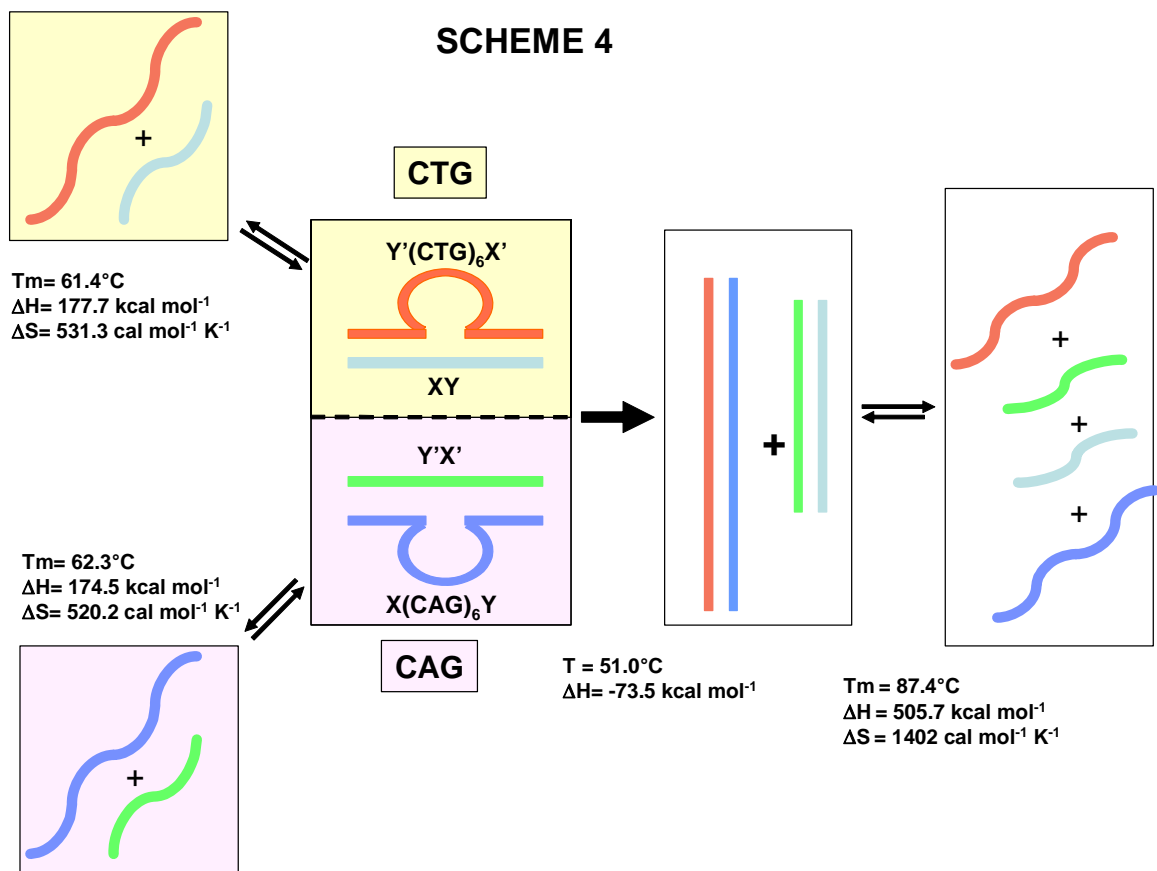


Supplementary Material

Scheme 4: Flowchart summarizing the measured thermodynamic data at the transition temperature for the strand exchange transformation and associated equilibria.



Scheme 5: Flowchart summarizing the measured thermodynamic data at the transition temperature for the 4 possible strand displacement transformations and associated equilibria

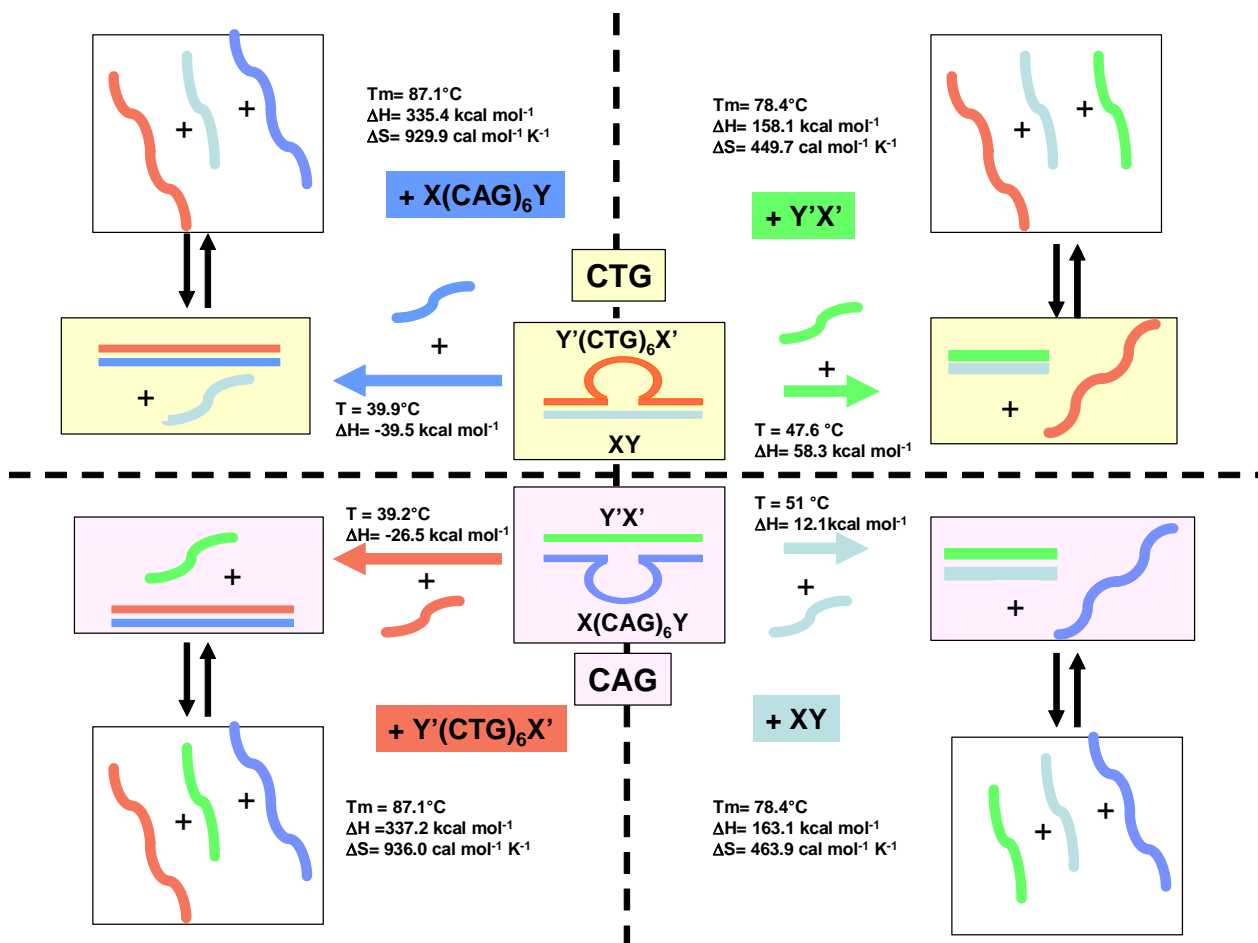


Figure 4B: Measured and calculated CD spectra for strand displacement of $X(\text{CAG})_6Y \cdot Y'X'$ with XY

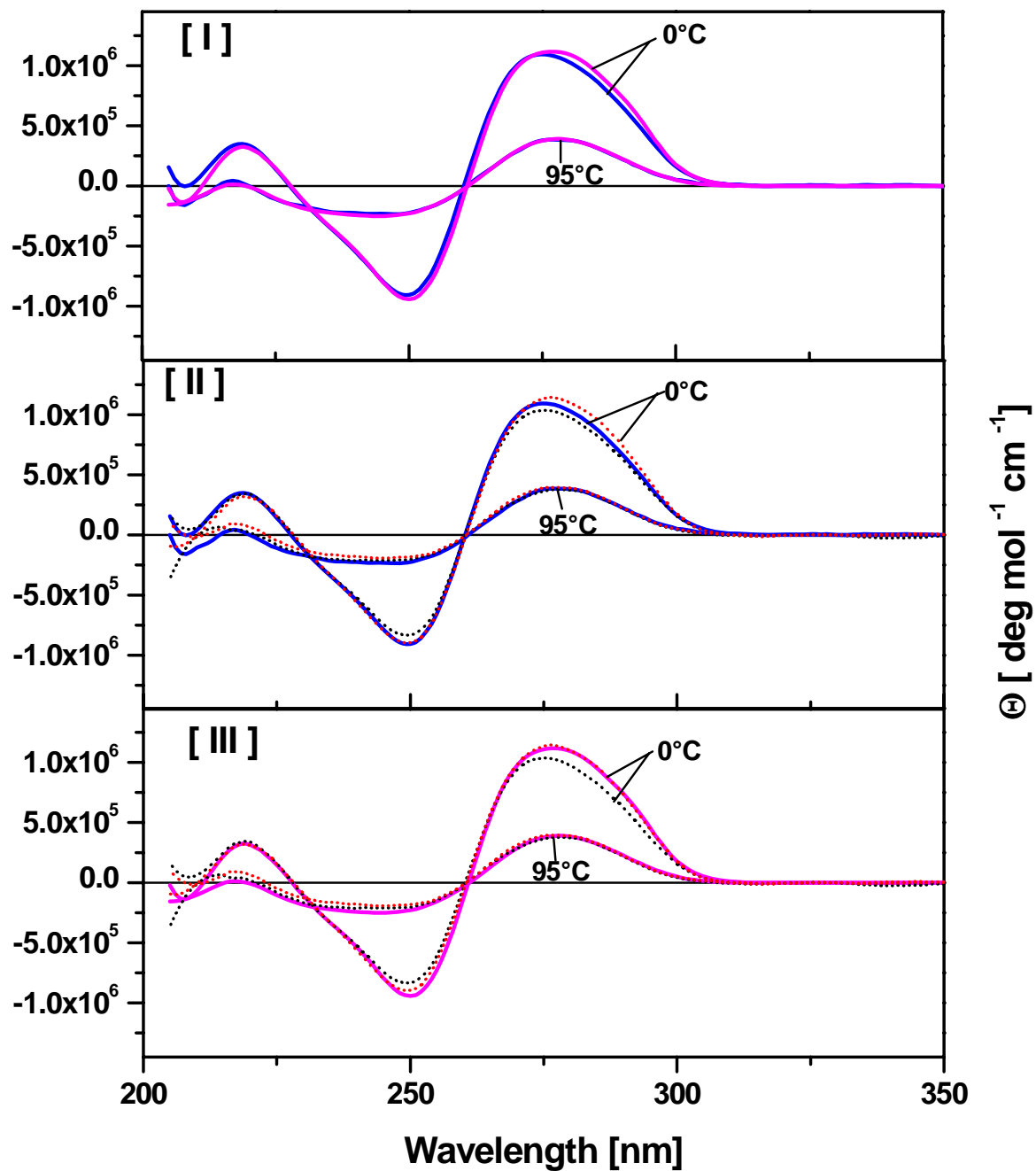


Figure 4C: Measured and calculated CD spectra for strand displacement of $Y'(CTG)_6X' \cdot XY$ with $X(CAG)_6Y$

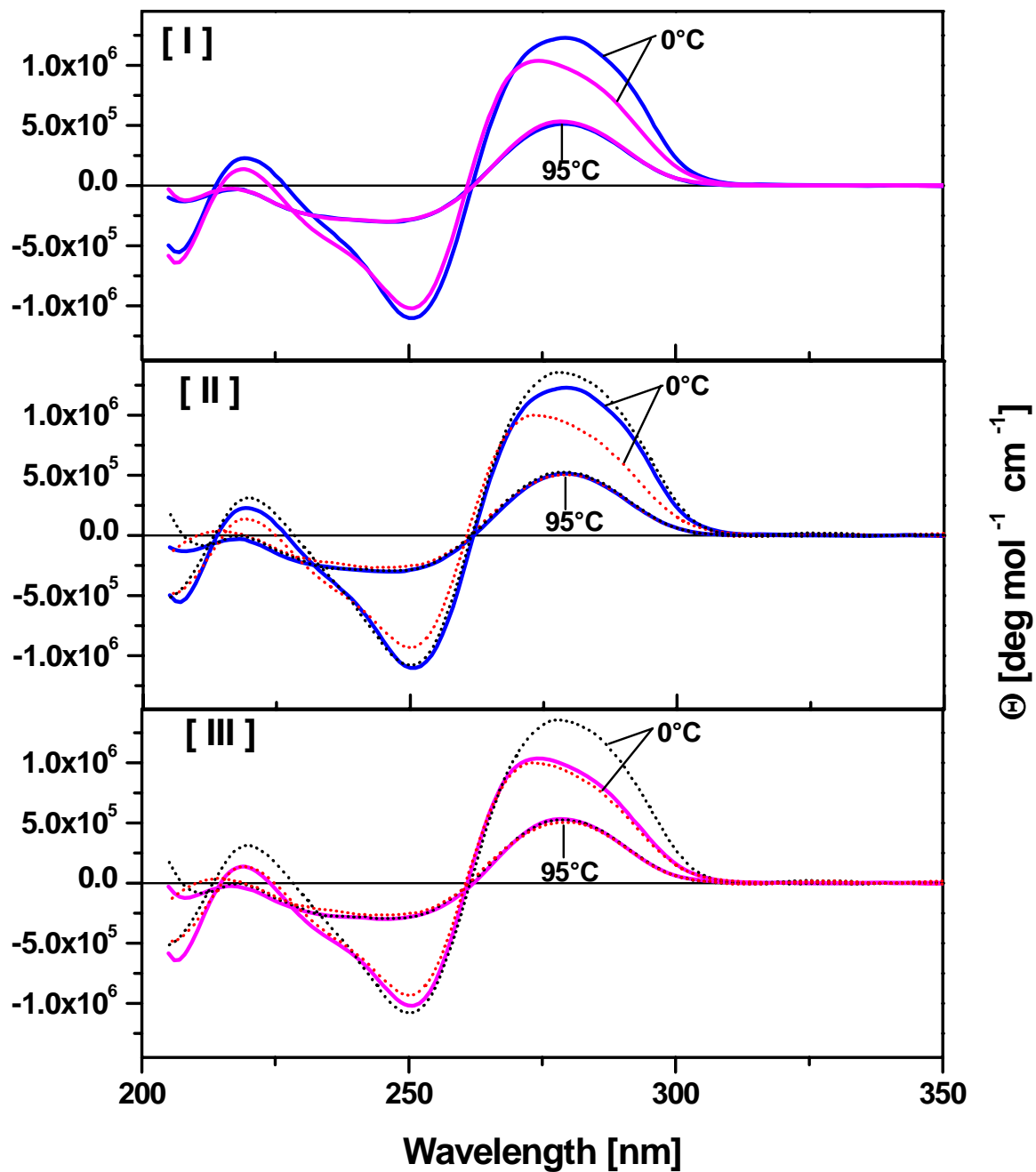


Figure 4D: Measured and calculated CD spectra for strand displacement of $Y'(CTG)_6X' \cdot XY$ with $Y'X'$

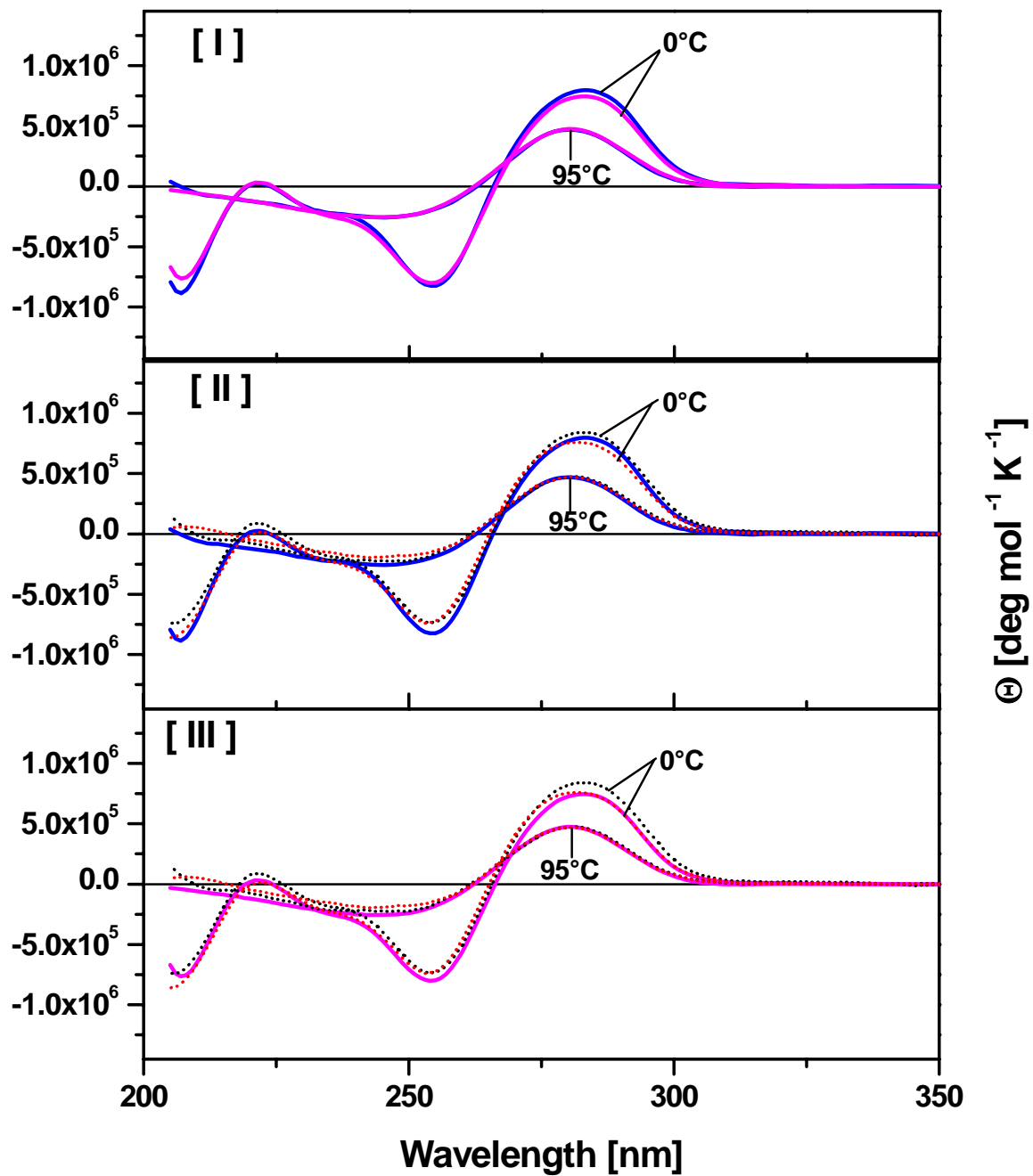


Figure 4 Supplementary : CD spectra measured for the 4 different strand displacement reactions shown in **scheme 3** and summarized in **scheme 5**

Panel I always shows the experimentally measured CD spectra of a 1:1 mixture of the Ω -DNA with the added single strand at 0°C prior to heating (blue, solid) and after heating to 95°C and cooling (magenta, solid). Also shown as internal controls are the CD spectra at 95°C (blue & magenta, solid). Panel II always compares the measured CD spectrum prior to heating to the spectra calculated for a 1:1 mixture of the Ω -DNA and added single strand (black stippled line), as well as for the 1:1 mixture of the resulting duplex and displaced single strand (red stippled line). Panel III always compares the corresponding experimental CD spectrum (magenta, solid) after heating to the calculated spectra (red & black, stippled line), as in panel II. Also shown as internal controls in panel II and III are the calculated (red & black, stippled) and measured (blue & magenta, solid) CD spectra at 95°C. Calculated spectra are derived by summation of experimentally measured spectra of the isolated individual components.

Figure 4A: $X(CAG)_6Y \cdot Y'X' + Y'(CTG)_6X' \rightarrow Y'X' + X(CAG)_6Y \cdot Y'(CTG)_6X'$ is shown in the main text

Figure 4B: $X(CAG)_6Y \cdot Y'X' + XY \rightarrow X(CAG)_6Y + XY \cdot Y'X'$

Figure 4C: $Y'(CTG)_6X' \cdot XY + X(CAG)_6Y \rightarrow XY + X(CAG)_6Y \cdot Y'(CTG)_6X'$

Figure 4D: $Y'(CTG)_6X' \cdot XY + Y'X' \rightarrow Y'(CTG)_6X' + XY \cdot Y'X'$

Figure 5B: Measured and calculated excess heat capacity curves for strand displacement of $X(\text{CAG})_6Y \cdot Y'X'$ with XY

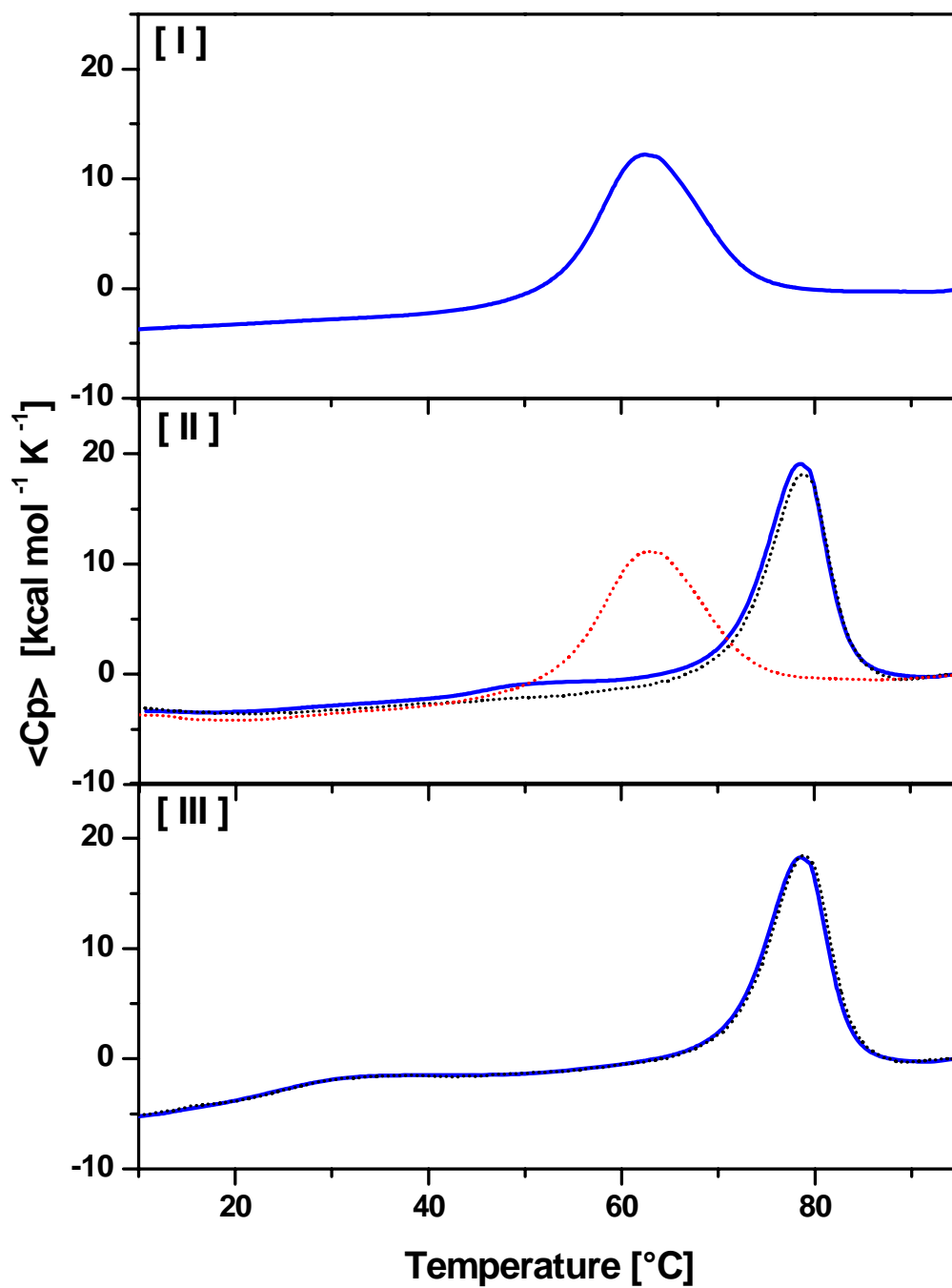


Figure 5C: Measured and calculated excess heat capacity curves for strand displacement of $Y'(CTG)_6X' \cdot XY$ with $X(CAG)_6Y$

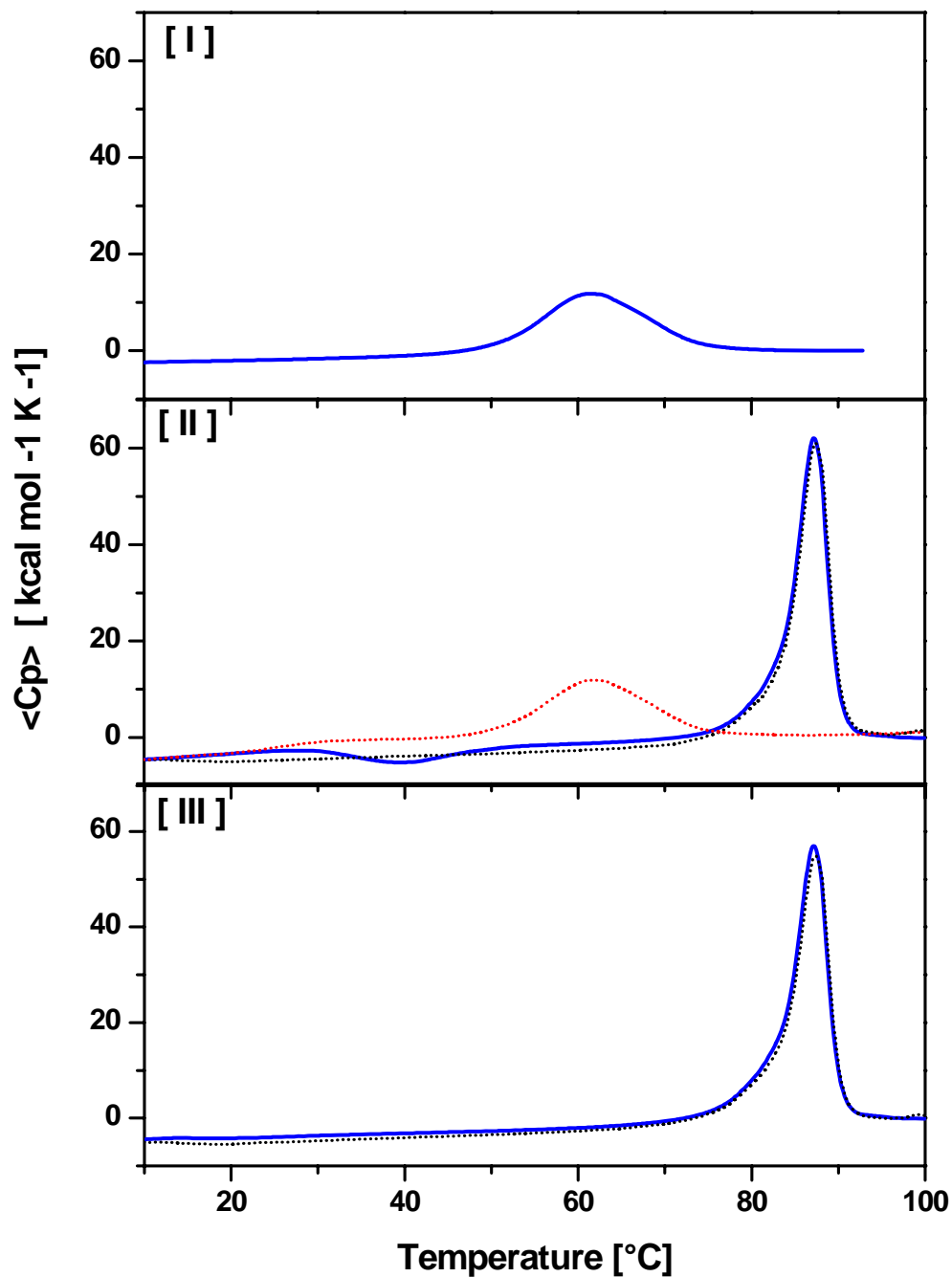


Figure 5D: Measured and calculated excess heat capacity curves for strand displacement of $Y'(CTG)_6X' \cdot XY$ with $Y'X'$

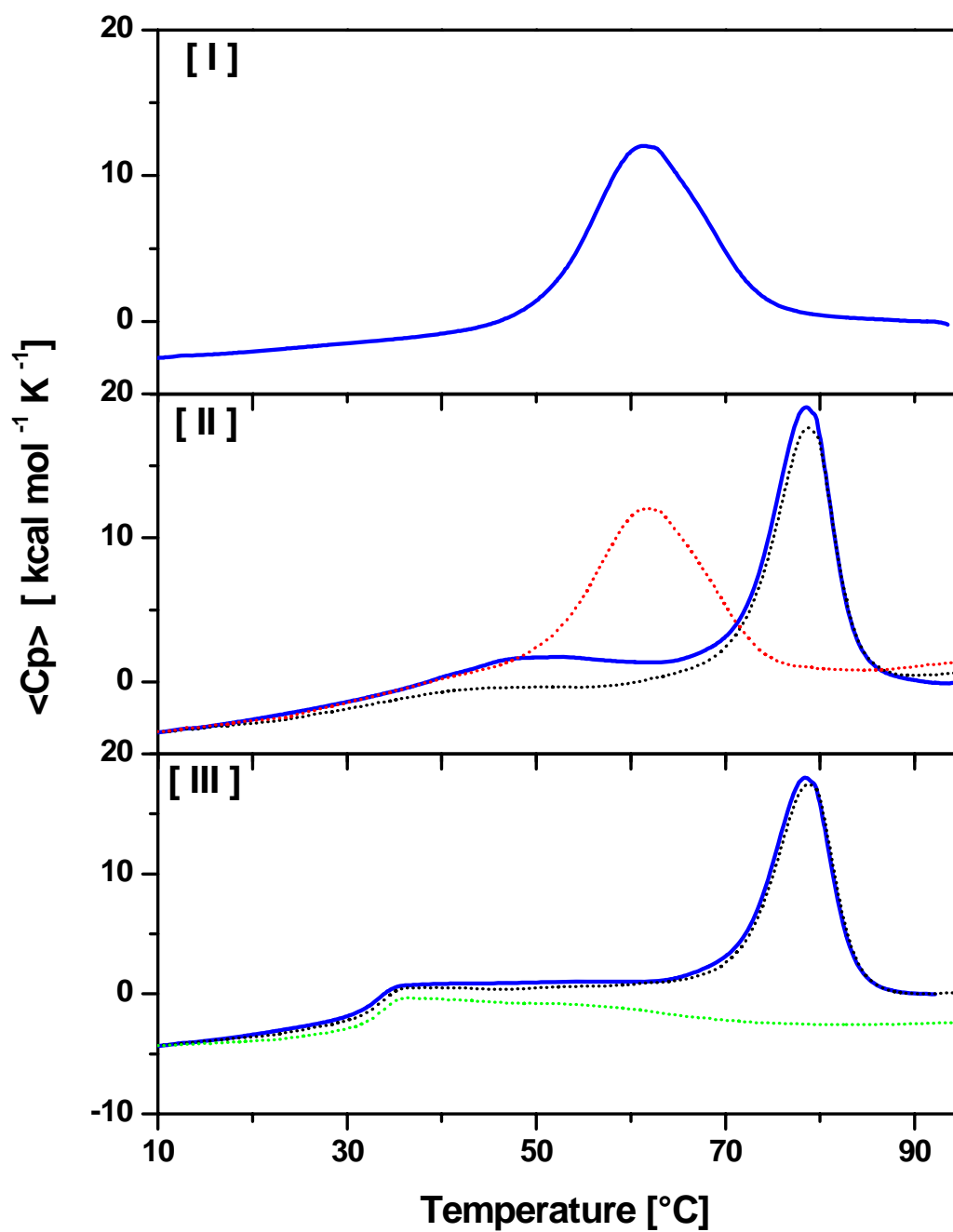


Figure 5 Supplementary – Excess heat capacity curves measured for the 4 different strand displacement reactions shown in **scheme 3** and summarized in **scheme 5**

Panel I always shows the measured excess heat capacity curves of the Ω -DNA's (blue, solid) in isolation. Panel II always shows the measured excess heat capacity curve of a 1:1 mixture of added single strand and Ω -DNA upon first heating (blue, solid line). Also shown are the corresponding calculated excess heat capacity curves of the 1:1 mixture of added single strand and Ω -DNA (red, stippled line), and the calculated excess heat capacity curve of a 1:1 mixture of the added single strand and the resulting duplex (black, stippled line). Panel III always shows the excess heat capacity curve of the 1:1 mixture of single strand and Ω -DNA (blue solid) upon all subsequent heating events. For comparison, the calculated excess heat capacity curves of the displaced single strand and resulting duplex are also shown (black stippled). To facilitate comparison the excess apparent heat capacity at 95°C is set equal for all curves. Panel III in **Figure 5D** also shows the melting of the Y'(CTG)₆X' single strand (green, stippled line) to help dissect the unusual shape of the of the low temperature excess heat capacity curve after strand displacement

Figure 5A: $X(CAG)_6Y \cdot Y'X' + Y'(CTG)_6X' \rightarrow Y'X' + X(CAG)_6Y \cdot Y'(CTG)_6X'$ is shown in the main text.

Figure 5B: $X(CAG)_6Y \cdot Y'X' + XY \rightarrow X(CAG)_6Y + XY \cdot Y'X'$

Figure 5C: $Y'(CTG)_6X' \cdot XY + X(CAG)_6Y \rightarrow XY + X(CAG)_6Y \cdot Y'(CTG)_6X'$

Figure 5D: $Y'(CTG)_6X' \cdot XY + Y'X' \rightarrow Y'(CTG)_6X' + XY \cdot Y'X'$