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Illuminated Manuscripts from Turfan Tracing Silk Road Glamour by Analyzing Pigments

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Abstract Investigation of manuscripts found in sites along the Silk Road shows the history of communication and exchange between Asia and the Mediterranean over the centuries. Different scripts and languages preserved on paper in certain styles provide information about traditions and provenance. To get an idea of the cultural background of the highly developed painting techniques obvious in some illuminated manuscripts, the material used for writing and painting is analyzed. Singular Sogdian and Tocharian manuscripts from Turfan show text on one side with a colorful painting on the other side of the document. In comparison with other productions of different periods or usages from the site, special features are worked out to characterize cultural roots or influence in the network of relationships. Details analyzed in the most elaborated documents show various influences led to the colorful masterpieces.

Statement of significance The bundle of manuscripts from Silk Road sites, distributed in different collections, is the most important source material for historic investigation and comparison of scripts, languages, and texts. It became obvious that not only originals, but also translations and copies are common. Some fragments are supposed to fit together. Knowing the composition of inks and paints is often the only way to determine that they have a common source. The highly developed writing material used in a specific place or region or for a special purpose, in particular, gives an extremely important hint about the cultural context of a document.

Keywords Pigment analysis of manuscripts/ micro-XRF-; VIS-; FTIR- spectroscopy/ Silk Road Cultures in Turfan and Dunhuang/ whitish surface/ arsenic and gold

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Introduction

At the beginning of the 20th century, A. Grünwedel and A. von Le Coq brought a great variety of manuscripts from the oasis of Turfan in Xinjiang, China to Berlin, Germany. These documents, written in different languages and scripts, reflect the cultural diversity of the people who travelled along trade roads between East and West. Philology has distinguished the manuscripts' origin from the 6th to the 13th century (Durkin-Meisterernst *et al.* 2004). Cultural contact, communication, and influence along the Silk Roads led to an exchange of ideas and material, manifested in the different scripts, texts, and inks used for writing. Elaborated for different purposes, non-local styles of manuscripts are thus common. Quite a few manuscript fragments show special manufactures with highlighted text on one side of a sheet of paper and colorful paintings on the other. Examination of their inks reveals special materials and compositions (Nöller

and Helman-Wazny 2013). Because the trade in precious pigments always played an important role along the Silk Road, the artists probably obtained their material also from non-local sources. In order to clarify the cultural background of these high standards, similar inks and techniques identified on the fragments are compared and exposed as possible markers of certain traditions in manufacture.

Experimental methods

From a compilation of nearly 200 fragile manuscript fragments selected for ink analyses, three fragments classified as Tocharian (THT 1685 a, d, h Figure 2, THT 1685 b + e, THT 3520)¹ and two as Sogdian (So 10100q, So 10237–10239 Figure 3) have written text on one side and remnants of a colorful painting on the other side. Manuscripts with corresponding special material characteristic for this high level technique of manufacture are detected in a wider cultural

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¹The manuscripts are published in the database of the Berlin- Brandenburg Academy of Sciences and Humanities (BBAW) <http://www.bbaw.de/forschung/turfanforschung/dta/index.html> and of the British Library, International Dunhuang Project (BL, IDP) http://idp.bl.uk/database/database_search.a4d.

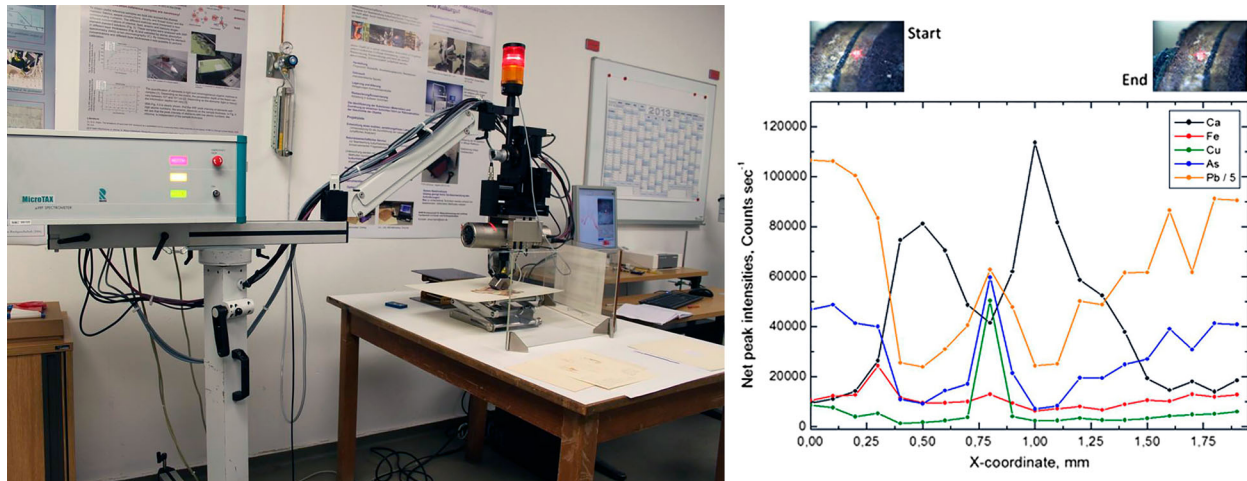


Figure 1 Non-invasive XRF investigation with ARTAX, Bruker Nano GmbH (left). Net peak intensities from different elements as a function from the line scan (right) of manuscript THT 1685a.

context on four Uighur (U 3832, U 3886, U 7123, U 7255), one Syrian (SyrHT 101), one Manichaean (M 660a-b) and two Tibetan (Tib 1366, Tib 1410) manuscripts from Dunhuang. Compared are furthermore two Buddhist illuminations from Chotscho in Sogdian script and an Indian Pustaka book format with long lines (MIK III 4932, Figure 6) and with short lines (MIK III 4984, Figure 7) for suggested similarities regarded as significant for tracing.

The manuscript fragments stored under glass covers in archives are opened for analysis, cleaned and carefully conserved between new covers again.

The analysis of the inks and pigments is conducted on site using non-destructive methods. It encompasses visible reflectance spectroscopy (VIS, Spectral Photometer SPM 100 Gretag Imaging AG company, Regensdorf, Switzerland, 1,5 mm spatial resolution, spectral resolution: 10 nm) for primary characterization of the colored materials (Fuchs and Oltrogge 1994). The method uses only the visible range of the electromagnetic spectrum (380 nm - 730 nm). Mobile X-ray fluorescence spectroscopy (XRF, ARTAX, Bruker Nano

GmbH, Berlin, Germany) was used for determination of elements. The instrument is equipped with an X-ray tube with a Mo target. An integrated CCD camera makes it possible to locate the X-ray precisely with a laser spot (Bronk *et al.* 2001). Measurements are done in line scans (Figure 1). Elements from Si (14) to U (92) are detected and evaluated using a software program. The semi-quantitative determination of the elemental composition of the inks gives a hint about their provenance and preparation technique, which is characteristic for regional workshops (Hahn *et al.* 2005). Fourier transform infrared spectroscopy (FTIR, EXOScan 4100, Polytec GmbH, Waldbronn, Germany) is used to determine molecular signatures. We use the ExoScan in a diffuse reflection mode (spatial resolution: 10 mm, spectral range: 600–4000 cm^{-1} , spectral resolution: 4 cm^{-1}).

Results and discussion

Whitish surface with text

Some manuscript fragments (Figure 2 and 3) show colourful illuminations on the verso side. Under the black



Figure 2 Tocharian THT 1685 a-h.



Figure 3 Sogdian So 10237–10239.

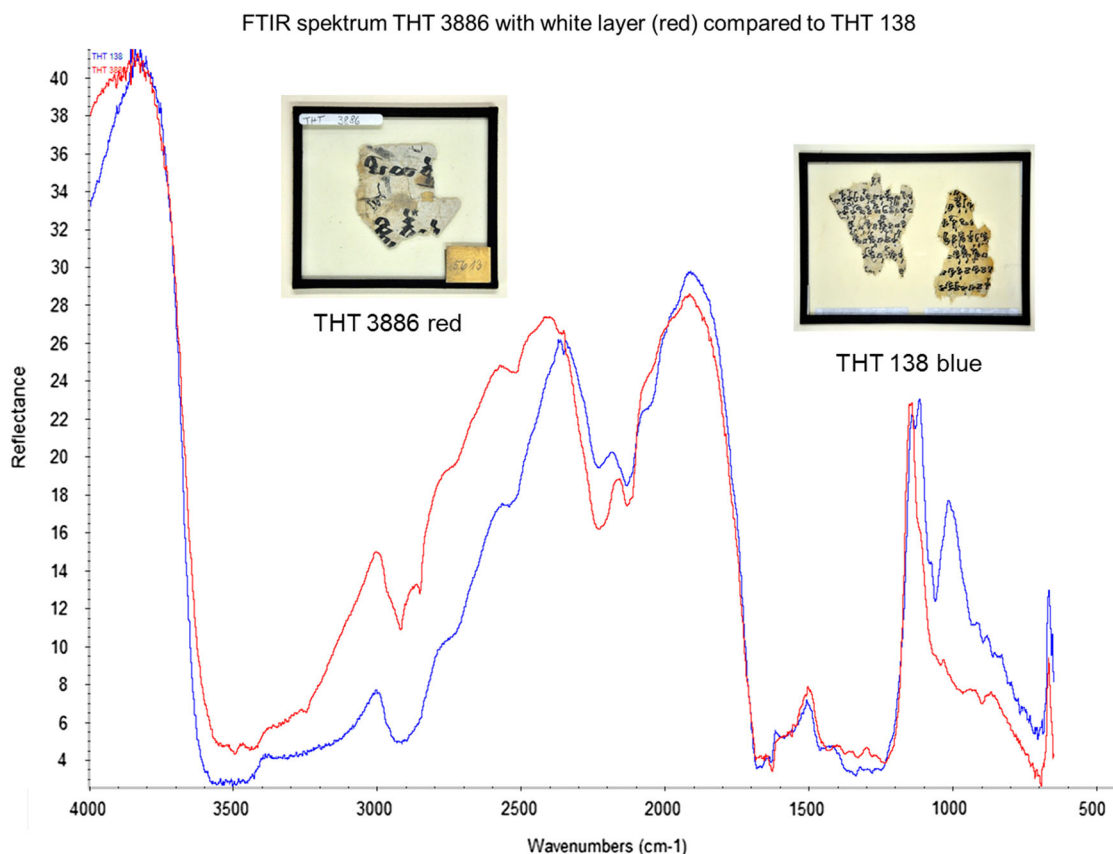


Figure 4 FTIR spectra of Tocharian manuscript fragments with a surface layer of gypsum.

letters of the writing, the paper is specially prepared with a whitish surface.

This white surface is identified with FTIR as gypsum (Figure 4). Such priming is detected on manuscripts mostly classified as Tocharian and, with a thinner or more diluted and sometimes hardly even perceptible

substrate, on some Sogdian fragments. Paper treated this way does not always show a painted illumination on the other side, so we infer that such a priming was not necessarily applied as barrier layer.

The whitish surface on the illuminated fragments is variable in thickness, but also in material. In addition to

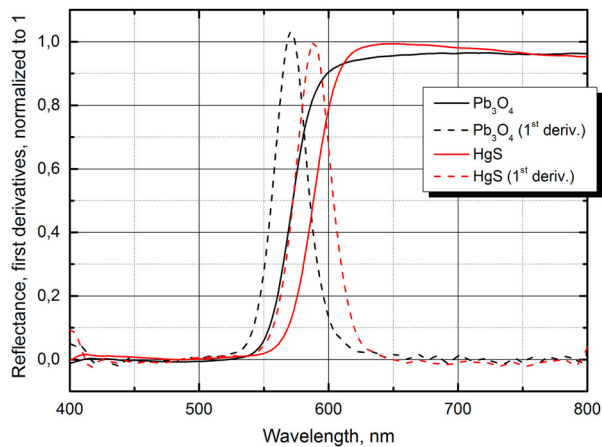


Figure 5 VIS-spectra (reflectance and 1st derivation) of cinnabar and lead red.

gypsum, chalk or starch was applied under black letters. The use of gypsum and chalk is seen as a further development of the treatment of paper with starch, a technique mentioned for Sanskrit manuscripts (Sander 1968).

The black ink used to write the texts of the Tocharian and Sogdian manuscript fragments reveals a significant common feature. The ink material was identified as carbon ink enriched with traces of a copper compound².

Pigments

Remnants of paintings on the verso side of the Tocharian and Sogdian documents THT 1685 a-h, THT 3520, So 10100q and So 10237–10239 with a whitish surface show a rich palette of colors. The pigments identified with XRF analyses are red lead (Pb_3O_4) on the Tocharian fragments in a rather uncommon brownish variation (Aze 2008) and additionally cinnabar (HgS) on the Sogdian fragments, confirmed by VIS spectroscopy (Figure 5). Red lead is identified furthermore on Tibetan and Syrian manuscripts in our analyses. Red ink made of cinnabar is very common in Asia (van Gulik 1981; Hulsewé and Loewe 1979). Used for writing, painting, or decoration, the pigment has been identified on various Chinese, Sogdian, Syrian and Tocharian manuscripts. Cinnabar inks mixed with a lead compound are analyzed in nearly all red Manichaean and Sogdian writings and are often used for painting ornaments or for accentuating special words or passages of the text³.

There is no blue on the Tocharian manuscripts, but the green copper chloride atacamite (CuCl_2) and the yellow arsenic sulfide orpiment (As_2S_3), detected by XRF analysis. Shining gold used for yellow coloration is confirmed with different trace elements.

Blue indigo is identified on a Sogdian fragment by its typical color curve. This kind of blue, as well as blue

and green copper pigments, are commonly applied along the Silk Road.

To learn more about the preferred pigments detected on the fragments that bear a painting and to clarify common features of similarly produced documents, additional manuscripts from Chotscho in Sogdian script with elaborated illumination on one and written text on the other side are analyzed (Figure 6 and 7).

The investigated black carbon inks contain a Cu compound identical to that found with XRF-analyses in all the fragments mentioned before. The pigments of the illumination MIK III 4932 are red cinnabar and a mixture of cinnabar with a lead compound, gold with traces of copper and white cerussite (PbCO_3). Arsenic is detected together with a lead compound in painted outlines. The VIS-spectrum shows blue lapis lazuli. Fragment MIK III 4984 has the same variety of colors with exception of the blue consisting here of indigo. Additionally, a green copper pigment is detected. In the yellow gold, mercury and arsenic occur as trace. Different workshops confirm this for these two Sogdian manuscripts.

Apart from the more common pigments applied in the illuminated fragments that were prepared partly with a whitish surface and the special black ink of the written texts, lapis lazuli- detected by VIS spectroscopy on one illumination and gold are the most precious ones. They have a long tradition of usage also in other regions (Jurado-Lopez *et al.* 2004; Plinius 2007). The most famous old deposits of lapis lazuli, a pigment detected here only on the illuminated Sogdian manuscripts MIK III 4932, are in the Pamir Mountains of Badakhshan, Afghanistan and Uzbekistan, mentioned in the context of trade along the Silk Road and already very early to Mesopotamia and the Indus Valley (Schmidt *et al.* 2009).

Data items concerning arsenic and gold

As arsenic is present in different compounds on all the mentioned fragments, it and gold are regarded as the most interesting materials for comparing workshops of manufacture in a wider cultural context.

The most common source of arsenic is the yellow pigment **orpiment** (As_2S_3), long used in China (Golas 1999; Needham 1972; Schafer 1955; Silbergeld and McNair 1988) and in other old cultural regions (Caley 1946; Colinart 2001; David *et al.* 2001; Kitchen 2001). It was applied interchangeably with gold for writing and painting. Orpiment applied as yellow pigment, resembles gold and in former times was traded at an equal price (Cennini 1437; Grundmann and Richter 2008). It is strongly associated with gold also because it can be found in the same natural formations and thus serves to find gold deposits (Ye Myint 2004). Arsenic, not necessarily as orpiment, thus occurs on manuscripts worked with gold.

²This indicates a special preparation or manufacturing technique, detected also on most of the Sanskrit and Syrian manuscripts.

³This type of ink is found more rarely on Chinese, Tibetan, Tocharian, and Uighur fragments.



Figure 6 Sogdian MIK III 4932.



Figure 7 Sogdian MIK III 4984.

Orpiment is often used as yellow pigment on **Uighur** and Tibetan manuscripts (Figure 8). The Uighur manuscript U 3832 is written in bright yellow script on indigo-colored dark blue paper, which is strongly related to the Tibetan design of Buddhist texts (Boisselier 1976). The ink is made of orpiment in the purest form, indicating its artificial preparation. On other manuscripts, the yellow pigment appears together with traces of Fe, Pb, or Hg. The more greenish yellow of fragment U 3886 is due to its Fe content. The light red in this fragment is a mixture of orpiment with cinnabar. Another manuscript, U 7123, shows yellow orpiment partly applied under red lead and black. A very special Uighur document painted on

textile, U 7255, shows yellow parts worked with pure gold over orpiment and red lead with arsenic as a trace element.

The high content of Fe and Pb or Hg together with As can be seen as a mixture of pigments (FitzHugh 1997; Wallert 1984), unless it is a hint that there were different natural sources of the raw material used on the Uighur manuscript fragments.

In the selection of **Tibetan** manuscripts, orpiment is commonly used as paint and ink for yellow coloration, though gold is known to have been used often in this cultural region. Imported to Tibet from India (Jackson 1976), gold was applied in the form of leaf on a base coat made of a mineral pigment. Gold

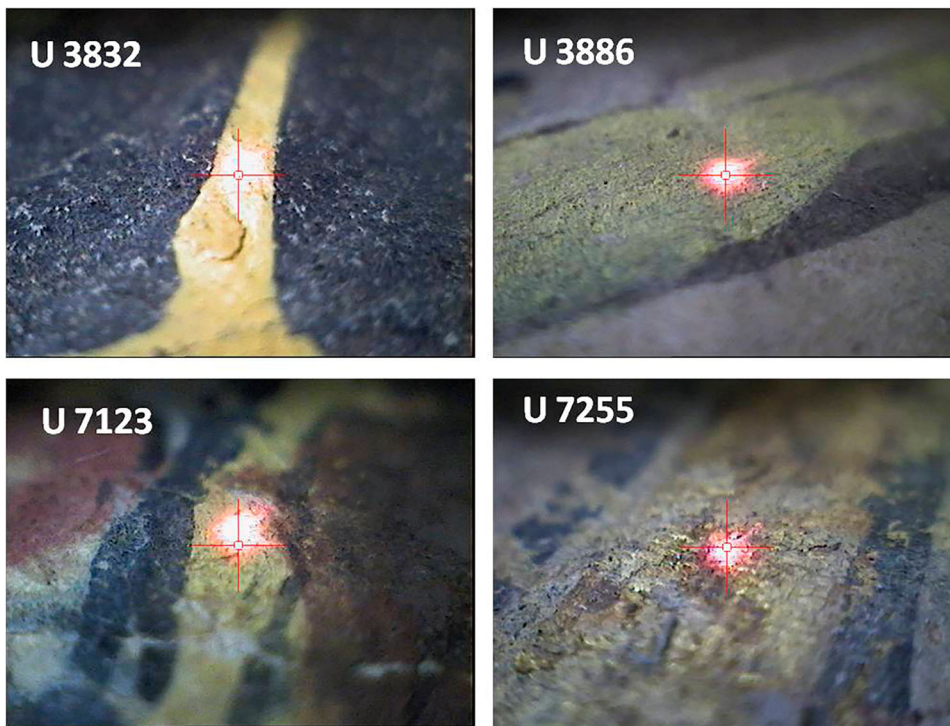


Figure 8 Photomicrographs from different manuscripts, showing different yellow areas selected by a laser spot for X-Ray analysis. Uighur U 3832: with pure orpiment; Uighur U 3886: orpiment with Fe; Uighur U 7123: orpiment and red lead; Uighur U 7255: orpiment under gold.

powder was traded as gold drops prepared with a binder; when used as paint, it is ground together with mercury and mixed with glue.

As in the Uighur fragments, orpiment on Tibetan manuscripts is identified with Hg, a trace element or mixture mentioned for Than-kas in Tibet (Duffy and Elgar 1997; Rötter *et al.* 2008) that originated in the eleventh century in Nepal from Hindi Indian cultural roots. Mixtures of orpiment with cinnabar (Hg) or ochre (Fe) are mentioned in the literature for producing brown colors (Giaccai and Winter 2005). But arsenic is strongly associated with mercury also in natural sources, as is well known for deposits in Tibet, so that a local origin of the pigment and a relation between Uighur and Tibetan sources may be possible.

The different yellow hues of the orpiment containing Hg on Tibetan manuscripts depend on the concentration of the pigment.

On the **Syrian** manuscript SyrHT 101 (Figure 9), yellow orpiment is applied in high concentration as a layer under a red-brown coat- possibly of lacquer with fissures. On this particular document, it is detected without trace elements, artificially prepared.

The use of orpiment together with varnish (lac dye or shellac) to make paper more opaque and light straw-colored is known from India also in later centuries (Schwarz 1912). Coating with arsenic is a widespread technique already applied in the 8th to 10th century on palm leaf manuscripts in western India and later in Nepal, identified for example on sheets of paper in Sanskrit sutras from Nepal (Masato *et al.* 2007). It was used before painting to soften and at

the same time to protect the surface of the paper, serving as insecticide and fungicide. This shows the abundance, easy availability, or low price of arsenic in this region. Exploitation of abundant raw material is well known from very old deposits in Persia (Iran), which may signify an influence from or trade relation with this region (Schafer 1955).

On the **Tocharian** manuscript THT 1685a, arsenic is also detected as yellow orpiment. The element arsenic is furthermore identified on THT 1685a and THT 1685 h together with a copper compound in the black line (Figure 1) and in red lead, showing a brownish coloration.



Figure 9 Photomicrograph with laser spot from orpiment as layer on paper, SyrHT 101.

Table 1 Manuscript fragments with white primer, arsenic or gold.

	Manuscript	white primer	As	Au	Composition (trace elements)
Uighur	U 3832		orpiment		As ₂ S ₃ (Hg, Cu)
	U 3886		orpiment		As ₂ S ₃ (Fe, Hg)
	U 7123		orpiment		As ₂ S ₃ (Pb)
	U 7255(textile)		orpiment	pigment	As ₂ S ₃ (Au-Pb) Au (Cu)
Syrian	SyrHT 101		orpiment layer		As ₂ S ₃
	Tib 1366		orpiment		As ₂ S ₃ (Hg)
Tibetan	Tib 1410		orpiment		As ₂ S ₃ (Hg)
	THT 1685 a, d, h	yes	Orpiment in Pb-brown	no	As ₂ S ₃ Pb (As,Cu)
Tocharian	THT 1685 b, e	yes	No	leaf/pigment on cerussite	Au (Cu)
	THT 3520	yes	in Au	flake/pigment	Au (Cu-Hg)
Manichaean	M 660a-b		in Pb-brown line		Pb (As)
	So 10100q	yes	in paper, in Pb-brown line	pigment	Pb (As) Au (Cu-Hg)
Sogdian	So 10237–10239	yes	in paper, in Hg-Pb-red	pigment	Hg-Pb (As) Au (Cu)
	MIK III 4984	no	in Au	flake	Au (Hg-As)
	MIK III 4932		in Pb-brown line	on cerussite	Pb (As-Hg) Au (Cu)

The **Sogdian** manuscript MIK III 4932 with text on one side and an illumination on the other side (Figure 6) shows the painting elaborated with outlines. The brown ink is composed of a lead component together with an arsenic salt and cinnabar (HgS). The yellowish brown coloration of the painting is made with metal gold on white cerussite (PbCO₃).

The ink of the brown outline of the Sogdian fragment So 10100q is prepared with a mixture with contains lead and arsenic. The element arsenic is detected also in other colors and in the paper. However, orpiment is not used as a pigment for yellow coloration on these Sogdian fragments. The paper of the Sogdian manuscript So 10237–10239 reveals the element arsenic. The red color is a mixture of cinnabar with a lead compound containing arsenic. The concentration of arsenic in the paper of Sogdian manuscripts is probably due to conservation treatment. The protection of paper with colorless arsenic compounds is mentioned in Asia from the 5th C on, but it may also

be a more recent treatment, since the technique was applied in collections as late as the 17th C.

The line on the **Manichaean** manuscript M660a-b is identified as having a similar composition consisting of black carbon ink with a lead compound and traces of arsenic.

Arsenic in the form of orpiment is applied on Uighur, Tibetan, Syrian, and less frequently Tocharian manuscripts. It is detected furthermore together with lead on Tocharian, Sogdian, and Manichaean manuscripts. A relation to the kind of **gold** detected on some of the fragments should give hints about possible common roots (Table 1). Primarily used for coloration, gold flecking – spreading particles of the metal – was applied to make the illumination shinier, a method known from Persian manuscripts (Laurie 1935).

There is only one **Uighur** document on textile in which gold was detected by XRF analysis together with orpiment in a painting. The gold contains traces of copper, which may give an indication to its



Figure 10 Photomicrographs with laser spot from different Tocharian manuscript fragments, showing different gold applications. Tocharian THT 1685b: gold leaf on white cerussite; Tocharian THT 3520: goldflake (containing Cu and Hg).

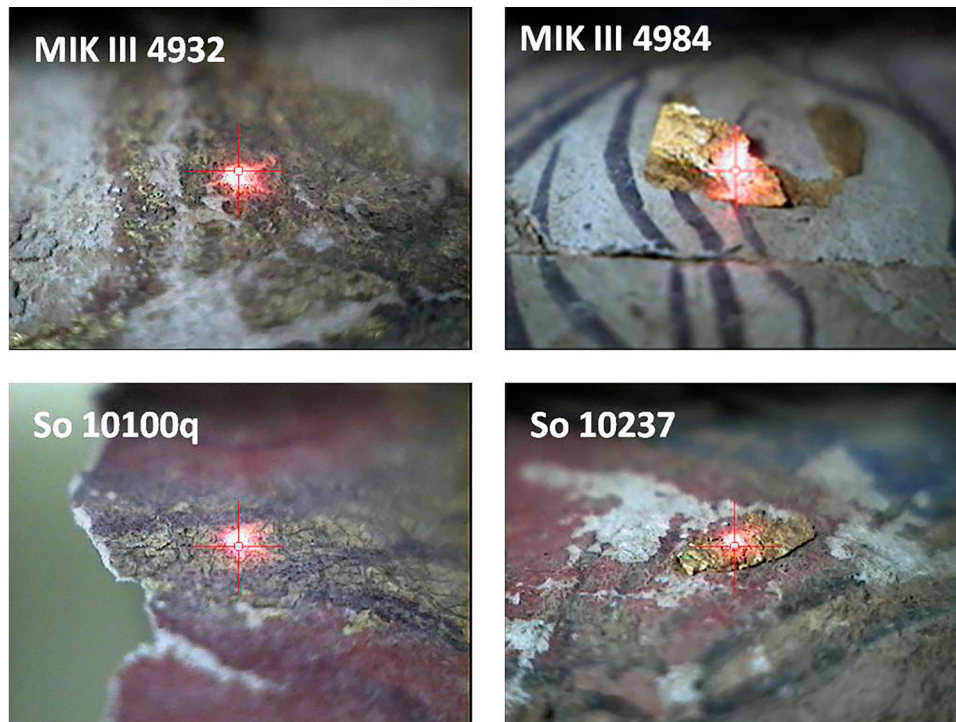


Figure 11 Photomicrographs with laser spot from different manuscript fragments, showing different gold applications. Sogdian MIK III 4932: powder gold (containing Cu) on white cerussite; Sogdian MIK III 4984: gold leaf (containing Hg); Sogdian So 10100q: powder gold (containing Cu and Hg); Sogdian So 10237: gold leaf (containing Cu).

provenance. Copper is endogenous in deposits of gold, for example in Azerbaijan, but is also very often used in the production process of alloying gold.

In contrast to the other illuminations of this compilation THT 1685 a-h (Figure 2), the yellow color of the **Tocharian** fragments THT 1685b + e is identified as gold with traces of copper. Only the paintings elaborated with gold show a white primer made of gypsum or chalk under the text written in black on the other side (Figure 10).

The analysis of the gold of THT 1685b + e showed a high lead content, attributed to white cerussite (Pb) under the yellow color, which is supposedly applied to fix the metal more firmly on the paper's surface. The white lead compound without metals as trace elements is artificially produced, unlike red lead, which is mixed with arsenic. On another Tocharian manuscript, THT 3520, a yellow flake of gold is applied without cerussite. It shows traces of Cu and Hg (Figure 10), thus indicating a different preparation and provenance.

There are some **Sogdian** fragments with gold as powder or leaf applied with binder (Figure 11). On manuscript MIK III 4932, gold is fixed with white cerussite (Pb) as a base coat using the same technique as that detected on the Tocharian fragment THT 1685b + e; here, however, the other side of the paper received no whitish surface treatment (Figure 6). The gold detected on this manuscript is associated with Cu as a trace element, which is also identical to that found in THT 1685b + e and in the unique Uighur

fragment U 7255, where gold was applied together with orpiment on textile. Another Sogdian fragment with gold applied in form of leaf, MIK III 4984, shows the whitish surface treatment with cerussite (Pb) also on the side of the paper written in black (Figure 7). Here, gold is always associated with mercury as trace element. On the Sogdian manuscript So 10100q, gold with Cu-Hg as trace elements is detected for yellow coloration. The shinier gold flake of another Sogdian manuscript, So 10237–10239, shows traces of Cu, similar to the Tocharian and Uighur fragments mentioned before, indicating the same provenance.

In addition to the application of gold with cerussite (Pb) detected as a special technique to fix gold on the surface of the paper on some Sogdian and Tocharian manuscript fragments, traces of Cu or Hg-As thus indicate different origins of the native gold. These characteristics, however, are not specific to only one of the cultural usages (Table 1), which underlines a close relation between the two groups, communication with the exchange of trade goods, or recycling of the precious material.

Conclusion

Based on the few illuminated fragments from the collection of Turfan manuscripts classified as Sogdian and Tocharian, special traditions in the production and provenance of the manuscripts are indicated by details found in the investigation. Even if their whitish

surface treatment combined with the application of gold is part of an exclusive technical development, in the light of analyses of other documents from Silk Road cultures, the pigments and manufacture give hints about the original applications as well as overlapping criteria due to cultural influence. This validates at least that some people communicated trans-regionally and exchanged more than others, as shown by the script, language, content, or reason for the writing.

The whitish surface of the paper, obvious under the black letters, is detected as a layer of gypsum on Tocharian and, in a thinner form, on Sogdian fragments. This treatment is not necessarily combined with a painting on the other side. The colorful illuminations are sometimes elaborated with gold.

The application of precious gold is also known from Tibetan and Uighur manuscripts.

Trace elements associated with gold, such as Cu and Hg on Sogdian and Tocharian, Cu on a Sogdian and Uighur, and Hg on Tibetan fragments, indicate different origins of the raw material. A special technique to fix the gold on paper through the application of cerussite indicates that some of these manuscripts come from the same workshop tradition. It is detected for only a few Sogdian and Tocharian fragments, revealing a closer relationship in their manufacture compared to other manuscripts.

Yellow orpiment (As_2S_3) appears artificially prepared on Tocharian, Uighur, Syrian, and, from local deposits, Tibetan fragments. Orpiment is assumed to be of Western provenance, a region with a long tradition in its application.

Independent of the use of a whitish paper, the Uighur tradition is linked to a Sogdian-Tocharian tradition in the use of Cu-bearing Au and to a Tibetan-Tocharian-Syrian tradition in the use of orpiment. This selection of manuscript fragments provides no hint of a Chinese or Sanskrit tradition in using whitish paper, gold, or orpiment.

Though red inks have often been used and exchanged, analysis of types underlines a strong relation between Tocharian and Chinese manufactures in the use of cinnabar (HgS), as well as Tibetan, Tocharian and Syrian manufactures in the use of red lead (Pb_3O_4). The preference for red lead with traces of arsenic on Tocharian and Sogdian manuscripts is thus seen as a tradition different from the use of red cinnabar. Sogdian and Manichaean seem to have common features in the use of both as mixed inks, indicating various influences.

The black carbon inks analyzed with a copper component are detected mostly on Tocharian, Sanskrit, and Syrian manuscripts. Traces of arsenic in black ink on Manichaean and Uighur fragments, sometimes together with mercury, are seen as characteristic for a certain regional production.

The results point out tendencies in a temporary, limited cultural-regional preference for special inks, pigments, and preparation techniques, valuable for the selected manuscripts. The highlighted types of

manufacture constitute a basis for further culture-specific investigations using material analyses.

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Author biographies

Renate Nöller is research associate in the field of inorganic chemistry/ mineralogy and ethnology. She held a position as scientist at the BAM Federal Institute for Materials Research and Testing, Division Analysis of Artefacts and Cultural Assets, funded by the German Research Foundation (DFG) with projects on investigation of inks and pigments on manuscripts. The thematic priority of her work is the analysis of material in relation to cultural questions with a focus on mineral pigments.

Oliver Hahn received his PhD in Chemistry in 1996. After a stint spent as a research associate in the Department for Restoration and Conservation of Books, Graphic Arts and Archival Materials at the University of Applied Sciences in Cologne, he now works for the BAM Federal Institute for Materials Research and Testing in Berlin. His areas of special interest include the analysis and the preservation of the country's cultural heritage. Hahn is head of the institute's Division 4.5 "Analysis of Artefacts and Cultural Assets". Since 2014 he is Professor at the University of Hamburg, Arts Faculty.

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