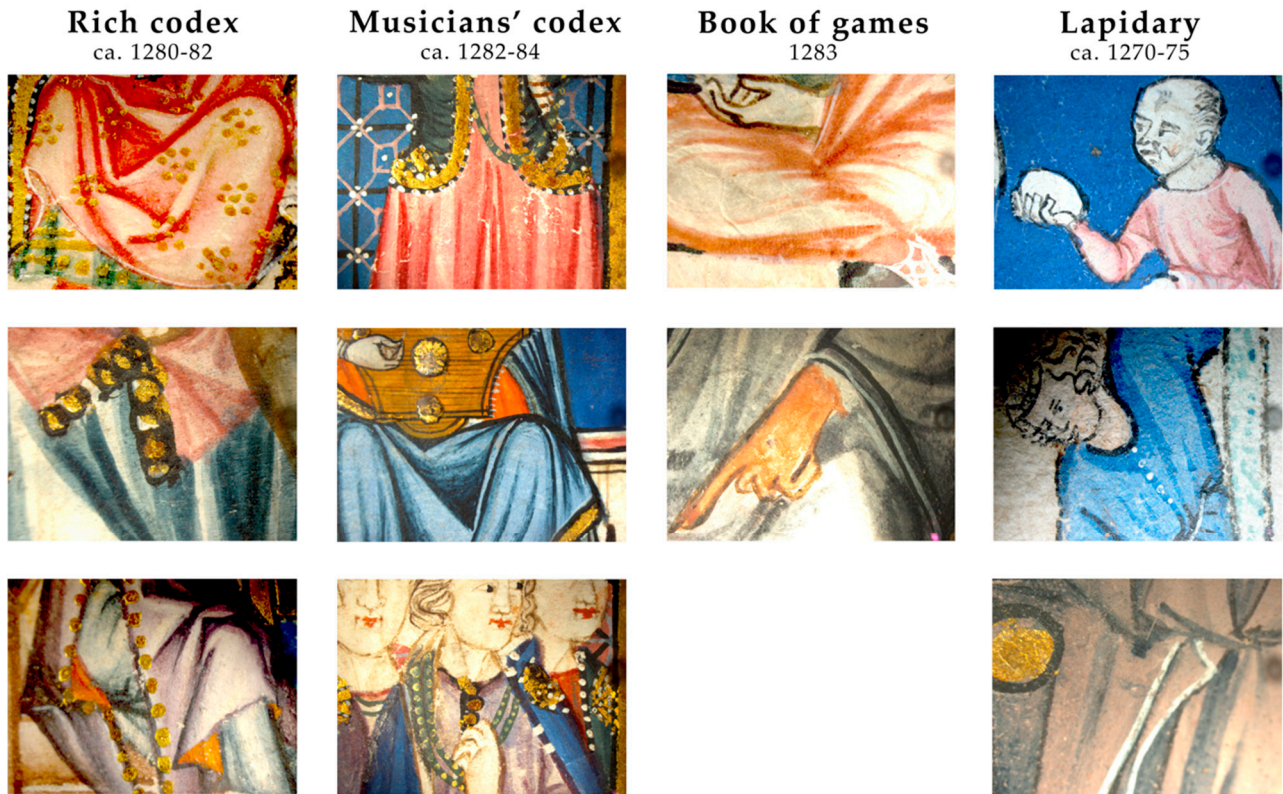


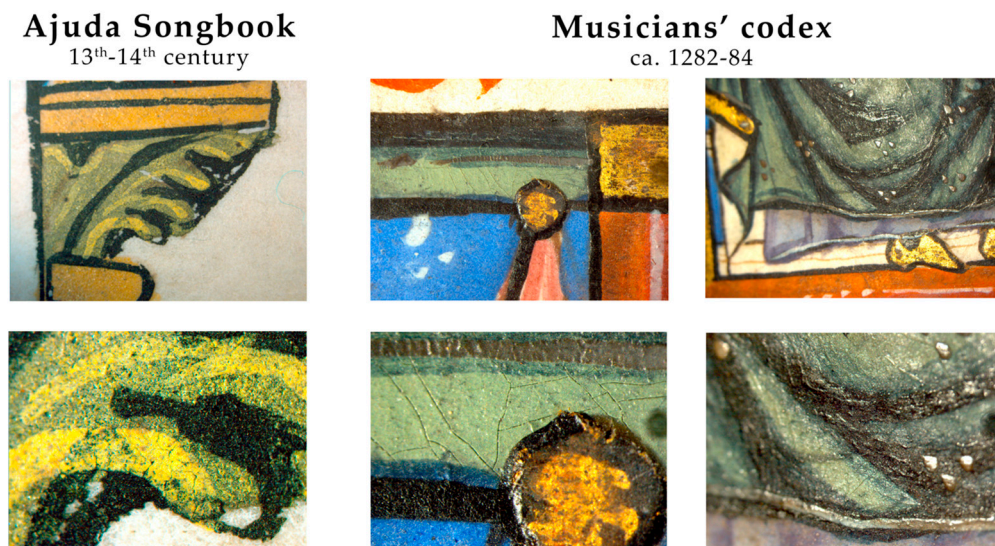
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### Supplementary file S3. Painting technique: singularities and specificities

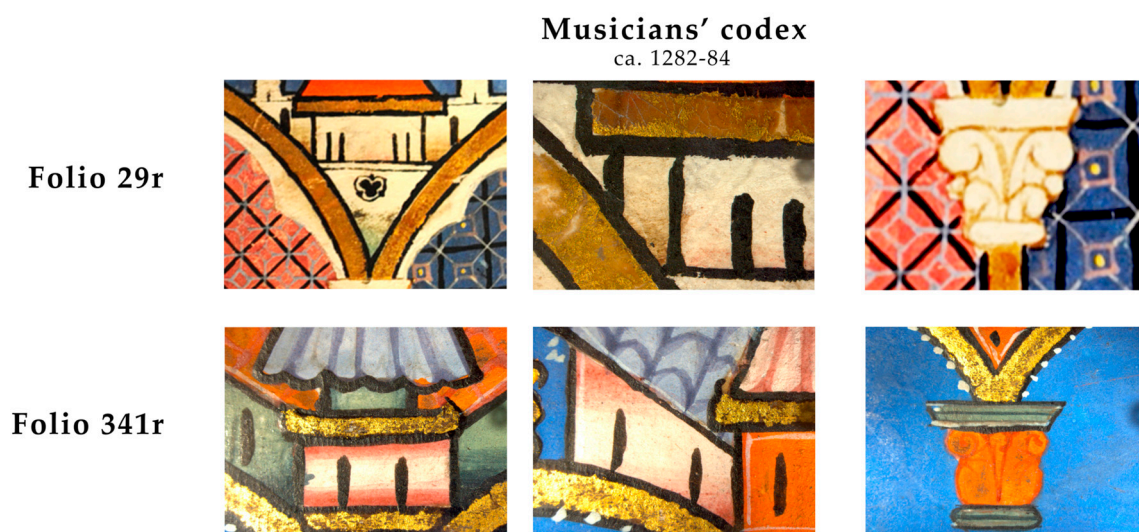
This supplementary material presents the information of each manuscript and details acquired with the Leica microscope, from which conservation condition observations were extracted.



**Figure S12.** Details of pink (top), blue (middle), and purple (bottom) vestments found in Alfonso's manuscripts. The difference in application is clear between the *Rich codex* and the *Musicians' codex* and *Lapidary*. Even though *Book of Games* is also different from the *Rich codex* in terms of style and various aspects of the pictorial technique, it was possible to see some similarities in the construction of colors, like the pinks and blues. In these, the illuminator took advantage of the parchment or white support for the brighter tones.



**Figure S13.** Details of green *vergaut* resulting from a homogeneous mixture found in the frames and vestments of *Musicians' codex*, compared to the color construction of the *vergaut* found in *Ajuda Songbook*, obtained through layering of orpiment above three other colorants.



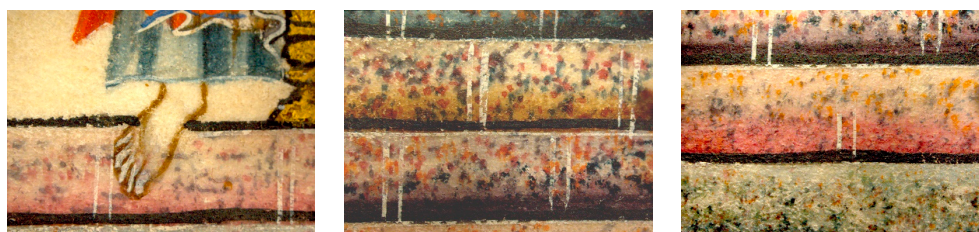
**Figure S14.** Details of the architecture found in folio 29r and 341r of *Musicians' codex* illustrating the difference between the detailed motifs of the first and the coarser application on the second.

### S3.1. Rich codex

Place of origin: Kingdom of Castilla and Leon/ Date: ca. 1280-82/ Script: French goth of the 13<sup>th</sup> century/ Language: Galician-Portuguese/ Dimensions: 326x490 mm / Support: Parchment / Folios: 257

#### Painting technique

Besides the faces and hands presented in the main manuscript, the architecture or surrounding elements can give information on the painting techniques used. For example, the stones that compose the stairs in folio 32v result from a first thin layer of paint, applied in degradé, from the bottom until the top of the stone, **Figure S15**. Above this first layer, small points were then added, in different colors, that seem to be chosen according to the base color of the stone. This color game gives the impression that the characters are stepping on a marble floor, composed of big stones, whose separation is simulated by thin white lines.



**Figure S15.** Details of stairs found in division 6 of the illumination in folio 32v of the *Rich codex*.

Contrary to the presented stairs, the frames were made with geometrical elements painted with uniform pure colors, **Figure S16a**. These elements are of special importance in the illumination, as they were used to separate the different actions of the *Cantiga* represented. It is of notice that, within the wide range of colors in the frames, one yellow of

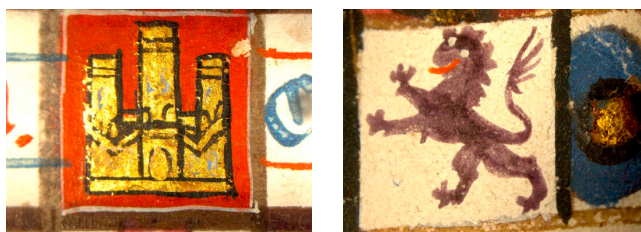


organic origin was also found, whose identification was not yet possible. In the interception of the columns that constitute the frames, it is always possible to find either a golden castle in a red vermillion background or a purple lion in a lead white background, **Figure S16b**.

a



b



**Figure S16.** Details of a) colors found in the frames of folio 212r and b) golden castle and purple lion found in the interception of frames of folio 192r.

**Conservation condition:** The manuscript is in a good state of preservation. Very few degradation problems can be detected, and the ones observed are mainly due to physical degradation such as abrasion. The gold leaf, in some areas, has disappeared probably due to abrasion. Only fragments of the lead and the brown layer of preparation have remained. It is common to find paints applied above the gold leaf, that in the case described have also detached from the support.

In the case of the paints, in some areas, there is crackling, probably provoked by the folding of the folio and the thickness of the paint, and in other areas the paints have detached completely leaving small gaps. The lead white and red lead applied in the details, and as a consequence in thicker layers, suffer from this problem the most. The bottle-green presents small cracks and gaps resulting from the detachment of the paint. In some points of this paint, small opaque dots can be observed.

Lead-based pigments do not show any darkening in the analyzed folios, which might be related to the minimal presence of orpiment, a pigment responsible for the formation of dark lead oxides.

### S3.2. Musicians' codex

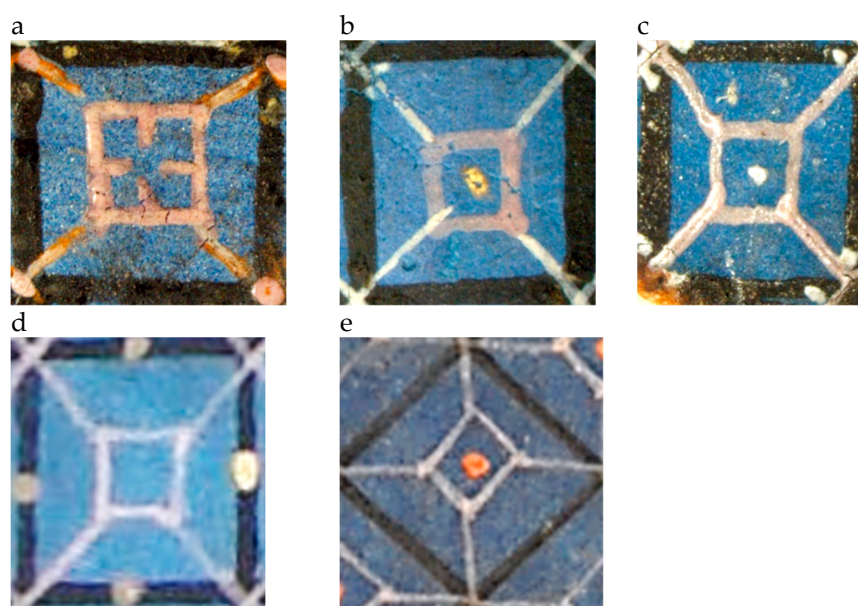
Place of origin: Kingdom of Castilla and Leon/ Date: ca. 1282-84/ Script: French goth of the 13<sup>th</sup> century/ Language: Galician-Portuguese/ Dimensions: 274x404 mm / Support: Parchment / Folios: 361

#### Painting technique

In Musicians' codex, one aspect of the illuminations stands out in terms of the painting technique used: the backgrounds. As an example, only the blue backgrounds will be discussed. In some of the folios analyzed, like folio 29r, 71v, 140v, 176v, and 305v, the background was painted in an elaborate pattern. Above the blue or pink base color, little squares with inside geometrical appointments were drawn, with at least 3 or 4 different colors. However, in folio 89r and 341r the background was painted in pure lapis lazuli, without any pattern. This might point to one hypothesis: as seen for the faces, the illuminators responsible were different and followed different painting techniques and rules.

Within the patterns applied to the blue backgrounds, it is possible to see that the squares are contoured with black paint and the interior has details in more bright colors. Just on folio 29r, which represents the king surrounded by his court, three different patterns were found for the blue background. In this folio, for the first pattern, the inside seems to have been drawn first with an orange paint and in this same paint, a pink color was applied above, **Figure S17a**. In the second pattern, the inside square was drawn in pink and the lines that connect it to the black exterior square are white. In the middle, a small yellow point was painted, **Figure S17b**. In the last pattern, the inside square and connecting lines were both drawn in pink. The middle point and additional details at the extremes of the lines were painted in white, **Figure S17c**. For the pattern found in folio 140v, another way of applying was used, where a simple pink square and lines were drawn, **Figure S17d**. In addition, golden details were added to the black contour. The application of a different color in the middle of the square, like the second pattern in folio 29r, was again used in folio 305v, **Figure S17e**. The square and lines were applied using a bright pink and the middle was done using orange.

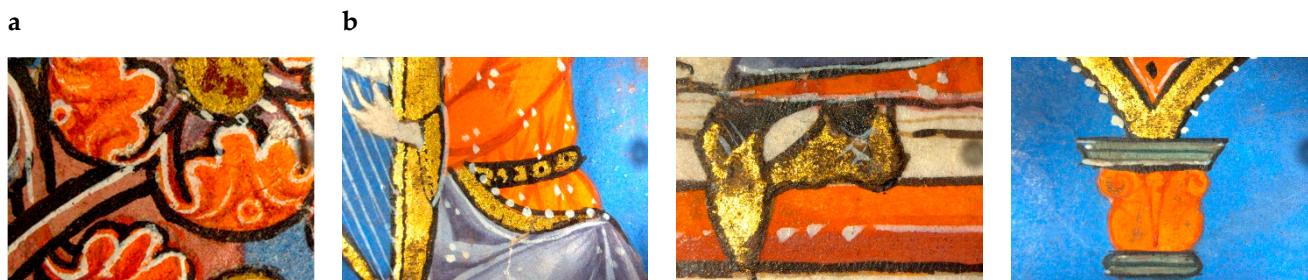
The faces and hands, being good indicators of the work of different illuminators, do not show any difference in these folios. This might indicate that the patterns do not result from different hands but were made on purpose to be different and provide variety to the background.



**Figure S17.** Details of patterns found in the blue backgrounds of folio 29r in a, b, and c, in folio 140v in d, and folio 305v.

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Another color that shows the delicate and professional work of the illuminators is orange. This orange was applied to the vestments, architecture, and initials. What makes it stand out, however, is the application of what seems to be an unidentified organic red to control the shades, **Figure S18a** and **b**. The same was verified for the Lapidary. The shades acquired with an organic red are very similar to what is found in Portuguese manuscripts of monastic production, where lac dye was applied over the vermilion or red lead.



**Figure S18.** Details of orange applied in the (a) initial of folio 29r and in the (b) the vestments and architecture of folio 341r. Red paint is used to control the shades and contour the shapes.

**Conservation condition:** The manuscript is in a good state of preservation and as observed for Rich codex it also presents two degradation problems due to physical action: the erosion and loss of gold leaf and paint layers. The folding of the folios encourages further the increase of these problems. Besides these problems, in some areas, the paints were dragged and scattered due to physical interaction, and in small details, they seem to have been dragged still wet or after entering in contact with water.

The lead-based pigments like lead white and red lead present darkening when applied near orpiment or *vergaut* (mixture of orpiment and blue).



### S3.3. Lapidary

Place of origin: Kingdom of Castilla and Leon/ Date: ca. 1270-75/ Script: French goth of the 13<sup>th</sup> century/ Language: Castilian/ Dimensions: 291x402 mm / Support: Parchment / Folios: 119

#### Painting technique

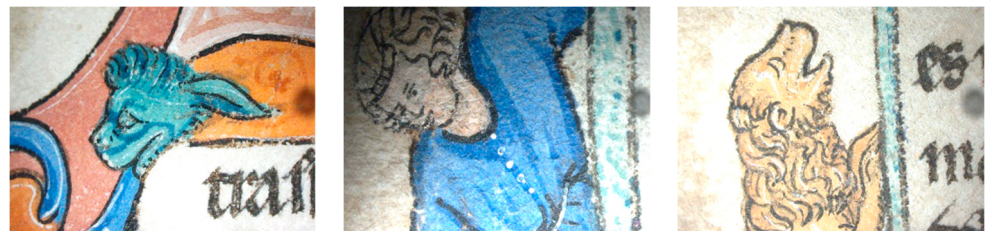
Lapidary is the manuscript that presents the highest variety in the making of all colors, with different mixtures and ways of paint application. Just as observed for the Musicians' codex, the oranges presented the application of varnish-like colors, not as red as the previous, that gives texture to the elements.

The bottle-green, contrary to what is observed for the other manuscripts, is applied in thin layers but presents the same level of degradation. The serpents are an example of the mastery of this application, which also presents the application of a diluted red to paint the belly, giving the sensation of movement, **Figure S19**.



**Figure S19.** Illumination with the bottle green serpent of folio 8r, where is visible the control over the thickness of the green and the application of the light pink for the movement.

Another interesting aspect of this manuscript that was not found in the others is the presence of drolleries. In folio 8r, there is a scene starring a lion reaching for a man that crabs into the tail of a dragon, trying not to be eaten, **Figure S20**.



**Figure S20.** Details of drollery found in the left margin of folio 8r, with a scene composed of a dragon, a man, and a lion.

As this manuscript is dedicated in part to the description of stones, these are thoroughly illustrated in the illuminations, with a high level of mastery. They were painted

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with texture and volume and give the illusion of the typical shine of a stone, either painted in violet or in blue, **Figure S21**.



**Figure S21.** Details of violet and blue stones found in the historiated initials of folio 87r.

**Conservation condition:** The overall state of the preservation of the manuscript is good, however, folio 8r presented the paints in the most fragile state of conservation compared to the other folios. Two degradation problems due to physical action are visible, similar to the previous manuscripts: the erosion and loss of gold leaf and paint layers. The loss of the gold leaf in folio 49r uncovered a green preparation layer similar to bottle-green.

As expected, the bottle-green paints show advanced crackling and consequent detachment from the parchment. In isolated cases, the paints form already opaque areas.

### S3.4. Libro del Ajedrez, Dados y Tablas

Place of origin: Kingdom of Castilla and Leon/ Date: ca. 1283/ Script: French goth of the 13<sup>th</sup> century/ Language: Castilian/ Dimensions: 280x400 mm / Support: Parchment / Folios: 97

#### Painting technique

This manuscript is particularly interesting for the everyday objects represented, in **Figure S22**. For example, in folio 18r, where three maids are seen playing a board game, simple details were painted in great detail like the shoes, the painted nails and tattooed hands, the necklaces and earrings, the half-drunk bottle of wine and glass (made with red color and silver that is now oxidated).



**Figure S22.** Details of everyday objects, accessories, and manners, represented in folio 18r.

**Conservation condition:** This manuscript presents degradation at a level not found in the previous ones, as seen in **Figure S23**. In some folios, it is possible to verify the degradation of red lead, and the loss of adhesion of some of the pigments like lapis lazuli, lead white, and bottle-green, and in folio 48r two of the figures were erased possibly by water.

Folio 1r



Folio 1r



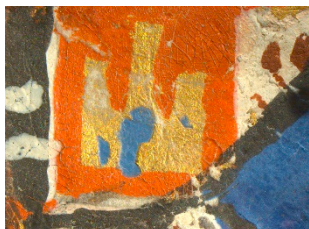
Folio 18r



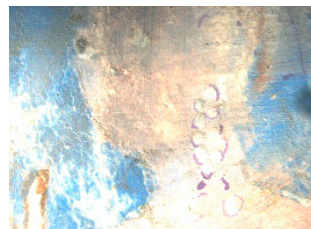
Folio 48r



Folio 65r



Folio 48r



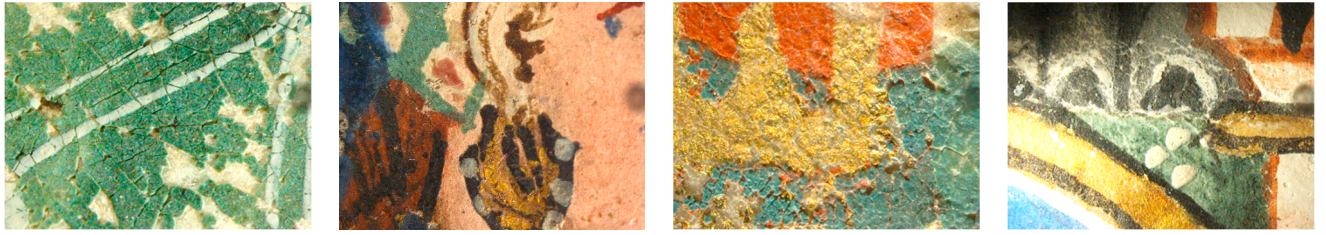
Folio 48r



**Figure S23.** Details of colors in an advanced state of degradation: loss of adhesion of the green and layers of preparation, degradation of red lead, and possible damage by water.

For the greens applied in the architecture and in the vestments the bottle-green used presents several gaps resulting from the loss of adhesion, however, the color remains intense, **Figure S24**. In some cases, the green is opaque, as seen for the Portuguese monastic bottle-green in an advanced state of degradation.





**Figure S24.** Details of greens applied in the architecture, vestments accessories of folios 1r, 18r, 48r, and 65r.