Long-Lived Self-Entanglements in Ring Polymers

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The entanglement of ring polymers remains mysterious in many aspects. In this Letter, we use electric fields to induce self-entanglements in circular DNA molecules, which serve as a minimal system for studying chain entanglements. We show that self-threadings give rise to entanglements in ring polymers and can slow down polymer dynamics significantly. We find that strongly entangled circular molecules remain kinetically arrested in a compact state for very long times, thereby providing experimental evidence for the severe topological constraints imposed by threadings.

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The dynamics of polymer melts and concentrated polymer solutions are dominated by the topological constraints imposed by entanglements between chains. Entanglement dynamics is a complex many-body problem and is commonly converted to a tractable one-body problem by replacing constraints exerted by neighboring chains with a confining tube. The tube model conceived by de Gennes, Doi, and Edwards [1–4] in which free ends—the only chain segments capable of randomizing their direction without constriction of the tube—play an essential role in relaxation mechanisms, can effectively describe the dynamics of entangled linear and branched polymers [5–7]. A remaining challenge in polymer physics is to fully understand the dynamics of ring polymers, which have no chain ends and lie outside the framework of the tube model.

Experiments with pure polystyrene rings have shown that ring polymers exhibit self-similar dynamics and follow a power-law stress relaxation with no entanglement plateau [8,9]. Theoretical studies on stress relaxation in ring polymer melts are based on rings assuming conformations of branched, double-folded loops, or "lattice-animal" conformations [10–12]. The lattice-animal model can capture the power-law stress relaxation, but is unable to describe the slow relaxation modes observed in the terminal relaxation regime [8,9,13], leading to the conjecture that inter-ring threadings have an important effect on slowing down ring polymer dynamics [14–17]. Recently, Michieletto and Turner [18] demonstrated through simulations that randomly pinning a small fraction of rings in concentrated ring polymer solutions can limit the center of mass diffusion of the unfrozen rings, thereby immobilizing the rings in a network of threadings and inducing a kinetically arrested state known as a "topological glass." Given sufficient threadings in the system, the glassy dynamics can be observed at any arbitrary temperature and monomer density [18,19].

The main challenge in studying concentrated ring polymer solutions and melts experimentally is that even a small fraction of linear contaminants can alter the dynamics significantly [8,20,21]. The difficulty in studying inter-ring threadings is further compounded by the challenge of defining and identifying such topological interactions both computationally and experimentally. By providing a platform for direct visualization of molecule topology and conformation, single molecule techniques are thus well suited to the investigation of ring polymer dynamics [22]. In this Letter, we use single molecule DNA experiments to study the dynamics of *self-entangled* ring polymers, which serve as a minimal system for studying chain entanglements. We perform experiments to induce self-entanglements in relaxed circular DNA with electric fields [23] and gain understanding of the entangled state by examining the expansion dynamics back to equilibrium. Our results demonstrate experimentally, for the first time to our knowledge, that threadings give rise to entanglements in ring polymers and lead to a slow down in polymer dynamics. We observe that highly self-entangled ring polymers can remain arrested in an entangled state for timescales longer than 2000 relaxation times.

Circular 114.8 kbp DNA molecules were prepared by replication of bacterial artificial chromosome constructs in *Escherichia coli*, followed by extraction, purification and enzymatic treatment [24,25]. DNA samples were stained with YOYO-1 at a base pair to dye ratio of 4:1 and diluted into the experimental buffer containing 4 vol% β-mercaptoethanol, 0.1% 10 kDa polyvinylpyrrolidone and 24% sucrose in 0.5X Tris-boric acid-EDTA (TBE) solution (ionic strength $\approx$ 27 mM, DNA concentration $\ll$ overlap concentration c^*). The molecules were observed in 2 μm tall, 100 μm wide, and $\sim$1 cm long straight channels constructed in polydimethylsiloxane (PDMS). We selected only relaxed circular and linear DNA molecules for experiments. Molecules were compressed using 10 Hz ac (alternating current) square-wave electric fields of varying root-mean-square electric field strengths (E_{rms}) and durations [23].

Each ensemble consists of 20–100 molecules. Single DNA molecules were visualized using an inverted Zeiss Axiovert microscope with a 63X 1.4 NA oil-immersed objective and images were recorded using a Photometrics Prime 95B sCMOS camera. See Supplemental Material (SM) [26] for experimental details.

The radius of gyration (R_g) of the molecules was determined as the square root of the trace of the radius of gyration tensor [31] and measured to be 0.75 ± 0.01 μm and 0.95 ± 0.01 μm (95% confidence interval) for the circular and linear molecules respectively, consistent with values reported by Robertson *et al.* [25] The longest relaxation times, obtained by fitting single-exponential decays to the rotational autocorrelation functions [31], were determined to be $\tau_c = 1.3 \pm 0.1$ s for the circular molecules and $\tau_l = 4.9 \pm 0.2$ s for the linear molecules. The ratio between the linear and circular DNA relaxation times is consistent with that reported by Li *et al.* [32] See SM for details of data processing and statistics.

Since the conformations of the compressed molecules cannot be resolved by conventional fluorescence microscopy, [23,33–35] we investigate the nature of the compressed state by following the expansion of the initially compact molecules after turning off the field (Movie S1). The nature and duration of the expansion process back to equilibrium provides insight into the type and degree of entanglement in the molecule. Figure 1 displays the time evolution of the ensemble average R_g for circular and linear molecules compressed using electric fields of $E_{\mathrm{rms}} = 600$ V/cm and $E_{\mathrm{rms}} = 900$ V/cm for durations of 1 s and 10 s (see SM for error bars). The dashed lines represent the equilibrium average R_g for circular and linear molecules. We make two key observations regarding the expansion process. First, for any given field strength and duration, circular molecules show a faster average expansion back to equilibrium compared to linear molecules (i.e., less time to expand to the dashed line), suggesting a lesser extent of entanglement. This is most evidently seen by comparing the circular and linear ensembles subjected to an electric field $E_{\mathrm{rms}} = 600$ V/cm for 1 s (red). While the average R_g of circular molecules reaches equilibrium within 15 s, the corresponding trace for linear molecules remains below equilibrium after 60 s, a similar timescale taking into consideration the ratio of relaxation times. Second, we note that the average R_g of circular molecules subjected to an electric field $E_{\mathrm{rms}} = 900$ V/cm for 10 s (orange) does not attain equilibrium even after 60 s ($\sim 50\ \tau_c$). The slow ensemble average expansion shows that circular molecules, like linear molecules, can become self-entangled.

To gain further insight into the nature of the self-entangled state, we probe the expansion dynamics of individual molecules. Tang *et al.* [23] categorized the expansion of linear molecules into two pathways with distinct dynamics: type I represents a continuous increase

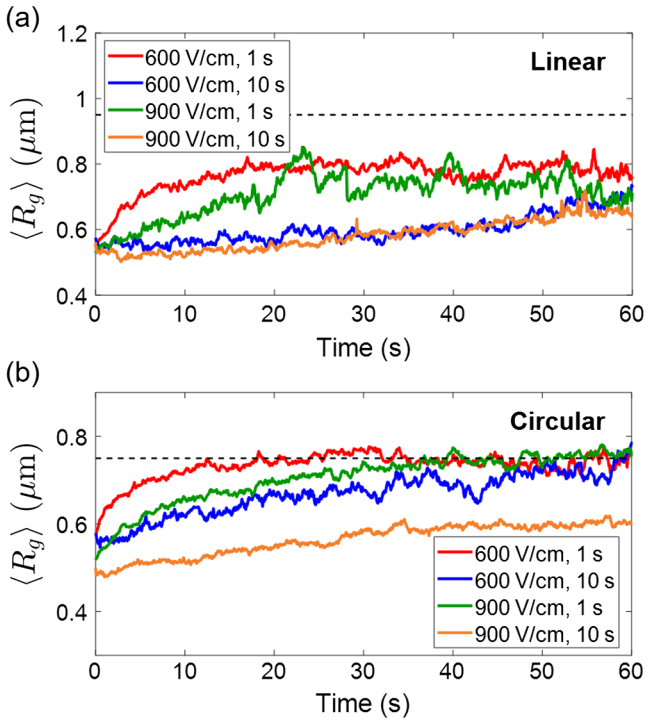

FIG. 1. Ensemble average expansion of (a) linear and (b) circular DNA molecules compressed by a 10 Hz ac square-wave electric field of various field strengths and durations. The dashed line indicates the equilibrium average radius of gyration $\langle R_g \rangle_{\mathrm{eq}} = 0.95$ μm for linear molecules and $\langle R_g \rangle_{\mathrm{eq}} = 0.75$ μm for circular molecules.

in R_g following compression, while type II characterizes a three-stage expansion process. In the first stage of type II expansion, the molecule remains arrested in a compact globular state, until a nucleation event in which chain ends disentangle from the globule initiates the second stage. The second arrested stage involves untangling the intramolecular knots, with complete disentanglement and return to equilibrium being marked by the third stage. Type I expansion is attributed to unentangled or weakly entangled molecules, and type II expansion to intramolecular knots induced by the electric field, with chain ends playing a pivotal role in the relaxation process. We can also broadly categorize the expansion of circular molecules into type I and type II expansion. Figure 2 shows representative individual traces of R_g vs time and snapshots of circular molecules that exhibit type I and type II expansion (see SM for more individual traces and proportion of molecules undergoing each type of expansion). The main difference between type II expansion for a linear and circular molecule is that circular molecules typically undergo a two-stage expansion process with a single arrested stage. This is likely due to the lack of chain ends that are responsible for initiating the second arrested state in the expansion pathway of self-entangled linear molecules [23]. Nonetheless, the existence of an arrested state affirms the presence of self-entanglements in circular molecules.

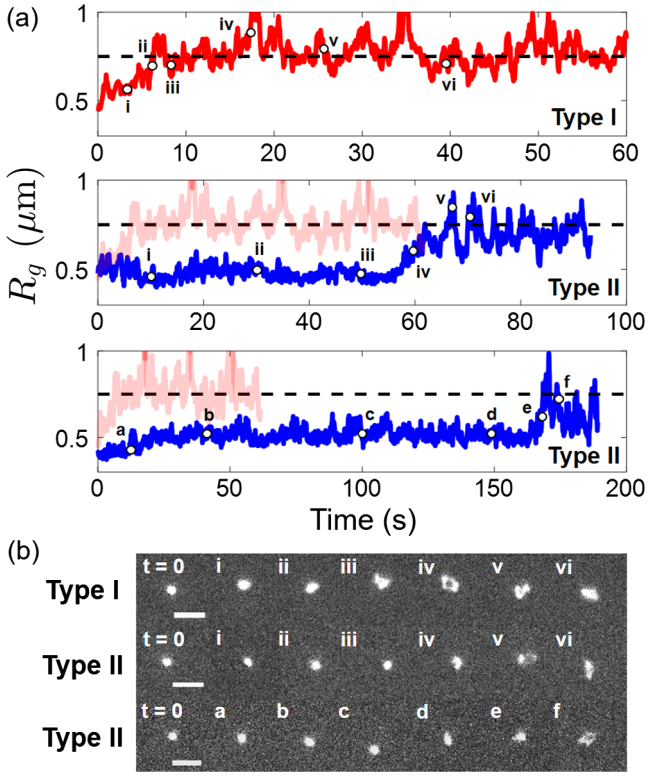

FIG. 2. Expansion of individual circular molecules compressed by ac square-wave electric fields. (a) Representative individual traces of R_g vs time for different expansion pathways. The dashed lines indicate the equilibrium average radius of gyration $\langle R_g \rangle_{\mathrm{eq}} = 0.75\ \mu$m. The type II traces are underlaid by the representative type I trace to facilitate comparison of timescales. (b) Snapshots of circular DNA conformation during expansion corresponding to the time points labeled in (a). Scale bars represent 5 μm.

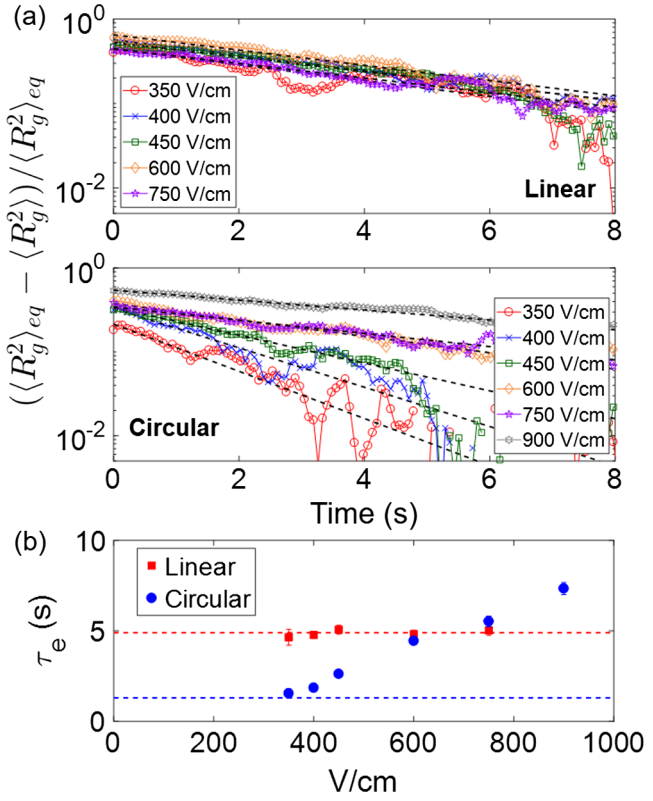

FIG. 3. (a) Normalized mean square R_g as a function of time for linear and circular molecules that are compressed with various field strengths for 1 s and exhibit type I expansion. The dashed lines are single exponential fits to the data. (b) Time constants extracted from single exponential fits to data in (a) as a function of field strength for circular (blue circle) and linear (red square) molecules. The dotted lines represent the relaxation times $\tau_c = 1.3$ s (blue) and $\tau_l = 4.9$ s (red) for circular and linear molecules respectively. Error bars represent 95% confidence interval. If not visible, the error bar is smaller than the symbol.

The question that then arises is, what do self-entanglements in circular molecules entail? While linear molecules self-entangle by passing chain ends through loops along the same chain to form intramolecular knots, [23,33] circular molecules cannot change topological states. Given that our system comprises individual unknotted circular molecules, the only way self-entanglement can occur is via *self-threadings*—double-folded segments opening up and penetrating double-folded segments on the same chain [36]. We imagine that a circular molecule that threads itself multiple times can give rise to an entangled state (see SM for examples of macroscale self-threaded circular chains). It should be noted that our definition of self-threadings encompasses known entanglements, such as slipknots, that might form on a circular molecule [32]. Although it is possible that circular molecules also self-entangle by forming double-folded knots, such an entangled configuration requires both halves of the molecule to align into a double-folded linear structure and is energetically unfavorable [37].

We first examine in detail the dynamics of type I expansion. The expansion dynamics of unentangled linear DNA have been shown to be governed by the longest relaxation time of the molecule, such that $(\langle R_g^2 \rangle_{\mathrm{eq}} - \langle R_g^2 \rangle) \sim \exp(-t/\tau)$ [23]. We consider the expansion of circular and linear molecules that are subjected to 1 s pulses of varying field strengths and undergo type I expansion. Figure 3 shows the normalized mean square R_g as a function of time for linear and circular molecules, and the time constants τ_e extracted from single exponential fits to the data (see SM for error bars). While the values of τ_e for linear molecules are consistent with the relaxation time τ_l over a range of field strengths, circular molecules exhibit a dependence of τ_e on field strength. As seen from Fig. 3(b), the greater the field strength used to compress circular molecules, the longer the timescale associated with expansion to equilibrium. The value of τ_e for circular molecules is bounded below by the relaxation time τ_c.

We highlight that the proportion of linear molecules that exhibit type I expansion at any given field strength is smaller than that of circular molecules. For example,

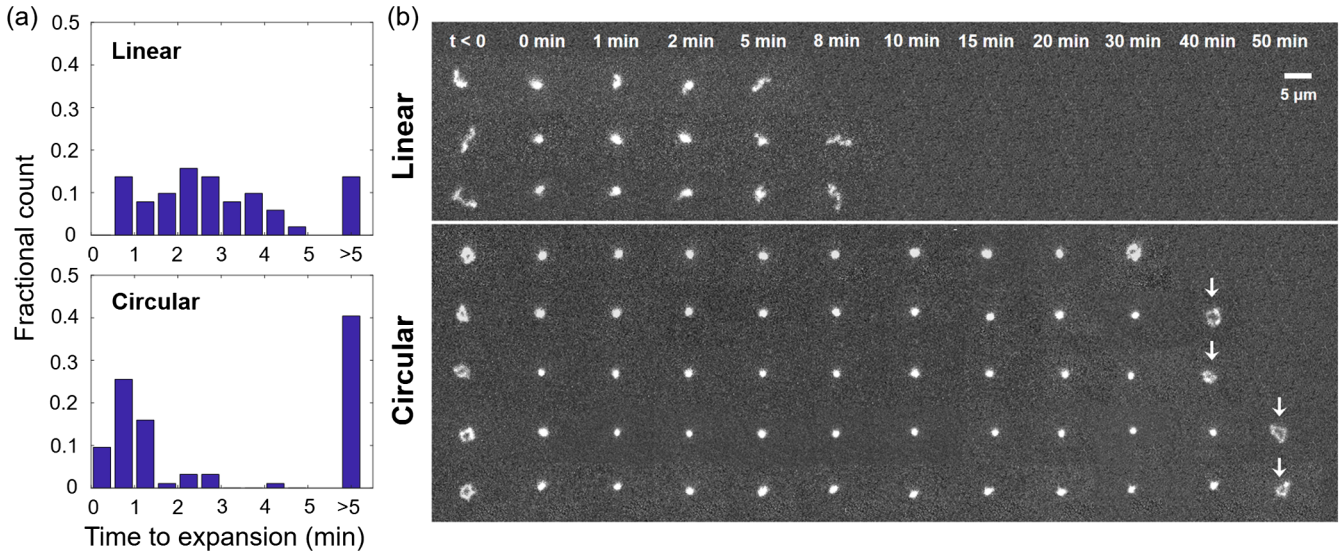

FIG. 4. (a) Distributions of time required to expand back to equilibrium for the ensemble of linear and circular molecules subjected to an electric field $E_{\mathrm{rms}} = 900$ V/cm for 10 s. (b) Snapshots of three linear (top) and five circular (bottom) molecules expanding back to equilibrium over time following compaction by an electric field $E_{\mathrm{rms}} = 900$ V/cm for 10 s. The electric field is turned off at time $t = 0$. Four of the five circular molecules expand back to equilibrium only after a 900 V/cm, 0.5 s pulse is applied (indicated by arrows).

among the molecules subjected to an electric field of $E_{\mathrm{rms}} = 600$ V/cm for 1 s, 40% of the linear ensemble exhibit type I expansion compared to 86% of the circular ensemble. This points to a different entanglement mechanism between the linear and circular molecules. Since circular molecules undergo type I expansion characterized by a field-dependent timescale, we postulate that compressed circular molecules that exhibit type I expansion can also be self-threaded. The self-threadings are not strong enough to arrest the molecule's expansion to equilibrium, but sufficient for slowing down relaxation dynamics. The greater the electric field strength, the more self-threadings induced in the molecules, hence the longer the relaxation process. The expansion timescale for circular molecules compressed at the highest field strength of $E_{\mathrm{rms}} = 900$ V/cm for 1 s is $\sim 5\ \tau_c$, an indication of the degree to which self-threadings can slow down polymer relaxation. This observation agrees well with computational studies that report the slowing down of polymer dynamics with inter-ring threadings, with such threadings having survival times much longer than the relaxation time of the polymer [15,16].

Next, we consider the dynamics of type II expansion, focusing specifically on the ensemble of molecules subjected to an electric field $E_{\mathrm{rms}} = 900$ V/cm for 10 s, as this contains the largest proportion of molecules that exhibit type II expansion. To further investigate the entangled states, we examine the times required for expansion to equilibrium (Fig. 4(a)). The distribution of expansion times for linear molecules is relatively uniform and can be attributed to randomly occurring nucleation events that initiate the unthreading of intramolecular knots [23]. On the other hand, the distribution of expansion times for circular molecules appears to be bimodal, with most molecules either expanding to equilibrium within 90 s ($\sim 60\ \tau_c$) or remaining arrested for longer than 5 min ($\sim 230\ \tau_c$). As discussed previously, circular molecules that exhibit type I expansion are not necessarily unentangled, but not entangled to the extent of arresting the expansion of the molecule. The bimodal distribution observed thus suggests that there is a certain threshold past which self-threadings in a circular molecule can result in a long-lived arrested state.

We can track molecules arrested in an entangled state beyond the typical observation period of a few minutes, until expansion to equilibrium is observed. Figure 4(b) shows snapshots of several linear and circular molecules that were observed to take the longest times to relax to equilibrium (see SM for additional snapshots of circular molecules). The linear molecules observed all expanded to equilibrium within 8 min ($\sim 100\ \tau_l$), similar to timescales reported in unknotting studies with linear DNA by our group [38,39]. The circular molecules, however, persisted in a compact, arrested state for >30 min. We note that four of the five circular molecules expanded to equilibrium only after being perturbed by a 900 V/cm, 0.5 s pulse, meaning that the molecules would have remained arrested for timescales longer than the observed 40–50 min ($\sim 2000\ \tau_c$). The reproducible finding of circular molecules relaxing to equilibrium shortly after a short pulse from the electric field is indication that the molecules do not form double-folded knots; intramolecular knots simply tighten and become more difficult to disentangle upon additional compaction.

Although circular molecules can remain in long-lived arrested states for long times, expansion to equilibrium occurs relatively quickly, typically over ~60 s. We postulate

that the disentanglement of self-threaded circular molecules is an all-or-none process that is initiated by the removal of one or more critical threadings. The field-induced unthreading is then due to the removal of such self-threadings, which consequently allows the molecule to relax to equilibrium. Our hypothesis is consistent with the conjecture put forth by Michieletto and Turner [18] that threadings form a hierarchical structure, such that the system is arrested by an ordered sequence of constraints. The system can thus relax only when the threading at the top of the hierarchy, or the critical threading, is released. This process bears similarity to the process in which string figures, such as Cat's Cradle and Jacob's Ladder, unravel completely upon release of the correct loop [40].

The very long times that circular molecules can remain entangled shows that self-threadings present severe topological constraints on the relaxation process and can slow down polymer dynamics dramatically. Such long-lived states are not observed with entangled linear molecules. While linear molecules always have chain ends to facilitate disentanglement, circular molecules have no standard mechanism for releasing threadings when strongly entangled. Although the entanglements were introduced by a non-equilibrium method in our experiments, the dynamics observed are solely a consequence of the number of entanglements, and we can imagine concentrated systems of ring polymers also containing numerous entanglements.

In this Letter, we studied the expansion dynamics of circular DNA entangled by electric fields. We demonstrated experimentally that circular molecules entangle via self-threadings, which can dramatically slow down polymer dynamics. Circular molecules are less easily driven into an arrested, entangled state compared to linear molecules. However, strongly entangled circular molecules exhibit long-lived compact states that do not relax on timescales greater than ~2000 relaxation times.

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