Supplementary Information for

Improved Spectral Imaging Microscopy for Cultural Heritage through Oblique Illumination

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Figure S1. Photograph of the OI spectral imaging microscope, with red arrows indicating the two possible rotation angles for the illumination arm and the sample stage (α and β, respectively).
Figure S2. A 1951 US Air Force Target imaged to show data processing steps for a single wavelength of 545 nm. Images acquired at a single illumination angle are first registered and then the A) average and B) minimum composite images are calculated. These composite images are compared to darkfield and brightfield images captured on a standard commercial microscope. The average generates an image similar to the commercial dark field microscope which captures interreflections at the edges of chrome features whereas the composite minimum produces the inverse of the brightfield image.
Table S1. A summary of experiments conducted on the cross-section taken from *La Miséreuse accroupie*. Pigment suggestions were made based on elemental information from SEM-EDS analysis, the identification of the listed fingerprint peaks in Raman Spectroscopy and spectral matches with reference mixtures for microscopic spectral imaging.

| Layer     | SEM-EDS                              | Raman Spectroscopy (Raman Shift, cm\(^{-1}\)) | OI Microscopic Spectral Imaging          |
|-----------|--------------------------------------|---------------------------------------------|-----------------------------------------|
| 4 (Blue)  | Lead White (Pb) Prussian Blue (Fe)   | Prussian Blue (2153 cm\(^{-1}\)) Ultramarine (550 cm\(^{-1}\)) | Prussian Blue Ultramarine              |
|           | Possible Ultramarine? (Si, Al)       |                                             |                                         |
| 3 (Red)   | Lead White (Pb) Vermilion (Hg)       | Vermilion (255 cm\(^{-1}\),344 cm\(^{-1}\)) | Vermilion Cadmium Yellow (particle)     |
|           | Calcium Sulfate (Ca,S) Cadmium particle |                                             |                                         |
| 2 (Purple)| Lead White (Pb) Ultramarine (Al, Si, Na) | Ultramarine (550 cm\(^{-1}\)) | Ultramarine Cochineal Lake             |
|           | Calcium based filler Layer is Aluminum rich, possible organic red lake? |                                             |                                         |
Table S2. Pigment content for each layer of the reference cross section.

| Layer | Pigments                                    | Weight % |
|-------|---------------------------------------------|----------|
| 4     | Cadmium Yellow (CdS)                        | 75       |
|       | Vermilion (HgS)                             | 25       |
| 3     | Prussian Blue \((\text{Fe}_4[\text{Fe(CN)}_6]_3)\) | 25       |
|       | Zinc White (ZnO)                            | 75       |
| 2     | Ultramarine \((\text{Na}_{6-10}\text{Al}_6\text{Si}_6\text{O}_{24}\text{S}_2-4)\) | 50       |
|       | Madder Lake                                 | 50       |
| 1     | Ultramarine \((\text{Na}_{6-10}\text{Al}_6\text{Si}_6\text{O}_{24}\text{S}_2-4)\) | 25       |
|       | Zinc White (ZnO)                            | 75       |

Movie S1 (separate file). This movie animates ten aligned and reconstructed RGB images captured at progressive azimuthal angles of illumination (every 36°) of the cross-sectional sample removed from Picasso’s *La Miséreuse accroupie* (1902). The effects of different angles of oblique illumination on the observed reflectance is evident, particularly from microfacets.