

研究生前沿教材书系

Glassy Materials and Disordered Solids

An Introduction to Their Statistical Mechanics

玻璃质材料和无序固体

——它们的统计力学导论

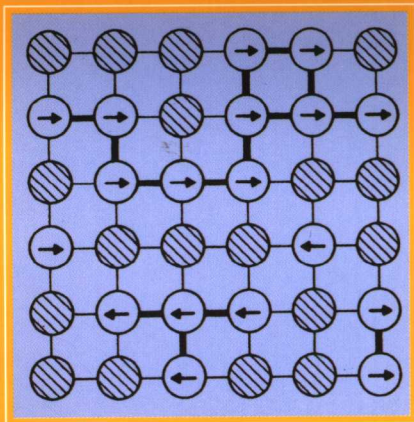
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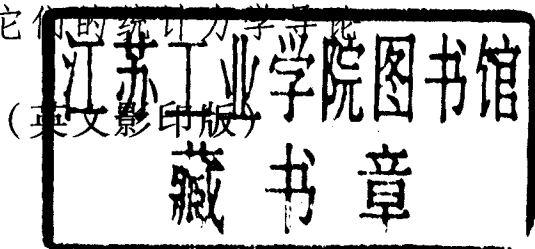
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Kurt Binder, Walter Kob

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复旦大学出版社出版英文影印版《研究生前沿教材书系》，主要基于以下几点考虑。

1. (新加坡)世界科技出版公司以出版科技专著闻名于世,同我社已有 10 多年的友好交往。从 20 世纪 90 年代以来,尤其是 1995 年该公司并购了伦敦帝国学院出版社(Imperial College Press) 51% 的股份(近年已完成了 100% 的股份收购)之后,这两大出版机构在潘国驹教授的集中指挥下,充分发挥了编辑学术委员会的职能,使得出书范围不断拓宽,图书层次逐渐丰富,因此从中遴选影印图书的空间就更大了。再加上该公司在上海设有办事机构,相关工作人员工作细致,服务周到,给我们两个单位的合作交流带来极大的便利。

2. 研究生教育是创新人才培养的关键,教材建设直接关系到研究生科学水平和创新能力的培养。从 2003 年开始,我社陆续出版了 *Fudan Series in Graduate Textbooks* 这套丛书,国内的读者反响很好。但限于作者人力,这套丛书涵盖的学科和门类都较为不足。为此,我们想到再借助国外出版力量,引进一批图书作为硕士研究生的补充教材,(新加坡)世界科技出版公司与我社的合作,恰好提供了这样一个良好的机会。我们从该公司提供的大量近期书目中,遴选出 30 多本样书,经过专家审读后,最终确定了其中的 11 种作为首批《研究生前沿教材书系》影印出版。这 11 种图书的作者来自美、英、法、德、加拿大 5 个国家的 10 多所高校或研究部门,他们既是相关学科科研的领军人物,又是高年级本科生和研究生教学的杰出教授(详见各书的作者介绍)。各门教材既考虑到深入浅出的认知规律,又突出了前沿学科的具体应用,每本书都有充实的文献资料,有利于读者和研究人员深入探索。其中 6 本教材配有习题,

还包括一本具有物理背景的人员都需要了解的高级科普读物——《理解宇宙——从夸克到宇宙学》。

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4. 原版书价格较贵,大大超出读者的购买能力,即使图书馆或大学资料室也会受到经费不足的制约。出版影印本的书价不到原价的十分之一,无疑会给需要这些书的研究生和图书馆带来真正的实惠,这也是(新加坡)世界科技出版公司与我们合作的目的之一。

5. 考虑到物理类图书是(新加坡)世界科技出版公司的第一品牌,我们首次引进的 11 本书,都属大物理的范畴。这一尝试如果得到读者和专家的认可,今后我们将陆续开辟其他学科的影印渠道。

欢迎读者批评指正,并提出有益的建议。

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内 容 简 介

对于研究生而言,玻璃质材料和无序固体是比结晶固体更加广泛、更加具有挑战性的研究领域,这也是迄今为止学术界还没有形成普遍共识和前后统一的理论的一个领域。本书的两位作者都是该领域的知名专家,成书之前第一作者曾在德国美因茨(Mainz)的 Johannes Gutenberg 大学讲课多年。可以说,这是填补该领域教科书空白的一部力作。

全书共分5章,第一章以教科书的方式,简要评述无序物质的模型和它的统计力学的普遍概念。第二章讲述无序物质的结构和动力学。第三章为无序结构的模型,其中以无规行走来讨论柔性聚合物构型的模型,从渗滤理论出发讨论分形结构等。第四章讲述无序物质的普遍概念和物理特性,包括聚合物动力学的 Rouse 模型、应用渗滤问题处理物理系统、分形结构的元激发、无定形固体的物理性质、自旋玻璃体等。第五章讲述超冷流体和玻璃体相变,包括形成玻璃体的唯象模式、慢弛豫模型和玻璃体相变的模-耦合理论等。各章之后都有大量参考文献,供深入研究参考。

这本可读性较强的教科书将紧紧抓住学生的心,让你懂得如此广泛的领域具有许多共同的特征,亦即表明在统计力学的框架内可以给予很好的描述。本书同样适用于作为研究无定形材料的研究人员、理论工作者和实验人员的标准教本。

Kurt Binder

德国 Mainz 的 Johannes Gutenberg 大学理论物理教授,德国物理学会会员、欧洲物理学会会员,德国高等教育研究联合会会员。

1944 年出生于奥地利,1962 年进入维也纳技术大学学习技术物理,1965 年、1967 年分别取得该校学士、硕士学位。1967 年进入奥地利原子物理研究所攻读博士,1969 年取得博士学位。1969 年进入德国慕尼黑技术大学物理系从教,其中有一年时间去瑞士苏黎世的 IBM 研究实验室做博士后研究,1974 年以研究顾问身份访问位于美国新泽西的 Bell 实验室。1974 年 10 月至 1977 年 9 月,德国 Saarland 大学理论物理教授。1977 年 10 月至 1983 年为德国科隆大学教授,之后一直是 Mainz 的 Johannes Gutenberg 大学物理研究所的理论物理教授。这期间曾兼任 Mainz 大学的材料研究中心协调委员会主任,德国国家基金会支持的“非金属无定形材料的玻璃态和玻璃相变”专题研究组的发言人,马克斯·普朗克学会外籍会员、奥地利科学院通讯会员。1993 年获德国物理学会马克斯·普朗克奖章。出版学术专著 5 本。他是 *International Journal of Modern Physics* 等 10 多种杂志的编委,又是 *Phys. Rev. Lett.* 等大量杂志的审稿人,还是德国科学基金会等很多基金组织的专家组成员。

业余爱好:钢琴伴奏

Walter Kob

法国 Montpellier II 大学教授,研究无序系统(超冷液体、块体玻璃、Potts 玻璃、电泳、离子导电玻璃、无定形表面)的统计力学特性。

1985 年 10 月毕业于瑞士 Basel 大学凝聚态理论物理专业,1989 年取得该校物理研究所凝聚态理论物理博士学位。之后在该所作了一年玻璃相变的博士后研究工作,从 1990 年至 1994 年,又去美国斯坦福大学化学系作博士后研究,专业方向是“超冷液体的动力学行为”。之后到德国 Mainz 大学物理所作助教,研究无序系统的统计力学特性。2000 年开始,成为法国 Montpellier II 大学教授。

从 1994 年至今,培养博士生 16 名,发表学术论文 80 多篇,出版专著 5 本。他是法、德两国相关领域重大课题组的科学顾问、项目负责人或协调员,还是欧美各国科学基金会的顾问和 *Chemical Physics* 等众多杂志编辑部的编委或审稿人。

序 言

理解无序材料的物理性质及弄懂从超冷液体转变为无定形固体的本质都是极为困难的,同时也是当代最有挑战性和最有魅力的问题。因此,针对这个课题开展了大量的科学活动:通过实验的、经过解析理论的,以及采用计算机模拟的。撇开这些努力不谈,至今对这一问题还没有出现被普遍认可和前后一致的看法。这也是造成迄今为止还没有关于这一课题的有代表性的教科书产生的原因,这同凝聚态物理的其他大多数课题(结晶固体、简单流体以及聚合物复杂流体——液晶)形成了鲜明对照。大多数已有的教科书讲述范围都相当狭窄,有些只强调无序的结构模型,有些只涉及唯象热力学方面,甚至只热衷于对这些系统提供复杂弛豫现象的文献资料。

因而,我们想用我们这本专题论述来填补这一空白,重点是讲述玻璃体相变和材料的无定形状态的统计力学方法,同时对无序固态物理学中的重要概念(渗滤、分形,诸如自旋玻璃等无序磁性模型系统等)提出符合教学要求的论述。我们感到本书的题材选取是十分自然的,因为无序固态与形成玻璃系统的统计力学包括很多共同的内容,简单问题的许多概念都能够解释出现在玻璃系统中的更加复杂的现象。比如,人们不断努力试图用渗滤理论去解释玻璃体结构和玻璃体相变问题,同样地,人们还试图采用从自旋玻璃有关问题中获得的洞察力去描述结构玻璃的物理学。

因此,本书在重点突出无序凝聚态系统的那些共同性的方面,强调以统计热力学作为普遍的理论框架,考虑到发展切实可行的解析方法的困难,以计算机模拟(Monte Carlo 和分子动力学方法)的经典方法作为实际研究工具的核心。注意到在处理同二级相变有关的临界现象等问题时,人们已经建立了诸如平均场理论和重整化

群等等很好的方法,这些方法都能够在定性,甚至定量的基础上描述现象。与此形成对照的是,关于玻璃体和玻璃相变的情况,我们还没有能够准确描述现象的平均场理论。

不管怎么说,由于过去数十年的大力研究,我们对玻璃体和其他无序固态的了解已经大大地进步了,因此本书就要努力反映这些进展,并对现在确立的基本事实作出前后一致的描述。反过来说,那些还停留在猜测水准上的很多概念并未作仔细讨论。同样,像电子结构和电子激发、无定形固态的光学特性等等许多相关方面,本书也未加讨论。因此,本书的选材在一定程度上反映了专家和作者的偏爱,因此这里未加讨论的论题不一定就是无关紧要的。本书选材范围的限制,反而有利于已经掌握基本统计热力学基础知识的研究人员和博士后研究人员更易理解,因为没有必要去读更专门的知识了。事实上,作者之一(K. B.)已经就这一课题在美因茨(Mainz)的 Johannes Gutenberg 大学已经讲过很多次课了,因此我们认为这是类似课程中的一本极有用的基础教程。同时,本书各章末尾的大量文献,即使对于高级研究人员而言也是极有用的参考资料。实际上,我们希望本书在某种程度上将会对玻璃体物理的深入研究产生深刻的影响,因此书中归纳性的事实描述和模型描述的有关知识有利于为将来的研究工作提出正确的问题。

最后,我们很高兴对合作者表示极大的谢意,他们对我们在这一个课题中的研究工作作出了重大的贡献,很多同事的观点和卓有成效的讨论使我们受益匪浅,最后但不是不重要,我们要感谢 A. Chase 女士,感谢她在打印手稿中对我们的宝贵帮助。

Kurt Binder 和 Walter Kob

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Chapter 1

Introduction

1.1 Models of Disordered Matter: A Brief Overview

Although this text deals with the physics of condensed matter, its contents differs strongly from the one of most standard textbooks on this subject in that it focuses on materials such as window glass, plastics, rubber, amorphous metals, porous materials, magnets with frozen-in disorder, etc., i.e. on systems that are referred to as “amorphous materials” or “ill-condensed matter”. Despite the fact that these materials have been used by man since ancient times, presumably the first glass was produced in ancient Egypt several thousand years ago, and are nowadays ubiquitous in daily life, most textbooks on the theory of condensed matter ignore this type of matter almost completely. The reason for this negligence is the lack of a coherent and elegant theoretical description for these systems, which in the case of crystals can be obtained by exploiting the periodicity of the crystal structure. Therefore, analytical theories for the properties of such strongly disordered matter are comparably scarce, and much of the theoretical knowledge that we have so far stems from computer simulations. Despite this lack of a well founded theoretical description, the physics and chemistry of glasses has become a prominent topic of research in the last decades: The structure of the glassy state has become a subject of intense experimental investigation and numerical simulations, and in particular the “problem of the glass transition”, i.e. the phenomenon that many liquids can be cooled below their melting temperature and solidify into an amorphous solid, is considered as one of the grand challenges in condensed matter physics, and hence studied intensively.

In this situation, the present book tries to fill a gap in the present literature by presenting an introduction to the subject under the point of

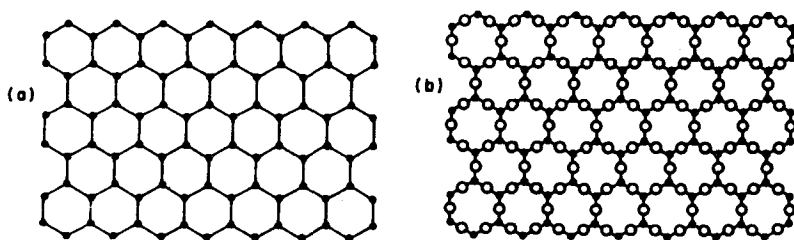


Fig. 1.1 (a) The honeycomb lattice and (b) the decorated honeycomb lattice. The topological structure of the honeycomb is the same as that of a layer of carbon atoms in graphite or of a layer of crystalline arsenic. The topology of the decorated honeycomb occurs in the structure of layers that make up crystalline As_2S_3 and As_2Se_3 . The sulfur (or selenium) atoms are represented by open circles. After Zallen (1983).

view of statistical thermodynamics. We feel that statistical thermodynamics statistical thermodynamics is a particularly useful framework for the description of amorphous solids and the understanding of their properties as we will outline in this Introduction. More details on the various topics can be found in the subsequent chapters.

As we will see later, the nature of the structural disorder depends strongly on the type of disordered solids one considers, and obtaining a proper characterization of these structures is one of the main topics of this book. (More detailed discussions on structural disorder can be found in the texts of Cusack (1986) and Elliott (1983).) A second main topic is to understand the implications of this “frozen-in disorder” for the various physical properties. In addition to this frozen-in disorder, we have of course also the normal thermal disorder, which is one of the standard problems considered by statistical thermodynamics, but which wins new facets here in its interplay with the quenched disorder.

These general remarks can be illustrated with a specific example, the “Continuous Random Network” (CRN) model of covalent glasses, which dates back to Zachariasen (1932), cf. Figs. 1.1 and 1.2. In such an amorphous solid, one encounters disorder in the “equilibrium” positions of the atoms even if the local chemical order is the same as the one in the corresponding crystalline structure.

We put the word “equilibrium” in quotation marks, since we assume that the real equilibrium structure is the ordered crystal of Fig. 1.1, which has a lower free energy. However, often the rates for the nucleation and growth of crystal from the amorphous structure are so low, that for most

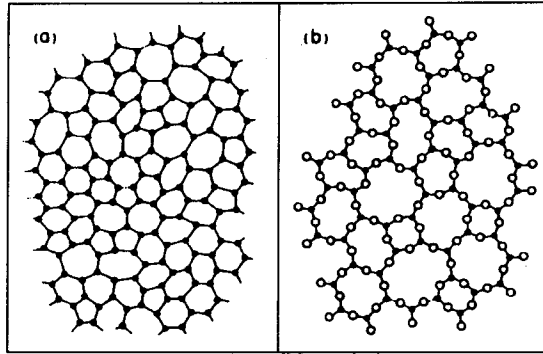


Fig. 1.2 Two-dimensional continuous random networks. A sketch of a (hypothetical) threefold-coordinated elemental glass is presented in (a), while Zachariassen's (1932) diagram for an A_2B_3 glass is shown in (b). After Zallen (1983).

practical purposes the latter is metastable phase that is essentially indistinguishable from the thermal equilibrium.

The difference in the geometrical structure between the crystal (Fig. 1.1) and the glass (Fig. 1.2) has immediate consequences for the spectrum of elementary excitations of the respective solids, and the resulting thermodynamic properties at low temperatures. In the crystal as well as in the amorphous solid we find small-amplitude vibrations of the atoms around their equilibrium positions, which can be treated as harmonic oscillations at low temperatures, at least as a first approximation. Now the lattice periodicity of the crystal has an important physical consequence: the vibrations can be decomposed into non-interacting plane waves with a wave-vector \vec{k} and a frequency $\omega(\vec{k})$, i.e. \vec{k} is a "good quantum number". As one can read in more detail in any standard text book on theoretical solid state physics, one finds a homogeneous density of eigenstates \vec{k} in the first Brillouin zone of the crystal, and this has an important consequence on the phonon density of states $\zeta(\omega)$ at small $k = |\vec{k}|$. Recalling the dispersion relation for acoustic phonons, $\omega = ck$, where c is the velocity of sound (for simplicity we ignore the distinction between longitudinal and transverse phonons here), one concludes immediately

$$\zeta(\omega)d\omega = \frac{\partial\omega}{\partial\vec{k}} d^d\vec{k} \xrightarrow{k \rightarrow 0} \text{const} \cdot k^{d-1} dk \propto \omega^{d-1} d\omega \quad , \quad (1.1)$$

where d is the dimension of the crystal. This yields as a contribution

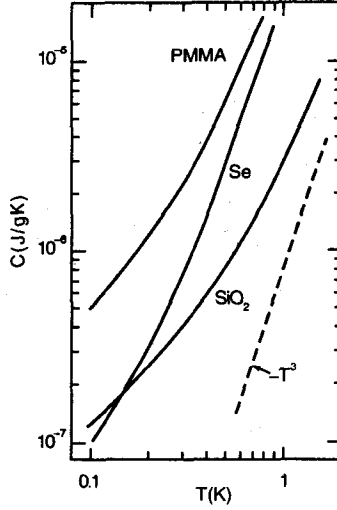


Fig. 1.3 Specific heat C_V of vitreous silica (SiO_2), amorphous selenium (Se) and polymethylmetacrylate (PMMA), also known as “plexiglass”) as a function of temperature. In the case of amorphous silica, the Debye contribution due to sound waves Eqs. (1.1)-(1.3) is included as a broken straight line on this log-log plot. The variation of C_V at low temperature roughly follows a relation $C_V \propto T^{1+n}$ with $0 < n \leq 0.3$ in the various amorphous materials. After Stephens (1976).

of the internal energy due to phonons (\hbar = Planck’s constant and k_B = Boltzmann’s constant)

$$U_{\text{phonons}} = \int \hbar\omega\zeta(\omega)[\exp(\hbar\omega/k_BT) - 1]^{-1} d\omega$$

$$\xrightarrow{T \rightarrow 0} \text{const} \cdot T^{d+1} \int_0^\infty x^d [e^x - 1]^{-1} dx, \quad (1.2)$$

where in the last step we have substituted $\hbar\omega/k_BT = x$ in the integral. From Eq. (1.2) we immediately recognize the famous T^3 law for the specific heat at low T (in $d = 3$ dimensions), since

$$C_V = (\partial U / \partial T)_V \propto T^d \quad (1.3)$$

(Here we have made the approximation that the internal energy U is given by U_{phonons} , an approximation which is reasonable if the contributions from conduction electrons or magnons can be neglected.)

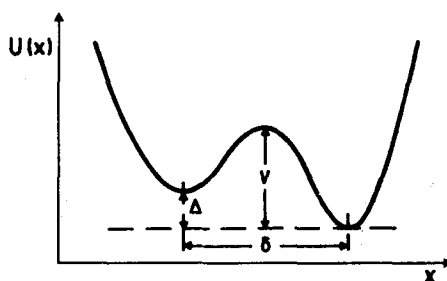


Fig. 1.4 Double-well potential with an asymmetry Δ , a potential barrier of height V , and width δ .

These considerations are no longer valid in amorphous materials: Since there is no crystal lattice, there is no Brillouin zone, and \vec{k} is no longer a good quantum number. While acoustic waves with the dispersion relation $\omega = ck$ still exist for $k \rightarrow 0$, these waves are not dominant in the density of states for small ω . Instead the temperature dependence of the specific heat at low T , see Fig. 1.3, seems to hint for the presence of more or less localized excitations that contribute to the density of states at low ω . In this context the model of the so-called “two-level systems” (TLS) has gained wide popularity to describe this excess. Within this model one postulates the existence of particular degrees of freedom (x) which experience a potential $U(x)$ with the shape of an asymmetric double well and which undergo a tunneling motion between the two wells, see Fig. 1.4. As we will see later, the TLS model (Anderson *et al.* 1972; Phillips 1972, 1981; von Löhneysen 1981; Kovalenko *et al.* 2001) is quite successful and widely used to account for many low temperature properties of various kinds of amorphous materials. E.g., if one assumes that the density of such TLS is independent of the gap Δ for $\Delta \rightarrow 0$, one can rationalize a linear variation of C_V with T . However, although the concept of two level systems in glasses is already more than 30 years old, it is still controversial what the physical meaning of the variable x in Fig. 1.4 really is. Figure 1.5 gives speculative examples of structures in which two-level systems could be present (Hunklinger and Arnold 1976). However, a microscopic theory that describes the precise physical nature of these two-level systems is still lacking. We will return to a more detailed discussion of these problems in Chap. 4 of this book.

While so far we have mostly considered solids with a disordered structure resembling the one of a liquid, as they are typically formed by super-