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## Characterization of the Chemical Nature of the Black Ink in the Manuscript of *The Gospel of Jesus's Wife* through Micro-Raman Spectroscopy

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second mu of ΜΑΡΙΑΜ in line →3 (which would surely indicate a modern forgery). Further conservation of the papyrus is required to confirm this beyond doubt. It is, however, clear that some issues that have been brought forward as evidence of forgery are apparent only on the digital images that were originally made available: this primarily concerns cases in which holes in the papyrus appear to be ink in the image<sup>12</sup> and where pooling of ink is not apparent on the papyrus itself.<sup>13</sup> The text in line →7 is also clearly under the “blob” of foreign matter (some type of wax?).<sup>14</sup> The “oblique stroke” before ΠΕΧΕ in →4 is more likely the remains of a letter than a mark of punctuation.<sup>15</sup> One can also note that the lack of ink on the left two-thirds of the “back” is clearly caused by the loss of most of the upper layer of fibers at this point and that, while the top edge of the papyrus does give the appearance of having been cut, not broken,<sup>16</sup> such a clean straight break is not unknown in genuine papyri.<sup>17</sup>

Overall, if the general appearance of the papyrus prompts some suspicion, it is difficult to falsify by a strictly paleographical examination. This should not be taken as proof that the papyrus is genuine, simply that its handwriting and the manner in which it has been written do not provide definitive grounds for proving otherwise.

## ■ Characterization of the Chemical Nature of the Black Ink in the Manuscript of *The Gospel of Jesus's Wife* through Micro-Raman Spectroscopy

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### *Brief Summary*

Date of study: March 11–12, 2013

A research team at Columbia University consisting of Professor James T. Yardley of the Department of Electrical Engineering and Alexis Hagadorn, Head of Conservation, Columbia University Libraries, with the support of Dr. David

<sup>12</sup> See, e.g., the first alpha of αρια in line →3, where there is a tiny hole in the papyrus at the bottom right of the alpha.

<sup>13</sup> E.g., to the bottom right of the first alpha of αρια in →3 and in the diagonal stroke before ΠΕΧΕ (→4), where I can see neither the hole that was noted in the draft edition, nor ink pooling around it.

<sup>14</sup> See especially the first ε of εΤΡΕ.

<sup>15</sup> Note that the scribe varies letter forms elsewhere (compare the *upsila* in → 4 and 5 [ηάγ], ξώω] with that in →6 [ε·οοογ]), and something like ]·μηοογ or ]·μηάγ ] might be considered.

<sup>16</sup> On the “front,” there appear to be no remains of a line above line →1: a trace of ink on a partially detached fiber above the second alpha of ΗΑΔΥ probably comes from the alpha itself.

<sup>17</sup> Such cuts are commonly made in modern times (e.g. to cut up a text to sell to different buyers), but similar breaks also occur in papyri discovered in archaeological context.

Ratzan, Curator of the Papyri Collection, has utilized micro-Raman spectroscopy to investigate the chemical composition of pigments for selected regions on both sides of the manuscript fragment known as the *Gospel of Jesus's Wife* (*GJW*) and also for an additional fragment from the Gospel of John. These manuscripts were provided for the purposes of this study through Professor Karen King of Harvard University. Most dyes or pigments exhibit characteristic Raman spectra.<sup>18</sup> Micro-Raman spectroscopy constitutes a non-destructive technique for characterizing the chemical composition of inks and pigments.<sup>19</sup> In the Raman scattering process molecules within the exciting laser beam emit light with photon energy reduced (and therefore wavelength increased) by the amount of a characteristic vibrational motion of the molecule. Therefore the measurement of scattered light intensity as a function of wavelength provides direct information about the inks under investigation through the display of characteristic vibrational resonances or peaks. Black ink pigments generally fall into the categories of carbon black (many variations),<sup>20</sup> iron gall,<sup>21</sup> hematite,<sup>22</sup> magnetite,<sup>23</sup> and various iron oxides.<sup>24</sup> Modern black inks are formulated using complex dye molecules that typically

<sup>18</sup> Robin J. H. Clark, "Pigment Identification on Medieval Manuscripts by Raman Microscopy," *Journal of Molecular Structure* 347 (1995) 417–27; idem, "Pigment Identification by Spectroscopic Means: An Arts/Science Interface," *Comptes Rendus Chimie* 5 (2002) 7–20; Gregory D. Smith and Robin J. H. Clark, "Raman Microscopy in Archaeological Science," *Journal of Archaeological Science* 31 (2004) 1137–60; Lucia Burgio, Robin J. H. Clark, and Richard R. Hark, "Raman Microscopy and X-ray Fluorescence Analysis of Pigments on Medieval and Renaissance Italian Manuscript Cuttings," *Proceedings of the National Academy of Sciences of the United States of America* 107 (2010) 5726–31.

<sup>19</sup> Ian M. Bell, Robin J. H. Clark, and Peter J. Gibbs, "Raman Spectroscopic Library of Natural and Synthetic Pigments (pre- Approximately 1850 AD)," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 53 (1997) 2159–79; Lucia Burgio and Robin J. H. Clark, "Library of FT-Raman Spectra of Pigments, Minerals, Pigment Media and Varnishes, and Supplement to Existing Library of Raman Spectra of Pigments with Visible Excitation," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 57 (2001) 1491–521.

<sup>20</sup> Eugenia P. Tomasini et al., "Micro-Raman Spectroscopy of Carbon-Based Black Pigments," *Journal of Raman Spectroscopy* 43 (2012) 1671–75.

<sup>21</sup> Alana S. Lee, Peter J. Mahon, and Dudley C. Creagh, "Raman Analysis of Iron Gall Inks on Parchment," *Vibrational Spectroscopy* 41 (2006) 170–75; Alana S. Lee, Vincent Otieno-Alego, and Dudley C. Creagh, "Identification of Iron-Gall Inks with Near-Infrared Raman Microspectroscopy," *Journal of Raman Spectroscopy* 39 (2008) 1079–84; Marina Bicchieri et al., "All That is Iron-Ink is Not Always Iron-Gall!," *Journal of Raman Spectroscopy* 39 (2008) 1074–78; Marina Bicchieri et al., "Non-Destructive Spectroscopic Investigation on Historic Yemenite Scriptorial Fragments: Evidence of Different Degradation and Recipes for Iron Tannic Inks," *Analytical and Bioanalytical Chemistry* 405 (2013) 2713–21.

<sup>22</sup> A. Rosalie David et al., "Raman Spectroscopic Analysis of Ancient Egyptian Pigments," *Archaeometry* 43 (2001) 461–73.

<sup>23</sup> Lubomir Slavov et al., "Raman Spectroscopy Investigation of Magnetite Nanoparticles in Ferrofluids," *Journal of Magnetism and Magnetic Materials* 322 (2010) 1904–11.

<sup>24</sup> Dimitris Bikaris et al., "Ochre-Differentiation through Micro-Raman and Micro-FTIR Spectroscopies: Application on Wall Paintings at Meteora and Mount Athos, Greece," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 56 (2000) 3–18.

exhibit characteristic sharp spectral features.<sup>25</sup> For black pigments based on forms of carbon black, the detailed spectral characteristics are somewhat dependent on specific pigment preparation as well as excitation wavelength.

For these manuscripts, we obtained micro-Raman spectra using a conventional commercial instrument (Renishaw inVia). We used 633 nm laser excitation (10 mWatt maximum power) using 0.5%–10% of the laser power focused onto the sample through a conventional  $\times 100$  microscope objective. We collected approximately 140 Raman spectra from selected regions of the two sides of the two manuscripts, examining in detail the Raman shift range from  $150\text{ cm}^{-1}$  to  $1900\text{ cm}^{-1}$ . In a parallel study we have examined Raman spectra from over fifteen papyrus manuscripts from the Columbia collection covering the time period from 500 B.C.E. to +1000 C.E.<sup>26</sup> The conclusions of this study for the *GJW* manuscript are as follows:

- 1) The inks used in this manuscript are primarily based on carbon black pigments such as “lamp black.” The observed Raman spectra are very similar to those of the carbon-based inks studied for a wide variety of manuscripts including many dating from the early centuries of the Christian era.
- 2) From the observed Raman spectra, we find no evidence for any constituents of ink or types of ink other than carbon black in the selected regions.
- 3) The ink or inks used in *GJW* are similar to, but distinct from, the ink used for the Gospel of John manuscript.
- 4) Within the available accuracy of our measurements, our data are consistent with a single ink composition for each individual side of the *GJW* manuscript.
- 5) The Raman spectra obtained from the “recto” side and from the “verso” side are indistinguishable within our experimental error.

<sup>25</sup> Laurence C. Abbott et al. “Resonance Raman and UV-Visible Spectroscopy of Black Dyes on Textiles,” *Forensic Science International* 202 (2010) 54–63; Irina Geiman et al. “Application of Raman Spectroscopy and Surface-Enhanced Raman Scattering to the Analysis of Synthetic Dyes Found in Ballpoint Pen Inks,” *Journal of Forensic Sciences* 54 (2009) 947–52; Racheal E. Littleford et al., “Surface-Enhanced Resonance Raman Scattering of Black Inkjet Dyes in Solution and *in Situ* Printed onto Paper,” *Applied Spectroscopy* 57 (2003) 977–83; James D. Womack, Thomas J. Vickers, and Charles K. Mann, “Determination of Azo Dyes by Resonance-Enhanced Raman Spectroscopy,” *Applied Spectroscopy* 41 (1987) 117–19.

<sup>26</sup> We thank Michael Ryan, head of the Columbia Rare Book and Manuscript Library, for making these papyri available.