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Supporting Information: Visualization of heated latent fingerprints by synchrotron radiation soft X-ray photoemission electron microscopy

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Fingerprint testing is routine for personal identification, but damaged prints, such as those exposed to heat in fire cases, are difficult to visualize. Currently, there is no method available for severely heat-damaged fingerprints. We developed a visualization technique using synchrotron radiation (SR) soft-X-ray photoemission electron microscopy (PEEM). Fingerprints on silicon, stainless steel, aluminum, brass, and glass were heated at 400 °C for an hour and then examined. Ridge patterns could not be seen with optical microscopy, laser microscopy, or scanning electron microscopy, except on silicon. Even then, SR PEEM revealed ridge patterns by detecting sodium particles ($\sim 1 \mu\text{m}$) clustered into 10 μm spots. Multiple images were combined to successfully produce a clear 5 mm-wide ridge pattern. For the glass substrate, prior titanium vapor deposition prevented electric discharge, resulting in clear ridge pattern observation by SR PEEM. Unlike conventional methods, SR soft-X-ray PEEM detects sodium sensitively and specifically, producing clear ridge patterns in heat-damaged fingerprints, which is promising for advanced criminal fingerprint testing.

1 Additional experimental data

1.1 Laser microscopy

For untreated fingerprints on the silicon plates, oil-droplet-like swelling particles of 210 μm size and 2080 nm of height were observed lining up to form friction ridges (Fig. S1(a)). Some particles, even though few, were also observed around the furrow region between the two lining ridges. For the untreated fingerprints on the glass, stainless steel, aluminum, and brass plates, views similar to those on the silicon plate were observed (data not shown). Untreated fingerprints can be detected on nonporous substrates by ordinary fingerprint detection technologies, because organic and inorganic fingerprint components are left on the substrates. For heat-treated fingerprint on silicon plate, instead of oil-droplet-like particles, many round aggregates of pale-colored particles of 1030 μm size and 2060 nm height, and occasionally black particles of 1030 μm size and about 1 μm height, forming ridge lines, were observed (Fig. S1(b)). These remaining materials should be inorganic compounds resistant to heating. However, for the heat-treated fingerprints on the glass, stainless steel, and aluminum plates, the traces of fingerprints could be barely as-

certained. This disappearance of the fingerprint residues may be derived from the combustion loss of organic materials. For that, on the brass plate, discolored swelling particles of 1050 μm size and 30300 nm height were observed forming ridge lines (Fig. S1(c)). In Fig. 6, the surfaces of the plates are shown as observed by a laser microscope. It is conceivable that residues derived from untreated fingerprints could be observed clearly on all the substrate plates examined, but the heat-treated fingerprints on the silicon and brass plates could be faintly observed due to leaving heat combustion residues on flat silicon surface and heat-reacting brass surface, and those on the stainless steel and aluminum plates could not be observed due to severe substrate plain roughness.

1.2 PEEM

The non-heated fingerprints on the nonporous silicon plate could be ascertained by a light microscope. In PEEM, under UV irradiation, a clear ridge pattern was observed, where not only sodium salts but also organic fingerprint materials were detected (Fig. S3(a)). Under the X-ray irradiation, more clearer ridge pattern was observed, where only sodium salts were detected (Fig. S3(b)). The heated fingerprints on the nonporous silicon plate could barely be ascertained by a light microscope. In PEEM, under UV irradiation, a clear ridge pattern was observed, where only sodium salts were detected (Fig. S4(a)). Under the X-ray irradiation, more clearer ridge pattern was observed, where only sodium

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salts were detected (Fig. S4(b)). Compared to non-heated fingerprints, the heated fingerprints could be more clearly ascertained for the ridge pattern. This is because sodium salts are masked by organic fingerprint components such as lipids in non-heated fingerprints, and in heated fingerprints, sodium salts were exposed to the surface by heat dropout of fingerprint components, leading to an increased efficiency of photoelectron emission. The same situation was observed for SEM-EDX. Fig. S5 and Fig. S6 show the SR PEEM Na mapping images of the heated fingerprint on the aluminum and brass plates, respectively. Although the heated fingerprints on nonporous but high surface roughness stainless steel, aluminum, and brass plates were not observed by laser microscopy, by PEEM under UV and X-ray irradiations, a clear ridge pattern was observed. The fingerprints on nonporous and insulating glass plates could not be analyzed by PEEM because of severe electric discharge. By vapor deposition of titanium metal on the glass plate, it was possible to perform PEEM by avoiding electric discharge. The glass plate used was soda-lime glass, and sodium was included in the glass by several percent. Fig. S7 shows the SR PEEM Na mapping images of the heated fingerprint on the glass plates. In PEEM, it was challenging to clearly identify fingerprint ridge patterns due to the influence of sodium in the glass plate. However, we were able to confirm the presence of sodium-containing particles and establish that PEEM can map the location of NaCl on glass. This finding suggests the potential for detecting fingerprints on glass using PEEM. In Figures S3-S7, artifacts caused by charging, as seen in Fig. 4, are also visible. In particular, in the case of the glass shown in Fig. S7, the insulating particles are too large, distorting the PEEM image.

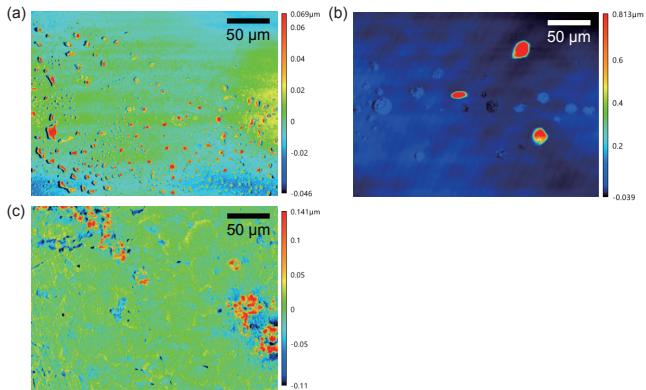


Fig. S1 Microscopic view of the fingerprints. Fingerprints on the silicon (a, b) and brass (c) plates were untreated (a) or heat-treated (b, c), and observed by laser microscope, of the used plates.

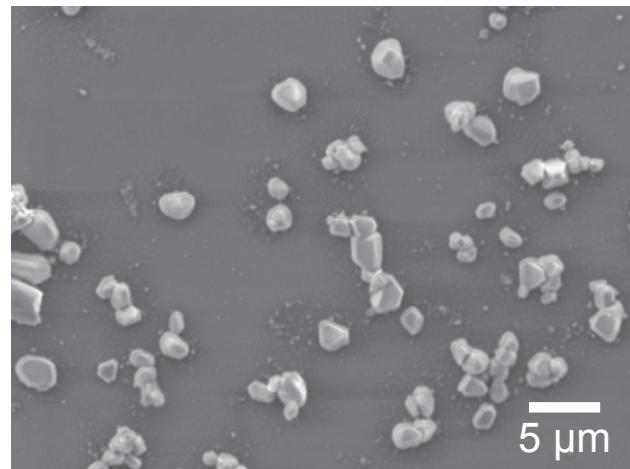


Fig. S2 SEM image of a heated fingerprint on a silicon plate at the same measurement position as in Fig. 3.

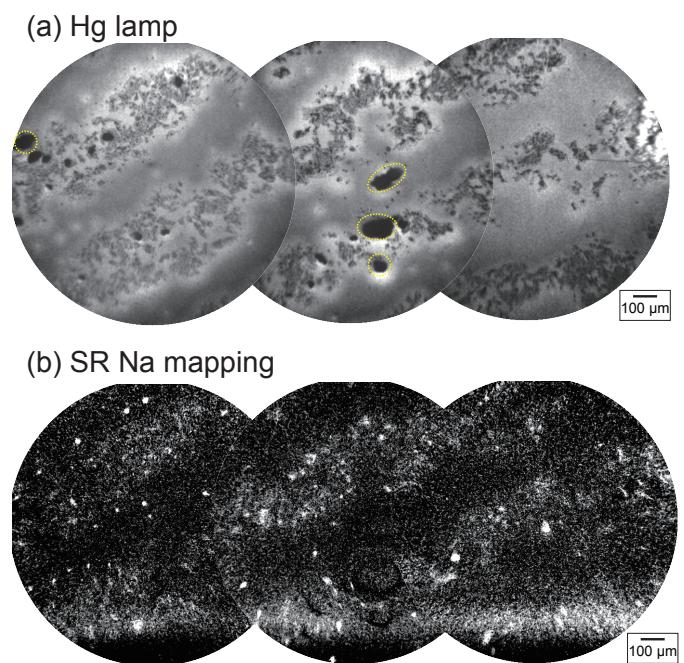
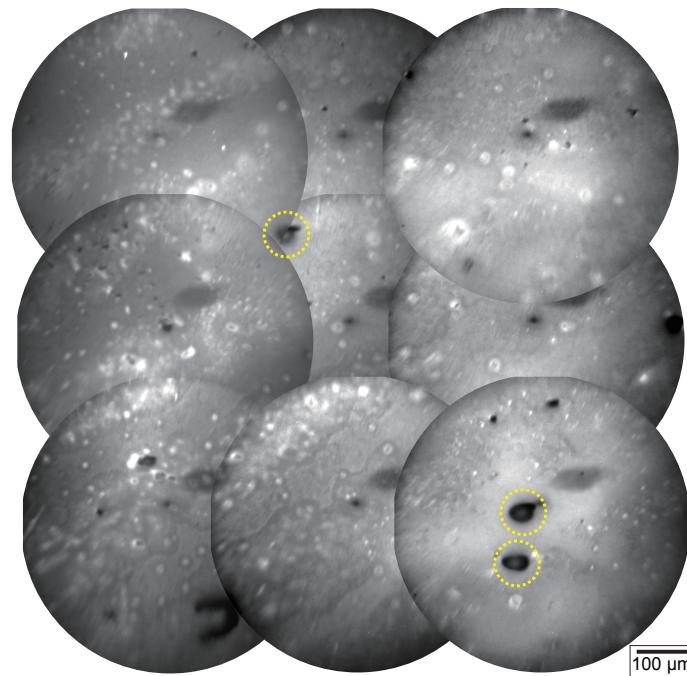


Fig. S3 (a) UV-PEEM images of the non-heated fingerprint on the silicon plate. (b) SR PEEM Na mapping images. The yellow dotted lines in this figure indicate artifacts caused by the charging of insulating particles, such as dust, on the sample surface. Only the most prominent artifacts are marked.

(a) Hg lamp



(b) SR Na mapping

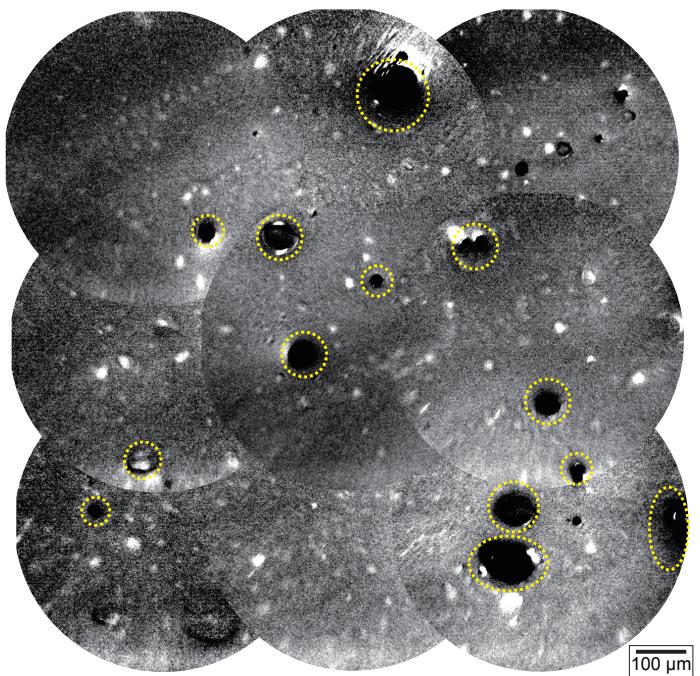


Fig. S4 (a) UV-PEEM images of the heated fingerprint on the silicon plate. (b) SR PEEM Na mapping images. The yellow dotted lines in this figure indicate artifacts caused by the charging of insulating particles, such as dust, on the sample surface. Only the most prominent artifacts are marked.

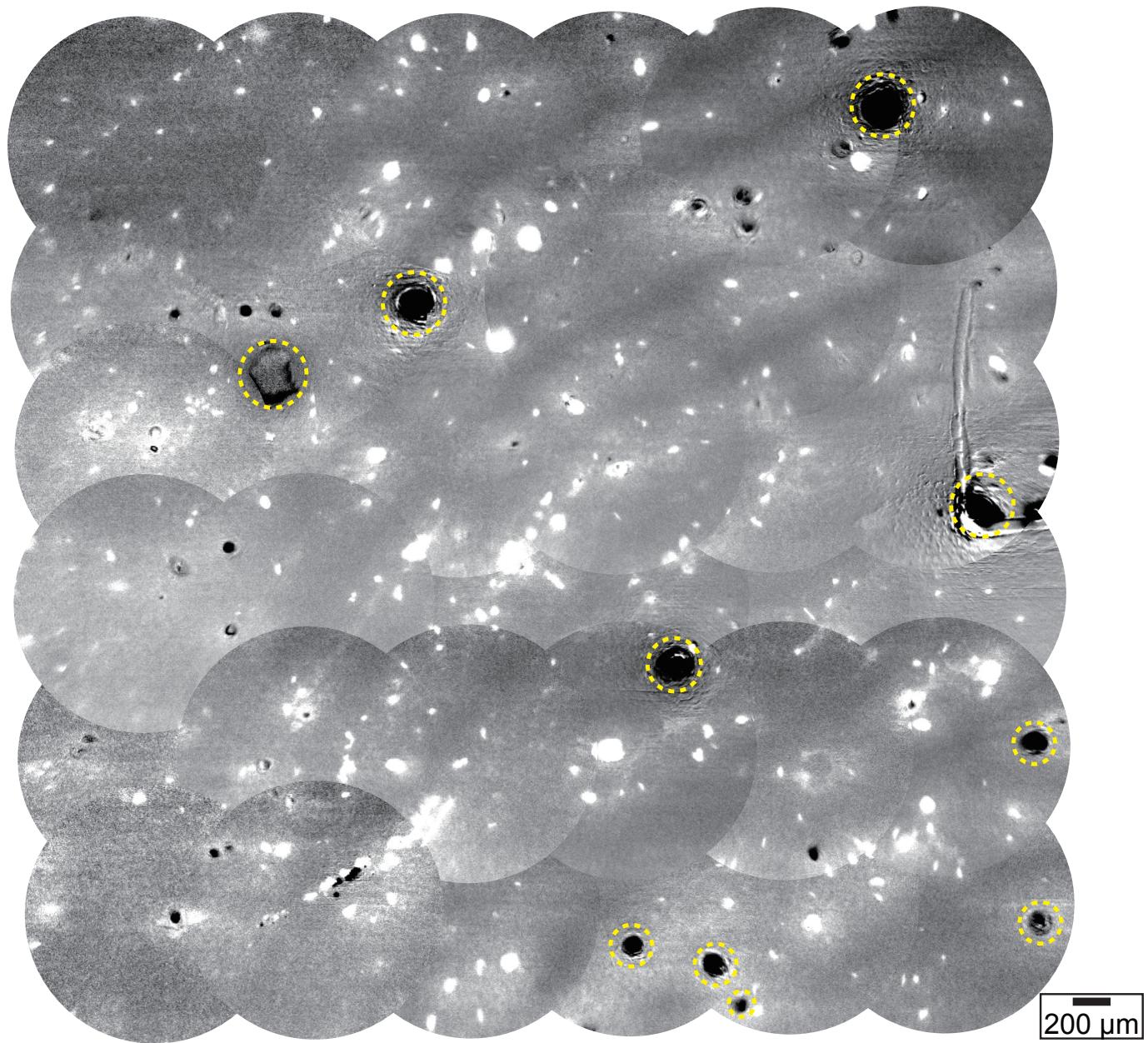


Fig. S5 Wide-field SR PEEM Na mapping images of the heated fingerprint on the aluminum plate. The yellow dotted lines in this figure indicate artifacts caused by the charging of insulating particles, such as dust, on the sample surface. Only the most prominent artifacts are marked.

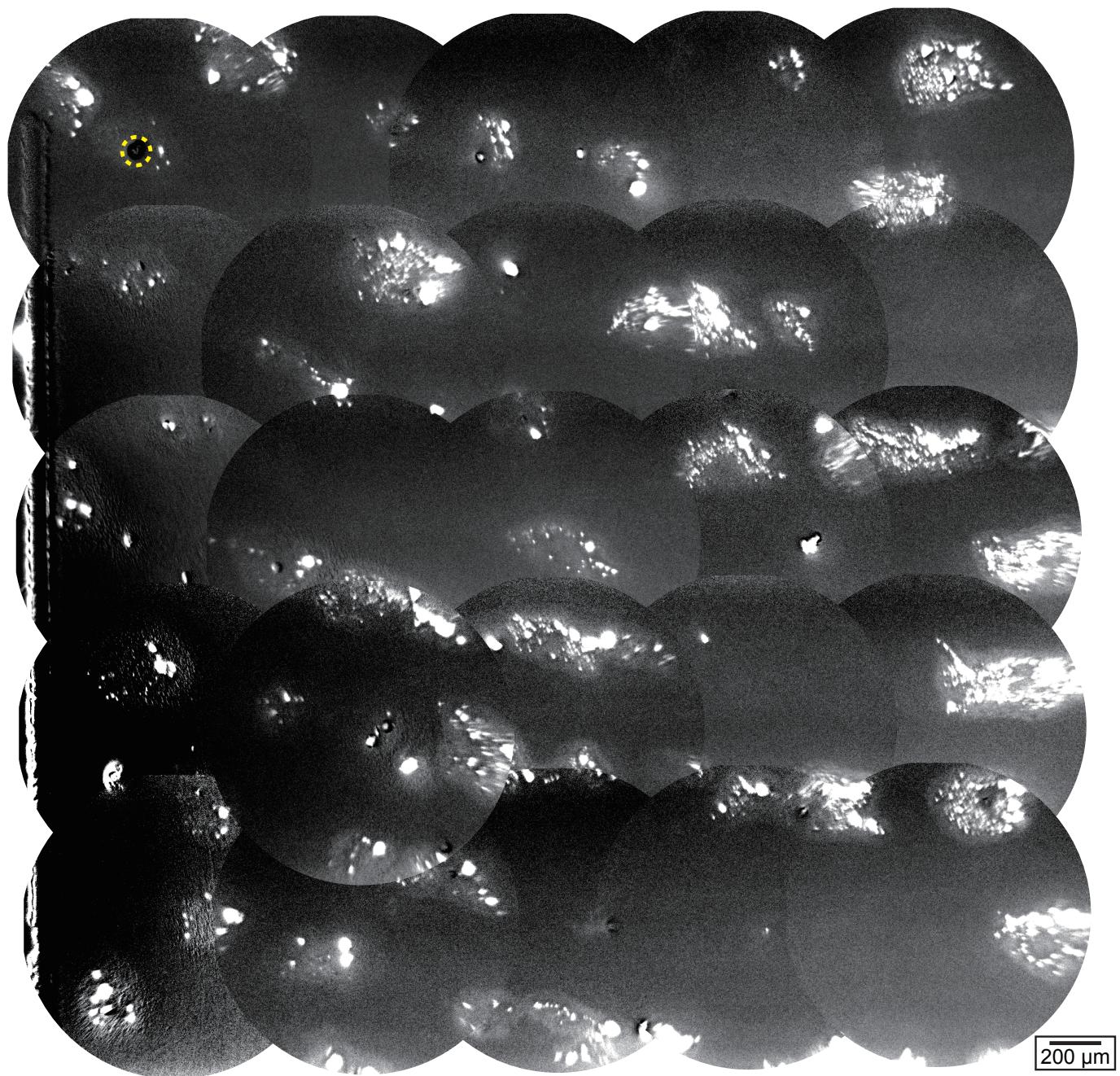


Fig. S6 Wide-field SR PEEM Na mapping images of the heated fingerprint on the brass plate. The yellow dotted lines in this figure indicate artifacts caused by the charging of insulating particles, such as dust, on the sample surface. Only the most prominent artifacts are marked.

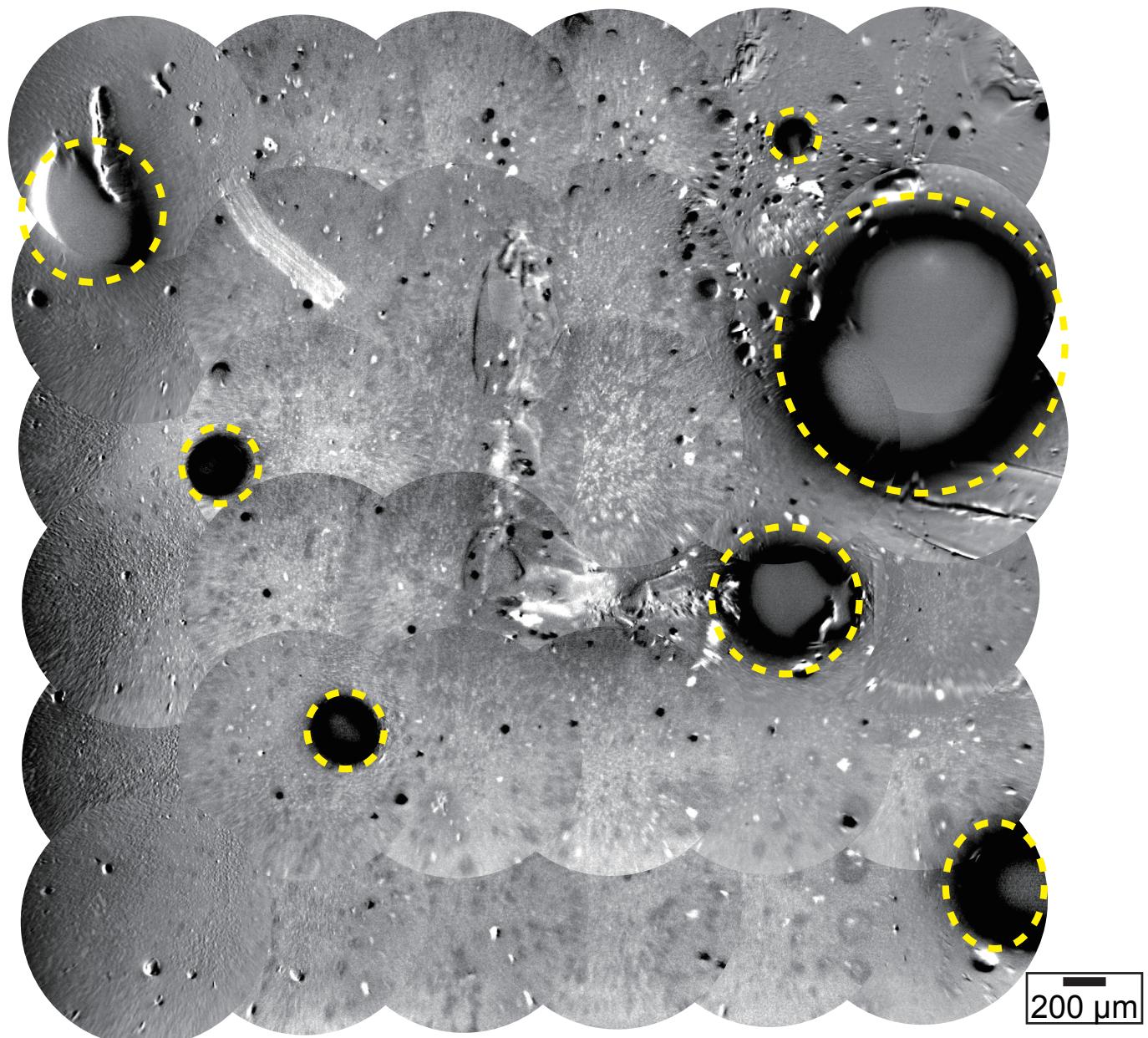


Fig. S7 Wide-field SR PEEM Na mapping images of the heated fingerprint on the glass plate. The yellow dotted lines in this figure indicate artifacts caused by the charging of insulating particles, such as dust, on the sample surface. Only the most prominent artifacts are marked.