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Ultrafast Time-resolved Fluorescence Study on Formation and Excitation Dynamics of Human Telomeric and Homo-oligomeric i-Motifs at Neutral pH

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Abstract

Asides from the common double stranded structure, nucleic acids depending on the base content and environmental condition may take many non-canonical secondary structures, including the four-stranded intercalated i-motif structure formed from cytosine-rich sequences that are abundant throughout the human genome.[1,2] The structures of i-motifs are known to be stable at slightly acidic condition, whether they can also be formed at the physiologically relevant neutral pH condition remains elusive and currently one of the frontier issues in the study of non-canonical nucleic acid structures.[3,4]

Herein we present a comparative steady-state and ultrafast time-resolved fluorescence investigation of a homo-oligomeric and human telomeric cytosine-rich sequence at both the acidic and neutral pH.[5] Our study obtained the intrinsic fluorescence and allowed for, the first time, a direct observation of the dynamics of the intrinsic fluorescence from i-motifs formed from the two types of sequences. The results observed formation of the i-motifs at neutral pH but with efficiency of the formation varied strongly from the homo-oligomeric to the human telomeric sequence. Our work also unveiled in real time the excitation dynamics and a multi-channel nonradiative pathway exclusive to the i-motif structures, highlighting a crucial role of the Hoogsteen pairing interaction mediated charge transfer and proton transfer during the course of deactivation. This provides important insight into the interplay between the excitation dynamics and the secondary structure of nucleic acids.

Keywords: Ultrafast spectroscopy, Excitation dynamics, i-Motif DNAs, Charge transfer, Proton transfer

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