marble cylinder isostatically annealed at  $T = 700^{\circ}$ C and  $P_c = 300$  MPa for 3 hours. c) Corresponding strain distribution for each strain map, it can be observed that the mean is always 0 but the spread of the distribution is smaller the larger *n* is.



Figure 7: Strain maps for the three components of the 2-D strain tensor for a sample of Carrara marble deformed at  $T = 600^{\circ}$ C,  $P_c = 300$  MPa to 11% shortening strain at  $3x10^{-5}$ s<sup>-1</sup>. The strain tensor was computed using the *n* point technique with n = 9. The grain boundaries are overlaid over the strain maps. Positive strains correspond to shortening strains.





Figure 8: Experimental set-up for studying creep of Carrara marble, San Carlos olivine, and fine grained Solnhofen limestone at high temperatures and  $P_c$  = 300 MPa in a Paterson Instruments gas apparatus. In (a) the composite half-cylinder is composed of two pieces, one is gridded using photolithography, the other is polished and sputtered with a window shape metal layer. Figures (b) and (c) show the composite set-ups used when the patterning involved e-beam lithography. San Carlos olivine was deformed using set-up (b): a patterned 1-mm thick disk was introduced between two short olivine samples. Solnhofen limestone was deformed using the setup in (c) : a patterned 1-mm thick rectangular slab was introduced in-between two half-cylinders.



675 Figure 9: Strain maps at different scales for a Carrara marble split cylinder deformed at  $T = 600^{\circ}$ C,  $P_c = 300$  MPa, to 11% shortening strain at  $3x10^{-5}$  s<sup>-1</sup> (a). The component along the shortening direction of the strain tensor, ε<sub>11</sub>, was computed at different scales. In (b), ε<sub>11</sub> was computed over the whole sample with a resolution of 3-mm (macro-scale). In (c), ε<sub>11</sub> was computed over a smaller section with a 20-µm resolution (micro-scale) and is mapped with the overlaying grain boundaries. Positive strains correspond to shortening strains.



Figure 10: Strain maps for  $\varepsilon_{11}$  computed with a *n* point technique (with *n*=9) for samples of Carrara marble deformed at  $T = 400^{\circ}$ C, 500°C, 600°C and 700°C,  $P_c = 300$  MPa,  $3x10^{-5}s^{-1}$  to 11% shortening strain. The strain component along the shortening direction is mapped with the overlaying grain boundaries. Positive strains correspond to shortening strains. Note the concentration of strain along grain boundaries and intra-granular features as *T* increases.



Figure 11: Strain maps for  $\varepsilon_{11}$  computed with a *n* point technique (with *n*=9) for samples of Carrara marble deformed at  $T = 600^{\circ}$ C,  $P_c = 300$  MPa,  $3x10^{-5}s^{-1}$  to 11%, 22% and 36% shortening strain. The strain component along the shortening direction is mapped with the overlaying grain boundaries. Positive strains correspond to shortening strains.

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Figure 12: Secondary electron images of a gridded fine-grained San Carlos olivine after deformation at  $T = 1100^{\circ}$ C,  $P_c = 300$  MPa to 5% shortening at  $1 \times 10^{-5}$ s<sup>-1</sup>. The deformed lines show clear offsets at grain boundaries. 735 The dotted orange lines put in evidence the deformation of the lines in the grid. The orange arrows show grain boundary displacement put in evidence by the grid.



Figure 13: Strain inversion on recovered grid in fine-grained sample of San Carlos olivine deformed at  $T = 1100^{\circ}$ C,  $P_c = 300$  MPa to 5% deformation at  $1x10^{-5}s^{-1}$ . a) Secondary electron image of the recovered area and b) superposed strain map of  $\varepsilon_{22}$ , a strain tensor perpendicular to the shortening direction.



Figure 14: Atomic force micrograph of deformed grid in synthetic forsterite deformed at T = 1110°C, P<sub>c</sub> = 1 atm
to 5% shortening. The chromium lines are about 110-nm thick and are clearly visible in the height map. The grain structure is also clearly visible and both the height difference and the offset of the lines show evidence of grain displacement. Amanda Dillman performed the deformation experiment and the AFM observations for this sample.



Figure 15: Secondary electron micrographs of a patterned Solnhofen limestone sample (a) before deformation and (b) after deformation at  $T = 700^{\circ}$ C,  $P_c = 300$  MPa, to 9% shortening strain at  $3x10^{-4}$ s<sup>-1</sup>. Before deformation the pattern consisted on 200-nm width lines spaced every 5-µm and of a grid of 200-nm lines spaced every 2-µm. The lines were made of 110-nm thick chromium. After deformation, the lines are clearly deformed but the grain structure is not apparent.