附件一

行政院國家科學委員會補助專題研究計畫 ☑ 成果報告 □期中進度報告

（計畫名稱）: 多種材料之彈性性質及生物物理的一些課題

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執行單位：淡江大學物理系

中　華　民　國　97　年　9　月　28　日

中文摘要及中文關鍵詞

關鍵詞：彈性性質，細長桿，生物高分子，序列相關，自發曲率，螺旋，構型，穩定性。

中文摘要：

由於其成功的描述了一些微觀物體(如碳奈米管和生物高分子)的構形與力學性質，近年來細長彈性桿的性質之研究受到了廣泛的重視。同時，DNA 的性質在相當程度上取決於其鹼基對的排列順序，但是這種序列相關性的理論研究迄今仍不多見。我們研究了序列相關的自發曲率對半柔軟的生物高分子物理性質的影響。我們嚴格證明了如果隨機的自發曲率只有短程關聯的話，其效應可以用其平均曲率以及適當修正後的駐留長度來取代，而無需考慮其具體的分布。我們導出了自發曲率只有短程關聯的二維半柔軟的生物高分子在無外力時的分布函數的解析表達式。我們求得的二維半柔軟的生物高分子之彎摺角的分布形狀，結果與具鹼基對長程關聯的雙股 DNA 的實驗結果符合得很好。另外，我們還證明了對很長的半柔軟的生物高分子而言，其序列相關的隨機駐留長度可以用其平均值來取代。然而對短的半柔軟的生物高分子而言，其物理性質將與駐留長度的具體分布有關，因此無法用幾個簡單的巨觀量來描述。

我們研究了具均勻自發曲率的二維細長彈性桿的力學性質，推導出了其形狀方程，並求得形狀方程的解析解。我們發現在強外力作用下並具有正的初始方位角時，二維細長桿的方位角將是弧長的雙值函數。在其他條件下方位角則是弧長的單值函數。在有限溫度下，我們推出了在強拉力和弱拉力時二維細長桿之外力與伸長之關係的解析解。我們發現當自發半徑與駐留長度之大小可比時，自發曲率對彈性性質將有重要影響。

我們推導出了細長彈性桿在外力和外力矩作用下的一組用尤拉角表示的形狀方程，並求得了在力學實驗中形成螺旋桿所需要的特殊邊界條件。我們證明了由於其特殊邊界條件，除了在一些特殊情況下，細長彈性桿沒有近螺旋解。我們詳細研究了螺旋桿在不同條件下的彈性行為。我們的解析結果證明了當固定外力矩及具有適當的彈性係數時，在拉力和壓力下螺旋桿的伸長都可能出現不連續的跳躍。我們的結果定量地解釋了化學成分確定的脂質液體(chemically defined lipid concentrate，CDLC)中受拉伸的螺旋的實驗結果，並預期在固定外力時，受扭曲的螺旋的伸長亦可能出現不連續的跳躍。

Jarzynski 等式(Jarzynski equality，JE)能利用從一個平衡態到另一個平衡態的過程中外力所做的功來計算這兩個平衡態間的自由能之差，在平衡態與非平衡態間架設了一條橋樑。但我們發現某些特殊系統可能不滿足 Jarzynski 等式的條件，因此 Jarzynski 等式有可能需要拓廣。我們的計算機模擬證實了我們的推測。我們亦提出了一個可能用於證實我們的推測的實驗。

英文摘要及英文關鍵詞

Keywords： elasticity, filament, biopolymer, sequence dependence, spontaneous curvatures, helix, conformation, stability.

Abstract：

The study of a filament has increasing importance due to it can account for elasticity of some microscopic objects, from carbon nanotubes to biomaterials. Moreover, It has been known that the local sequence-dependent properties of biopolymers play crucial and active role in many biological processes. We have studied the effects of sequence-dependent spontaneous curvatures for a semiflexible biopolymer with short-range correlation in spontaneous curvatures. We showed exactly that for such a system, the effects of disorder on spontaneous curvatures can be incorporated into a model with well-defined mean spontaneous curvature and renormalized persistence length. We obtained the exact expression for the distribution function of a two-dimensional semiflexible biopolymer with short-range correlation in spontaneous curvatures and free of extern force. Our analytical results for the bending profiles agreed well with experimental observations for dsDNA with long-range correlation in base pairs. On the other hand, we showed that for a long biopolymer with large mean persistence length, the sequence-dependent persistence lengths can be replaced by their mean. However, for a short biopolymer or for a biopolymer with small persistence lengths, inhomogeneity in persistence lengths tends to make physical observables very sensitive to details and therefore less predictable. Furthermore, we investigated the mechanical properties of a two-dimensional filament with spontaneous curvature and under uniaxial applied force. We derived the equation that governs the stable shape of the filament and obtain analytical solutions for the equation. We found that for a long filament with positive initial azimuth angle and under large stretching force, the azimuth angle must be a two-valued function of arclength. Otherwise, the azimuth angle is a monotonic function of arclength. At finite temperature, we obtained closed-form expressions on the force-extension relation for a filament at low force and for a long filament under strong stretching force. Our results showed that the effect of a nonvanishing spontaneous curvature may become important when the spontaneous radius is comparable to the persistence length of the filament.

Meanwhile, we derived the shape equations in terms of Euler angles for a uniform elastic rod. We found that due to the special requirements on the boundary conditions, a static slightly distorted helix cannot exist in this model except for some special cases. We studied the elasticity and stability of a helical filament under different conditions. We showed analytically that the extension of a helix may undergo a one-step sharp transition when we fix the torque. This agrees quantitatively with experimental observations for a stretched helix in a chemically defined lipid concentrate (CDLC). We predicted further that the extension of a twisted helix in CDLC may also undergo a one-step sharp transition. We found that a negative twist tends to destabilize a helix.

Jarzynski equality (JE) can be used to extract the free energy difference between two equilibrium states from the non-equilibrium work performed on the system in the process between these two states. We pointed out that the crucial condition in the derivation of the JE from the fluctuation theorem is that the time integral of the phase space contraction factor is exactly expressed as the entropy production resulting from the heat absorbed from the thermal bath by the thermal system. We suggested that for the system violating this condition, a more general form of JE may exist. This conjecture was supported by our computer simulation on a simple system.

內容目錄

一、 前言(Introduction and Background)

The topics on elasticity of various materials are basically the continuous and extension of my previous three-year plan (2002-2005).

The study of a long thin rod has a long history in mechanics and engineering dating back to Euler and Lagrange. It has increasing importance due to the recent experiments and theories revealed that it can account for elasticity of some microscopic objects, from carbon nanotubes to biomaterials. For instance, The wormlike chain model which regards a rod as an inextensible chain with certain bending rigidity but with vanishing radius of the cross section, and wormlike rod chain model which regards a rod as a chain with certain bending rigidity and spontaneous twist and circular cross section, have been applied to predict biopolymer's [such as double-stranded DNA(dsDNA)] elastic response successfully up to moderate force and torque. The conditions to form a helix from a rod and its relevant stability and elasticity are in particular interesting topics since the helix is one of most simple filamentary structures found in nature. It is more or less a surprise that after several century studies, a picture of the elasticity for a helical rod is still far from complete. It was reported that a helical rod is not stable under stretching, and may undergo a multi-step sharp transition of extension, from a free-standing helix to a strongly distorted helix. However, recent experiments for a helix in the chemically defined lipid concentrate (CDLC) observed a one-step reversible sharp transition of extension from an almost perfect helix, to an almost straight line. Whether the elastic model can describe such observations is therefore a very intrigue question. **In my previous plan, we had obtained some preliminary results in this topic.** We have derived the general shape equations in terms of the Euler angles (θ, ϕ and ψ) for a uniform rod with spontaneous torsion and curvatures. We found that to form a helix, ψ must be a constant. Furthermore, our analytically basic calculations showed that a helical rod may undergo a one-step reversible discontinuous transition under a stretching force, agree quantitatively with the experimental observations for a helix in the chemically defined lipid concentrate. We found that the larger the twisting rigidity, the larger the jump in the extension. On the other hand, the effect of torque is dependent on the value of the spontaneous torsion. In contrast, increasing spontaneous torsion encourages the continuous variation of the extension. However, our stability analysis demonstrated that a static helix is in general unstable under small force. **In this plan, we provided a full picture for the elasticity of a helical filament under different conditions. We also provided a better stability criterion so solved the stable problem of a helix under low force.**

dsDNA is a double-helical biopolymer in which two chains of complementary nucleotides wind around a common axis to form a double-helical structure. As the genetic material, dsDNA molecule is of fundamental importance in living organisms, and therefore a thorough understanding of dsDNA molecule is a great challenge of our time. Despite definite progress made recently, our knowledge of DNA is still far from complete, especially in theoretical aspect concerning its conformations, deformation, replication, combination and denaturation. The sequence-specific property of dsDNA affects to a considerable extent many important biological processes involving interactions between DNA and proteins, especially the recognition of specific nucleotide sequences by various regulatory proteins, (for instance, DNA/RNA polymerases, *lac* repressor, TATA box-bonding protein, p53 protein, etc.) and the subsequent binding of such proteins on these recognized sequences. Recent progresses in experimental techniques such as laser or magnetic tweezers and atomic force microscopy, make it possible to manipulate and observe single biomolecule directly, and so can make a better comparison between theoretical expectations and experimental observations. In theoretical studies, a semiflexible biopolymer is often modelled by a filament. Based on the elastic model, two effects of sequence-disorder have to be considered. First, the local structure yields variations of the bending rigidity along the filament, and results in the s-dependent persistence length $l_p(s)$, where s is the arc length. **In this plan, we presented a general proof that for a long semiflexible biopolymer, such as dsDNA, without long-range correlation (LRC) in $l_p(s)$, this effect can be accounted by a replacement of the $l_p(s)$ by its proper average. However, for a short dsDNA, inhomogeneity in $l_p(s)$ tends to make physical observables divergent.** Second, the structural inhomogeneity of the biopolymer can be characterized by the sequence-dependent spontaneous curvature $c(s)$. It has been demonstrated that, for dsDNA, the effect of $c(s)$ is dependent on the degree of correlation in base pairs (bp). **In this plan, we showed rigorously and generally that without correlation or the correlation is short-range, the effect can be also reduced into a renormalization of the uniform persistence lengths.** However, with LRC, the simple correction to the uniform persistence length is invalid because the biopolymer develops a macroscopic intrinsic curvature. On the other hand, it was demonstrated by computer simulations that the mean spontaneous curvature, rather than the details of its distribution, determines many physical properties for a three dimensional semiflexible biopolymer with short-range correlation (SRC) in $c(s)$. **In this plan, we provided a general and rigorous proof so confirmed the conclusion obtained from computer simulations.** Moreover, at first sight, the SRC in $c(s)$ should result from the SRC in sequences, so that the above conclusion would have little significance. However, this is not always true. For instance, for a homopolymer, the correlation in sequences is 100%, but it can be described by a constant spontaneous curvature so can be regarded as no correlation in $c(s)$, since it corresponds to the limit case of Gaussian distribution with vanishing variances. **In the more general case, LRC in sequences tends to make neighbor sequences have similar bending so to develop a macroscopic intrinsic curvature, and the local $c(s)$ may have only a small random deviation from its mean, it in turn leads to the Gaussian distribution for $c(s)$ or $c(s)$ has SRC, at least in the first approximation, as we have demonstrated in this plan**. Moreover, the experimental observations are often conducted in two dimensional environment. Consequently, **it is a significant topic to study the effects of sequence-dependent spontaneous curvatures on the properties of a two-dimensional semiflexible biopolymer, and this was also one of the focus in this plan**.

二、 研究目的(Goal)

To have a comprehensive understanding of the conformal and elastic properties of biopolymers, such as DNA and protein. To develop the elastic theory for a filament.

三、 研究方法(Methodology)

We used both analytical approaches and computer simulations to explore the properties of biopolymers. Our analytical approaches are based on elastic model of filament, statistical mechanics and technique of path integral. We also constructed a sequence-dependent model for two-dimensional semiflexible biopolymer and used Brownian dynamics to simulate the conformal and mechanical properties of the biopolymers.

四、主要結果與討論（Main results and discussions）

1. We studied the effects of sequence-dependent spontaneous curvatures for a two-dimensional semiflexible biopolymer with short-range correlation in spontaneous curvatures. We showed exactly that for such a system in constant extension ensemble, the effects of disorder on spontaneous curvatures can be incorporated into a model with well-defined mean spontaneous curvature and renormalized persistence length. We obtained the exact expression for the distribution function. Our analytical results for the bending profiles agreed well with experimental observations for dsDNA with long-range correlation in base pairs.
2. Using path integral technique, we showed exactly that for a semiflexible biopolymer in constant extension ensemble, no matter how long the polymer and how large the external force, the effects of short range correlations in the sequence-dependent spontaneous curvatures and torsions can be incorporated into a model with well-defined mean spontaneous curvature and torsion as well as a renormalized persistence length. Moreover, for a long biopolymer with large mean persistence length, the sequence-dependent persistence lengths can be replaced by their mean. However, for a short biopolymer or for a biopolymer with small persistence lengths, inhomogeneity in persistence lengths tends to make physical observables very sensitive to details and therefore less predictable.
3. We studied the mechanical properties of a two-dimensional filament with spontaneous curvature and under uniaxial applied force. We derived the equation that governs the stable shape of the filament and obtain analytical solutions for the equation. We found that for a long filament with positive initial azimuth angle and under large stretching force, the azimuth angle must be a two-valued function of arclength, decreases first and then increases with increasing arclength. Otherwise, the azimuth angle is a monotonic function of arclength. At finite temperature, we obtained closed-form expressions on the force-extension relation for a filament at low force and for a long filament under strong stretching force. Our results showed that the effect of a nonvanishing spontaneous curvature may become important when the spontaneous radius is comparable to the persistence length of the filament.
4. We derived the shape equations in terms of Euler angles for a uniform elastic filament with isotropic bending rigidity and spontaneous curvature, and study within this model the elasticity and stability of a helical filament under uniaxial force and torque. We found that due to the special requirements on the boundary conditions, a static slightly distorted helix cannot exist in this system except in some special cases. We showed analytically that the extension of a helix may undergo a one-step sharp transition when we fix the torque. This agrees quantitatively with experimental observations for a stretched helix in a chemically defined lipid concentrate (CDLC). We predicted further that under twisting, the extension of a helix in CDLC may also exhibit similar behavior. We found that a negative twist tends to destabilize a helix. Finally, we found that there is not sharp transition for the extension of a helical filament when one uses the supercoiling degree as independent variable.

5. We constructed a model to simulate the elasticity of two-dimensional semiflexible biopolymers with constant spontaneous curvature by Browian dynamics computer simulation and obtained some preliminary results.
6. Above results make some significant progresses on the conformal and elastic properties of semiflexible biopolymers, such as DNA and some proteins. However, there are still some unsolved problems which are worth a further investigation. For instance, effects of looping and excluded volume effects on the mechanical properties of semiflexible biopolymer with spontaneous curvature, as well as the elastic response to the moderate force at finite temperature for a semiflexible biopolymer are not yet well understood. Moreover, our knowledge on the thermal effect on the mechanical properties of a helical filament is far from complete. Similarly, the dynamical behavior of the above systems is still an open question. Our works provided a foundation to approach these intrigue problems.
7. Jarzynski equality (JE) can be used to extract the free energy difference between two equilibrium states from the non-equilibrium work performed on the system in the process between these two states. The crucial condition in the derivation of the Jarzynski equality (JE) from the fluctuation theorem is that the time integral of the phase space contraction factor is exactly expressed as the entropy production resulting from the heat absorbed from the thermal bath by the thermal system. For the system violating this condition, a more general form of JE may exist. Our computer simulation on a simple system supports this conjecture. We expected that with the aid of the nanotechnology, one can justify our conjecture in experiment.
8. A novel technique, gold nanoparticle-assisted single-drop microextraction (SDME) combined with atmospheric pressure matrix-assisted laser desorption/ionization mass spectrometry (AP-MALDI-MS) for the identification of peptides has been described. The SDME of peptides from aqueous solution was achieved using gold nanoparticles prepared in toluene as the acceptor phase. A simple phenomenon of isoelectric point (pI) of the peptides has been utilized successfully to extract the peptides into a single drop of nanogold in toluene. After extraction, a singledrop nano gold solution was directly spotted onto the target plate with an equal volume of matrix, α-cyanohydroxy cinnamic acid (α-CHCA) and analyzed in AP-MALDI-MS. The parameters, such as solvent selection, extraction time, agitation rate, and pH effect, were optimized for the SDME technique. Using this technique, in aqueous solution, the lowest concentration detected for Met- and Leuenkephalin peptides was 0.2 and 0.17 *í*M, respectively. In addition, the application of this technique to obtain the signal for the selected peptides in a mass spectrum in the presence of matrix interferences such as 1% Triton X-100 and 6.5 M urea has been showed. The application was extended to identify the peptides spiked into urine.

五、計畫成果自評(self-evaluation)

原計畫在三年左右的時間在國際期刊上發表至少 7 篇研究論文。自從 94 年 8 月 1 日本計劃啟動以來，我們已經在 SCI 期刊上發表了 5 篇論文，另有 1 篇相關研究論文已向 SCI 期刊投稿，還有 1 篇論文已基本完成，正處於寫作修改階段。期刊論文的數目雖然只達到原計畫的最低目標，但注意到其中有兩篇論文的篇幅較長(參考文獻中的期刊論文之第二，三兩篇論文) ，分別為 12 與 18 頁。另外，我們還在國內外研討會上發表了 16 篇研究報告，並指導 3 位碩士班研究生完成了畢業論文。因此總而言之，進展尚屬差強人意。

原計畫要用三年左右的時間研究: 1. 細長桿的彈性性質; 2. 研究 DNA 微環構型及其穩定性以及 DNA 局域變性的序列相關性，以及蛋白質摺疊的動力學過程; 3. 計算非定型態半導體材料的彈性系數，由此得出非中心力系統在臨界滲透區的軟模相變現象的基本圖像。

上述第一項內容進展順利，已經在 SCI 期刊上發表了 3 篇論文，有 1 篇論文已投到 SCI 期刊，另有一篇相關論文的主要工作已基本完成，正處於撰寫階段。

上述第二項內容中的 DNA 微環構型及其穩定性與第一項內容實際上是相關的，因此進展亦算順利。但其餘內容則因為缺乏足夠的時間與良好訓練的人力而處於資料收集與分析階段。由於是在私立大學，本人的教學負擔相當繁重，平均每週有 10 個鐘點左右的課，因此不可能親自從事需要大量處理收集資料以及大運算量的工作。如果有較高質量的研究生或博士後，應該可以克服這個問題。因此，缺乏經過良好訓練的人力，以致不得不放棄一些很有意義但又有時間緊迫感的題目，是近年來本人研究工作中的最大缺憾。

上述第三項內容已有一篇相關論文的主要工作已基本完成，正處於撰寫階段。而主要工作所需要的計算機模擬程式已經完成，正進入調試階段。但這部分內容亦因人力問題而進展緩慢。

另外有兩篇研究論文不在原計畫目標內。其中有一篇指出非平衡統計力學中的 Jarzynski 等式有可能需要拓廣。第二篇描述了一種利用奈米金粒子幫助，並整合質譜儀及毛細管電泳儀的一滴溶劑微萃取法的新技術。這兩篇論文主要是涂展春與蘇德喜兩位博士後研究員的貢獻，及與本校化學系老師的合作成果。兩位博士後的到來對本人有相當大的幫助。

總而言之，本計畫的方向及其進展基本上符合預期。

六、參考文獻(publications)

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5. **Zicong Zhou**, "Elasticity and stability of a helical filament", International Conference on Recent Advances in the Interdisciplinary Applications of Statistical Physics, 20-22 September 2006, Beijing
6. **Zicong Zhou**, Béla Joós, and Pik-Yin Lai, "Elasticity of a helical filament under twisting", the Fifth Joint Meeting of Chinese Physicists Worldwide (OCPA5 Conference), June 27-30, 2006, Taiwan University, Taipei, Taiwan
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III. **國內研討會論文**:

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3. 碩士班學生: 何柏樺,「排斥體積效應對具有均勻自發曲率的二維半柔軟生物高分子的彈性性質之影響」(Effect of excluded volume on the elasticity of two-dimensional semiflexible biopolymers with constant spontaneous curvature)。2008 年 6 月畢業