

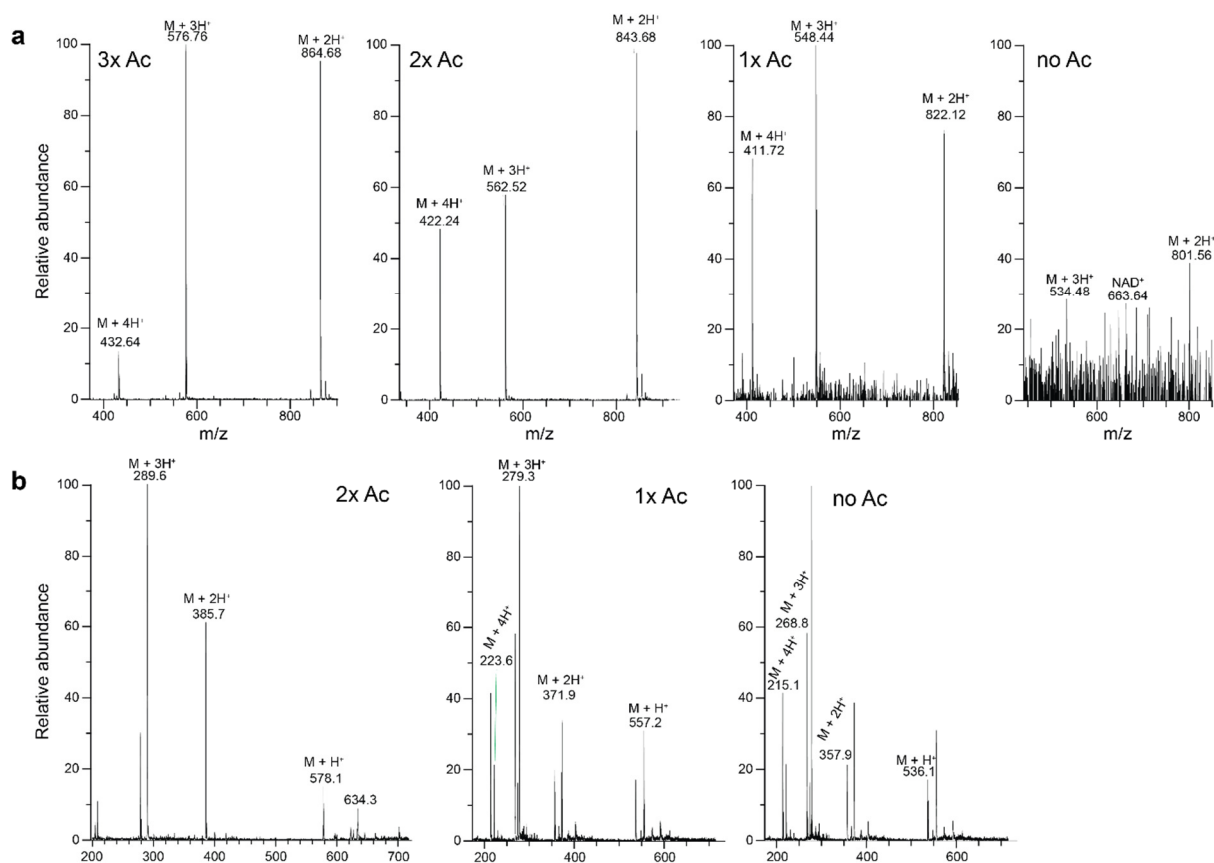
## Supplementary Information

### Shape transformations in peptide-DNA coacervates driven by enzyme-catalyzed deacetylation

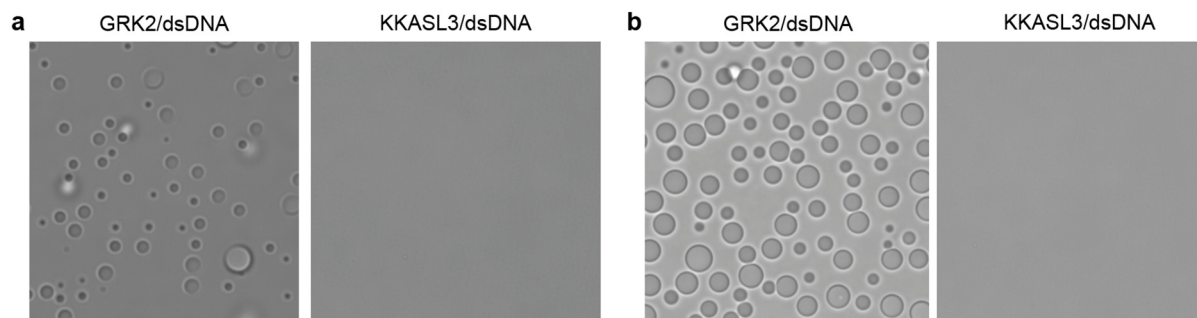
Merlijn H. I. van Haren<sup>1</sup>, Nienke S. Helmers<sup>1</sup>, Luuk Verploegen<sup>1</sup>, Viveca A. C. Beckers<sup>1</sup> and Evan Spruijt<sup>1\*</sup>

<sup>1</sup>Institute for Molecules and Materials, Radboud University, Heyendaalseweg 135, 6523 AJ

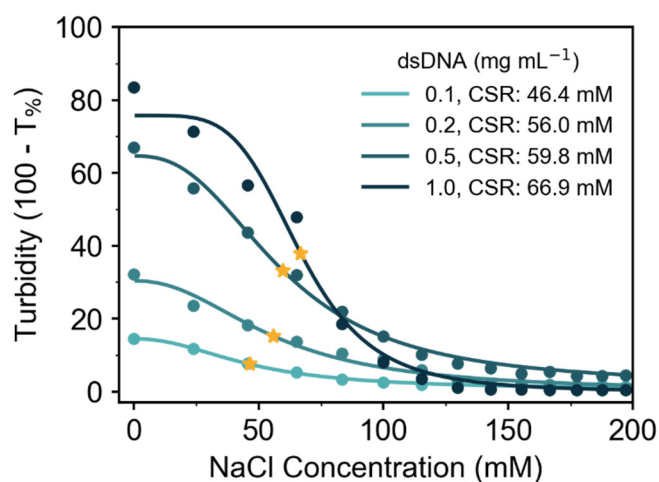
Nijmegen, The Netherlands. \*Email: [e.spruijt@science.ru.nl](mailto:e.spruijt@science.ru.nl)



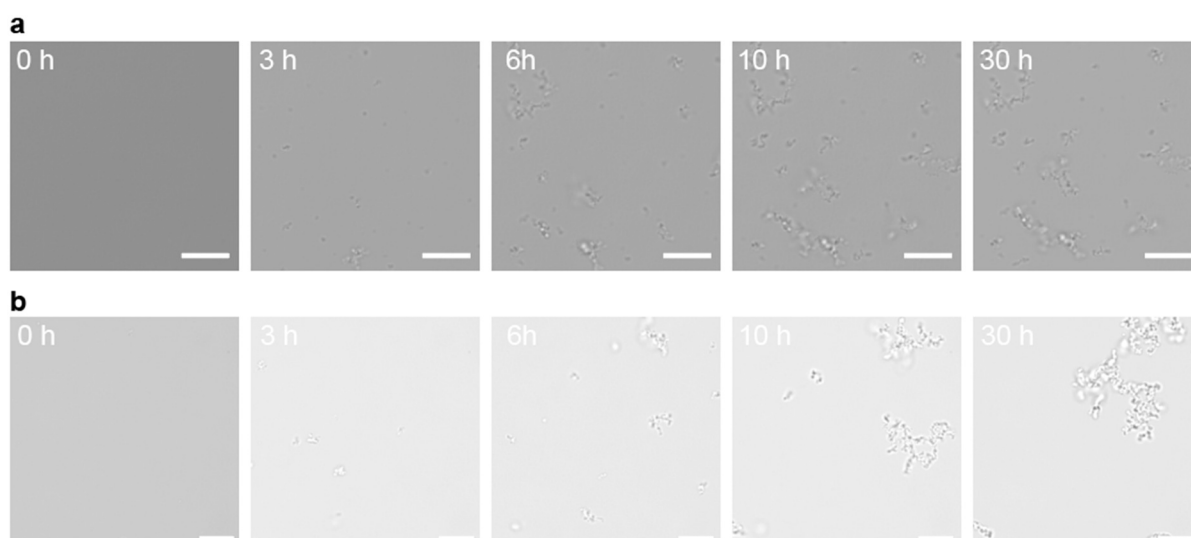
**Supplementary Figure 1** Mass spectrum of KKASL3 and GRK2 after deacetylation by SIRT3. **a** mono-, di-, tri- and fully deacetylated KKASL. **b** mono-, di- and fully deacetylated GRK2.



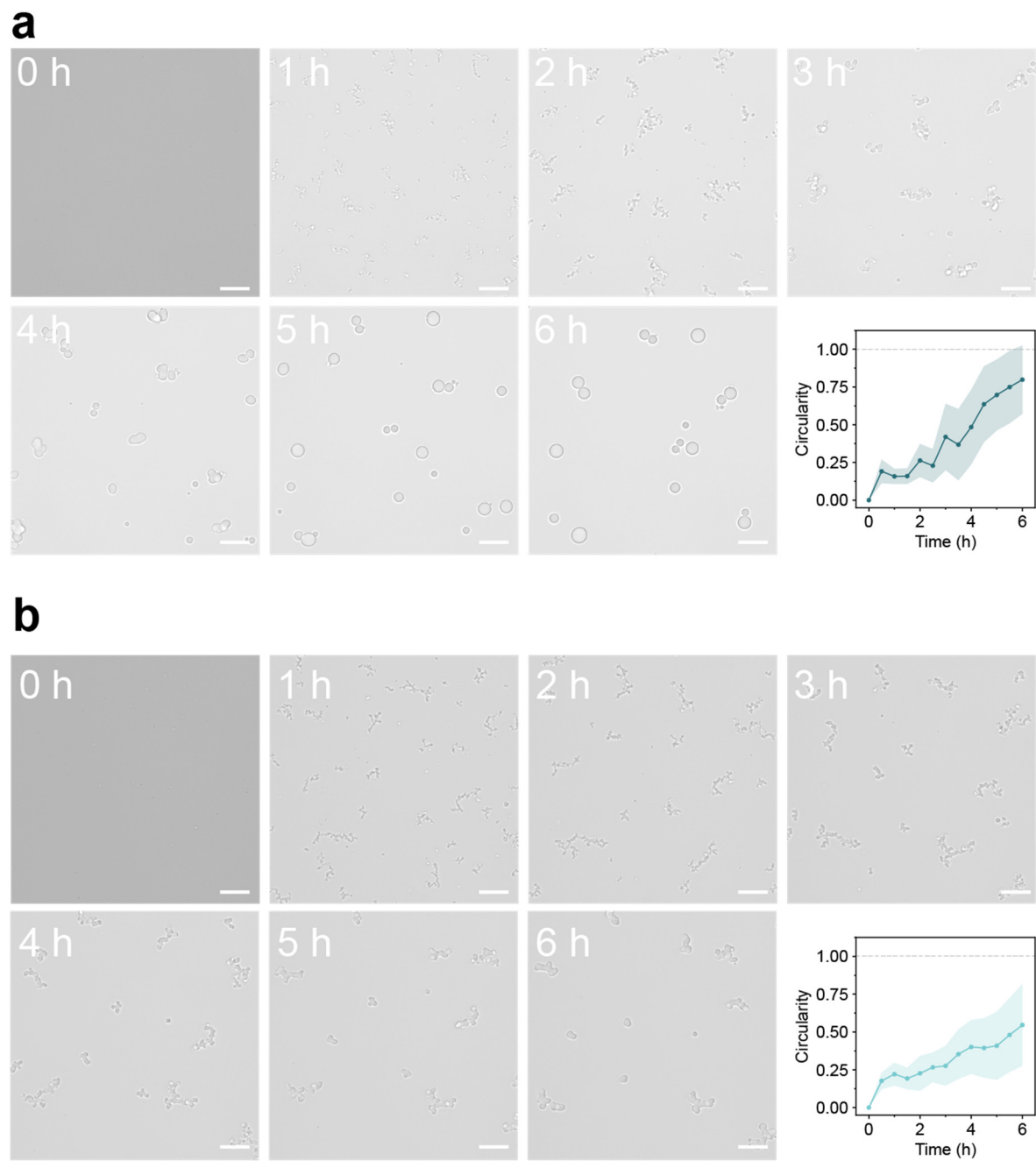
**Supplementary Figure 2** Coacervate formation of GRK2 with dsDNA and KKASL3 with dsDNA, before (a) and after (b) incubation with SIRT3.



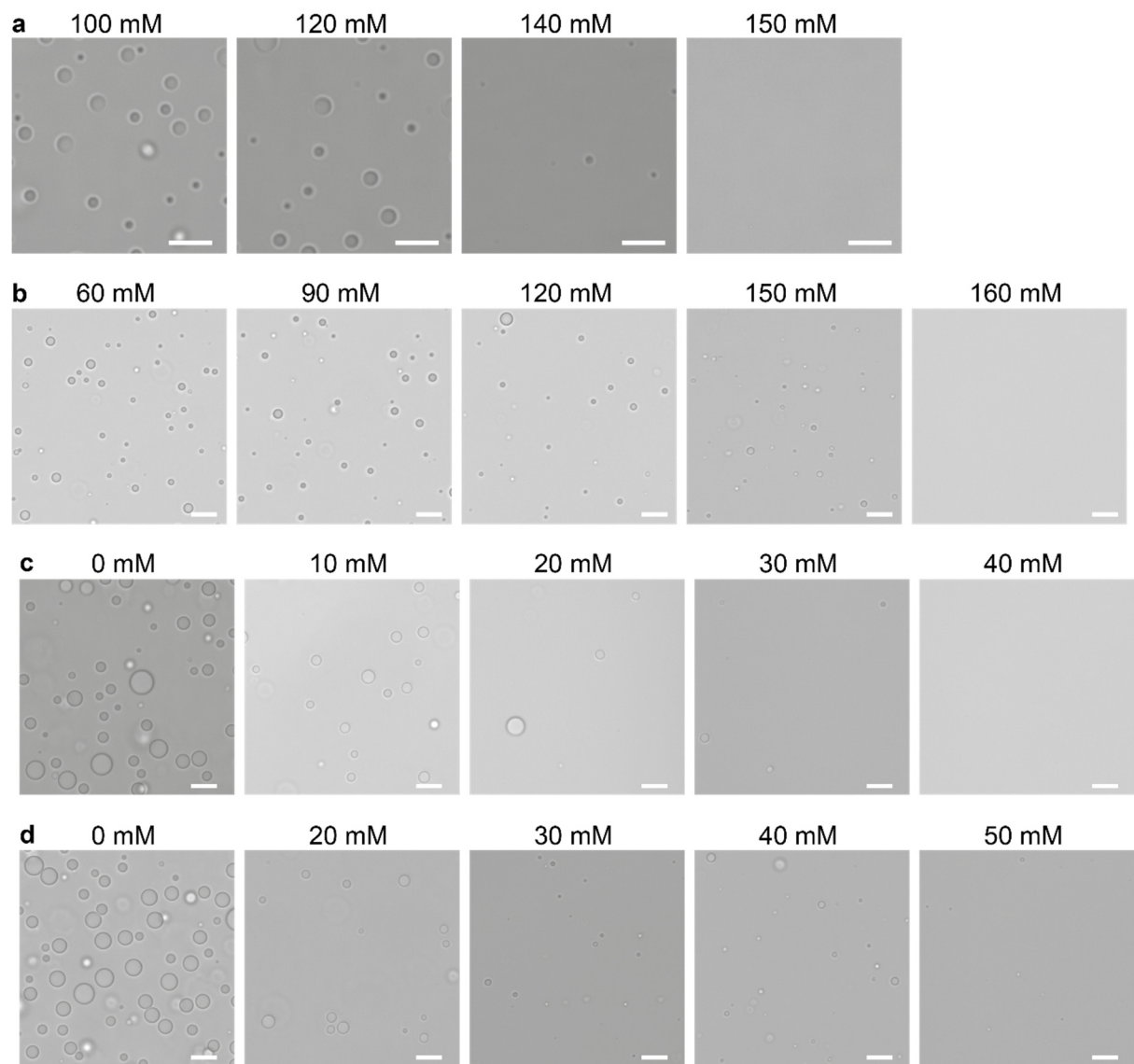
**Supplementary Figure 3** Absorbance measurements are converted to turbidity and fitted with a Hill equation (see methods). Yellow stars indicate the coacervate salt resistance at the half-maximum of the fitted Hill equation. Sample consists of 2 mM GRK2, 20 mM NAD<sup>+</sup>, 1 mM DTT, 2.5 μM SIRT3 in 50 mM HEPES pH 7.4.



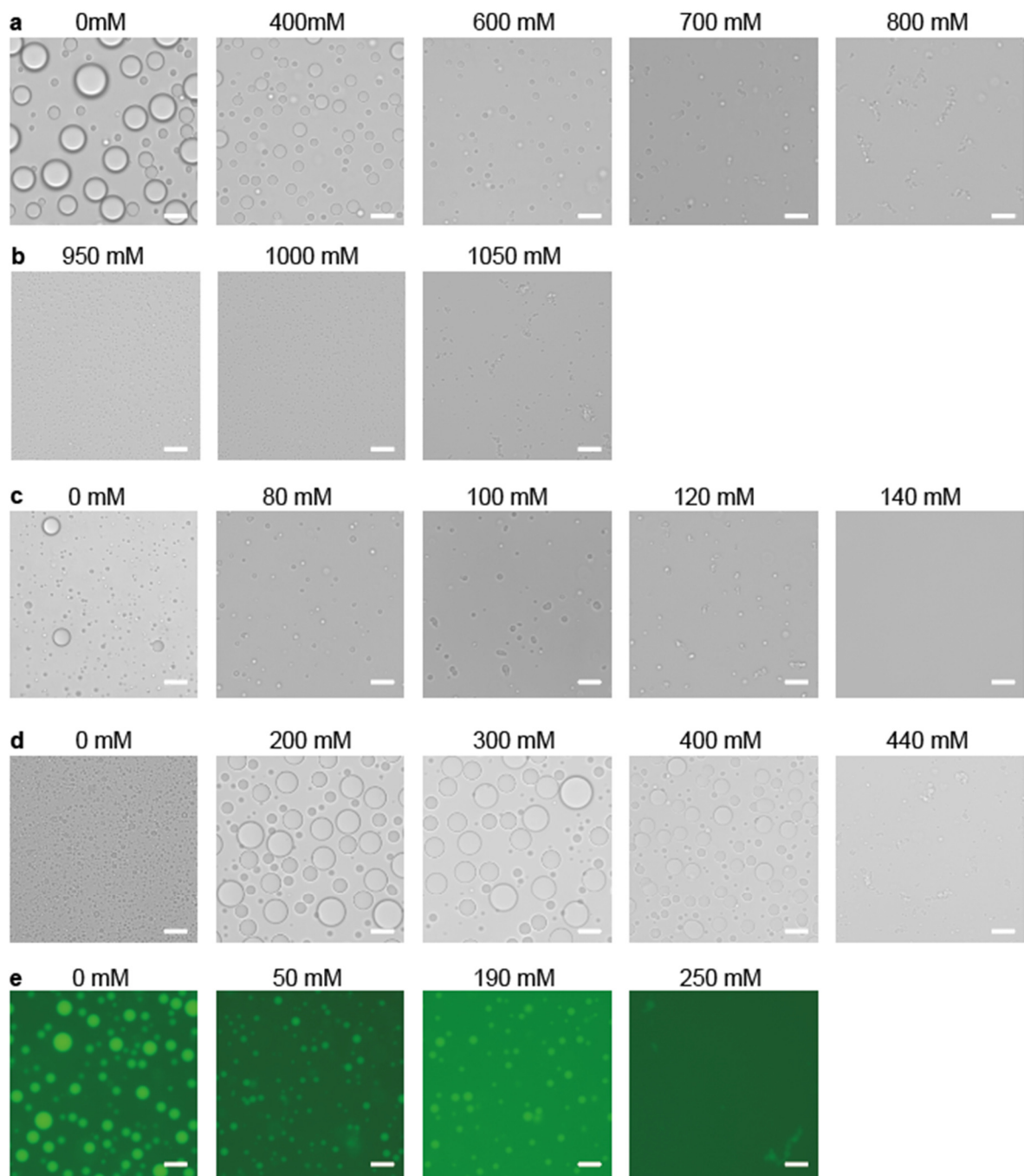
**Supplementary Figure 4** Control reactions without NAD<sup>+</sup> (a) and inactivated SIRT3 (b). SIRT3 was inactivated by 5 minute incubation at 100 °C. Scale bar is 10 μm.



**Supplementary Figure 5** Shape transformations during enzymatic reaction with different SIRT concentrations. **(a)** First 6 hours of the deacetylation reaction containing 1.5  $\mu\text{M}$  SIRT3. **(b)** First 6 hours of the deacetylation reaction containing 0.5  $\mu\text{M}$  SIRT3. Scale bar is 20  $\mu\text{m}$ .



**Supplementary Figure 6** Phase separation of GRK2 with polyU (a) Scale bar 5  $\mu\text{m}$ . polyA (b),  $D_{100}$  (c) and  $E_{100}$  (d) at increasing NaCl concentrations, scale bar 10  $\mu\text{m}$ . No amorphous aggregates were found close to the CSR.



**Supplementary Figure 7** Phase separation of dsDNA with polycations  $K_{20}$  (**a**),  $K_{100}$  (**b**), SRRRR (**c**), PDDA (**d**), ELP- $K_{72}$  (**e**) at increasing NaCl concentrations. Amorphous gel-like aggregates were found close to the CSR. Scale bar is 10  $\mu\text{m}$ .