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Introduction

Mankind has always been fascinated by optical properties of solids. Besides manufacturing and availability aspects it has been the color of metals like gold, silver, and bronze that defined their outstanding ornamental and decorative value in history. The same is true for gemstones and minerals due to their colorful appearance and their intense reflections caused by a high index of refraction.

Whereas most gemstones are insulators, the precious metals are more or less good conductors. Only fairly recently another class of materials, the semiconductors, have caught the interest of researchers, engineers, and technicians. Semiconductors are situated somewhere in between metals and insulators, even though the exact boundaries are often not that well defined. Many aspects of our modern civilization are based on semiconductors. Virtually all technical equipment for everyday use contains semiconducting material in one form or another.

The first semiconductor applications used their unique transport properties. Contacts of semiconductors with metals were used in rectifiers and diodes. Then the invention of the transistor led to fabulous developments, most prominently in computing and information technology. The application of optical and optoelectronic semiconductor properties was somewhat delayed, mainly because of the fact that the initially dominant semiconductors, silicon and germanium, have indirect optical gaps and are thus not well suited for optical devices. Today, however, compound semiconductors like GaAs and its relatives are basis materials for optoelectronic applications, e.g., in light-emitting diodes, semiconductor lasers, etc. Besides the GaAs-like materials that are composed of elements from groups III and V of the periodic system, also II–VI compounds, as well as ternary, or even quaternary systems are becoming increasingly relevant.

Over the years, fundamental research on the optical properties of semiconductors has revealed a large number of fascinating effects. However, a real boost came with the development of techniques that allow for controlled atomic growth and thus nanoscale structuring of semiconductor materials.

Using epitaxial growth techniques and modern lasers that emit ultrashort light pulses in the range of femtoseconds makes it now possible to study optoelectronic linear and nonlinear processes on ultrafast time scales in structures characterized by length scales down to a few nm. Using structuring on nanometer (nm) scales the spatial dimension became a relevant physical parameter. Effectively zero-, one-, and two-dimensional systems can be fabricated and studied by growing suitable semiconductor heterostructures. These nanostructures may resemble atoms (quantum dots), wires (quantum wires), films (quantum wells), superlattices (periodic arrays of quantum wells), etc.

Using ultrafast laser pulses, electronic processes can be monitored on time scales that are comparable to typical interaction times among the electronic excitations. It is also possible to investigate the dynamics of the coherently excited many-particle system on time scales shorter than typical decoherence times. This coherent regime has attracted the interest of researchers most recently, as the concepts of "ultrafast switching", "coherent control", or "quantum computing" rely on processes which are destroyed by phase-breaking interactions.

By now, a large amount of experimental and theoretical work on optical properties of semiconductors in the coherent ultrafast time regime exists. The interpretation of the experimental findings requires a thorough treatment of the semiconductor as a many-particle system, where the often dominant processes result from the Coulomb interaction between the optically excited charge carriers. In addition, realistic semiconductor nanostructures always show a certain degree of disorder, which has a profound influence on their optoelectronic properties. One often applies external ac or dc electrical or magnetic fields to semiconductor structures. Such fields lead to new dynamical processes which might be useful for future applications.

1.1 Coherence

The dynamics of a classical plane wave with given wave vector and frequency is determined by the solution of the relevant wave equation. Generally, the wave is described by a spatially and temporally varying field $\Psi(\boldsymbol{r}, t)$. The propagation of this wave function in free space is called *coherent* if, by knowing the amplitude and phase at a given point in space at a given time, we can immediately tell the amplitudes and phases at all other times and space points.

In this book, we are dealing exclusively with temporal, not spatial coherence. A formal definition of temporal coherence of an arbitrary function $f(t)$ is given by the autocorrelation function

$$F(t) = \lim_{T\to\infty} \frac{1}{T} \int_0^T f^*(t')f(t'+t)dt\,. \tag{1.1}$$

The complex conjugate has been used in this definition in case the function is complex valued. If we find that $F(t)$ decays in time, we say that the coherence

of the function $f(t)$ is characterized by a finite coherence time, i.e., the function $f(t)$ is subject to *dephasing*.

In most experiments, it is not the function $f(t)$ which describes, e.g., an electric field, that is measured. In a statistical ensemble, the expectation value of the field is often zero, i.e.,

$$\langle f(t) \rangle = 0\,. \tag{1.2}$$

In such a case, one may still record the related intensity I_f, which is given by

$$I_f(t) = \langle |f(t)|^2 \rangle. \tag{1.3}$$

The intensity is an equal-time autocorrelation function and can be nonzero even if $\langle f(t) \rangle = 0$.

Consider that $f(t)$ is the superposition of two functions $f_1(t)$ and $f_2(t)$.

$$f(t) = f_1(t) + f_2(t). \tag{1.4}$$

One then measures

$$\langle |f_1(t) + f_2(t)|^2 \rangle = \langle |f_1(t)|^2 \rangle + \langle |f_2(t)|^2 \rangle + 2\Re\langle f_1^*(t) f_2(t) \rangle. \tag{1.5}$$

Here, the third term on the right-hand side describes the interference of the two fields f_1 and f_2. This term is finite only if f_1 and f_2 are at least to a certain degree coherent to each other. Thus, in order to observe interference the temporal evolution of the phases of f_1 and f_2 must be determined by the coherent dynamics governed by, e.g., a wave equation. If, however, the dynamics of either f_1 or f_2 is dominated by incoherent processes, e.g., scattering events, interference is absent.

In nature, particles and waves interact with the surrounding media. The resulting *scattering processes* depend both on the nature of the particle or wave and that of the medium. In order to determine the trajectory of a classical particle, one has to insert all the (possibly time-dependent) forces into Newton's equation of motion and solve for $\boldsymbol{r}(t)$, i.e., compute the particle's position as a function of time. In many cases one does not have the full information about all the acting forces, but only knows some statistical properties. In such a case, one has to resort to a statistical description yielding distributions of the dynamical variables of the particle.

In the case of waves we can distinguish two different cases:

(i) Imagine a source that emits a stationary wave into free space. At some point, this wave enters a medium that is characterized by static scattering centers. The wave is then scattered in such a way that a distorted wave pattern is formed. For a stationary situation this wave pattern is also stationary and is therefore still coherent. It can be computed from the wave equation, where the stationary scattering processes have to be included. This means, in particular, that the phases in all points are rigidly

correlated. Examples are Bloch waves of electrons in atomic lattices and electron waves in disordered solids as well as light waves in photonic crystals and in disordered dielectrics. Also, waves in random stationary media are coherent in this sense, although their wave pattern can be very complicated.

(ii) If the medium possesses scattering centers that have some temporal dynamics, e.g., such as those of a gas of particles, we could still try to include this dynamics into the wave equation and attempt a solution for the scattered wave. However, often the trajectories of the scattering particles are not known in detail. Only their statistical properties can be specified, e.g., by giving the temperature and the density if the scattering medium is a thermal (heat) *bath*. Since we do not have full knowledge of the dynamics of the bath particles, we cannot hope to obtain all possible information about the scattered wave. Only some average quantities are accessible in such a situation. As the intensity of the scattered wave is related to some kind of local energy density, it seems reasonable to assume that we can still obtain decent information about the spatial and temporal behavior of the amplitude. However, the information about the phases is partially or completely lost. In this case, one says that the phase has suffered phase-breaking interactions or dephasing. In other words, the wave-like excitation is no longer coherent. Since the information about the intensity is less sensitive, we arrive at a description where some local object, e.g., a maximum of the intensity, moves in space. This is strongly reminiscent of the dynamics of a classical particle. In fact, dephasing destroys the wave-like features that are introduced into the description by using quantum-mechanical concepts, and we are left with a classical description of particle motion.

In the coherent regime, on the other hand, we retain the full coherent nature of the excitation, which this book focuses on. We will see, however, that even in this coherent regime *optical phase coherence* can be limited if this notion is defined in terms of certain properties of experimentally determined transient signals.

1.2 Basic Optical Principles

The measurement of optical properties of solids often follows the scheme where one obtains information about the material system by applying an external field and recording the response of the system to this field. In many cases, one is interested in the equilibrium properties of the material system. These properties can be investigated by experiments in the linear response regime, where the response linearly depends on the excitation field. Examples are linear optical absorption spectra, refractive index, etc.

Modern semiconductor optics, however, often investigates nonlinear optical properties. In particular, if dynamical processes are of interest, one often

applies sequences of ultrashort laser pulses to the system and records the subsequent dynamical response. Examples are pump–probe and four-wave-mixing experiments. The nonlinear response can either be measured in the frequency domain (bleaching, induced absorption, etc.) or in the time domain (decay, echoes, quantum beats, etc.).

An interesting application of ultrafast optical measurements is related to transport properties. As the laser field generates populations of electrons and holes, the subsequent spatio-temporal dynamics of these particles can be investigated. For these studies, local excitation and detection schemes can be applied, e.g., using near-field optical microscopy. Such studies yield information about the interesting and intricate interplay between structural disorder and many-particle interactions and their influence on the spatio-temporal dynamics of the optically generated excitations. Alternatively, coherent-control schemes make it possible to induce particle currents on an ultrafast time scale by homogeneous optical excitation of the semiconductor system.

Besides optical excitation, also externally applied electric and magnetic ac and dc fields lead to interesting dynamical signatures. Examples are Bloch oscillations in dc-biased superlattices, dynamical localization in an ordered semiconductor induced by an ac electric field, and Aharonov–Bohm oscillations of excitons and biexcitons in semiconductor rings subjected to a magnetic flux. Since these dynamical effects are related to moving charges, the system emits electromagnetic radiation. The upper time limit at which the mentioned coherent dynamical processes can be observed is determined by phase-breaking interactions. As these occur typically on time scales in the picosecond range for the case of excitonic excitations in typical semiconductors, parameters have to be chosen such that the relevant periods of the coherent oscillations are shorter than this time limit. Considering picosecond periods, the emitted signals thus have frequencies in the terahertz range.

1.3 Relevant Material Systems

Most of the experiments mentioned so far have been performed mainly on semiconductor nanostructures with reduced effective spatial dimensions. Here, one-dimensional quantum wires and two-dimensional quantum films and arrays composed of these structures are widely used examples. The III–V system GaAs and similar III–V systems are prototype materials. However, II–VI systems and some wide-gap III–V compounds like GaN have larger excitonic binding energies. Therefore, these materials are considered to be useful candidates for some applications. However, the majority of experiments have been performed on GaAs-type semiconductor heterostructures. These also dominate most applications in information technology. In this book, we therefore predominantly take fundamental electronic properties of this class of materials as a basis for the parameters of our schematic model. These include

effective electron and hole masses, exciton binding energies, optical selection rules, etc. However, extensions to other material parameters are often straightforward.

In the main part of our calculations, we concentrate on a one-dimensional model, the evident realization of which would be quantum wires. However, in many cases the predictions of the model do not strongly depend on the dimensionality of the system. Therefore, they can, *mutatis mutandis*, be taken as a guideline also for experiments on two-dimensional or three-dimensional heterostructures like quantum wells and superlattices.

1.4 Related Systems

As mentioned above, the results obtained on the basis of the schematic one-dimensional model can be easily transferred to one-dimensional heterostructures like quantum wires. One could also attempt to apply the predictions to other, more natural one-dimensional systems like polymeres, liquid crystals composed of disc-shaped molecules (discotic crystals), and even to biological systems. However, these systems belong to the class of highly correlated electron systems, and the theoretical approach used here is often not directly applicable. The linear optical spectra of such systems are studied in ongoing research and the investigation of dynamical processes of optical excitations is a fascinating field that appears to be far from being settled yet.

1.5 Aim of the Book

Although considerable progress has been achieved in the theoretical description of coherent optical properties of semiconductor structures, a full theoretical treatment taking into account all details of the real heterostructure, the full many-particle Coulomb interaction, disorder, coupling to lattice vibrations and external fields is beyond the capability of even the largest modern computing facilities. Therefore, one is forced to simplify the theoretical model by neglecting certain complications or by considering a simplified electronic or atomic structure. Even then, a numerical simulation of the dynamical processes often presents a formidable problem. In particular, the many-particle interaction leads to a hierarchy of equations or to an infinite number of relevant terms.

In this book, we present a discussion of coherent semiconductor optics starting from the simplest possible model of a semiconductor. The advantage of this approach, besides leading to mathematically transparent equations and numerically tractable simulation schemes even for situations including external electric and magnetic fields and disorder, lies in the fact that the physical principles of the various dynamical processes can be clarified and introduced without being distracted by numerous details necessary to describe

a real semiconductor structure. On the other hand, one should not expect that such a schematic model quantitatively accounts for all of the detailed features observed in experiments on a particular structure.

Nevertheless, for the time being, a number of new predictions and interpretations concerning dynamical processes of optical excitations in the coherent regime is only possible on the basis of such a schematic model. Although it is conceivable that these theories will be substantiated for realistic situations once the next generation of computers is available, the underlying general physical principles can most easily be perceived on the basis of the schematic model.

The presentation of this book is based on a one-dimensional tight-binding model for a semiconductor with finite length. If periodic boundary conditions are applied, this model represents a ring-like structure. The theoretical treatment will in most cases make use of a real-space representation, which allows us to incorporate disorder in a most natural way and to consider structures with a finite length.

There is also a tutorial reason for working with such a model system. Coherent optical properties were first observed for atomic and molecular systems because of their much longer phase relaxation times compared to solids. The theoretical models used in this context were based on systems having only a few single-particle energy levels. These few-level systems allow for an easy and transparent introduction of many dynamic processes initiated by ultrashort laser pulses. The transfer of the developed concepts to a solid is then relatively straightforward. Complications and fascinating differences to few-level systems arise *mainly* from the many-particle Coulomb interaction which has to be implemented into the model. Coulomb effects are responsible for the mutual repulsion of carriers with equal charges and for new resonances due to the attractive interaction between the oppositely charged electrons and holes.

When introducing students to the field of semiconductor optics, we realized that, although there is a large amount of theoretical work on dynamical processes of optical excitations in the coherent regime, a presentation that summarizes the fundamental principles and explanations without going into material-specific details is lacking. In this book, the material is therefore presented at a level useful for students intending to work in the field of semiconductor optics. It is also suitable for researchers more interested in applications of optoelectronic devices as it provides a basis for a fundamental understanding of optical properties of semiconductor heterostructures in the coherent regime.

This book consists of three parts. In Part I we develop the theoretical concepts and the equations of motion, which form the basis of the following two parts. Part II deals with applications of the theory to level systems and the semiconductor model, while Part III is devoted to special dynamic properties of semiconductors that can be studied by coherent optical experiments.

1.6 Necessary Prerequisites

In the first place this book is written for students intending to learn the principles of modern semiconductor optics. They should have some knowledge of quantum mechanics, including the basics of second quantization. In the introductory chapters describing few-level systems we have, however, avoided using second quantization and apply the Dirac bra and ket notation. This is sufficient since many-particle interactions are not considered here. However, for the discussion of many-particle interaction effects in the chapters dealing with the semiconductor system, second quantization techniques are needed.

Some background knowledge of solid state physics is helpful, but is not absolutely necessary. As far as mathematics is concerned, the reader should be familiar with simple differential equations and with the Fourier transformation.

1.7 Suggested Reading

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