

Neutron Diffraction

Edited by H. Dachs

Neutron Diffraction

Edited by H. Dachs

With Contributions by

P. Chieux P. Coppens H. Dachs J. B. Hayter

P.-A. Lindgård D. Petrascheck W. Prandl

H. Rauch W. Schmatz G. Zaccai

With 138 Figures

Springer-Verlag Berlin Heidelberg New York 1978

Professor Dr. Hans Dachs

Hahn-Meitner-Institut für Kernforschung Berlin GmbH,
D-1000 Berlin 39, Fed. Rep. of Germany

ISBN 3-540-08710-9 Springer-Verlag Berlin Heidelberg New York
ISBN 0-387-08710-9 Springer-Verlag New York Heidelberg Berlin

Library of Congress Cataloging in Publication Data. Main entry under title: Neutron diffraction. (Topics in current physics ; 6). Bibliography: p. Includes Index. 1. Neutrons-Diffraction. I. Chieux, P. II. Dachs, H., 1927-. III. Series. QC793.5.N4628N48 539.7'213 78-2969

This work is subject to copyright. All rights are reserved, whether the whole or part of the material is concerned, specifically those of translation, reprinting, re-use of illustrations, broadcasting, reproduction by photocopying machine or similar means, and storage in data banks. Under § 54 of the German Copyright Law where copies are made for other than private use, a fee is payable to the publisher, the amount of the fee to be determined by agreement with the publisher.

© by Springer-Verlag Berlin Heidelberg 1978.
Printed in Germany.

The use of registered names, trademarks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.

Offset printing and bookbinding: Konrad Tritsch, Graphischer Betrieb, Würzburg
2153/3130-543210

Preface

Since the great discovery made by Laue, x-ray diffraction has become the most important method for the investigation of atomic structure in condensed matter. Certain investigations, however, are difficult or totally impossible to conduct using x-rays, for example, the localization of atomic nuclei or atoms having only a few core electrons, and the observation of magnetic moments. The investigation of these important areas is made possible by neutron diffraction. Thus this method has developed into an important supplement to x-ray investigations. An ever-increasing demand is put on the method by research areas, old and new. Neutron diffraction has completely reformed the subject of solid-state magnetism; especially the area of chemical binding has received a new impetus by the union of x-ray and neutron diffraction. An exchange of different isotopes of the same element gives rise, as a rule, to a change in the strength of the neutron diffraction. Due to this effect it is possible, by means of deuteration, to make visible a single chain in a solid high polymer. Thus neutron small-angle scattering is important in protein research and for the biology of macromolecules. Of equal importance is the application of neutron diffraction in metallurgy.

There already exist several excellent books which discuss the results obtained by means of neutron diffraction. It is not the purpose of this book to compete with these in the area of completeness; it is our purpose to stress the technical aspects of the subject. The book is intended to help researchers who are engaged in neutron diffraction, and students beginning their research work. It is meant as a help in deciding if neutron diffraction can be used in the solution of a certain problem, and to simplify the preparation.

Many discussions at Institut Laue Langevin, Grenoble, were important in the preparation of this book. I wish to express my thanks to all my friends there, and especially to Mr. Jacrot who made important contributions to Chapter 7.

Berlin, March 1978

Hans Dachs

Contents

1. *Principles of Neutron Diffraction*. By H. Dachs (With 22 Figures) 1

1.1 Introduction 1

1.2 Background 2

1.3 VAN HOVE Theory in the Classical Approximation 3

1.3.1 Neutron Spectroscopy 8

1.3.2 Neutron Diffraction 8

1.4 Interaction of Neutrons with Matter 10

1.4.1 Scattering from the Atomic Nucleus 10

1.4.2 Incoherent Scattering 12

1.4.3 Magnetic Scattering 14

1.5 Optimization of Scattering Experiments 15

1.5.1 Description of the Reactor Spectrum 15

1.5.2 Focusing Methods 20

1.6 Theory of the Neutron Spectrometer 25

1.6.1 Monochromator Crystals 25

1.6.2 Single-Crystal Measurements 27

Half-Width of Rocking Curves 28

Systematic Errors in the Measurement of Integrated Intensities 31

Resolution of Single-Crystal Spectrometers 32

Measurement Problems for Crystals with very Large Lattice Constants 34

Intensity Measurements Using Fixed Samples 36

1.6.3 Powder Diffractometer 37

References 38

2. *Polarized Neutrons*. By J. B. Hayter (With 6 Figures) 41

2.1 Overview 41

2.2 Theoretical Background 42

2.2.1 Definition of a Polarized Neutron Beam 44

2.2.2 Polarization-Dependent Cross Section 46

2.2.3 Neutron Optical Effects 49

2.3	Production and Handling of Polarized Beams	51
2.3.1	Devices Using Field Gradients	52
2.3.2	Polarizing Filters	53
2.3.3	Use of Neutron Optical Effects	53
	Magnetic Mirrors and Guides	53
	Polarizing Multilayer Structures	56
2.3.4	Neutron Spin-Turn Devices	56
	The Leningrad Flipper	57
	The MEZEI Spin-Turn Coil	58
2.4	Some Applications of Polarized Neutron Scattering	60
2.4.1	Topological Problems	60
2.4.2	Flipper Choppers	62
2.4.3	Neutron Spin Echo	62
2.4.4	Resonance-Modulated Diffraction	64
2.5	Summary	67
	References	67
3.	<i>Combining X-Ray and Neutron Diffraction: The Study of Charge Density Distributions in Solids.</i> By P. Coppens (With 10 Figures)	71
3.1	Background	71
3.2	The Electron Distribution in Crystals and the Pertinence of Neutron Diffraction	73
	3.2.1 Charge Density Functions	73
	3.2.2 The Importance of Neutron Diffraction	77
3.3	Experimental Aspects	79
	3.3.1 Survey of Experimental Requirements	79
	Essential Conditions for Density Analysis	81
	Desirable Conditions	81
	3.3.2 Extinction	82
	3.3.3 Multiple Reflection	87
	3.3.4 Thermal Diffuse Scattering	88
	3.3.5 Differences Between X-Ray and Neutron Thermal Parameters ...	89
3.4	Statistical Analysis of the Errors in X-N Maps	92
3.5	Survey of Selected Results	96
	3.5.1 Analysis of Lone-Pair Densities	97
	3.5.2 Analysis of Bond Densities	101
	3.5.3 Quantitative Comparison of Theory and Experiment	103
3.6	Results Based Exclusively on X-Ray Data; Can They Replace the X-N Technique?	105
	References	108

4. *The Determination of Magnetic Structures.*

By W. Prandl (With 7 Figures)	113
4.1 Magnetic Neutron Scattering: Elementary Treatment	113
4.1.1 Unpolarized Neutrons	113
4.1.2 Polarized Neutrons	117
4.2 Magnetic Structures: Phenomenology	118
4.3 Generating Models	121
4.4 The Application of Magnetic Groups	122
4.4.1 Magnetic Point Groups	123
4.4.2 Magnetic Translation Groups	125
4.4.3 Magnetic Space Groups (SHUBNIKOV Groups)	127
4.4.4 The Construction of Models	128
4.4.5 Limitation of the Method and New Developments	134
4.5 Irreducible Representation of Space Groups	136
4.6 Magnetic Neutron Scattering: The General Case	143
4.6.1 Unpolarized Neutrons	143
4.6.2 Polarized Neutrons	145
4.7 Instruments / Measurements / Data Analysis	146
4.8 Nomenclature and Publications	147
References	147
5. <i>Disordered Structures.</i> By W. Schmatz (With 30 Figures)	151
5.1 Summary	151
5.2 Scattering Laws	154
5.3 Experimental Techniques	167
5.4 Small-Angle Neutron Scattering (SANS)	171
5.5 Diffuse Elastic Scattering by Nonmagnetic Crystals	179
5.6 Scattering by Disordered Magnetic Systems	186
References	193

6. *Phase Transitions and Critical Phenomena.*

By P.-A. Lindgård (With 10 Figures)	197
6.1 Overview	197
6.2 Theory of Phase Transitions	200
6.2.1 The Mean Field Theory, Introduction of Critical Exponents ..	201
Simple Example	201
6.2.2 The Landau Expansion	205
Range of Validity of the Landau Theory	207
The Dimensionality n of the Order Parameter	209
6.2.3 Competing Order Parameters and Multicritical Points	211

6.2.4	Phase Transitions in Random Systems	213
	Spin-Glass and Order in Amorphous Material	214
	Competing Magnetic and Quadrupolar Order Parameters	216
6.3	Theories of Critical Phenomena	216
6.3.1	Series Expansions	216
6.3.2	Scaling and Renormalization Group Theories	217
6.3.3	Crossover Phenomena and the Critical Equation of State	220
6.3.4	First-Order Transitions	222
6.3.5	The Correlation Function in the Critical Region	223
	Spatial Anisotropic Critical Scattering	224
6.4	The Neutron Scattering Cross Section	224
6.5	Critical Phenomena at a Second-Order Phase Transition	226
6.5.1	One-Dimensional Systems ($d = 1$)	226
6.5.2	Two-Dimensional Systems ($d = 2$)	229
6.5.3	Three-Dimensional Systems ($d = 3$)	230
6.6	Phase Diagrams: Multicritical Points and First- and Second-Order Transitions	233
6.6.1	Uniform Magnetic Systems	233
6.6.2	Other Transitions	236
6.6.3	Mixed Systems and Alloys with Competing Order Parameters ..	236
6.6.4	Spin-Glass Order	237
6.7	Discussion and Outlook	238
	References	239
7.	<i>Application of Neutron Diffraction to Biological Problems.</i>	
	By G. Zaccaï (With 14 Figures)	243
7.1	Background	243
7.1.1	The Ambition of Molecular Biology	243
7.1.2	Structure	244
7.1.3	Why Neutron Diffraction is an Attractive Technique in Biology	244
7.2	Theory	246
7.2.1	Contrast Variation	246
7.2.2	Low Angle Scattering from Solution	248
7.2.3	The Interpretation of the Scattering Curves at Higher Angles	250
7.2.4	Selective Deuteration	251
7.3	Single-Crystal Analysis	252
7.3.1	Proteins	252
7.3.2	Hydrogen Atoms in Proteins	253
7.3.3	Very High Resolution	254
7.3.4	Phase Determination	254

7.3.5	Crystals of Large Complexes at Low Resolution	255
7.4	Oriented Systems	255
7.4.1	Myelin	255
7.4.2	Oriented Systems	255
7.4.3	Disorder	256
7.4.4	Artificial Ordering	257
7.4.5	Model Membranes	257
7.4.6	Retinal Rod Outer Segment (ROS) Membranes	258
7.4.7	Calcified Tendon	260
7.5	Particles in Solution	262
7.5.1	Spherical Viruses	263
7.5.2	Ribosomes Studied by Contrast Variation	264
7.5.3	Ribosomes Studied by Triangulation	266
7.6	Conclusions	267
	References	267
8.	<i>Liquid Structure Investigation by Neutron Scattering.</i>	
	By P. Chieux (With 8 Figures)	271
8.1	Overview	271
8.2	The Basic Equations	272
8.2.1	Van Hove Scattering Law	272
8.2.2	The Static Approximation and the Structure Factor	273
8.2.3	Polyatomic Systems and Partial Structure Factors	275
8.2.4	Molecular Systems	276
8.2.5	Relation Between Structure Factor and Thermodynamics	277
8.3	Neutron Scattering Experiment	278
8.3.1	The Machines	278
8.3.2	How to Optimize an Experiment	279
8.3.3	Data Analysis	283
8.4	Monatomic Liquids	285
8.5	Binary Systems	287
8.5.1	The Normalization and the Thermodynamic Limit	287
8.5.2	The Isotopic Substitution Method	290
8.5.3	The Concentration Method — The "Zero Alloys"	292
8.5.4	Magnetic Systems	294
8.5.5	Conclusion	296
8.6	Molecular Liquids	296
8.6.1	The Angular Correlation Function	296
8.6.2	The Inelasticity Correction	298
8.6.3	The Intramolecular Structure Factor Determination	298

8.6.4 The Spherically Symmetric Part of the Molecular Correlation Function	299
8.6.5 The Orientational Correlation Function	299
8.7 Solutions	300
References	301
9. <i>Dynamical Neutron Diffraction and Its Application.</i>	
By H. Rauch and D. Petrascheck (With 31 Figures)	303
9.1 Basic Equations	304
9.1.1 One-Beam Approximation	308
9.1.2 Two-Beam Approximation	309
9.2 Solution for a Plane Slab	311
9.2.1 Laue Case	313
9.2.2 Bragg Case	316
9.2.3 The Directions of the Neutron Current	318
9.3 Spatial Intensity Profiles and the Spherical Wave Theory	320
9.3.1 Ray Considerations	320
9.3.2 Spherical Wave Theory	323
9.4 Influence of Absorption	326
9.5 Magnetic Crystals	331
9.6 Applications	334
9.6.1 High Angular- and Energy-Resolution Experiments	334
9.6.2 Vibrating Crystals	339
9.6.3 Neutron Topography	340
9.6.4 Neutron Interferometry	342
References	348
<i>Subject Index</i>	353

1. Principles of Neutron Diffraction

H. Dachs

With 22 Figures

1.1 Introduction

The small velocities of thermal neutrons make possible the investigation of atomic motion in crystals and liquids. This application of neutron scattering, which is probably the most important, is treated in [1.1]. The present volume deals with the investigation of the structure of condensed matter using elastic and quasielastic neutron scattering. The dominating method in this area is the diffraction of x-rays (for recent treatments, see [1.2,3]). Chapter 1 discusses the differences between x-ray and neutron diffraction. The first section deals with the general theory of neutron diffraction. Sections 1.3 and 1.4 treat the differences in the experimental techniques. Most applications of neutron diffraction can be deduced from the different types of interaction of x-rays and neutrons with substances, dealt with in Section 1.2. Chapter 2 by HAYTER continues the introduction and discusses the production and possible applications of polarized neutrons.

It is not the purpose of this volume to discuss in great detail all possible areas of applications of neutron diffraction. This has already been done excellently in the book of BACON [1.4]. The purpose here is to emphasize the methodical aspects by treating in detail certain special areas. The area of crystal structure determination, for example, is represented by x-ray and neutron synthesis as discussed in Chapter 3. In this synthesis, exact electron densities can be determined by a combination of neutron and x-ray measurements. These investigations require high accuracy of measurement so that all methodical questions in the structure determination are important.

The magnetic moment of the neutron makes possible the determination of the arrangement of the magnetic moments in magnetically ordered materials. Such magnetic structure determinations are based largely on symmetry considerations which are an exciting development of the classical methods in crystallography. Chapter 4 is an introduction to this area.

Neutrons offer a series of important advantages for the investigation of disordered systems: the diffuse inelastic scattering can be isolated, the small angle scattering can be handled simply, and by change of isotopes a dyeing of certain-

components can be achieved. The theory and practice of disorder scattering is treated in Chapter 5.

The study of phase transitions is a particularly active research field at the present time, and many theoretical physicists have turned to this area. Their interest is especially in magnetic systems which can be exactly described. The experimental investigation of magnetic phase transitions and the accompanying critical fluctuations is a large and active field for neutron scattering. Lindgård, a theorist, gives a survey of this field which should be useful for experimentalists.¹

Chapters 7 and 8 treat diffraction in biological systems and liquids. In both cases, the use of isotope replacement is of great importance.

The volume closes with a chapter (Chap.9) on diffraction phenomena connected, specifically, with the use of perfect crystals. As these enable the construction of neutron interferometers [1.5], they are of particularly current interest. Since this chapter is the first review of this subject, an attempt was made to be as complete as possible.

1.2 Background

The neutron, which was discovered in the early 1930s, has been used in scattering experiments on crystalline solids since 1946 when the pioneering work of WOLLAN and SHULL started (see SHULL [1.6]).

Neutrons have no electrical charge, a mass almost the same as the proton, a nuclear spin of $1/2$, and a magnetic moment. The energies of neutrons ejected in the fission of uranium nuclei lie between 0.5 and 3 meV. In nuclear reactors these fission neutrons are thermalized or slowed down by collisions with light atoms in a moderator and used to fission further uranium nuclei. A chain reaction takes place. For neutrons thermalized to 20°C, the average energy is 0.025 eV, corresponding to a velocity of 2.2 km/s. From the de Broglie relationship $\lambda = h/mv$, the wavelength can be calculated. For 20°C neutrons the wavelength is 1.8 Å.

Thermal neutrons have, as in the case of x-rays, wavelengths of the same order of magnitude as the diameter of atoms and are, thus, also suited for the study of condensed matter. There are, however, enough differences between the two types of radiation to make neutron scattering an interesting complement to the x-ray technique.

¹A more detailed treatment will be presented in two forthcoming volumes of this series under the editorship of H. THOMAS and K.A. MOLLER.

1) The exchange forces which produce the scattering are different for the two cases: for neutrons they are due to nuclear and magnetic interactions, and for x-rays they are electrical.

2) The dependence of wavelength on energy is fundamentally different for the two cases. Thermal neutrons have velocities of the same magnitude as those of thermal waves in crystals, and this can give rise to a measurable Doppler shift of the scattered neutrons. This property makes possible the investigation of the dynamics of atomic systems. Essential differences in the scattering of neutrons and x-rays also exist if one is only interested in space ordering of atoms in systems such as liquids. The kinematics of the scattering process for neutrons must be treated more generally than that for x-rays.

The application of neutrons to study condensed matter is treated in a number of textbooks [1.4, 7-19]. Many references to the literature on neutron diffraction are to be found in the book of BACON [1.4] and in a special reference publication [1.20].

1.3 VAN HOVE Theory in the Classical Approximation

In the description of the interaction of neutrons with matter, concepts are used which are partly in accord with the wave picture and partly with the particle picture.

The particle density d is defined as the number of particles/volume, and the flux density ϕ as the number of particles moving through a unit area in unit time. These two densities are related by $\phi = d \cdot v$, where v is the particle velocity, since all particles in a cylinder of length v pass through the end of the cylinder in unit time. The various types of interaction are described by means of an effective cross section.

$$\sigma_i = \frac{\text{number of events of type } i/\text{time}}{\text{flux density}}$$

Cross sections have the dimensions of area, and as unit the barn ($1 \text{ barn} = 10^{-24} \text{ cm}^2$) is used.

For every special process there is a particular cross section, for example, there is one for absorption and one for scattering. In the case of scattering, a distinction is made between *total* and *differential* cross section. The calculation of the differential scattering cross section includes only those particles scattered into a particular element of solid angle and sometimes also those having frequencies in a certain interval.

The scattering can also be described in terms of the wave picture. Let

$$\psi = e^{2\pi i \frac{x}{\lambda}} = e^{i\mathbf{k}\mathbf{r}}$$

be the wave function of the incident neutrons. The neutron density is then $\psi \psi^*$, and the flux density is then

$$\phi = \psi \psi^* v$$

The wave function of the neutrons scattered from a single fixed nucleus is

$$-b \frac{e^{i\mathbf{k}\mathbf{r}}}{|\mathbf{r}|}$$

The wavelength of thermal neutrons is much larger than the diameter of the nucleus and the scattering is thus isotropic, and b is a constant, the so called scattering amplitude or scattering length. The relationship between the total scattering cross section and scattering amplitude is given by

$$\sigma = \frac{\text{flux scattered through a sphere centered on scattering center}}{\text{flux of incident wave}}$$

$$\sigma = \frac{4\pi r^2 b^2 \cdot \frac{1}{r^2} v}{v} = 4\pi b^2 \quad (1.1)$$

In order to treat a system of atoms, we describe it in the classical approximation by a special density function $\rho(\mathbf{r}, t)$ which is time dependent since the atoms are in motion. The density function has peaks of the δ -function type at those positions where the atoms are located, the peaks being weighted with the scattering amplitudes of the atoms. The density function can be written in terms of a Fourier integral

$$\rho(\mathbf{r}, t) = \int F(\mathbf{Q}, \omega) e^{-i(\mathbf{Q}\mathbf{r} - \omega t)} dV_Q d\omega, \quad (1.2)$$

where the integration is over reciprocal space and over all possible frequencies. The scattering from a time-independent density

$$\rho(\mathbf{r}) = \int F(\mathbf{Q}) e^{-i\mathbf{Q}\mathbf{r}} dV_Q \quad (1.3)$$

can be easily calculated using the Born approximation. The amplitude of the total scattered wave can be found by summing the waves scattered by each volume element, taking into account the phase differences. If \underline{k}_0 is the wave vector of the incident wave, \underline{k} that of the scattered wave, and \underline{r} the displacement vector between two scattering centers, then the phase difference between the two scattered waves is

$$\frac{2\pi\Delta}{\lambda} = \underline{r}(\underline{k} - \underline{k}_0) \quad , \quad (1.4)$$

where Δ is the difference in path length. The amplitude A of the scattered wave is

$$\begin{aligned} A(\underline{k} - \underline{k}_0) &= \int \rho(\underline{r}) e^{i(\underline{k} - \underline{k}_0)\underline{r}} dV_{\underline{r}} \\ &= F(\underline{k} - \underline{k}_0) \end{aligned} \quad (1.5)$$

Since the scattering due to a time-independent density function is elastic, $k=k_0$, the above results can be formulated in the following way. The differential scattering cross section for neutrons with momentum change

$$\Delta \underline{p} = (\underline{k} - \underline{k}_0)\hbar$$

is given by $|F(\underline{Q})|^2$ with $\underline{Q} = \underline{k} - \underline{k}_0$. The above considerations lead to the Ewald construction and finally to the Bragg law. Scattering occurs only for those Fourier-components for which the Bragg law is satisfied.

From the measurement of the scattered intensity it is not possible to determine the density function without additional information, since such a measurement does not determine the phase of the Fourier coefficients. What can be determined is the pair-correlation function, i.e., the probability of finding a second particle displaced by \underline{r} from a given particle weighted by the product of the scattering amplitudes. The pair-correlation function is obtained from the density function as follows:

$$\begin{aligned} G(\underline{r}) &= \int \rho(\underline{r}') \rho(\underline{r}' + \underline{r}) dV_{\underline{r}'} \\ &= \iiint F(\underline{Q}) e^{i\underline{Q}\underline{r}'} \cdot F(\tilde{\underline{Q}}) e^{i\tilde{\underline{Q}}(\underline{r}' + \underline{r})} dV_{\underline{Q}} dV_{\tilde{\underline{Q}}} dV_{\underline{r}'} \\ &= \int F(\underline{Q}) \cdot F(-\underline{Q}) e^{i\underline{Q}\underline{r}} dV_{\underline{Q}} = \int |F(\underline{Q})|^2 e^{i\underline{Q}\underline{r}} dV_{\underline{Q}} \end{aligned} \quad (1.6)$$

$\hbar = h/2\pi$ (normalized Planck's constant)

In crystallography, the pair-correlation function is called the Patterson function, and is the Fourier transform of the quantities $|F(Q)|^2$ directly obtainable by intensity measurements. The density function can only be obtained from the pair-correlation function in special cases. There exist special methods for determining the phases experimentally, and they can also be obtained if it is known how many and what type of atoms the density function represents.

For the time-dependent part of the general density function, the components of the Fourier representation are traveling waves and not standing waves as in the time-independent case. A Doppler shift occurs and the energy of the neutrons is not conserved in the scattering process. In order to treat this case, a transformation is made to a coordinate system in which the density wave being considered is at rest, and in which the Bragg law is satisfied, i.e., the transformed velocities of incoming and outgoing neutrons are equal in magnitude. From Fig.1.1 it

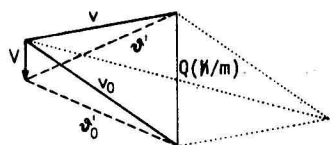


Fig.1.1 Velocity diagram for the scattering of a neutron from a density wave moving with velocity V . In the laboratory system the velocities of the incident and scattered neutron are v_0 and v , respectively. The magnitudes of these velocities are equal in the coordinate system moving with the density wave

is found that the changes in momentum and energy of the neutrons are determined, respectively, by the velocity and frequency of the density wave, i.e.,

$$\frac{v - v_0}{\hbar} m = \frac{v' - v'_0}{\hbar} m = Q \quad (1.7)$$

$$(v - v_0) (v + v_0) = 2QV \frac{\hbar}{m}$$

$$\frac{1}{2} m (v^2 - v_0^2) = \Delta E = \hbar \omega \quad (1.8)$$

The corresponding double differential cross section is

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega} = \frac{k}{k_0} |F(Q, \omega)|^2 = \frac{k}{k_0} S(Q, \omega) \quad (1.9)$$

where $|F(Q, \omega)|^2$ is the particle density of the scattered wave. The multiplication with $k:k_0 = v:v_0$ gives the flux.

The quantity

$$\int S(Q, \omega) e^{i(Qr - \omega t)} dV_Q d\omega = G(r, t) \quad (1.10)$$

is the time-dependent pair-correlation function, i.e., the weighted probability of finding a particle at r at time t , given another particle at the origin at time zero. The knowledge of this function is the maximum information which can be obtained from a scattering experiment. A more exact theory of the above processes, as developed by VAN HOVE [1.21], must also take into account the change of state of the scatterer in inelastic scattering. This theory is the starting point in discussions of all possible scattering experiments.

In a scattering experiment, the following quantities are to be determined:

- 1) the change of momentum of the neutrons
- 2) the energy change caused by the scattering
- 3) the scattered intensity, i.e., the frequency with which a scattering process occurs.

In order to determine the first two quantities above, the velocity of the neutrons must be measured before and after scattering.

Different types of scattering experiments can be classified according to the accuracy and range in which momentum and energy changes can be measured. The most general goal of a scattering experiment that can be imagined is to determine the space-time-correlation function for a system. In order to do this, the scattered intensities for a series of momentum and energy changes must be determined. The spread of the intensity measurements in Q and ω set boundaries on the resolution of the space variable r and speed, respectively, of the processes which can be determined.

The space-time-correlation function is used to describe the dynamics of liquids and other disordered systems. Because of the isotropy of liquids, the pair-correlation function depends only on $|r|$, and the corresponding scattering function is a continuously varying function of $|Q|$, i.e., $S(Q, \omega) = S(|Q|, \omega)$. Time-of-flight measurements are used to determine $S(|Q|, \omega)$. Two choppers (disk with slits) rotating with the same angular velocity but with a phase difference are used to generate constant-velocity neutron pulses of short duration. These pulses impinge on a sample and the scattered neutrons are registered by detectors placed at a fixed distance from the sample. The detection of the scattered neutrons is made as a function of the flight time from sample to detector. By the use of a system of detectors, many different values of $S(Q, \omega)$ can be measured at the same time (Fig. 1.2).

For crystalline solids, $S(Q, \omega)$ splits into different branches corresponding to the acoustical and optical modes of vibration existing in these substances. Here one is interested in the dispersion relation which gives the relationship between frequency and wavelength of the phonons. In order to determine the dispersion relation it is not necessary to include the whole Q, ω space in the determination, but in principle only the part of this space known as a Brillouin zone. The dispersion relation in the Brillouin zone must be accurately determined, and a triple-