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The metallurgical texture of gold artefacts found at the Bronze Age rampart of Bernstorf (Bavaria) studied by neutron diffraction

Friedrich E. Wagner[a,*], Rupert Gebhard[b], Weimin Gan[c], Michael Hofmann[d]

[a] Physics Department E 15, Technical University of Munich, James-Franck-Straße 1, 85747 Garching, Germany
[b] Archäologische Staatssammlung München, Lerchenfeldstraße 2, 80535 München, Germany
[c] German Engineering Materials Science Center at MLZ, Helmholtz-Centre Geesthacht, Lichtenbergstraße 1, 85747 Garching, Germany
[d] Heinz Maier-Leibnitz Centre (MLZ), Technical University of Munich, Lichtenbergstraße 1, 85747 Garching, Germany

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ABSTRACT

The metallurgical texture of several of the gold artefacts found in 1998 at a Bronze Age fortification near Bernstorf in Bavaria was studied by neutron diffraction using the STRESS-SPEC instrument at the FRM II reactor of the Maier-Leibnitz Centre of the Technical University of Munich. The pieces consist of approximately 0.1 mm-thick gold sheets embossed with ornamentations. The purpose of the texture studies was to obtain information on the technique used to produce the thin gold sheets. All studied spots with about 5 mm diameter on the artefacts were found to exhibit a cube type {100}⟨001⟩ texture that is typical for many cold rolled and subsequently annealed and recrystallized face centred cubic metals. A cube texture similar to that of the Bernstorf artefacts was obtained by straight rolling followed by annealing at different temperatures and for different periods of time, but never with the purity observed in two of the Bernstorf artefacts. This leaves some uncertainty about the manner in which the Bernstorf objects were made. By comparison with laboratory-made reference samples, one can rule out hammering or cross-rolling with or without subsequent annealing for the manufacture of the gold foils. Beyond the interest in these results for the discussion of the authenticity of the Bernstorf gold finds, texture determinations using neutron diffraction are shown to be a non-destructive method for obtaining information on the techniques used to produce archaeological gold artefacts in general.

1. Introduction

Between 1994 and 1997, two amateur archaeologists, M. Moosauer and T. Bachmaier, conducted excavations at a Bronze Age rampart near the hamlet of Bernstorf, approximately 30 km north of Munich in Bavaria, Germany. This Bronze Age fortification is situated on a sand and gravel escarpment about 55 m above the valley of the Amper River. The wall surrounding the fortification was approximately 1.6 km long and consisted of a wooden framework filled with gravel and earth, and presumably covered with mudplaster. For unknown reasons, this wall was destroyed in a devastating fire around 1300 BCE. The fire is well documented by studies of the earth heated to temperatures up to 1100 °C and remains of charred wood and charcoal, which allowed ^{14}C dating (Gebhard, 1999; Gebhard et al., 2004; Gebhard and Krause, 2016).

In the summer of 1998 the landowner began to use part of the area near the rampart for commercial gravel extraction. Consequently, trees were felled, their trunks were uprooted, and the topsoil was removed. During a last survey of the site, several gold artefacts were found on the surface by the two amateur archaeologists, who notified archaeologists at Bavarian State Archaeological Collection and the Bavarian Office for the Conservation of Historical Monuments. Further gold finds were made in the following weeks, most of them in the presence not only of the amateurs, but also of professional archaeologists from the two institutions mentioned above. A total of 21 pieces of gold artefacts were recovered, which are now in the possession of the Bavarian State Archaeological Collection, Munich. Some of these objects were found scattered on the ground and some attached to the roots of the tree trunks piled on a heap. The discovery of the finds has been described in several publications (Gebhard, 1999; Moosauer and Bachmaier, 2005; Bähr et al., 2012; Gebhard et al., 2014), most recently in a volume edited by Gebhard and Krause (2016).

Soon after the discovery, doubts arose about the authenticity of the artefacts, mainly because the finds were not made in a proper scientific excavation. Already in the first description of the finds, Gebhard (1999) reported that the gold was uncommonly pure, which he explained by

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[*] Corresponding author.

E-mail addresses: fwagner@tum.de (F.E. Wagner), rupert.gebhard@extern.lrz-muenchen.de (R. Gebhard), weimin.gan@hzg.de (W. Gan), michael.hofmann@frm2.tum.de (M. Hofmann).

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the use of cementation for its purification. More recently, the discussion became more animated when element analyses showed that the gold was of about 99.99% purity. The silver and copper in it was found to be quite inhomogeneously distributed with silver contents between about 20 and 200 μg/g and copper contents between 1 and 10 μg/g (Pernicka, 2014a, 2014b; Radtke et al., 2016).

Pernicka (2014a, 2014b) interprets the high purity of the gold as an indication that the objects are modern forgeries. There are, however, also good arguments for the authenticity of the objects, like the surface structure similar to other Bronze Age artefacts, alterations of the surface by burnishing and possibly by the formation of autigenic gold, ^{14}C-dating of a piece of wood embedded in one of the gold foils, and an unusually high gold content in the soil recovered together with one of the objects. These findings have been discussed in detail in the volume edited by Gebhard and Krause (2016) who attribute the high purity of the gold artefacts to a purification process in antiquity, and suppose that the gold sheets were made by hammering followed by burnishing to obtain smooth and shiny surfaces. However, these conclusions were again questioned by several authors (Reichenberger, 2017; Harding and Hughes-Brock, 2017; Pernicka and Wunderlich, 2017). The authenticity of the Bernstorf gold finds thus remains a subject of vivid dispute.

In this situation, any additional information on the material properties of the gold objects from Bernstorf would be of interest. One such property is the metallurgical texture of the gold, which should give indications to the production process. Texture in this context refers to the distribution of the orientation of the crystallites in the polycrystalline metal (Bunge, 2013; Suwas and Ray, 2014). Texture analysis is usually done by X-ray diffraction, but X-rays like the 8.04 keV Cu K_α radiation penetrate only about 2 μm deep into gold, thus showing only the texture in the uppermost layer of an object. The same is *a fortiori* true for methods based on the diffraction of backscattered electrons in scanning electron microscopes (Suwas and Ray, 2014). High-energy X-rays would penetrate deeper into the gold (Wcislak et al., 2002; Wenk and Grigull, 2003; Bastie et al., 2006). For 100 keV X-rays, for instance, the half-value thickness of metallic gold is about 0.15 mm, enough for studying the bulk texture of gold sheets like those from Bernstorf, which are about 0.1 mm thick. For thermal neutrons, however, the half-value thickness is about 2.5 mm. Thus neutron diffraction is ideal for the determination of the bulk texture even in rather thick gold artefacts. The STRESS-SPEC diffractometer at the FRM II neutron source of the Heinz Maier-Leibnitz Centre (MLZ) of the Technical University of Munich at Garching, Germany, is ideally suited for such measurements (Hofmann et al., 2006a; Hofmann et al., 2006b; Brokmeier et al., 2011). We therefore performed a texture study on several pieces of the Bernstorf gold artefacts and on suitable reference materials prepared in the laboratory to emulate various manufacturing processes.

Texture measurements have been used to study the production processes of a variety of archaeological metal objects, but not yet of gold artefacts. Thus copper and bronze axes were studied (Kockelmann et al., 2006a; Artioli, 2007; Arletti et al., 2008) as well as a variety of other bronze objects (Siano et al., 2004; Pantos et al., 2005; Cartechini et al., 2006; Kockelmann et al., 2006b; Siano et al., 2006; Griesser et al., 2016). Grazzi et al. (2009) studied copper and iron guard rings of Japanese swords, and silver coins were measured by various authors (Xie et al., 2004; Kockelmann and Kirfel, 2004; Kockelmann et al., 2006b; Sheedy et al., 2015).

2. Texture analysis

The deformation of polycrystalline metals results in typical texture properties (Hu, 1974; Suwas and Ray, 2014). Texture in this sense refers to the distribution of the crystallographic axes of the individual crystallites in a polycrystalline metal with respect to a coordinate system that is chosen according to the macroscopic shape of the specimen. An observation of this texture can reveal how the material was formed, for instance by forging, hammering, or rolling. Heat treatments usually result in recrystallization processes that alter the texture in typical ways (Humphreys and Hatherly, 2002). For archaeological artefacts, texture measurements should therefore yield information on ancient craft techniques used in the manufacture of the objects, and hence may be of value in discussions of the authenticity of gold finds.

The texture of plane specimens like rolled or hammered sheets is described as the probability distribution of the orientations of the crystallographic axes of the individual grains with respect to a coordinate system with two orthogonal axes within the specimen plane. For rolled specimens, these coordinates are the rolling direction (RD) and the transverse direction (TD) within the plane of the sheet, and the normal direction (ND) perpendicular to the specimen plane. In ancient artefacts, the methods of production of which are a priori unknown, the RD and TD axes have to be chosen arbitrarily, for instance on the basis of the geometry of the objects. If, for a rolled sheet, the crystallographic plane {hkl} is parallel to the rolling plane and the ⟨uvw⟩ crystal axis is pointing in the rolling direction, the orientation is described as {hkl}⟨uvw⟩ (Bunge, 2013; Suwas and Ray, 2014). The individual grains are usually not all oriented in the same way. The distribution of the orientations can then be described as a weighted average over several {hkl}⟨uvw⟩ orientations (Suwas and Ray, 2014). For measuring the distribution of the orientation of the grains, a beam of monochromatic radiation, e.g., thermal neutrons, is directed at the sample. For crystallites having the proper orientation, Bragg reflection takes place. Texture determinations are usually performed with strong Bragg reflections like the (111) and (200) reflections for face centred cubic (*fcc*) metals. The measurements then yield information on the orientation of the {111} and {200} lattice planes or the respective scattering vectors normal to these. When the sample is rotated in the incident beam, the intensity of the reflections changes, which allows one to determine the probability distribution of the grain orientations. The direction of the scattering vector for a grain marks a point, or pole, on a hemisphere with the sample in the centre of the equatorial plane. When the sample is rotated through all possible orientations with respect to the scattering vector of the diffractometer, the density of the poles on the hemisphere represents the information on orientation of the individual crystallites in the specimen.

Experimental texture data are often visualized as pole figures. A pole figure is a circular two-dimensional stereographic projection of the probability distribution of the poles in the specimen-based coordinate system (Suwas and Ray, 2014). A crystallographic axis oriented perpendicular to the plane of the specimen would yield a point in the centre of the pole figure, and an axis within the specimen plane would contribute to the intensity on the periphery of the pole figure. The distribution of the poles is usually depicted by contours of equal pole density or by a color code for the intensities. For the pole figures shown later in this paper, the latter representation has been chosen. A measure for the degree of orientation of the crystallites is the intensity of the pole figure expressed in multiples of that for a random distribution (mrd).

3. The STRESS-SPEC diffractometer at FRM II

The STRESS-SPEC diffractometer at FRM II (Hofmann et al., 2006a; Hofmann et al., 2006b; Brokmeier et al., 2011), is located at a beam port for thermal neutrons of the reactor. For the gold texture measurements, a Ge (311) single-crystal monochromator was used to select neutrons with a wavelength of $\lambda = 0.170$ nm, corresponding to an energy of 28 meV. For these, the 2θ scattering angles for the (111) and (200) Bragg reflections of gold are 42.4° and 49.3°, respectively. The diffracted neutrons are detected by a $25 \times 25\ \mathrm{cm}^2$ (256×256 pixel) two-dimensional position-sensitive ^{3}He detector located about 1 m from the sample, which allows for the (111) and (200) reflections to be observed simultaneously. The roughly circular sampling area on the gold foils had a diameter of approximately 5 mm. Fig. 1 shows the diffraction pattern of the two reflections of gold as observed by the position-