行政院國家科學委員會專題研究計畫　成果報告

生物材料之物理及非晶態半導體材料的軟模相變之研究

(3/3)

計畫類別：個別型計畫
計畫編號：NSC93-2112-M-032-006-
執行期間：93 年 08 月 01 日至 94 年 10 月 31 日
執行單位：淡江大學物理學系

計畫主持人：周子聰

計畫參與人員：林方庭，何國敏，江正雄，詹政諱，徐鵬英，陳彥銘，陳守鍊，曾華民，林佩怡，劉毓凡，吳坤達，蘇德喜

報告類型：完整報告
報告附件：出席國際會議研究心得報告及發表論文
處理方式：本計畫可公開查詢

中　華　民　國 94 年 12 月 7 日

國科會專題研究計畫成果報告撰寫格式

一、說明

國科會基於學術公開之立場，鼓勵一般專題研究計畫主持人發表其研究成果，但主持人對於研究成果之內容應負完全責任。計畫內容及研究成果如涉及專利或其他智慧財產權、違異現行醫藥衛生規範、影響公序良俗或政治社會安定等顧慮者，應事先通知國科會不宜將所繳交之成果報告蒐錄於學門成果報告彙編或公開查詢，以免造成無謂之困擾。另外，各學門在製作成果報告彙編時，將直接使用主持人提供的成果報告，因此主持人在繳交報告之前，應對內容詳細校對，以確定其正確性。

本格式說明僅為統一成果報告之格式，以供撰寫之參考，並非限制研究成果之呈現方式。精簡報告之篇幅（不含封面之頁數）以 4 至 10 頁為原則，完整報告之篇幅則不限制頁數。

成果報告繳交之期限及種類（精簡報告、完整報告或期中報告等），應依本會補助專題研究計畫作業要點及專題研究計畫經費核定清單之規定辦理。

二、內容格式：依序為封面、中英文摘要、目錄（精簡報告得省略）、報告內容、參考文獻、計畫成果自評、可供推廣之研發成果資料表、附錄。

(一)報告封面：請至本會網站（http：//www.nsc.gov.tw）下載製作（格式如附件一）。

(二)中、英文摘要及關鍵詞(keywords)。

(三)報告內容：請包括前言、研究目的、文獻探討、研究方法、結果與討論（含結論與建議）…等。若該計畫已有論文發表者，可以 A4 紙影印，作為成果報告內容或附錄，並請註明發表刊物名稱、卷期及出版日期。若有與執行本計畫相關之著作、專利、技術報告、或學生畢業論文等，請在參考文獻內註明之，俾可供進一步查考。

(四)頁碼編寫：請對摘要及目錄部分用羅馬字 I 、II、 III……標在每頁下方中央；報告內容至附錄部分請以阿拉伯數字 1.2.3.……順序標在每頁下方中央。

(五)附表及附圖可列在文中或參考文獻之後，各表、圖請說明內容。

(六)計畫成果自評部份，請就研究內容與原計畫相符程度、達成預期目標情況、研究成果之學術或應用價值、是否適合在學術期刊發表或申請專利、主要發現或其他有關價值等，作一綜合評估。

(七)可供推廣之研發成果資料表：凡研究性質屬*應用研究*及*技術發展*之計畫，請依本會提供之表格（如附件二），每項研發成果填寫一份。

三、計畫中獲補助國外或大陸地區差旅費、出席國際學術會議差旅費或國際合作研究計畫差旅費者，須依規定撰寫心得報告（出席國際學術會議者須另附發表之論文），以附件方式併同成果報告繳交，並請於成果報告封面註記。

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2. 格式: 中文打字規格為每行繕打（行間不另留間距），英文打字規格為 Single Space。
3. 字體: 報告之正文以中英文撰寫均可。在字體之使用方面，英文使用 Times New Roman Font，中文使用標楷體，字體大小請以 12 號為主。

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）

生物材料之物理及非晶態半導體材料的軟模相變之研究(1/3-3/3)

Physics of bio-materials and the floppy to rigidity transition of amorphous semiconductors(1/3-3/3)

計畫類別：☑ 個別型計畫　□ 整合型計畫

計畫編號：NSC 91－2112－M－-032－006, NSC 92－2112－M－032－009, NSC 93－2112－M－032－006－(周子聰)

執行期間：91 年 8 月 1 日至 94 年 10 月 31 日

計畫主持人：周子聰

共同主持人：無

計畫參與人員：林方庭，何國敏，江正雄，詹政諱，徐鵬英，陳彥銘，陳守鍊，曾華民，林佩怡，劉毓凡，吳坤達，蘇德喜

成果報告類型(依經費核定清單規定繳交)：□精簡報告　☑完整報告

本成果報告包括以下應繳交之附件：

□赴國外出差或研習心得報告一份

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處理方式：除產學合作研究計畫、提升產業技術及人才培育研究計畫、列管計畫及下列情形者外，得立即公開查詢

□涉及專利或其他智慧財產權，□一年□二年後可公開查詢

執行單位：淡江大學物理系

中　華　民　國 94 年 12 月 6 日

一、 中英文摘要及關鍵詞(keywords)

關鍵詞：DNA 構型與穩定性，相變，彈性性質，螺旋構型與穩定性，硬度滲透，非定型態半導體。

中文摘要:

DNA 分子的彈性性質與其生物功能有密切的關係，因此相關之研究有助于對生物體的遺傳和演化的深入了解。我們求得了一個可用於描述雙股 DNA(dsDNA)彈性性質的模型在基態時的嚴格解，還求得另一個類似模型在考慮外力和外力矩時形成近似於螺旋結構的條件。我們推導出一個考慮了鹼基對交互作用的 dsDNA 模型所對應的構型的方程，並用於研究 dsDNA 在外力和外力矩下從 B 構型到 S 構型的尖銳相變，發現其一階相變的性質可以由一個有效位能來清楚地描述。在經受外力和外力矩時，dsDNA 分子的非對稱的彈性性質是個饒有趣味的問題。我們用解析推導的方法論証了兩個用於描述 dsDNA 分子的非對稱彈性性質的模型的相容性。我們推導出了一個可用於描述巨觀長細桿、微觀奈米碳管及生物高分子彈性性質的理論模型所對應的形狀方程。我們求得了該模型在經受外力和外力矩時形成螺旋結構的條件。我們發現在外力和外力矩作用下，螺旋桿的伸長可能出現不連續的躍變，並用於成功地解釋了化學成分確定的脂質液體(chemically defined lipid concentrate, CDLC)中的螺旋形膽固醇的實驗結果。

我們用計算機模擬的方法研究了在外力作用下的隨機稀薄化(random diluted)的中心力網絡的臨界彈性性質。我們發現系統的臨界彈性性質對應力相當敏感。而無應力系統是相當特殊的，有一個狹窄的臨界區。非定型態半導體材料如Ge_xSe_{1-x}等在近年來吸引了理論和實驗物理學家的廣泛的關注。然而許多基本問題如其玻璃態的軟模相變的性質，其相關之彈性係數的計算及其Raman光譜之理論與實驗間的比較仍然未得到解決。我們正在用分子動力學計算機模擬的方法研究Ge_xSe_{1-x}玻璃態的軟模相變，並用我們最近才推導出的"平衡漲落法"公式計算其彈性系數。

我們導出了適用於模擬單元(simulation cell)之外形可變的分子動力學方程，可用於模擬一般的非等向壓力之等溫−等應力和等焓−等應力系集 (ensemble)。

我們用離子阱串聯質譜儀研究麴酸抗生素和氯化鐵的交互作用，以及$Ca^{2+}/Mg^{2+}/Zn^{2+}$, Cl^-和短桿菌肽(gramicidin)的交互作用。

Keywords：DNA conformation and stability, phase transition, elasticity, helical filament conformation and stability, rigidity percolation, amorphous semiconductors.

Abstract：

The novel elasticity of double-stranded DNA (dsDNA) has very important biological significance. Therefore, a thorough investigation of the elasticity of dsDNA will enable us to gain better insight on many important biological processes concerned with life and growth. We found analytically the conditions to form a nearly helical conformation for a model of dsDNA under external force and torque. We obtained exact results of the classical mechanical solution of the worm-like chain model, a model for dsDNA elasticity, under an external stretching force. We investigated the sharp extension of dsDNA from the B-form S-form under external force in the framework of a model of a double-stranded chain with basepair interactions and bending.

Shape equations governing the structure of the DNA under external forces and torques are derived using a classical mechanical approach. The 1st order nature of the B- to S-form transition is revealed clearly in terms of an effective potential with a barrier separating these two states. We present an analytical based proof for the consistency of two asymmetric elastic models of the double-stranded DNA (dsDNA) in the regime around the B-form. The elasticity of a filament is a significant issue, not only because its wide application in engineering and sciences, but also because the recent experiments and theories have revealed that it may account for some elastic properties of microscopic objects, from carbon nanotubes to biomaterials such as DNA. We derive the general shape equations for an elastic rod with spontaneous torsion and curvatures in terms of Euler angles. We find that to form a helix, ψ must be a constant. We find that the extension of a helical rod may undergo a discontinuous transition under stretching force. This agrees quantitatively with experimental observations for a helix in a chemically defined lipid concentrate.

We study the rigidity of two dimensional site-diluted central force triangular networks under tension. We calculate the shear modulus μ directly and fit it with a power law of the form, $\mu \sim (p-p^*)^f$, where p is the concentration of sites, p^* its critical value, and f the critical exponent. We find that the critical behavior of μ is sensitive to tension. The amorphous semiconductors have made a significant impact on the areas of information and energy. However, the critical behavior of the floppy to rigidity transition of amorphous semiconductors is not yet clear, and the relevant theoretical study at finite temperature is still unavailable. We are exploring, by using molecular dynamics simulation, the critical behavior of the rigidity transition of amorphous semiconductors, such as Ge_xSe_{1-x}, and calculate its elastic constants using the equilibrium fluctuation formulae, and compare the theoretical results with experiments.

We derive a new set of equations of motion to produce correct isothermal-isotension and isoenthalpic-isotension ensembles in molecular dynamics simulation for a system under arbitrary anisotropic loading.

We study the interactions of a peptide, gramicidin A with biologically important divalent cations, and the interactions of biomedically important antibiotics, kojic acid with iron, using tandem electrospary ionization mass spectrometry (ESI-MS).

二、研究目的、研究方法

目的: To have a comprehensive understanding of the elastic properties of various materials, from carbon nanotubes to biomaterials such as DNA. To obtain a full picture for the floppy to rigidity transition. Using mass spectrometry to analyze biologically relevant interactions and drug metabolites.

方法: Analytical approaches, computer simulations, and using modern mass spectrometry.

三、主要結果與討論:

1. We derive the general shape equations in terms of Euler angles for a uniform elastic rod with spontaneous torsion and curvatures and subjected to external force and torque. Our results based on an analytic formalism show that the extension of a helical rod may undergo a one-step discontinuous transition with increasing stretching force. This agrees quantitatively with experimental observations for a helix in a chemically defined lipid concentrate. The larger the twisting rigidity, the larger the jump in the extension. The effect of torque on the jump is, however, dependent on the value of the spontaneous torsion. In contrast, increasing the spontaneous torsion encourages the continuous variation of the extension. An "over-collapse" behavior is observed for the rod with asymmetric bending rigidity, and an intrinsic asymmetric elasticity under twisting force is found.
2. We present an analytical-based proof for the consistency of two asymmetric elastic models of the double-stranded DNA in the regime around the B-form. We derive the shape equations in terms of Euler angles for a uniform ribbon with noncircular cross section and spontaneous torsion. We study the conditions to form a helical ribbon for the model and find that the Euler angle ψ must be either 0 or $\pi/2$: We find that there is either a continuous or discontinuous transition of extension, from a twisted straight ribbon to a helical ribbon.
3. The peptide, gramicidin A (GrA), has been demonstrated to interact with divalent salts ($CaCl_2$, $MgCl_2$, and $ZnCl_2$) using electrospray ionization mass spectrometry (ESI-MS). The ESI-MS analysis revealed different complexes formed due to the interaction of Val-GrA and Ile-GrA with divalent salts: [Val or Ile-GrA-H+M]$^+$, [Val or Ile-GrA+MCl]$^+$ and [Val or Ile-GrA+M]$^{2+}$, where M is Ca or Mg or Zn. All these complexes have been subjected to collisionally activated dissociation (CAD). CAD of singly and doubly charged GrA and metal complexes exhibited the losses of water molecules, indicating the ligand preference of GrA. MS/MS and MS^3 of [Val or Ile-GrA+MCl]$^+$ resulted in the elimination of chloride ion and water, respectively. The tandem mass spectrometry data of the complex [Val-GrA+MCl]$^+$ suggest that chloride interaction is stronger in the presence of Ca than of Mg and Zn. This study reveals that GrA could interact with Ca, Mg, and Zn in metal ion form as well as in ion pair (MCl) form. The interactions of GrA with Ca support the proposal of a physical basis for the messenger role of Ca
4. Kojic acid, 5-hydroxy-2-(hydroxymethyl)-4*H*-pyran-4-one, has been used extensively as a clinical iron-chelating drug although the nature of the complexes of iron and kojic acid has not been established. We demonstrate the complexation of kojic acid with iron(III) chloride by using electrospray ionization mass spectrometry (ESI-MS). The ESI-MS analysis revealed different reactions between iron(III) chloride and kojic acid (M), and the mass spectrum exhibited four complexes: [Fe+2(M–H)]$^+$, [Fe+3(M–H)+H]$^+$, [Fe2+4(M–H)+Cl]$^+$, and [Fe_2+5(M–H)]$^+$. All these proposed complexes and the presence of chloride ion in one of the dinuclear complexes have been confirmed by isotopic patterns and fragmentation studies by means of tandem mass spectrometry (MS^n).
5. Using density-functional theory within the generalized gradient approximation the interaction

between atomic Si and the Cu(1 1 0) surface is investigated. Various structures of on-surface adsorption as well as surface-substitutional adsorption for a wide range of Si coverage are considered. Our results shows that the Cu(1 1 0) surface is active towards adsorption of Si. The energetically most preferred structure is the c(2×2)-Si/Cu(1 1 0) surface alloy. The reason that (2×2) Si chain can form on c(2×2)-Si/Cu(1 1 0) surface is due to the particular stability of c(2×2)-Si/Cu(1 1 0) surface.

6. The existing equations of motion in molecular dynamics for a system under anisotropic loading are inconsistent with the theory of finite elasticity due to the introduction of a special reference value, and so cannot work properly under high anisotropic stress. We derive a new set of equations of motion in conjunction with proper isothermalisotension and isoenthalpic-isotension ensembles to be used in molecular dynamics simulations for a system under anisotropic loading. The new equations of motion satisfy the requirement of invariance of physical properties with respect to the simulation cell transformation, and hence are more appropriate for use in studying a system undergoing a structural transformation. We also present the correct expressions for calculating the elastic constants in these ensembles.
7. We study the rigidity of two-dimensional site-diluted central force triangular networks under tension. We calculate the shear modulus μ directly and fit it with a power law of the form $\mu \sim (p-p^*)^f$, where p is the concentration of sites, p^* its critical value, and f the critical exponent. We find that the critical behavior of μ is quite sensitive to tension. As the tension is increased there is at first a sharp drop in the values of both p^* and f, followed by a slower decrease towards the values of the diluted Gaussian spring network. We find that the size of the critical region is also sensitive to tension. The tension-free system has a narrower critical regime with the power law failing for $p>.0.8$. In contrast, a small tension is sufficient to extend the power law to near $p=1$. The physical basis for these behaviors is discussed.
8. Exact results of the classical mechanical solution of the worm-like chain model under an external stretching force is derived. The locus of the chain under arbitrary stretching force and grafting conditions is calculated explicitly in terms of elliptic functions. In the long chain limit, the chain locus can be expressed in terms of elementary functions. In addition, the solution of the worm-like rod chain under a small twisting torque is also calculated perturbatively. The relative extension, writhe number and the locus of the chain are computed to leading orders in the reduced torque.

四、計畫成果自評

原計畫在三年左右的時間在國際期刊上發表 9 篇研究論文。自從 91 年 8 月 1 日本計劃啟動以來，我們已經在 SCI 期刊上發表了 13 篇相關的論文，論文的質量與數量均超出原計畫目標。

原計畫要用三年左右的時間研究: 1. dsDNA的彈性性質和dsDNA微環構型及其穩定性以及dsDNA變性(denaturation)的序列相關性; 2.蛋白質摺疊的非格點模型; 3. 用分子動力學

計算機模擬的方法研究Ge_xSe_{1-x}玻璃態的軟模相變，計算其Raman光譜，並用我們最近才推導出的"平衡漲落法"公式計算其彈性系數。

上述第一項內容已經大部分完成並已在 SCI 期刊上發表了 8 篇相關的論文，參見本報告的「主要結果與討論」及「參考文獻」等部份。必須指出在這方面還有相當多值得進一步深入研究的課題。

上述第二項內容由於缺乏足夠的時間與人手因此基本上限於停頓狀態。

上述第三項內容亦因為因缺乏足夠的時間與人力而進展不如預期。但有兩篇與此相關的論文已發表在Phys. Rev. E的 rapid communication部分，和Surface Science上。另有一篇相關的論文的主要工作已基本完成。用"平衡漲落法"公式計算Ge_xSe_{1-x}玻璃態的臨界彈性行為的計算機模擬程式亦已基本完成。因此這一部分的產出亦可算差強人意。

另外有三篇研究論文不在原計畫目標內。其中有一篇導出了適用於模擬單元(simulation cell)之外形可變的分子動力學方程，可用於模擬一般的非等向壓力之系集(ensemble)。第二篇用離子阱串聯質譜儀研究麴酸抗生素和氯化鐵的交互作用。第三篇用離子阱串聯質譜儀研究$Ca^{2+}/Mg^{2+}/Zn^{2+}$, Cl^-和短桿菌肽(gramicidin)的交互作用。後兩篇是與本校化學系老師的合作成果。

總而言之，儘管有一定偏差，但本計畫的方向大致符合預期，而研究成果的數量與質量則超過預期。必須說明的是，原定的三個方向都有相當多值得進一步深入研究的課題，我們亦已有足夠的經驗來挑戰這些問題。目前的主要制約在於缺乏有足夠經驗和能力的研究人員。

五、參考文獻：主持人於 91 年 8 月 1 日至 94 年 10 月 31 日期間著述：

I. 期刊論文（大部分可見於：國科會網站之學術著作（全文）登錄及上傳）：

1. **Zicong Zhou**, Pik-Yin Lai, and Béla Joós, "Elasticity and Stability of a Helical Filament", Phys. Rev. E **71**, 052801-1—052801-4(2005) **(SCI)**
2. Putty-Reddy Sudhir, Hui-Fen Wu, and **Zi-Cong Zhou**, "An application of electrospray ionization tandem mass spectrometry to probe the interaction of $Ca^{2+}/Mg^{2+}/Zn^{2+}$ and Cl^- with gramicidin A", Rapid Commun. Mass Spectrom. **19**, 1517-1521(2005) **(SCI)**
3. **Zicong Zhou**, "Elasticity and stability of a helical ribbon with noncircular cross section", Modern Physics Letters B **19**, 249-267(2005) **(SCI)**
4. **Zicong Zhou** and Pik-Yin Lai, "On the Asymmetric Elasticity of dsDNA", Physica A **350**, 70-83(2005) **(SCI)**
5. **Putty-Reddy Sudhir, Hui-Fen Wu, and Zi-Cong Zhou**, "A study on kojic acid complexation with iron by using electrospray ionization mass Spectrometry", Rapid Commun. Mass Spectrom. **19**, 209–212(2005) **(SCI)**.
6. Pik-Yin Lai, G. Rowlands, and **Zi-cong Zhou**, "Worm-Like Chain under External Forces: Some Analytic Results", Chinese Journal of Physics **42,** 410-422(2004) **(SCI)**
7. Guo-min He, Shu-ping Li, and **Zicong Zhou**, "Si adsorption on Cu(1 1 0) surface from ab initio calculation", Surface Science **553**, 126-132(2004) **(SCI)**
8. **Zicong Zhou**, "Equations of motion and calculation of elastic constants for a system under anisotropic loading", Chinese Journal of Physics **42,** 21 (2004) **(SCI)**.
9. **Zicong Zhou**, Béla Joós and Pik-Yin Lai, "Elasticity of randomly diluted central force networks", Phys. Rev. E **68**, 055101(R)(2003) **(SCI)**.
10. **Zicong Zhou** and Pik-Yin Lai, "Nearly helical conformations of an elastic dsDNA ribbon", Int. J. Mod. Phys. B **17**, 3021-3032(2003)**(SCI)**
11. Pik-Yin Lai and **Zi-cong Zhou**, "Stretching a double-stranded DNA: Nature of the B-form to S-form Transition", J. Chem. Phys. **118**, 11189-11199(2003)

12. Pik-Yin Lai and **Zicong Zhou**, "B- to S-form transition in double-stranded DNA with basepair interactions"**,** Physica A **321**, 170-180(2003)
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II. 國際研討會論文:

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