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New insights into Zwischgold application from a multi-analytical survey of late medieval polychrome sculptures at the Swiss National Museum

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ABSTRACT

Zwischgold, or part gold, is a bi-layered metal foil made from gold and silver, popular in European sculptures from the 14th century. It is notoriously difficult to unambiguously identify and to study in detail, due to its close appearance to gold leaf (when still well preserved) and complicated individual situations. Although Zwischgold is nowadays commonly viewed as an economical substitute for gold leaf, arguments regarding its function have circulated for decades. In order to understand the application and related functions of Zwischgold in late medieval sculptures, a survey of 162 objects was recently implemented at the Swiss National Museum Collection Centre, focusing on polychrome wooden sculptures mainly dated 1400–1550 and produced in southern Germany and Switzerland. Handheld X-ray fluorescence (XRF) measurements show a high rate of correlated silver and gold signals mainly in a proportion range of 1:5–3:1, indicating Zwischgold in extremely diverse areas such as hair, beard, crown, gown fold, bordure, dalmatic, undergarment and attribute of saint statues, as well as altar background, frame and framework. This non-invasive XRF analysis is a first step to efficiently locate Zwischgold applied areas and obtain basic statistics regarding the Zwischgold applied artefacts. It provides objective evidence showing that the reasons for medieval Zwischgold application are not limited to cost-saving, but include other factors such aesthetics, symbolism, workshop preference and workability. We discuss such factors with some illustrative case studies, in which visual light microscope (VLM) and scanning electron microscopy coupled with energy dispersive X-ray (SEM-EDX) measurements on samples exhibit certain technological features of Zwischgold of that epoch, including its foil structure, thickness and materials compositions.

1. Introduction

Zwischgold (German) [1–4] or *part-gold* (English) [5,6] is a special gilding material popular in late medieval works of art. As a bi-layered metal foil made from a thin layer of gold over a silver backing [1–7], it is distinct from a gold-silver alloy, where gold and silver atoms are evenly mixed. Zwischgold technology allows the gold layer of the foil to be made thinner and thus become cheaper than regular gold leaf of that time [9,10], is therefore commonly regarded as an economical alternative to gold leaf [1,8,11,12]. The application of Zwischgold on artworks started in panel paintings in the beginning of the 13th century and became popular in German sculptures from the 14th century [8]. It has also been frequently observed in other forms of art from this era, such as wall paintings and book illuminations [13–15]. The disadvantage of using Zwischgold lies in the fact that it tends to oxidize quickly in atmosphere due to the vulnerability of its silver base against invasive species such as sulphur and chlorine, causing surface darkening and loss of metal lustre. It is therefore required by many medieval guilds to be coated with protective varnish [16,17].

The purpose of applying Zwischgold on medieval works of art is an interesting topic in the research history of art technologies. In addition to the widely accepted economic reason, other motives such as aesthetics and symbolism are discussed by art historians and researchers in medieval gilded objects [12,18,19]. However, such arguments are mainly based on individual cases and don't provide sufficient scientific details to be conclusive. Further, while Zwischgold is frequently mentioned in historical documents such as guild regulations, city statutes, purchase orders or workshop contracts, the production of Zwischgold

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are rarely recorded in medieval artists' treatises [8,20], indicating that it has been strictly kept as a commercial secret. This secrecy, on other hand, supports the argument that motivations for the application of Zwischgold is not restricted to the cost-savings, especially in late medieval periods the value of craftsmanship already gained much significance [21].

A survey of 162 polychrome wooden artefacts, including 13 altars, has been recently implemented at the Swiss National Museum (SNM) - Collection Centre. This object survey lays a foundation of basic statistics regarding medieval Zwischgold application and provides scientific evidence for arguments regarding the function and purpose of Zwischgold application in late medieval works of art. As a non-invasive elemental analysis instrument, the handheld X-ray fluorescence analyser (HHXRF) has been proven to be a reliable and practical tool for the preliminary identification of Zwischgold in our previous study [22], and it was therefore used for quickly identifying the application areas of Zwischgold on the surveyed objects. In addition to Zwischgold, the HHXRF measurements were also expected to detect other metal foils such as gold, silver and tin. The identification of the distribution of different metal foils helps to define and summarize the gilding practice in medieval sculptures. Meanwhile, elemental signals obtained through the HHXRF can provide evidence of the presence of certain pigments involved in the construction of the sculpture polychromies. In addition to the non-invasive HHXRF measurements, samples were taken from some objects that present specific functions of Zwischgold and/or significant features for further materials analysis such as visible light microscopy (VLM) and scanning electron microscopy coupled with energy dispersive analysis (SEM-EDX), in order to study technological details of Zwischgold including its foil structure and thickness, ageing mechanisms and chemical compositions. Such data can also support attributing a master or workshop to the artworks. Since medieval masters often learned in the same workshop or school and their works hence appear similar [23], observing the art style alone is often insufficient for an accurate attribution.

2. Methods

2.1. Non-invasive measurements on artefacts applied with Zwischgold

The object survey focuses on the late medieval polychrome wooden sculptures and altarpieces that are currently stored in the SNM - Collection Centre. Based on museum catalogues and on-site inspections, 162 objects in form of bust (7), relief (15), figure (111), figure group (16) and altar (13), mainly dated in the time period of 1400–1550, were selected for the HHXRF measurements. Museum archives regarding the conservation history of certain objects were also checked, in order to ensure that the object surfaces examined have not been, or at least minimally, altered by previous restoration treatments.

The *Niton XL3t GOLDD+* XRF analyser was used in the survey for the HHXRF measurements, with the X-ray tube at 200 mW (50 kV, 40 μA) and the 8 mm aperture. The built-in measurement mode “Mining Cu/Zn”, which has been successfully used in our previous study for the identification of Zwischgold models and artefacts [22], was chosen. In this mode, an AlFe-filter ("main range") and a Mo-filter ("high range") work sequentially during the measurements; the former works more efficiently for the analysis of gold element (Au) and the latter for silver (Ag). The measuring time was pre-set to 5 s per filter, a total measuring time of 10 s is thus required for obtaining both Au and Ag signals in order to quickly identify Zwischgold. More information about this “Mining Cu/Zn” mode is presented in Supplementary Materials: S1. A minimum of 2 measurements were performed for each targeted area of the surveyed objects and a total of 2288 measurements were completed for the 162 objects. Flush contact between the measuring aperture of the analyser and the object surfaces was managed for safe and accurate measurements. Since most of the gilded surfaces examined are curved and slippery, and thus the HHXRF cannot be held firmly onto the surfaces during the measurements, two small pieces of *ethafoam* (ca. 1 mm thick) were attached on the front face of the analyser, above and below the measuring aperture, in order to increase contact friction and stabilise the measurement position.

The built-in software *Thermo Scientific NDT 8.4.3* were used for data transfer and analysis. The Ag:Au ratios of XRF signals were calculated based on the relative concentration (weight%) of Ag and Au elements shown in the data table created through the built-in software [24]. Details regarding data analysis are presented in Supplementary Materials: S1 and S4. It is important to point out that the calculated Ag:Au ratio can vary significantly, depending on the relative thickness of the gold and silver layers, their stratigraphic arrangement, and the composition and thickness of any overlayers that might be present. Such variation is caused by the attenuation of the excitation X-ray beam as it penetrates the sample layers and of the fluorescent X-rays trying to escape from different depths within the sample. For example, if a Zwischgold foil is flipped over (i.e. orientated Ag side up), then signal coming from the Au layer would be suppressed due to attenuation by the upper Ag layer and the measured Ag:Au ratio would increase [25]. In addition, the presence of surface corrosion also leads to a small increase of Ag:Au ratios due to the corrosion driving diffusion of silver from the base layer, through to the foil surface [26]. However, test measurements on a Zwischgold model only showed a slightly higher (ca. 10%) Ag:Au signal ratio in the corroded area than that in the uncorroded area of the model, and so the effect is clearly not significant enough to hide the presence of Zwischgold. Details of the test are presented in Supplementary Materials: S1. Similar to the surface corrosion, a superficial layer (e.g. protection varnish, colour glaze) could also alter the Ag:Au signal ratio by acting as an extra attenuation layer. The alteration magnitude is, however, strongly dependant on the materials composition and thickness of this superficial layer for each individual object.

In addition to the foil structure of Zwischgold, corrosion and surface layer stratigraphy, many other factors can also influence the XRF signals, such as the measurement angle, distance and duration. Therefore, the HHXRF measurements were only used for preliminary, qualitative identification of Zwischgold applied areas, not for quantitative analysis.

It is worth to point out that, depending on the applied measurement mode, certain elements could be falsely shown in the data table created through the built-in software. For example, with the “Mining Cu/Zn” mode, mercury (Hg) signals can be shown as Au in the data table, while the spectra still exhibit Hg peaks. Therefore, when red paint (e.g. cinnabar HgS) is involved in the measurements, it is suggested to check both data table and spectra.

2.2. Materials analysis on Zwischgold samples

For certain representative objects, micro-samples were taken in relevant areas that have been identified with the HHXRF, but preferably on spots invisible to viewers in order to maintain the aesthetic integrity of the artefacts. Therefore, micro-XRF was used to further screen the samples to ensure that elements of interest such as Au, Ag, and/or Sn are included. Cross-sections were used for materials analyses including VLM and SEM-EDX. FTIR and Raman spectroscopy were also performed on the same sample cross-sections but are not the focus of this article. As supportive data, experimental conditions for Micro-XRF, FTIR and Raman measurements are presented in Supplementary Materials: S2.

Samples were embedded in Araldite 2020 epoxy resin. Sample cross-sections were pre-trimmed with a *Reichert-Jung TM60* and then cut with a *Leica UC7* ultra-microtome. A *DiATOME trimtool 45* diamond knife was used with a clearance angle of 6°.

A *Leica DM4000M* VLM, coupled with a *uEye UI144xSE-C* camera, was used to observe and image the stratigraphy of the sample cross-sections in both bright field (BF) and dark field (DF) modes.

The SEM-EDX measurements were implemented through the high-resolution SEM *Hitachi Regulus 8230* at the Paul Scherrer Institute.

Experimental conditions of 5 keV voltage and 10 μA current were applied for both electron imaging and EDX analysis. Depending on samples, the *Condenser Lens 1* of 5–7 with high current, *Low Angle Detector with 100 SE Supress* (*LA100(U)*) and working distance (WD) of 2.5–3.5 mm were used for backscattered electron images (BSE). For EDX, *Cond. Lens 1* of 2–4, *X-Max detector* and WD of 6 mm were used. The sample cross-sections were coated with a thin layer (e.g. 6.5 nm) of chromium, to reduce charging issues. The built-in software *Hitachi Regulus* was used for SEM imaging and the software *Oxford – Aztec 3.3* for EDX analysis. A factory standard (5 kV set) was used for EDX quantification analysis.

3. Results and discussions

3.1. Results

Based on the Ag:Au signal ratios obtained through the HHXRF measurements, 108 objects (including 95 sculptures and 13 altars) out of the 162 surveyed objects have been preliminarily identified with Zwischgold application and an additional 9 objects, which show Ag signals in hair areas, are expected to contain Zwischgold. Since it is very rare to observe silver applied in hair, it is assumed that such objects were initially applied with Zwischgold and so they are included in the analysis. Details regarding the art historical background of these 117 objects are presented in Supplementary Materials: S6.

On the Zwischgold applied objects, the XRF signals of Ag and Au have mainly been observed in a broad but still reasonable range of Ag:Au proportion of 1:5–3:1 with the distribution peak occurring near the ratio of 1:2, as shown in Fig. 1. Note that this histogram excludes 10 measurements with Ag:Au ratios between 1:42 and 1:10, and 2 measurements between 10:1 and 14:1. Signals outside of the main range (1:5–3:1) might indicate some unusual Zwischgold application or overlaying of different metal foils (e.g. gold overlaying Zwischgold and vice versa), which usually occurs in the bordering areas during the initial application, or by later replenishment of new metal foils during the restoration treatments [22].The HHXRF Ag:Au ratio range for each identified object is presented in Table S4 (Supplementary Materials: S6).

Zwischgold application has been identified in extremely diverse areas of the 117 objects such as crown, mitre, hat, hair and/or beard, hair band, gown or cloak (especially side and rear part, and fold trough), dalmatic or dress, undergarment, fur coat, bordure of garments, fibulae and button decoration, saint's attribute, fruit, sword handle, altar frame, side wall and ceiling, altar framework and decoration, and even eye pupil. Within individual figure sculptures, Zwischgold is frequently observed on hair, bordure, fur coat, dalmatic, dress, as well as in the form of small square or rectangular patches on the golden gown (Fig. S2a in Supplementary Materials: S3.1); while in altars, Zwischgold is mainly present on the altar background directly behind the altar figures (Fig. S2b in Supplementary Materials: S3.1) and on the side wall. It is worth to point out that an extensive Zwischgold application is very rare to be observed in areas of saints' gown, which is dominated by gold. The identified Zwischgold areas of each object are presented in Table S4 in Supplementary Materials: S6.

Some interesting combinations such as Zwischgold above tin press brocade [7,27,28], and Zwischgold as the metal base of sgraffito [13,29], are frequently observed in areas of saint's undergarment or bishop's dalmatic. Such combined use of Zwischgold and surface decoration techniques will be further discussed with illustrative cases in Section 3.2.

Most of the Zwischgold surfaces identified appear darkened and matte (Fig. S2a-c in Supplementary Materials: S3.1) due to their corrosion. Only a few of them exhibit golden colour tones that are slightly different from gold surfaces, indicating relatively well-preserved states. One example is a saint statue named “Woman of the Apocalypse” (LM16701.2) (Fig. S2d in Supplementary Materials: S3.1), which is also one of the very few objects exhibiting extensive Zwischgold application in the gown area.

The area of hair, beard and fur coat is one of the foci in the survey. 127 surveyed objects that include such areas have been extensively measured with the HHXRF. Although the hair and/or beard on some of the objects were over-painted with black or ochre-coloured paint, significant signals of Au, Ag or both can still be obtained. Amongst these 127 objects, 67 were identified to have Zwischgold due to the acquisition of both Au and Ag signals; 9 objects were possibly applied with Zwischgold based on the Ag signals obtained. Signals of tin (Sn) have been observed in 70 objects; amongst them, 49 showed the presence of Au, Ag and Sn together. FTIR and Raman measurements on sample cross-sections have clarified that the Sn signals detected in the hair areas were mainly from lead-tin yellow present in the layers under Zwischgold, bound with oil-based media. An example case is presented in Supplementary Materials: S3.2. Lead-tin yellow is a typical yellow pigment widely used in medieval artworks [30]. As a lead pigment, it could function as a siccative during the drying process of oil in oil/mordant gilding [31]. The combined application of Zwischgold and lead-tin yellow could possibly come from the artists' perception of the colour influence of the substrate on the gilded surface: i.e. the yellow substrate would enhance the cool, pale tonality of Zwischgold, suitable for imitating blonde hair. Such perceptions are even popular nowadays, mainly due to the common misunderstanding that metal leaf/foil is too thin to block the light reflection from the coloured substrate [32]. However, gold leaf thicker than 50 nm has no significant transparency

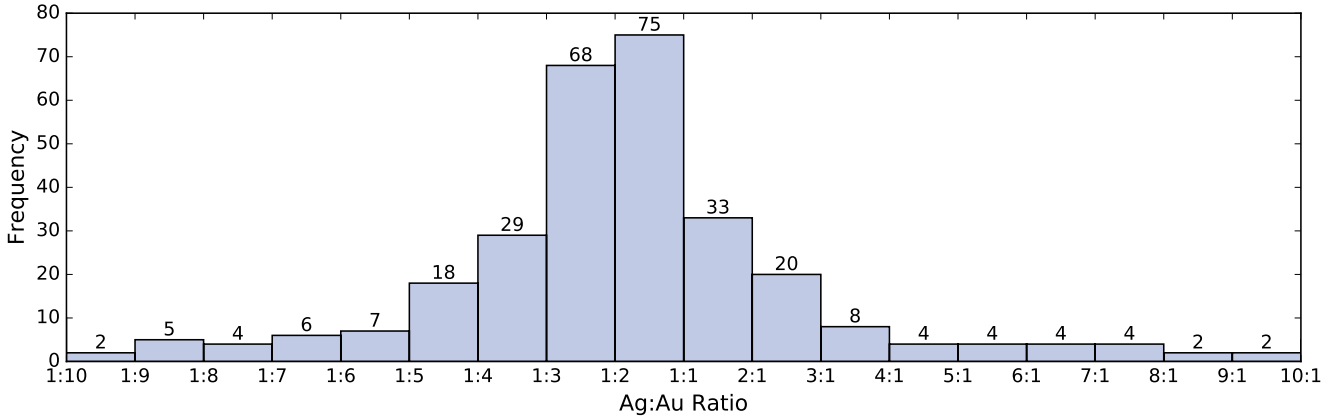

Fig. 1. Histogram of the HHXRF Ag:Au signal ratios observed in Zwischgold applied objects. The most commonly observed ratios occurred near 1:2.

[33].

It is interesting to observe that another type of gold imitation - vermeiled silver (i.e. silver surface coated with yellow varnish) [7,8,34] was also applied on a couple of objects. In our visual observations, surfaces applied with such vermeiled silver exhibit a similar colour tonality to Zwischgold in well-preserved states (i.e. slightly paler than gold) and can be thus easily misidentified as Zwischgold to the naked eye. In addition to vermeiled silver, brass leaf, which was a common economical substitute of gold leaf in recent times [34,35], was also observed in the fold troughs of some figure gowns, where clear copper (Cu) and zinc (Zn) signals were obtained through the HHXRF. Such surfaces appear slightly darker than the surrounding gold surfaces and thus could be falsely identified as slightly corroded Zwischgold. Example artefacts applied with brass are presented in Supplementary Materials: S3.3.

As the main focus of this article, the function and purpose of Zwischgold application will be further discussed with illustrative cases in Section 3.2, together with an example showing a combined use of vermeiled silver and Zwischgold. Some interesting technological details observed through samples taken from the example objects are also presented.

3.2. Further discussions with examples

Six examples involving 10 objects were discussed in this case study section, combining their art historical background, output of HHXRF measurements, and technological details observed through samples. Table 1 presents a summary of Zwischgold applied areas of each example object, together with its functions and hypothesized purposes. Detailed information regarding the analysis of HHXRF data for specific Zwischgold applied areas of each example object is presented in Supplementary Materials: S4. Object descriptions, some of the object images, and other supportive data are presented in Supplementary Materials: S5.

3.2.1. Example 1: relief Last Supper (AG61)

The relief (Fig. 2a), titled *Last Supper*, was previously exhibited on the inner side of a retable wing from the Cloister Rheinau, Canton Zurich, dated in early 16th century [36]. The relief was attributed to Hans Thomann, a master from Memmingen, southern German, active in the beginning of 16th century [36].

The relief exhibits a visual colour palette of gold, brown, blue and black: gold is exclusively used for the gowns; brown for table, cutlery, lamb and the undergarment of the lower, right apostle; blue is partly applied on the under garment of the lower, left apostle. Except for the sleeping John whose hair appears slightly brownish gold, all other figures have black hair and/or beard. Through HHXRF measurements, a more abundant and deliberately selected colour set has been revealed (Fig. 2b): while the gowns of all figures were confirmed to be applied with gold leaf as we have expected, the wine cups, plates and knifes, as well as the gown lining of the lower, left apostle were identified as silver. The bread, as a gift from God and the symbol of the body of Christ [37], was applied with gold, while the Passover lamb inside of the central big plate showed no metal application.

Zwischgold was identified on John's hair and the undergarment of the two lower apostles (Fig. 2b), functioning as the metal base of sgraffito. A blue paint layer was partly applied above the Zwischgold surface of the lower left apostle. The HHXRF spectra with the “High range” filter show strong Cu peaks alongside with Au and Ag signals, indicating the use of copper-containing pigments for the blue coating (Fig. 2c). FTIR and Raman analysis of a sample taken from the underside of the right sleeve of the lower, left apostle further indicated azurite bound with protein-containing media (Fig. S6a-b in Supplementary Materials: S5.1). The SEM-EDX analysis of the same sample confirmed the presence of Zwischgold, in which the silver layer (below the gold layer) was strongly corroded and transformed into silver corrosion products (mainly silver sulphide) (Fig. 2e). On the undergarment of the lower, right apostle, a red-brownish half-transparent colour layer was observed above the Zwischgold surface. The correlated rate of silver and gold XRF signals detected in both lower apostles exhibited a high-level similarity (mainly in range of 1:2–1:3), indicating the application of the same Zwischgold foils.

The reason why Zwischgold was applied specifically on the undergarment of two lower apostles is uncertain. Since Zwischgold was much cheaper than gold leaf of that time, it was possible from an economic consideration. Further, the slightly different golden colour tone of Zwischgold could give viewers a visual contrast against the extensive use of gold in the object. However, through a close observation on the gown collar of Jesus and other apostles, which is decorated with black-coloured sgraffito, a slight colour difference between the golden base of the “collar sgraffito” and that of the “undergarment sgraffito” can be discerned (Fig. S5b-d in Supplementary Materials: S5.1). The HHXRF measurement identified gold as the metal base of the “collar sgraffito”. Therefore, the usage of gold and Zwischgold for sgraffito in this case also shows a materials hierarchy: gold for gown and Zwischgold for undergarment.

3.2.2. Example 2: altar shrine St. Magnus with sponsor and dragon; virgin and child with St. Anna; St. Francis (DEP64)

The use of different metal foils for sgraffito was also observed in the altar shrine *St. Magnus with sponsor and dragon; Virgin, Child and St. Anna; St. Francis,* which was dated 1520 and attributed to Jörg Lederer, a well-known master from southern Germany [38].

The HHXRF measurements identified Zwischgold to be extensively applied in diverse areas such as the hair of Mary and the baby Jesus (Fig. S7a in Supplementary Materials: S5.2), the flesh of the pomegranate (Fig. S7d in Supplementary Materials: S5.2) held by Mary's hand, the top part of Magnus's hat, small patches in fold troughs of saint gowns, the inner altar frame, the grape vines in the frame decoration, and even the bird eyes (Fig. S7e in Supplementary Materials: S5.2). The application of different metal foils for sgraffito was observed on Anna's veil and wimple: Zwischgold for veil and silver for wimple (Fig. S7b-c in Supplementary Materials: S5.2). The most likely reason for such application could be aesthetic contrast, since there is no obvious symbolic hierarchy between veil and wimple. Likely from the same consideration, Zwischgold was applied on the flesh part of the pomegranate while its skin is applied with gold leaf. The same situation was also observed on the altar frame decoration: the grape vines were applied with Zwischgold, while the grape leaves, the bird bodies and the crown were applied with gold. Zwischgold was also observed to be applied to the entire inner altar frame, which is not readily visible, indicating obvious economic considerations. Zwischgold application for the cost saving reason is also presented in the figure group of St. Magnus: while a clear materials hierarchy was observed - gold gown of Magnus, silver gown of the sponsor and painted body of the dragon, Zwischgold was observed on the top part of Magnus's hat that is not normally visible to viewers and economic considerations seem more convincing for this situation than symbolic reasons.

Despite different corrosion levels of Zwischgold surfaces, the correlated XRF signal ratios of Ag and Au in all Zwischgold applied areas (except for the bird eyes) are very close (1.5:1–1:2), indicating the usage of same Zwischgold foils. The XRF signals obtained for the bird eyes in the frame decoration showed a much higher Ag:Au ratio (7.5:1–3:1). However, since the 8 mm aperture was used for the HHXRF measurement, signal from the surrounding gold surface would have been included and it is possible that the birds’ eyes were applied with silver.

3.2.3. Example 3: sculpture St. Bishop Theodore (LM10557)

The HHXRF measurements on the relief-patterned dalmatic of Sculpture *St. Bishop Theodore* (Fig. 3a), who is recognized by his attribute “demon and bell” set by his right foot [39], showed both Au and

Table 1
Summary of example objects including their art historical background, identified Zwischgold areas, functions and hypothesized purposes of Zwischgold application.

Example	Object title	Object type	Dating	Production region	Attribution	Zwischgold applied areas	Function of Zwischgold	Hypothesized application purpose	Significant features
1	*Last supper*	Altar wing relief	1500–1510	Southern Germany	Hans Thomann	Undergarment of two lower apostles, hair of John the apostle	Imitation of blonde colour tonality, metal base of sgraffito	Cost saving, colour nuance, aesthetic contrast, materials hierarchy	Well preserved Zwischgold sgraffito
2	*St. Magnus with sponsor and dragon; Virgin, Child and St. Anna; St. Francis*	Altar shrine	1520	Southern Germany	Jörg Lederer	Bordure of Anna's veil, flesh of pomegranate, hair of Mary and baby Jesus, altar frames, frame decorations, St. Magnus's hat (top part), small patches on saints' gown	Metal base of sgraffito, imitation of blonde colour tonality, substitute of gold leaves, patching for poorly accessible areas	Aesthetic contrast, colour nuance, cost saving, higher workability	Very diverse application areas and purpose of Zwischgold
3	*St. Ulrich*	Altar shrine figure	1500	Unknown	Unknown	Bishop dalmatic	Gilding above tin press brocade, golden base of coloured glazing	Materials hierarchy, cost saving	Zwischgold gilded tin press brocade, coated with colour glaze
4	*Nailing to the Cross; Descent from the Cross; Resurrection of Jesus*	Altar wing reliefs	1520	Eastern Switzerland	Unknown	Hat, shirt and doublet of soldiers; undergarment of Mary and her companions	Gilding for non-saint figures, gilding in areas with lower symbolic meaning for saints	Materials hierarchy or symbolism	Emphasized symbolic meaning of Zwischgold; special foil structure of Ag-Au-Au-Ag; extra-thick Ag layer
5–1	*Grieving Mary; Grieving John*	Figures of a crucifix group	1470	Unknown (perhaps matching *St. Martin*)	Unknown (perhaps matching *St. Martin*)	Bordure of gown, veil and undergarment, decorations on garment	Gilding for all golden areas, substitute of gold leaf?	Possibly artisans' preference, cost-saving?	Exclusive Zwischgold application, no gold observed; similar technological features as *St. Martin*
5–2	*St. Martin with the beggar*	Altar pinnacle figure	1500	Southern Germany	Niklaus Weckmann, Ivo Strigel	Bordure of gown, veil and undergarment, decorations on garment, hair of John	Gilding for all golden areas, substitute of gold leaf?	Possibly artisans' preference, cost-saving?	Exclusive Zwischgold application, no gold observed; similar technological features as *Grieving Mary* and *John*
6	*Adoration of the Magi; St. Roch; St. Sebastian*	Altar retable	1535	Southern Germany	Unknown	Hair and beard of the second Magi from left, hair of St. Sebastian	Imitation of blonde colour tonality for young saints	Possibly artisans' intention for colour nuance	Vermeiled silver and Zwischgold in hair areas: vermeiled silver for the older saint, Zwischgold for the younger

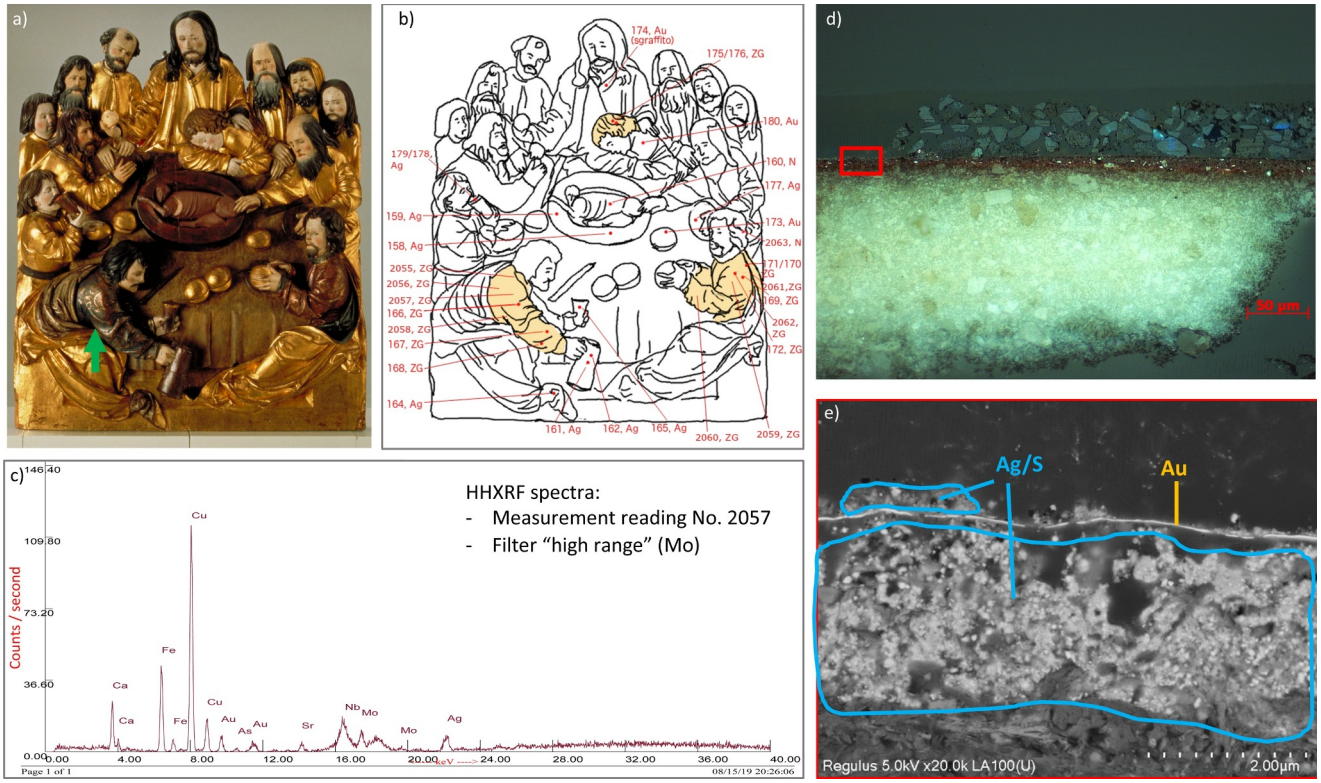

Fig. 2. (a) Overview image of Relief *Last Supper*; (b) map of HHXRF measurement positions with reading numbers (i.e. measurement indices) and detected metal signals (Au for gold, Ag for silver, ZG for Zwischgold, N for non-metal), Zwischgold applied areas are highlighted in yellow; (c) HHXRF spectra of reading No. 2057 (in the blue areas of the undergarment shoulder of the left lower apostle) showing strong Cu signals alongside with Au and Ag signals; (d) VLM image (BF, mag. 20x) of a sample cross-section taken from the underside of the sleeve of the left lower apostle (sample taking position indicated with a green arrow in Fig. 2a); (e) SEM-BSE image of one part of the sample (inside of the red frame in Fig. 2d) showing a thin gold layer and Ag corrosion, scale bar: 2 µm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Ag signals together with a strong Sn signal, indicating the application of Zwischgold over tin press brocade. Sn signals obtained from tin press brocade on dalmatics are usually much greater than those from lead-tin yellow in hair areas (see Table S2 in Supplementary Materials: S4).

Through the HHXRF measurements, it is very interesting to observe that the green mitre was applied with silver and the green dalmatic was applied with glazed Zwischgold. Detailed image of the green glazed dalmatic is presented in Fig. S8b in Supplementary Materials: S5.3. The reason for such a differentiated application is uncertain. Since both mitre and dalmatic belong to the bishop costume, there should not be a significant difference in their symbolic meanings. The undergarment of *St. Theodore* is not shown under the dalmatic, but its sleeves can be seen, where the HHXRF measurements identified the application of silver. Meanwhile, Theodore's cloak was observed to be applied with gold and his "demon" attribute was only painted with colour. The combined use of different metal foils in different areas of the saint statue and no metal application for saint's attribute, which we have also observed on St. Magnus in the previous example (Example 2), indicates a clear materials hierarchy. However, the possibility that Zwischgold was used here as an economical substitute for gold leaf cannot be fully excluded: if a lower-cost metal foil, such as Zwischgold, shows a similar golden colour tone and is protected by a coloured glaze, there is indeed no need for gold leaf.

The SEM-EDX analysis (Figs. 3c–f) confirmed the application of Zwischgold on the dalmatic of St. Theodore. It is also interesting to observe two thin gold layers below the relatively thick corrosion layer (mainly silver sulphide, mixed with small amount of silver chloride), indicating a possible application of multi-layered Zwischgold, which is a very rare type of Zwischgold, containing multiple, alternating gold and silver layers [40]. Further discussion of the multi-layered Zwischgold is in Section 3.2.5.

3.2.4. Example 4: relief series nailing to the cross (LM18014), descent from the cross (LM18015), resurrection of Jesus (LM18016)

These three reliefs (Figs. S9a–c in Supplementary Materials: S3.4) originally came from the inner sides of a winged retable in the Canton Freiburg [41], in which the symbolic meaning of the Zwischgold application is obvious. While gold was applied on the loincloth or robe of Jesus and also on the gown of saints such as Mary and her companions, Zwischgold was widely applied on the hat, shirt and doublet of soldiers, and also on the underdress of saints (Figs. S9d–f in Supplementary Materials: S3.4). This obvious materials contrast can be observed especially clearly in *Resurrection of Jesus*. It is also interesting to observe that John's hair was applied with Zwischgold in *Descent from Cross,* while the blonde hair of soldiers shows vermeiled silver in the same relief (Table S2 in Supplementary Materials: S4).

An unusual application of Zwischgold has been observed on certain figures in the reliefs. For example, a sample was taken from the lower part of the doublet of the second figure from right in Relief *Descent from the Cross* (Fig. 4a), and the SEM-EDX analysis of the sample exhibits a Zwischgold foil containing two gold layers with relatively thick silver corrosion layers above and below the gold layers (Fig. 4c). The initial foil structure is uncertain: it could be multi-layered Zwischgold with Ag-Au-Au-Ag structure, or a single folded Zwischgold foil with silver side facing outwards. Compared to the Zwischgold foils observed in most of the identified objects, having such thick silver layers and also with gold layers between them are both very unusual.

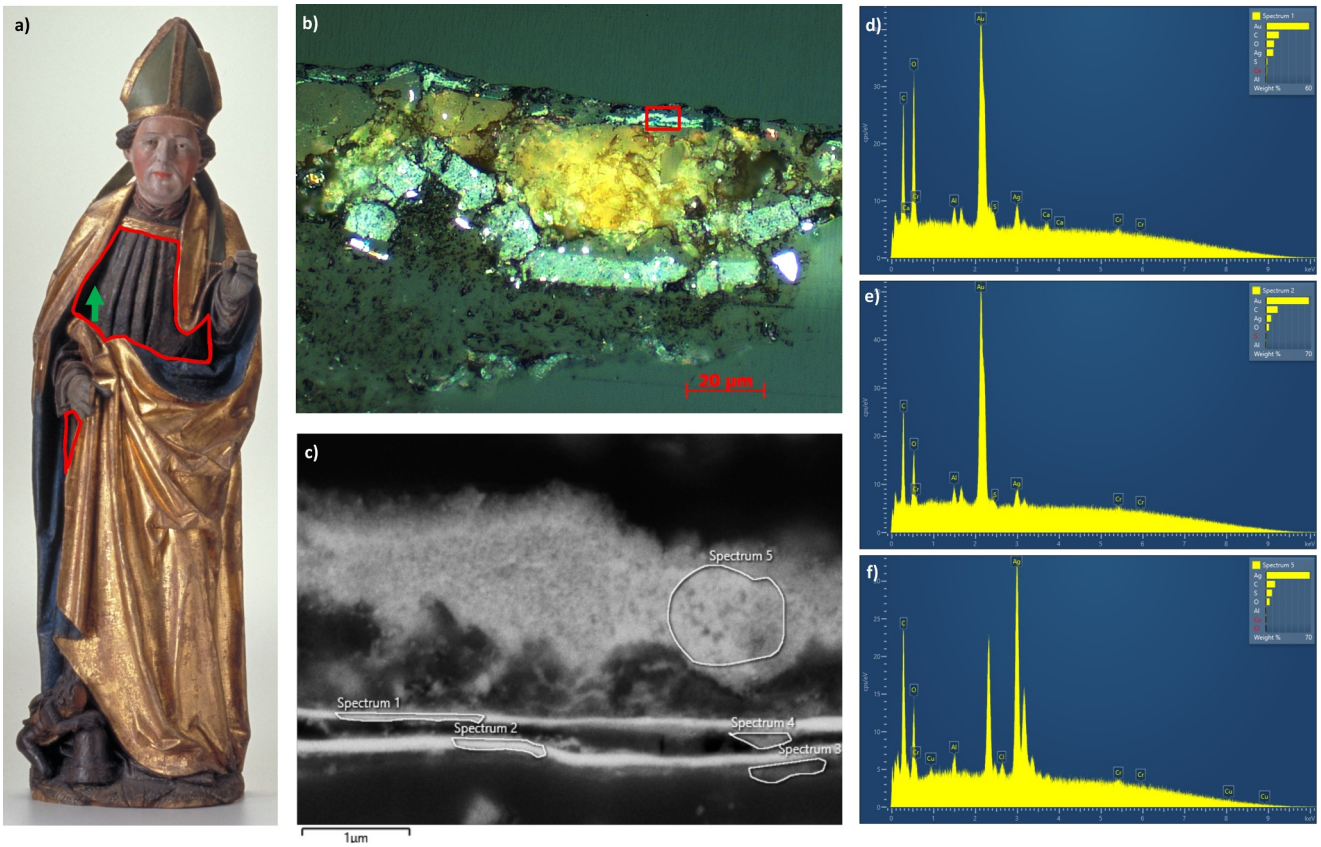

Fig. 3. (a) Overview image of Sculpture *St. Theodore*, Zwischgold applied areas indicated with red frames; (b) VLM image (BF, mag. 50x) of a sample cross-section taken from the dalmatic of the bishop (sample taking position indicated with a green arrow in Fig. 3a); green glaze is not attached on this sample; (c) SEM-BSE image of one part of the sample (inside of the red frame in Fig. 3b), scale bar: 1 µm; (d–f): EDX spectra of spectrum 1, 2 and 5 indicated in Fig. 3c. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.2.5. Example 5: sculptures Grieving Mary (LM9730), Grieving John the apostle (LM9731) and St. Martin with the beggar (LM9489)

The sculptures *Grieving Mary* and *Grieving John the Apostle,* dated 1470, were originally from Pleiv, Canton Graubünden [42]. No clear attribution was previously given to these sculptures and the museum catalogue states that the production region is "unknown, possibly Lombardy" without evidence [42].

The HHXRF measurements indicate that Zwischgold was exclusively applied in all golden areas of both sculptures, including the bordure of garments and the round, golden decorations on the undergarment (Fig. S10a-d in Supplementary Materials: S5.5). No gold leaf was observed on either sculpture. The hair of St. John was also observed to be applied with Zwischgold but now appears brownish due to corrosion (Fig. 5a). Meanwhile, tin signals were obtained in John's hair and in the golden decorations on the undergarment. The presence of Zwischgold was further confirmed through the SEM-EDX analysis of samples taken from the bordure of Mary's veil (Figs. S11a–b in Supplementary Materials: S5.5) and hair of John (Figs. 5c–d).

The exclusive application of Zwischgold in golden areas has been also observed in some other surveyed objects. For example, Sculpture *St. Martin with the beggar*, a pinnacle figure of a retable, also shows Zwischgold application together with underneath paint layers containing lead-tin yellow particles (confirmed through SEM-EDX by obtaining both Sn and Pb signals) for all golden bordures and the hair. The object is dated 1500 and attributed to Niklaus Weckmann together with Ivo Strigel, two well recognized masters from southern Germany who

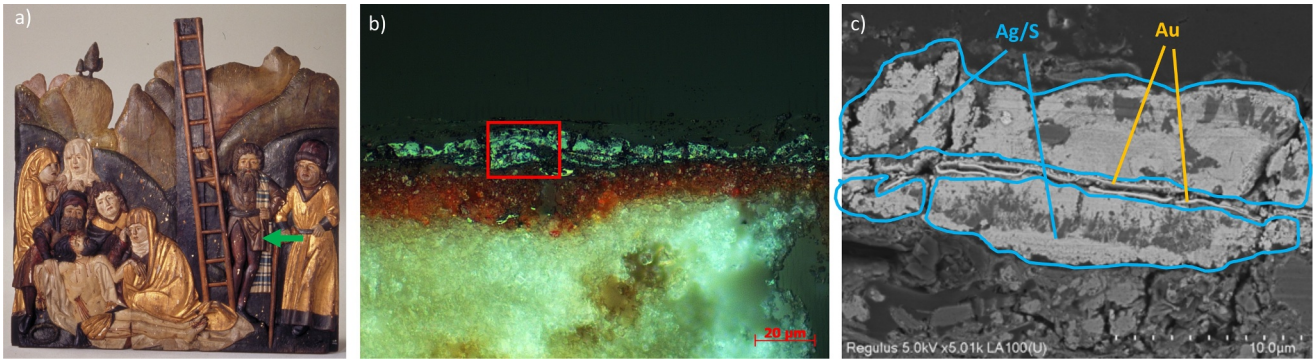

Fig. 4. (a) Overview image of Relief *Descent from the Cross*; (b) VLM image (BF, mag. 50x) of a sample taken from the doublet of the second figure from right in Relief *Descent* (sample-taking position indicated with a green arrow in Fig. 4a); (c) SEM-BSE image of the same sample with the measurement area inside of the red frame in Fig. 4b, scale bar: 10 µm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

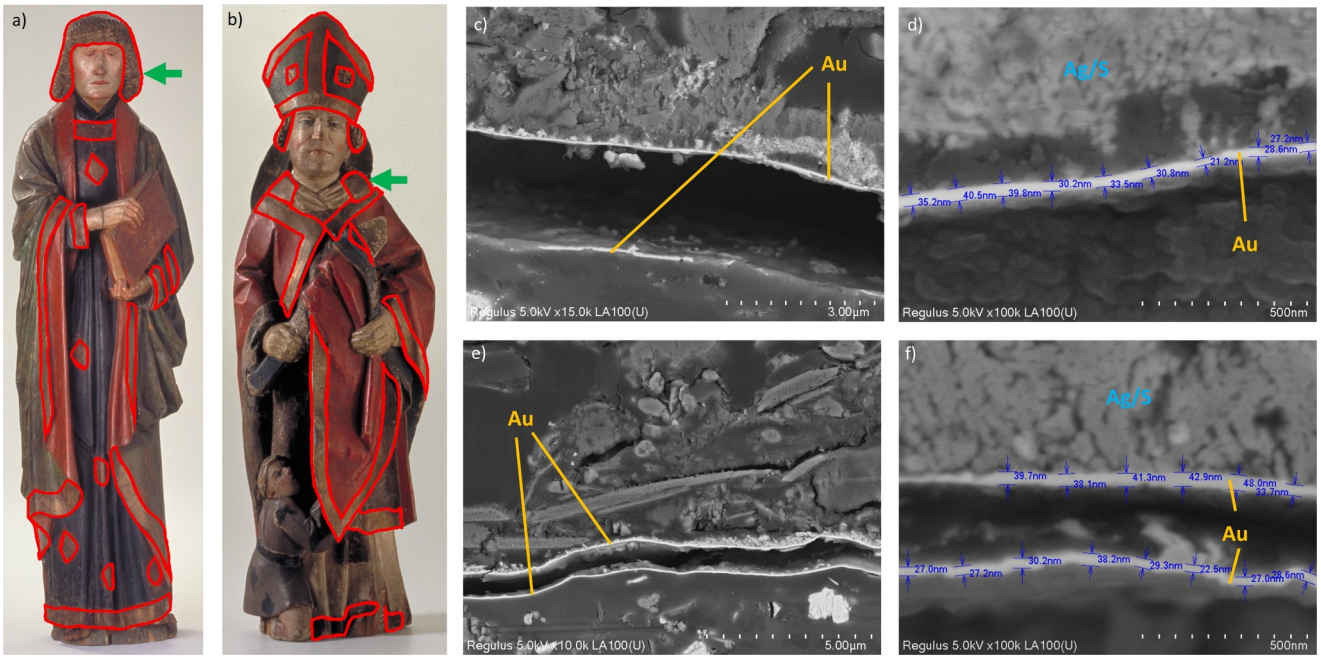

Fig. 5. Overview images of Sculptures (a) *Grieving John the Apostle* and (b) *St. Martin with the beggar*. Zwischgold is identified to be exclusively applied in all golden areas (indicated with red frames); (c–d): SEM-BSE images of a sample cross-section taken from the hair of St. John (sample taking position indicated with a green arrow in Fig. 5a) with two magnifications: 15kx and 100kx; (e–f): SEM-BSE images of a sample cross-section taken from the gown collar of St. Martin (sample taking position indicated with a green arrow in Fig. 5b) with two magnifications: 10kx and 100kx. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

were active in the turn of the 16th century [43]. Morissen, in Canton Graubünden, was recognized as the original exhibition place of *St. Martin* by some art historians, based on its art style and some research studies correlating artworks exhibited in Morissen to those produced in Memmingen [43]. It is interesting to point out that the geographical distance between Pleiv (the original exhibition place of *Grieving Mary* and *Grieving John*) and Morissen is only 3 km. In addition, the art style and polychromy of *St. Martin* and the two "Grieving" sculptures show a high-level agreement. Based on such observations, a question has been raised: is there any correlation between these three sculptures regarding the attribution?

The SEM-EDX analysis of the samples taken from *Grieving John* and *St. Martin* gives a positive answer by showing similarity in many of the technological features of the applied Zwischgold, including the same foil structure – a sandwich-like multi-layered Zwischgold type containing two gold and one silver layers (with structure of Au-Ag-Au) (Figs. 5c and e), similar gold layer thickness (mainly 25–40 nm) (Figs. 5d and f), very low copper content (Cu:Au weight ratio < 2%) in the gold layer, and similar main component in substrate e.g. lead-tin yellow. From SEM imaging, it is clear to observe a gap between two gold layers. The formation of such a gap was mostly likely caused by silver having diffused through the grain boundaries of the gold layer and then transformed into silver corrosion products [26]. Supportive SEM-EDX data for these two samples, regarding the Cu content in the gold layer and materials composition in substrate, are presented in Supplementary Materials: S5.5.

3.2.6. Example 6: altar retable adoration of the magi; St. Roch; St. Sebastian (IN66)

Vermeiled silver can be easily misidentified as Zwischgold by the naked eye, since it also appears slightly paler than pure gold when it is still well preserved. Vermeiled silver has been observed in the surveyed objects with a very low frequency. An example is the shrine of the altar retable *Adoration of the Magi; St. Roch; St. Sebastian* (Fig. S13b in Supplementary Materials: S5.6), in which vermeiled silver was observed to be applied on the hair and beard of one of the Magi, who is kneeling towards Mary and the baby Jesus while presenting a gift (Fig. S13a in Supplementary Materials: S5.6). It is interesting to observe that the beard of the second Magi, who is standing to the right of the kneeling one, was applied with Zwischgold (Fig. S13c in Supplementary Materials: S5.6). (Zwischgold is also observed to be applied on the hair of St. Sebastian on the right wing of the retable.) The different colour tones between vermeiled silver and Zwischgold in such hair and beard areas seem quite subtle, with the vermeiled silver slightly paler. The reason for such special application is uncertain, possibly it was the artist's intention to differentiate two Magi in different ages and the elder Magi was intended to have more whitish hair and beard than the younger one.

In addition to the above mentioned six examples, it is worthwhile to give some attention to the darkened Zwischgold patches in the folds on saints' gowns. Many researchers believe that such application is mainly for cost savings, however our experience during the production of gold and Zwischgold models in a previous study [22] provides an alternative: the high workability of Zwischgold. Once Zwischgold is applied onto the wet bole [44] in water gilding [32,45]), its application position can be slightly adjusted, which is impossible during the application of gold leaf that will tear easily. Zwischgold is therefore easier to apply than gold leaf for positions that are difficult to access or need high accuracy and this would have provided motivation for its application in such areas.

4. Conclusions

This object survey at the Swiss National Museum focusing on the historical application of Zwischgold provides the first significant body of data regarding this special gilding material and its use in late medieval sculpture. The abundant application of Zwischgold in extremely diverse areas on more than 100 surveyed objects (more than two thirds of the total surveyed objects) has been proven with spectroscopies and imaging. Such large-scale, first-hand data helps to understand why and how Zwischgold was applied in artworks of that time, which correspondingly provides us many opportunities to understand the material

and the society in which it was created and evaluated. Meanwhile, the application of Zwischgold with similar technological features such as foil structure, gold layer thickness and materials compositions in the surrounding layers provides reliable supportive data for the attribution of medieval sculptures.

Based on the survey results, we believe that the application of Zwischgold in medieval sculptures should not be merely regarded as an economical substitute for gold leaf, despite its lower cost than gold leaf of that time. The functions and purposes of Zwischgold application can be concluded in several aspects, with individual situations determining their relative weight, and hence presented either solely or in an integrated manner. In other words, there is no absolute evaluation criteria about which factor is more important, and which is less. Some common purposes of Zwischgold observed in the surveyed artefacts include:

- Economic reason: a typical situation from this aspect is that Zwischgold was applied in areas invisible to viewers, including the side and rear of the figure gown, altar background directly behind figures, altar side wall and ceiling. In most of such areas, Zwischgold exhibits strongly corroded appearance possibly due to less care, and therefore is easy to recognize. This, on the other hand, explains why Zwischgold is commonly regarded as the economical version of gold leaf in the literature. Another common example based on the cost reason is that Zwischgold surfaces were coated with colour glaze.
- Aesthetic reason: the most representative case for this reason is Zwischgold application in hair and beard areas. Due to its paler colour tone than pure gold, Zwischgold was preferentially used by artisans for imitating blonde hair. The fact that the hair (and beard) of more than half of the surveyed saint statues were applied with Zwischgold demonstrates a high popularity of such application in medieval sculptures. Other cases from the aesthetic aspect are the contrasted application of Zwischgold and other metal foils.
- Symbolic reason: this is a very common and understandable motive in medieval works of art. As we have observed in the survey, the wide application of gold in gown, silver in undergarment and Zwischgold in dress or dalmatic shows a clear materials hierarchy. A good example is the deliberately separate application of gold and Zwischgold on the clothing of Jesus and soldiers in the scene of *the Passion of Jesus*.
- Workability: isolated dark Zwischgold patches in the fold troughs of gowns are commonly perceived as economical rescue practice for damage during the application and/or burnishing process of gold leaves. However, comparing the amount and size of such patches to most of gold gowns, this explanation is not convincing. A more likely reason is that the relatively thicker Zwischgold allows the foil's position to be adjusted on the wet substrate in a way not possible with the more fragile gold leaf and so it is easier to be applied in difficult to access positions.
- Preference of artisans or workshops: the two “Grieving” sculptures and “St. Martin” for example show correlated technological features, suggesting they originate from the same workshop. These sculptures also show the same usage of Zwischgold despite the variations seen across the survey, which could be a demonstration of the preference of the workshop.

As a non-invasive analysis method, HHXRF demonstrated itself to be practical and reliable during the preliminary identification of Zwischgold and other metal foils applied on sculptures and large-scale altars. The accuracy of HHXRF analysis has been confirmed with materials analysis on samples such as SEM-EDX. However, researchers should be aware that many factors influence the XRF signals, and therefore the HHXRF measurements can be only used for qualitative analysis or preliminary investigation. Meanwhile, situations such as the ageing or restoration of historical objects need to be considered in order to prevent false interpretations.

CRediT authorship contribution statement

Qing Wu: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Visualization. **Tiziana Lombardo:** Conceptualization, Formal analysis, Investigation, Resources, Writing - review & editing, Supervision. **Vera Hubert:** Formal analysis, Investigation, Writing - review & editing. **Erwin Hildbrand:** Formal analysis, Investigation, Writing - review & editing. **Peter Wyer:** Investigation, Resources, Writing - review & editing. **Frithjof Nolting:** Conceptualization, Methodology, Resources, Writing - review & editing, Supervision. **David Ganz:** Conceptualization, Methodology, Writing - review & editing, Supervision, Project administration.

Declarations of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

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