MODERN PROBLEMS IN CONDENSED MATTER SCIENCES

General Editors:
V.M. AGRANOVICH and A.A. MARADUDIN

VOLUME 8

OPTICAL ORIENTATION

Volume Editors:

F. MEIER and B. ZAKHARCHENYA

North-Holland

OPTICAL ORIENTATION

F. MEIER
ETH-Hönggerberg
CH-8093 Zürich, Switzerland

B.P. ZAKHARCHENYA
A.F. Ioffe Physico-Technical Institute
USSR Academy of Sciences
Leningrad 194021, USSR

1984

NORTH-HOLLAND
AMSTERDAM·OXFORD·NEW YORK·TOKYO

ISBN: 0444 86741 4

Published by:

North-Holland Physics Publishing

a division of

Elsevier Science Publishers B.V.
P.O. Box 103
1000 AC Amsterdam
The Netherlands

Sole distributors for the U.S.A. and Canada:

Elsevier Science Publishing Company, Inc.

52 Vanderbilt Avenue
New York, N.Y. 10017
U.S.A.

Library of Congress Cataloging in Publication Data
Main entry under title:

Optical orientation.

(Modern problems in condensed matter sciences ; v. 8)
Bibliographies: p.
Includes indexes.
1. Semiconductors--Optical properties. 2. Spin-lattice relaxation. 3. Photoemission. I. Meier, F. (Felix), 1943- . II. Zakharchenia, Boris Petrovich. III. Series.
QC611.6.O6066 1984 537.6'22 84-14705
ISBN 0-444-86741-4

Printed in The Netherlands

OPTICAL ORIENTATION

MODERN PROBLEMS IN CONDENSED MATTER SCIENCES

Volume 8

NORTH-HOLLAND
AMSTERDAM·OXFORD·NEW YORK·TOKYO

MODERN PROBLEMS IN CONDENSED MATTER SCIENCES

Vol. 1. SURFACE POLARITONS
V.M. Agranovich and D.L. Mills, *editors*

Vol. 2. EXCITONS
E.I. Rashba and M.D. Sturge, *editors*

Vol. 3. ELECTRONIC EXCITATION ENERGY TRANSFER IN CONDENSED MATTER
V.M. Agranovich and M.D. Galanin

Vol. 4. SPECTROSCOPY AND EXCITATION DYNAMICS OF CONDENSED MOLECULAR SYSTEMS
V.M. Agranovich and R.M. Hochstrasser, *editors*

Vol. 5. LIGHT SCATTERING NEAR PHASE TRANSITIONS
H.Z. Cummins and A.P. Levanyuk, *editors*

Vol. 6. ELECTRON–HOLE DROPLETS IN SEMICONDUCTORS
C.D. Jeffries and L.V. Keldysh, *editors*

Vol. 7. THE DYNAMICAL JAHN–TELLER EFFECT IN LOCALIZED SYSTEMS
Yu.E. Perlin and M. Wagner, *editors*

Vol. 8. OPTICAL ORIENTATION
F. Meier and B.P. Zakharchenya, *editors*

Oh, how many of them there
are in the fields!
But each flowers in its
own way —
In this is the highest achievement
of a flower!

Matsuo Bashó
1644 - 1694

PREFACE TO THE SERIES

"Modern Problems in Condensed Matter Sciences" is a series of contributed volumes and monographs on condensed matter science that is published by North-Holland Publishing Company. This vast area of physics is developing rapidly at the present time, and the numerous fundamental results in it define to a significant degree the face of contemporary science. This being so, it is clear that the most important results and directions for future developments can only be covered by an international group of authors working in cooperation.

Both Soviet and Western scholars are taking part in the series, and each contributed volume has, correspondingly, two editors. Furthermore, it is intended that the volumes in the series will be published subsequently in Russian by the publishing house "Nauka".

The idea for the series and for its present structure was born during discussions that took place in the USSR and the USA between the former President of North-Holland Publishing Company, Drs. W.H. Wimmers, and the General Editors.

The establishment of this series of books, which should become a distinguished encyclopedia of condensed matter science, is not the only important outcome of these discussions. A significant development is also the emergence of a rather interesting and fruitful form of collaboration among scholars from different countries. We are deeply convinced that such international collaboration in the spheres of science and art, as well as other socially useful spheres of human activity, will assist in the establishment of a climate of confidence and peace.

The General Editors of the Series,

V.M. Agranovich A.A. Maradudin

PREFACE

This contributed volume represents the first comprehensive survey of studies of the optical orientation in semiconductors performed in the recent 15 years by scientists of France, Switzerland, the USA and the USSR.

The history of research into the optical orientation of the electron and nuclear spins in semiconductors is expounded in sufficient detail in the introductory chapters, therefore we shall not dwell on it in this preface.

The volume consists of two parts.

The first part covers studies dealing with the effects of optical orientation and alignment of electrons and excitons, as well as studies of the optical polarization of nuclei originating from the hyperfine interaction of spin-polarized electrons with nuclei of the crystal lattice.

The second part includes studies of the optically oriented photoelectrons emitted from crystals.

In commenting on the first part, it should be stressed that many effects observed under optical polarization in semiconductors are in their physical nature very close to the phenomena revealed in gases under optical pumping. At the same time in a system of strongly coupled electron and nuclear spins oriented by light one observes effects which do not have counterparts in the optical pumping of gases. This manifests itself particularly clearly in very weak external fields. This problem is considered in detail in several chapters of this volume. We believe, however, this point to be sufficiently important to warrant mentioning in the very beginning of the book.

Although in the pioneering experiments of G. Lampel in 1968 optical orientation in silicon was revealed by the conventional NMR technique, the main method of detecting spin orientation under optical pumping in semiconductors has now become the study of the degree of polarization of recombination radiation. R.R. Parsons was the first to use this method to advantage.

The method of optical orientation combined with the optical detection of polarized electrons, excitons and holes provided a possibility to measure band structure parameters with a high precision, and to study picosecond-scale processes occurring in a system of free carriers and excitons. Optical orientation studies have led to the development of very sensitive methods for the optical detection of magnetic resonances. This permits one to observe resonances in extremely small samples, e.g. in very thin epitaxial semiconducting

films. It is this technique that has revealed forbidden resonant transitions whose detection by conventional classical methods would be difficult if at all possible.

The optical detection method is applicable only in cases where the lifetime of the light-excited electrons is less than their spin relaxation time. For this reason it has not yet been employed in studies of indirect-band semiconductors, e.g. silicon. Optical polarization in silicon is detected by conventional NMR. It was found that the NMR signal of optically polarized nuclei in specially doped silicon is hundreds of times that observed in the experiments of 1968. This provided a basis for development of optical polarization studies on silicon and for accumulation of extensive information on the spin relaxation of nuclei including their spin diffusion in semiconductors with both homogeneous and inhomogeneous distribution of impurities and defects. Unfortunately, no review of these works has been included in the present volume, the introductory chapter by Perel' and Zakharchenya containing not more than a brief summary of them.

One should mention here also the experiment of Vlasenko, Fleisher and Zavaritskii in 1983 who have succeeded in detecting with a quantum interferometer SQUID the polarization of optically oriented nuclei in silicon. Since it is a very recent experiment, it likewise has not been included in the book.

The second part of the book deals with applications of optical spin orientation in photoemission. In spite of the rapid development of photoelectron spectroscopy the effort to measure the spin polarization of the emitted electrons was undertaken only by few groups, although the feasibility of such experiments had already been shown by Siegmann and coworkers in 1968. The early spin polarized photoemission experiments dealt exclusively with magnetic materials where spin polarization is present already in the ground state of the system. Among the notable successes of this technique is e.g. the observation of negative spin polarization of the electrons at the Fermi surface of nickel. The year 1974 marked the beginning of a considerable expansion of the field of spin-polarized photoemission: it was shown that highly polarized electrons can be emitted also from materials with no spin order in the ground state. Then the polarization is obtained by the optical excitation process itself, i.e. by optical spin orientation.

Optically oriented photoelectrons were first observed with GaAs, many years after the mechanism of optical spin orientation in solids had been shown to be effective by other experimental techniques. This experiment had a rather remarkable impact also on other electron spectroscopies since it provided the experimentalists practically overnight with a very efficient source of polarized electrons. Applications of polarized electron beams in atomic, solid-state and high-energy physics are described in the chapter by Pierce and Celotta. Although the GaAs-source has proven useful in many instances, there is still a challenging problem left: to replace it by a device which yields not a 50% but a

100% polarized electron beam (all electron spins pointing into the same direction) without sacrificing the other qualities like high intensity and possibility of polarization reversal without changing the other beam parameters.

Soon after photoelectrons were optically oriented in GaAs it became clear that the method bore an exciting potential as a general spectroscopic tool. As will be discussed in the chapters by Wöhlecke and Borstel and by Meier and Pescia the polarization of the photoelectrons created in a particular transition is determined simply by the symmetry of the electronic states involved. When observing transitions between energetically closely spaced initial states (e.g. d-bands in transition metals) the sign of the polarization alone may be very useful to identify the symmetry character of an orbital. Increased resolution compared to conventional techniques will be obtained in the frequently encountered case where two adjacent transitions yield polarizations of opposite sign. The sensitivity of the polarization spectra with respect to the surface atomic arrangement has not yet been exploited at all, e.g. for observing structural phase transitions or adsorbate geometries. Perhaps the most important application of optical spin orientation is the experimental determination of the hybridization of energy bands. This is an important property of the electronic states which cannot be derived generally from simple energy- and angle-resolved photoemission.

It seems that the mechanism on which optical spin orientation rests is by now rather well understood. What still has to be done is to make full use of this still novel technique.

F. Meier
Zürich, Switzerland

B.P. Zakharchenya
Leningrad, USSR

CONTENTS

CHAPTER 1

Major Physical Phenomena in the Optical Orientation and Alignment in Semiconductors

V.I. PEREL' and B.P. ZAKHARCHENYA

A.F. Ioffe Physico-technical Institute
USSR Academy of Sciences 194027 Leningrad
USSR

Optical Orientation
Edited by
F. Meier and B.P. Zakharchenya

The basic principles of optical orientation have grown out of the brilliant studies by A. Kastler and his followers. A review of these studies can be found in Kastler's Nobel Prize Award lecture (1967).

In the 1920s, the pioneering works of Wood and Hanle revealed an effect of a weak magnetic field on the polarization of the resonance fluorescence of mercury and sodium vapors. The importance of these works was not recognized until 1949. In that year, a paper by Brossel and Kastler appeared and initiated a systematic investigation of the phenomena making up the branch of physics we now call optical pumping (see reviews by Happer 1972, and Aleksandrov 1973).

One can safely say that all processes in physical systems originate essentially from the same main source, to wit, unequal population of different states. This inequality under thermodynamic equilibrium comes naturally from the Boltzmann distribution by whose virtue the population of a state is the lower the higher is its energy. The population of states of the same energy, i.e. of degenerate states, is, however, the same under equilibrium, so that external action cannot induce transitions between them. Degeneracy is caused, as a rule, by symmetry, and in order to observe these transitions, one has to remove the degeneracy by applying an external field which changes the symmetry of the system. This underlies the classical methods of detection of electron spin and nuclear magnetic resonances which require application of sufficiently strong magnetic fields and low temperatures to raise the difference in the sublevel populations to values compatible with the equipment sensitivity.

An equilibrium distribution can be changed by acting upon the system by light from a narrow spectral region which induces transitions between levels by reducing the population of the lower level and increasing that of the higher level. We have in mind here widely separated levels with strongly differing populations under equilibrium. From the viewpoint of purely energy considerations, optical excitation leaves the population of degenerate states unchanged.

The main outcome of the investigations of Kastler and his coworkers was gaining a clear understanding and practical implementation of the fact that optical excitation results in a different population of degenerate or nearly degenerate sublevels which belong to levels coupled through an optical transition. The reason for this lies in the anisotropy of optical excitation, caused by the transversality of the electromagnetic wave. This anisotropy can be substantially enhanced by using for excitation light which is circularly or linearly polarized. This is what underlies the optical pumping method.

The efficiency of the method is due to the fact that in most cases relaxation mixing between degenerate or nearly degenerate states proceeds much slower than the processes returning a system from the excited to ground state.

Optical pumping provides the possibility of not only producing a difference in the population of degenerate states but of detecting this difference as well. With pumping carried out in an excited state, detection is obtained by the polarization of spontaneous radiation. When pumped in the ground state, bleaching of the medium occurs (fig. 1).

Optical pumping provides a remarkable possibility of optical detection of RF resonances. If we increase the separation between degenerate sublevels by applying, e.g., a magnetic field, and induce transitions between them by means of an RF field, the onset of resonance will manifest itself in a change of the luminescence polarization or in the absorption of light (when pumping in the ground state). In this case there is no need to perform measurements at low temperatures, and one may use comparatively weak magnetic fields (and, hence, low frequencies) since the magnetic field has not any more the function of creating a population difference.

When linearly polarized or unpolarized light is used for excitation, the so-called alignment of atoms occurs, which means that the electron shell is no more isotropic.

If the degenerate levels in question are associated with different orientations of the angular momenta and circularly polarized light is used for excitation, then optical pumping will result in the angular momenta becoming oriented. Optical orientation is a direct consequence of angular momentum conservation. When polarized photons are absorbed, their momentum is transferred to the medium.

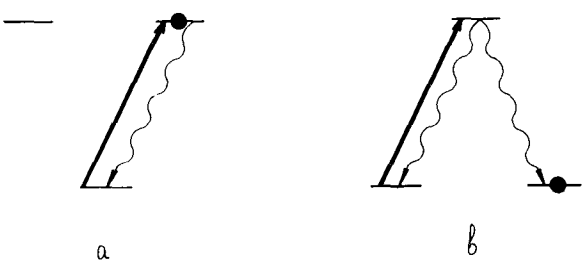

Fig. 1. Schematic of optical pumping of (a) excited and (b) ground state. Straight arrows indicate optical excitation. Wavy lines denote relaxation processes (including spontaneous emission) returning system into ground state. Point shows which of the two degenerate states is preferentially populated. Figure shows a limiting case, where only optical transitions between one pair of levels are allowed by the selection rules. In a general situation, all or almost all transitions are allowed, but with different intensities.

It is important that optical orientation is possible also in systems which interact only weakly with light, if at all. Indeed, while only the electron orbital momenta are oriented directly by light, the electron spins become polarized through spin–orbit interaction. Hyperfine interaction results also in orientation of the nuclear spins. The electron or nuclear spin orientation affects the luminescence polarization, thus making possible optical detection of electron spin and nuclear magnetic resonances.

We have already mentioned the effect of magnetic fields on the resonance luminescence polarization. Studying the magnetic field dependence of luminescence polarization (the Hanle effect) has become a traditional tool used frequently in optical pumping experiments. One could suggest more than one explanation of this phenomenon, the classical interpretation being the simplest.

Since the circular polarization of the luminescence originates from a transfer of angular momentum from electrons to light, the degree of this polarization is proportional to the projection of the average electron spin onto the direction of observation. Now if the excitation is collinear with observation (along the Z-axis) and the magnetic field is directed perpendicular to it (X-axis), then what will happen in steady state is as follows: the light will orient the excited electrons along the beam, with the magnetic field turning the spins, so that the spin distribution of the electrons in the $Z-Y$ plane will at any time have the form of fig. 2. As the magnetic field increases, the average projection of the spin onto the Z-axis decreases and, hence, the degree of circular polarization of the luminescence will likewise drop. The polarization practically disappears if the product of the angular rate of spin precession in the magnetic field and the spin lifetime becomes large. Such measurements can obviously yield the magnitude of the spin lifetime. Within the context of quantum theory,

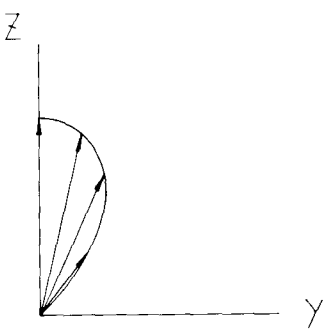

Fig. 2. Photoexcited electron spin distribution under steady-state excitation (excitation along Z-axis, magnetic field along X-axis). Arrows denote total spin of electrons excited previously. The earlier has been the excitation, the larger will be the angle of spin rotation and the smaller the magnitude of the spin, because of the process of spin relaxation and de-excitation involved.

circular polarization results from the spin-up and spin-down states being coherent. When the separation between corresponding states becomes greater than their width, the coherence disappears, as does also the circular polarization of the luminescence. It should be emphasized that only the homogeneous broadening of the levels, which is connected with the spin lifetime through the uncertainty relation, is essential here, while the various mechanisms of inhomogeneous broadening (such as the Doppler effect in gases, random crystalline fields) are of no importance. This provides the possibility of measuring homogeneous broadening parameters related to the lifetime and spin relaxation time under conditions where it is totally masked by inhomogeneous broadening.

The main aspects of the optical orientation phenomenon (or, in a more general sense, of optical pumping) are of a universal nature. However, optical pumping experiments have been carried out for a long time on isolated gas atoms only.

In solid state physics, optical orientation methods were originally employed under conditions close to the gas state: indeed, one studied transitions between impurity center levels in crystals (Karlov et al. 1963, Anderson et al. 1966, Mollenauer et al. 1968, and others). The specific direction of these studies is connected with the triplet exciton orientation in molecular crystals. One detected here (Maier et al. 1967) nuclear polarization originating from the dynamic polarization of the anthracene protons by oriented excitons.

Lampel in 1968 carried out an experimental investigation on electrons in semiconductors using silicon for this purpose. Because of an unfavorable relationship between lifetimes and spin relaxation time, the degree of electron polarization was small, however it was sufficient for polarizing the lattice nuclei. The nuclear spin polarization was detected by the conventional NMR technique.

In the subsequent years, all the major classical methods of optical pumping were used in the investigation of electrons in semiconductors:

- detection of the electron orientation by circular photoluminescence polarization and depolarization of radiation in a magnetic field, i.e., by the Hanle effect (Parsons 1969, Ekimov and Safarov 1970, Zakharchenya et al. 1971);
- optical detection of NMR (Ekimov and Safarov 1972, Berkovits et al. 1974) and ESR (Hermann and Lampel 1971);
- orientation of the majority current carriers, which is the counterpart of optical orientation in the ground state (Dyakonov and Perel' 1971a, Ekimov and Safarov 1971).

The specific behavior of the electrons and nuclei in semiconductors manifests itself in all the aforementioned phenomena. In crystals, processes of electron spin relaxation occur (Dyakonov and Perel' 1971b, Bir et al. 1976) which do not have counterparts in gases. The Hanle curve may exhibit distortions as a result of spin diffusion (Vekua et al. 1976).