Supporting Information

Synthesis, Characterization, and Stability Studies of
Ge-Based Perovskites of Controllable Mixed-Cation Composition,
Produced with an Ambient Surfactant-Free Approach

Shiyu Yue¹, Scott C. McGuire¹, Hanfei Yan², Yong S. Chu², Mircea Cotlet³, Xiao Tong³, and
Stanislaus S. Wong¹,*

¹Department of Chemistry, State University of New York at Stony Brook,
Stony Brook, NY 11794-3400

²National Synchrotron Light Source II,
Brookhaven National Laboratory, Upton, NY 11973

³Center for Functional Nanomaterials,
Brookhaven National Laboratory, Building 735, Upton, NY 11973
I. Introduction to Pb-free containing Perovskites:

Whereas perovskites often take on different chemical compositions, we are focused herein on structures associated with an ABX$_3$ structure, incorporating species such as a larger cation A, a smaller cation B, and an anion X. Traditional perovskites, such as but not limited to CaTiO$_3$, give rise to an orthorhombic crystal structure, possessing a *Pnma* space group. By contrast, common lead halide perovskites characterized by a formula of MAPbI$_3$ maintain either a tetragonal *I4cm* space group or a cubic phase *P4mm* space group.$^1$ These organometallic halide perovskites have shown very promising performance for energy conversion applications with measured power conversion efficiencies (PCE) routinely surpassing 20%. Several factors appear to play a role in this behavior, with one of the most important ones ascribed to the capability of tuning bandgaps$^2$ through externally controlled variables such as chemical composition.

As one strategy, the presence of Rb as a substitutional cationic dopant has been used to tailor the band gap energy to 1.73 eV with the resulting overall perovskite/silicon tandem cell configuration, achieving a power conversion efficiency of 26.4%.$^3$ As another complementary approach, that has been successfully pursued, the creation of novel ‘combinatorial’ composite materials, such as (FAPbI$_3$)$_{0.85}$(MAPbBr$_3$)$_{15}$, has been reported, based on theoretical calculations,$^4$ demonstrating that rational and systematic variations to either the A$^+$ or X$^-$ sites can yield practical consequences with respect to the magnitude of the resulting bandgap.

At present, many more synthesis protocols have been developed to fabricate lead-based perovskites. The majority of these procedures can be categorized generally as being based upon (i) ‘hot injection’, (ii) the ligand-assisted re-precipitation (LARP) method, and (iii) ultrasonication, respectively, though other types of efforts using less popular techniques also exist. Specifically, 4 to 15 nm-sized CsPbX$_3$ nanocubes have been reported using the ‘hot injection’ method in which Cs salt
and PbX₂ precursors were reacted at 140-200°C within an organic solvent.\(^5\) Similarly, nanowire motifs of CsPbBr\(_3\)\(^6\) and 2D-nanoplates of CsPbX\(_3\)\(^7\) could be produced using this technique. Apart from inorganic lead halide perovskites, analogous organic perovskites, such as FAPbBr\(_3\) nanocrystals with an average size range of 5 to 12 nm, have also been fabricated in this manner.\(^8\) Nevertheless, whereas ‘hot injection’ can give rise to a relatively homogeneous overall morphology, this protocol requires a fair amount of experimental skill to ensure reproducibility, and often requires the use of either high temperatures or ‘less-than-ideal’, problematically hazardous precursors.

Ligand-assisted re-precipitation denotes an equally promising route and moreover, it is arguably simpler and can be performed at lower temperatures. For example, nanosized MAPbBr\(_3\) can be synthesized via the LARP method at 80°C, and the resulting quantum yield of these materials was rather high.\(^9\) A ‘room temperature’ variant of this synthesis method was later applied with respect to MAPbX\(_3\) using MAX and PbX\(_2\) as the relevant precursors.\(^10\) Additional efforts have led to nanoscale formulations of both CsPbX\(_3\)\(^11\) and FAPbX\(_3\).\(^12\) The ultrasonication procedure is fundamentally based on LARP coupled with an additional ultrasonication step to accelerate precursor dissolution and further affect the growth rate.\(^13\) In this manner, perovskites incorporating MA\(^+\), FA\(^+\),\(^13\) Cs\(^+\),\(^14\) and different X\(^-\) species have been synthesized as nanocubes, nanowires, nanocrystals, and so forth. Moreover, whereas solvothermal\(^15\) and ball milling\(^16\) techniques have all been reported to form either CsPbX\(_3\) or FAPbBr\(_3\), LARP is particularly attractive for combining product size and morphological homogeneity with ease of fabrication.

Though lead perovskites evince excellent performance as light sensitizers, Pb itself is highly toxic, as it can be absorbed into the human body by either respiratory, dermal, gastrointestinal routes with transfer to the major organs through blood,\(^17\) thereby resulting in the possibility of severe damage to a number of important bodily organs and functional systems.\(^18\) Moreover, lead
perovskites are extremely unstable in the presence of air and moisture. Hence, the major challenge has been to seek viable and practical alternatives to lead without necessarily compromising upon the intrinsically high efficiency and favorable photophysical properties of this class of materials. After all, lead perovskites possess a large absorption coefficient ($\sim 10^3 - 10^6$ cm$^{-1}$), can be readily processed in solution, incorporate cheap and abundant starting materials, maintain advantageous direct bandgaps ($\sim 1.50$ eV), and are characterized by large diffusion lengths and high carrier mobilities. A strategy of chemical substitution for Pb would entail retaining as much of these attributes as possible, while mitigating for environmental toxicity issues. In theory, Pb within the perovskite structure could be replaced by many different transition metals, including but not limited to Ag, Ba, Sr, Ca, Mg, Zn, Ge, Cd, Sn, Be, Co, Cr, Cu, Fe, Ir, Mn, Mo, Ni, Os, Pd, Pt, Rh, Ru, Ti, V, and W.$^{19}$ Furthermore, some potential candidates have been proposed, wherein Pb is replaced with either one or two atoms, as seen in materials such as but not limited to Cs$_3$Sb$_2$I$_9$, $^{20-21}$ MABi$_2$I$_9$, $^{22}$ MA$_2$CuCl$_x$Br$_{4-x}$, $^{23}$ CsSnI$_3$, $^{24}$ La$_2$NiMnO$_6$, and La$_2$CoMnO$_6$. $^{25}$ However, many of these material alternatives often possess either poor stability or large bandgaps, thereby limiting their applications as solar absorbing materials.

Tin is located right above Pb within group 14, thereby rendering it as a possible replacement. Whereas MASnI$_3$ perovskites with lower bandgaps have indeed been reported, in general, the resulting perovskite-based cells delivered lower efficiencies and poorer performance metrics, $^{26}$ in part because Sn$^{2+}$ is relatively unstable. For example, measured power conversion efficiencies reduced by as much as 40% after 2 hours of operation under ambient conditions, $^{27}$ and these numbers were only slightly improved with the generation of mixed tin-lead hybrid perovskites, such as FA$_{0.75}$Cs$_{0.25}$Sn$_{0.5}$Pb$_{0.5}$I$_3$. $^{28-29}$ Furthermore, whereas Sn itself is relatively benign, tin iodide perovskites are potentially more harmful, $^{17}$ because their degradation process may introduce not only
possibly tetratogenic SnI\textsubscript{2} but also undesirable hydroiodic acid into the environment.\textsuperscript{18} A more promising candidate is Ge, due to its direct band gap of 1.88 eV which is comparable in magnitude with the 1.48 eV value reported for Pb-based perovskites. Moreover, as compared with elements such as Sn, Cd, and Be which possess similarly narrow bandgaps, Ge is less toxic, more earth abundant, and inherently more stable in its 2\textsuperscript{+} oxidation state. Therefore, in light of these stability and tolerance considerations, our approach in this study herein has been to substitute Pb with Ge.
II. Characterization Protocols:

**TEM imaging:** TEM characterization was employed to probe the morphology of as-prepared Ge-based perovskite samples. Specifically, aliquots of different samples were dispersed in hexane and subsequently sonicated. The solution was then drop cast onto a lacey carbon-coated copper grid. Afterwards, the sample was analyzed using a JEOL JEM-1400 transmission electron microscope, equipped with a 2048 × 2048 Gatan CCD camera, operating with an incident beam energy of 120 keV. Higher-resolution TEM images were collected with the help of a JEOL 2100F transmission electron microscope, incorporating a field-emission electron gun and equipped with a Gatan Tridiem energy imaging filter. Images were collected at energy levels of 200 keV.

**SEM imaging:** SEM has also been utilized to ascertain and confirm the morphology of as-prepared samples. Specifically, samples were prepared in a similar manner to that of the TEM, in a process involving drop casting a sample solution onto a silicon wafer substrate. In effect, the structure, motif, and size of these samples were probed using a Hitachi S-4800 Scanning Electron Microscope instrument, set at a 10 kV accelerating voltage with a ~10 µA emission current.

**X-ray Diffraction:** XRD patterns were taken so as to provide insight into the chemical composition and crystallinity of the as-prepared samples. A zero-background holder (i.e., MTI Corporation, Zero diffraction plate for XRD, B-doped, p-type Si, measuring 23.6 mm in diameter by 2 mm in thickness) served as the substrate onto which a hexane solution containing the sample was applied. Diffraction patterns were collected using a Scintag diffractometer, operating in the Bragg-Brentano configuration with incident Cu Kα irradiation (λ = 1.54 Å). To ensure consistency, these diffraction patterns were all collected in a range of 10º to 70º in the Bragg configuration, operating with a step size of 0.25º in 2θ degree intervals and a scanning speed of 10º per min. In this way, the relevant properties of Ge-containing perovskites possessing different A site cations were
individually analyzed. Moreover, the compositional and structural stabilities of both MAGeI₃ and FAGeI₃ as a function of time were also characterized in this way.

**XPS measurements:** X-ray photoelectron spectroscopy enables data collection on the elemental composition (and specifically the oxidation state) of the surfaces of samples. Moreover, we can also gain insights into the effects of X-ray irradiation by monitoring possible sample decomposition, based upon an investigation of as-collected spectra. Samples are prepared by drop casting aliquots of an analyte-containing solution onto a 1 cm × 1 cm silicon wafer. These were later placed into a vacuum chamber, adjusted to an ultrahigh vacuum (UHV) pressure of less than 5×10⁻⁹ Torr. XPS measurements were carried out using a home-built surface analysis system, equipped with a hemi-spherical electron energy analyzer (SPECS, XR50). Al Kα (1486 eV) source radiation was provided under current-voltage conditions of 13 kV and 30 mA, respectively. The angle between the analyzer and the X-ray source was set at 45º, and photoelectrons were collected along the sample’s surface normal vector. Acquired data were interpreted using commercial Casaxps software, incorporating well-established curve fitting algorithms.

**FT-IR measurements:** Methylammonium and formamidinium groups possess either characterizable organic functional groups or readily identifiable chemical bond fragments. As such, Fourier Transform infrared spectra were acquired using a Nexus 670 instrument (Thermo Nicolet), equipped with a Smart Orbit diamond ATR accessory for solid powder analysis, a KBr beam splitter, and a DTGS KBr detector. The ZnSe crystal surface served as the substrate onto which the solid sample was immobilized. Measurements were collected in transmittance mode over a wavelength range of 500-4000 cm⁻¹ with a spatial resolution of 4 cm⁻¹, and acquired data were subsequently interpreted using OMNIC software.
UV-Visible and Photoluminescence measurements: UV-visible spectra were collected at 1 nm intervals using a Lambda 950 UV-Visible spectrometer, equipped with a 100 mm diffuse reflectance and transmission integrating sphere accessory for all high-end Lambda systems. The dry powder associated with our sample was loaded inside a 100 mm Powder Cell sample holder, equipped with a quartz window. The background was removed in advance using a standard reference of barium sulfate powder, prior to data acquisition and interpretation. The subsequent spectral data were collected in the relevant range of 300 nm to 900 nm for each sample. Concurrently, room-temperature PL spectra were obtained on samples in solution using a Horiba Nanolog spectrofluorimeter, equipped with a xenon lamp for optical excitation and a photon multiplier tube for detection. The excitation wavelength of 350 nm was selected using an ‘in-house’ grating characterized by 300 groves/mm. The desired PL signal was collected using a front face geometrical configuration after optical filtering using a 420 nm long-pass filter (Semrock).

Thermogravimetric Analysis (TGA): TGA experiments were performed on a TGA Q500 (TA Instruments), using an environment consisting either “extra dry” air or ‘high-purity’ nitrogen. As-prepared samples were then prepared for TGA analysis by first loading more than 10 mg of the product powder within a Pt boat, followed by the application of a controlled heating profile. A typical heating profile involved a ramping up at a rate of 20°C per minute, from room temperature up to a final temperature of 600°C.

X-ray nanoprobe measurement: Samples were dispersed in ethanol and were drop cast onto a specially designed Si substrates with fiducialized grid lines (NCT4155P-III-Cr, Nocarda). Nanoscale scanning hard X-ray micrographs, combining fluorescence and differential phase contrast (DPC) imaging, was performed using the Nano-Mii (Nanoscale Multimodal Imaging Instrument) apparatus at the Hard X-ray Nanoprobe (HXN) beamline, situated at the National Synchrotron
Light Source II (NSLS-II) facility. Monochromatic X-rays with energies of 12 keV were focused down to a ~13 nm spot size using multilayer Laue lenses. Raster imaging was performed using continuous fly-scans, wherein both far-field diffraction images arising from the transmitted nanobeam through the sample and fluorescence spectra were acquired, while the motor was still moving. The former results in differential phase contrast imaging by analyzing the angular shift of the nanobeam, due to the sample, while the latter enables elemental maps, highlighting the chemical composition of the sample and yield a morphologic image showing the electron density distribution. For a more accurate measurement, spectral fitting and intensity normalization of the fluorescence datasets were performed using PyXRF software. Specifically, the process involved (i) first fitting the summed fluorescence spectra in order to identify constituent, component elements and then (ii) fitting the individual spectrum collected at each scanned position so as to separate and differentiate between overlapping peaks, associated with different elements. The DPC dataset was processed by the DPC analysis software at the HXN beamline.
III. Introduction to Solvent Effects for Perovskites.

In the case of Pb perovskites, the solvent used has been proven to play a key role in the crystallization process. The nature of interactions between the Pb salt precursor and the solvent in addition to the formation of the resulting crystalline, solvent-coordinated intermediates have been extensively studied, especially when DMF and dimethyl sulfoxide (DMSO) have been used as solvents for Pb perovskites.\textsuperscript{36-39} Specifically, in solution, PbI\textsubscript{2} will form PbI\textsubscript{6}\textsuperscript{4-} octahedra. However, solvent molecules may displace and replace the I\textsuperscript{-} ions, thereby yielding a structure of Pb\textsubscript{1-x}S\textsubscript{6-x}\textsuperscript{2-x}, wherein S is the solvent and ‘x’ is an integer between 0 and 5.\textsuperscript{40} These ‘coordination-type’ compounds have been detected by means of UV-visible absorption spectroscopy. Either perovskites containing defects can be produced via solvent coordination or the desired perovskites may not form at all, especially if the solvent molecules coordinate too strongly.

All of these observations have helped to elucidate the formation mechanism of lead iodide perovskites, which is expected to be similar to that of the related Ge-based perovskites analyzed herein. Specifically, the first step involves the creation of PbX\textsubscript{2} layers separated by A-site cations, followed by the generation of APbX\textsubscript{3}, formed as a perovskite structure.\textsuperscript{41} Obviously, it is essential that PbX\textsubscript{6}\textsuperscript{4-} octahedra in the form of PbX\textsubscript{2} layers are present within the precursor solution, as these units are the fundamental building blocks of the perovskite structure. However, as previously discussed, solvent molecules can replace the X\textsuperscript{-} ions to varying degrees, depending on the solvent, thereby resulting in ostensibly different products. A solvent which coordinates strongly onto Pb can almost entirely replace all of the X\textsuperscript{-} ions, or at the very least, greatly reduce the amount of PbX\textsubscript{6}\textsuperscript{4-} octahedra present in solution, thereby effectively inhibiting formation of the perovskite structure. Alternatively, a solvent that moderately coordinates with the Pb can still form a perovskite structure, but this will yield a structure containing X-site defects from whence the solvent molecules are
coordinated onto the Pb ‘core’. The presence of these X-site defects renders the perovskite as potentially more susceptible to degradation via hydration upon exposure to moisture in the air. As a result, it is of utmost importance to select a solvent with an appropriate and ‘balanced’ amount of coordination to the precursors in order to generate the desired perovskite with a minimal number of defects. This solvent effect has been relatively unexplored with respect to the formation Ge-based perovskites, and hence, it is not well understood. Therefore, we carried out experiments using different solvents to address this issue with respect to the formation of MAGeI$_3$ in particular.
IV. IR Discussion

In Fig. S3, a broad peak centered at 3090 cm$^{-1}$ could be assigned to a C-H stretch and was observed in all of the organic germanium perovskite samples. With respect to specific perovskites, a broad (though expected) peak located at 3450 cm$^{-1}$ was correlated with the N-H stretching mode for primary amines, a signal found only in association with the MA$^+$ cation (as in MAGeI$_3$). By comparison, a sharp peak situated at 1704 cm$^{-1}$ denoted a C=\(N\) vibrational stretch, whereas a related peak located at 1350 cm$^{-1}$ could be ascribed to the NH scissoring mode of secondary amine; both were characteristic of the FA$^+$ cation alone (as in FAGeI$_3$). Within the series of MA$_{1-x}$FA$_x$GeI$_3$ samples, we noted a sharp peak located at 1485 cm$^{-1}$ and assigned that to an asymmetric NH$_2$ bending mode for MAGeI$_3$ itself; this peak signal decreased in intensity, with increasing \(x\) and ultimately disappeared with pure FAGeI$_3$. We observed a similar pattern of behavior associated with two additional peaks positioned at 1408 cm$^{-1}$ and 1246 cm$^{-1}$, corresponding to the CH$_2$ scissoring and C-N stretching modes, respectively. Overall, these spectra suggested that the mixed MA$_{1-x}$FA$_x$GeI$_3$ perovskites had indeed formed as a stable entity and that the two cations were well incorporated within the underlying lattice. With respect to CsGeI$_3$, we did not find evidence for either the presence of organic ligands or pendant hydroxyl groups adsorbed on its surface, all of which were consistent with expectations.
V. Additional Synthesis Experiments and Insights

As a logical extension of our efforts presented herein, we want to highlight two key aspects worth emphasizing to underline the significance of this synthetic development. In this regard, we wish to emphasize (i) the large scale synthesis potential of our methodology coupled with (ii) the flexibility and generalizability of our paradigm to the production of related families of perovskites.

In terms of the potential capability of initiating a large-scale synthesis, we note that in the experimental section, we used 0.1 mmol precursors for our reported protocol. In a test run as a demonstration of principle, we tripled the quantities of precursors (and products) without any issue, confirming the potential viability of scaling up our methodology. Indeed, there are no practical limitations, hindering us from dramatically increasing the quantities of not only the GeI₂ and Al precursors used but also the solvent since the reagents are soluble within the reaction medium.

Moreover, we have emphasized that the methodology we have used to synthesize our Ge-perovskites does not depend on the identity of either the halide used for X site or the cation utilized for the A site. Indeed, in this study, we have already shown that the A site cation could be systematically tuned by changing the chemical nature of the precursor used. In subsequent work therefore, as a viable demonstration of principle, we have substituted the iodide anion at the X site with Br⁻. Specifically, we used GeBr₂ and either MAI or FAI to synthesize the corresponding MAGeBrₓI₃₋ₓ and FAGeBrₓI₃₋ₓ with ‘x’ = 1 or 2. The XRD patterns of our as synthesized materials are shown in Figure S10. The results are consistent with the reported XRD pattern of MAGeI₃₋ₓBrₓ with “x” = 0 ~ 0.33. We also compared data on as-obtained MA/FAGeBr₂I or MA/FAGeBrI₂ with the corresponding XRD patterns for MAGeI₃ or FAGeI₃, respectively, as shown in Figure S10.

Herein, we are confident that we have indeed generated the mixed halide Ge-perovskite. Following this logic, by using relevant MABr, FABr, MACl, and FACl precursors, we can likely
generate the corresponding Br and Cl perovskites in this manner and further demonstrates that this method could be generally applied to the production of other analogous systems on a larger scale.
Figure S1. Photographs of isolated MA$_{1-x}$FA$_x$GeI$_3$ products with ‘x’ = 0, 0.25, 0.5, 0.75, and 1 (from left to right). From left to right, the observed color of the as-prepared samples progresses and lightens in shade from dark red, red, orange-red, orange, and finally orange-yellow with increasing ‘x’ values (i.e., increasing FA$^+$ cation content).
Figure S2. Collected XRD patterns of as-prepared (A) MAGel$_3$; (B) MA$_{0.75}$FA$_{0.25}$GeI$_3$; (C) MA$_{0.5}$FA$_{0.5}$GeI$_3$; (D) MA$_{0.25}$FA$_{0.75}$GeI$_3$; and (E) FAGel$_3$ within the magnified range from 25º to 35º. The position of the yellow arrow implies a small, notable angular shift, with increasing FA$^+$ cation content (i.e., increasing ‘x’ values).
Figure S3. Infrared spectra of as-prepared CsGeI₃ (black curve), MAGeI₃ (red curve), MA₀.₇₅FA₀.₂₅GeI₃ (blue curve), MA₀.₅FA₀.₅GeI₃ (pink curve), MA₀.₂₅FA₀.₇₅GeI₃ (green curve), and FAGeI₃ (navy curve).
Figure S4. XPS survey spectra for (A) MAGeI₃; (B) MA₀.₇₅FA₀.₂₅GeI₃; (C) MA₀.₅FA₀.₅GeI₃; (D) MA₀.₂₅FA₀.₇₅GeI₃; (E) FAGeI₃; and (F) CsGeI₃ with all of the elements labeled.
Figure S5. UV-visible spectra (black) associated with the normalized PL spectra (blue) of (A) MAGeI₃; (B) MA₀.₇₅FA₀.₂₅GeI₃; (C) MA₀.₅FA₀.₅GeI₃; (D) MA₀.₂₅FA₀.₇₅GeI₃; and (E) FAGeI₃. (F) presents a summary of all of the five samples tested with the solid line, highlighting the UV-visible spectra and the dotted line, indicative of PL spectra, with black for MAGeI₃, red for MA₀.₇₅FA₀.₂₅GeI₃, blue for MA₀.₅FA₀.₅GeI₃, pink for MA₀.₂₅FA₀.₇₅GeI₃, and olive for FAGeI₃.
Figure S6. Analyzing the Solvent Effect. Normalized UV-visible absorption spectra of GeI₂, taken in anhydrous ethanol (red), THF (blue), DMF (purple), and acetonitrile (green).
Figure S7. TEM images of particulate samples subjected to strong electron beam damage. Images presented are of (A) MAGeI$_3$; (B) FAGeI$_3$; as well as of CsGeI$_3$ both (C) prior to and (D) after increasing beam brightness.
Figure S8. Normalized scanning X-ray microscopy images for a series of samples of MA$_{1-x}$FA$_x$GeI$_3$: (A-D) MAGeI$_3$; (E-H) MA$_{0.75}$FA$_{0.25}$GeI$_3$; (I-L) MA$_{0.5}$FA$_{0.5}$GeI$_3$; (M-P) MA$_{0.25}$FA$_{0.75}$GeI$_3$; and (Q-T) FAGeI$_3$. From left to right, the data columns denote Ge K edge images (A, E, I, M, & Q); I L edge images (B, F, J, N, & R); Ge K edge / I L edge signal images (C, G, K, O, & S); in addition to I L edge signal/ Ge K edge signal images (D, H, L, P, & T). Signals less than 5% of the average intensity were considered as noise.
Figure S9. Normalized scanning X-ray microscopy images for CsGeI$_3$. The data include images of the (A) Cs $L$ edge; (B) Ge $K$ edge, (C) I $L$ edge; (D) Cs $L$ edge / I $L$ edge; (E) Ge $K$ edge signal / I $L$ edge; and (F) I $L$ edge / Ge $K$ edge.
Figure S10. XRD patterns of a series of as-prepared Ge-Br/I perovskites, including (A) MAGeI₃; (B) MAGeBrI₂; (C) MAGeBr₂I; (D) FAGeI₃; (E) FAGeBrI₂; and (F) FAGeBrI₂.
Table S1. Analyzing the Solvent Effect. Measured absorbance peak locations of GeI₂ collected within various solvents.

| Solvent   | Absorbance peaks (nm) |
|-----------|-----------------------|
| Acetonitrile | 251, 281, 328          |
| DMF       | 268                   |
| THF       | 294, 367              |
| Ethanol   | 291, 360              |
| Technique | X-ray Energy | Estimated Photon Flux | Focus area | Estimated Flux Rate |
|-----------|--------------|-----------------------|------------|---------------------|
| XRD       | 8 keV        | $10^5$ photons/sec    | 65 mm$^2$ | $10^6$ photons/(sec·m$^2$) |
| XPS       | 1.3 keV      | $5\times10^{12}$ photons/sec | 1 cm$^2$ | $5\times10^{16}$ photons/(sec·m$^2$) |
| SHXM      | 12 keV       | $10^9$ photons/sec    | 156 nm$^2$ | $6.4\times10^{24}$ photons/(sec·m$^2$) |

Table S2. A comparison of voltage and irradiation conditions, spanning different characterization and imaging techniques utilizing X-ray sources.
Table S3. A comparison of current-voltage conditions, spanning different characterization and imaging techniques, utilizing electron-based sources. Parameters highlighted include a combination of real-time data of current and voltage operating conditions, complemented by additional information we extracted by carefully perusing the operating manuals of the relevant instruments used to acquire the data in question.

| Technique  | Current Conditions | Accelerating Voltage | Dose Rate               |
|------------|--------------------|----------------------|-------------------------|
| TEM/HRTEM  | 70 / 149 µA (beam) | 120/200 kV           | ~10 electrons/ (Å²·sec) |
| SEM        | 10 µA (beam)       | 10 kV                | ~20 electrons /(Å²·sec) |
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