Preface

The discovery of electron paramagnetic resonance (EPR) at Kazan State University by Soviet physicist Yevgeny Zavoisky in 1944 [1] represented the beginning of broad studies in the field of the energetic structure and dynamics of spin and pseudospin systems. In the short period of time that has passed since this fundamental discovery, the narrow area of research samples has been significantly enlarged to include almost all types of compounds containing paramagnetic centers [2].

The importance of Zavoisky's discovery is its real universal application in the study of the energy spectra of paramagnetic centers and the shapes of EPR lines, which are conditioned by spin–spin, spin–phonon, and other interactions in different systems: ionic [3–5] and molecular [6] crystals, coordination compounds in solid [4–7] and liquid [8] crystals, free radicals [6, 9, 10], chemical solvents [3, 11], color centers [12, 13], semiconductors [14–16], magnetic semiconductors [17–20], glasses (dielectric [3] and semiconductor [21, 22]), ordinary [14, 23, 24] and superconducting [25–27] metals and alloys, and spin-assigned biological systems [28], etc. This discovery also has a methodological importance, because the method used by Zavoisky to detect magnetic resonance based on changes in the field of electromagnetic radiation (instead of matter) during the interaction between the substance and the field has been widely applied to detect many other low-frequency resonances in condensed matter. The following list of phenomena observed and studied by the resonance method after the EPR discovery is a conclusive proof of the great importance of EPR, not only as a new physical phenomenon, but also as a new method for research: nuclear magnetic resonance (NMR) [29–34], nuclear quadrupole resonance (NQR) [35], acoustic paramagnetic resonance (APR) [3, 36–40], paraelectric resonance (PER) [41, 42], cyclotron resonance [43–45], spin resonance of conduction electrons in metals [23] and semiconductors [46–48], mixed resonance [44, 49], ferromagnetic resonance (FMR) [50–52], antiferromagnetic resonance (AFMR) [50], Pound–Overhauser double resonance [14], electron-nuclear double resonance (ENDOR) [4, 5, 53, 54], nuclear–nuclear double resonance [55–57], electron–electron double resonance (ELDOR) [58], electron–nuclear double magnetoacoustic resonance (acoustical ENDOR) [59], and more.

Some resonance phenomena among those listed above, due to the importance of the research results obtained by using these methods and their valuable practical applications, have triggered the development of new research directions in solid state physics. Thus, the phenomenon of APR [36], discovered in 1952 by Al'tshuler, later became the starting point for the development of direct methods for the research of spin interactions in paramagnetic centers and nuclei with phonons in condensed matter, including both magneto-dispersed and magneto-condensed systems with magnetic ordering. The ensemble of these methods forms modern quantum acoustics, with several independent research directions.

The reciprocal influence of one domain of spectroscopy on others as well as the unitary physical character of similar phenomena can be extended further. For example, it is easy to see that the coupled magnetoelastic waves in ferromagnetic materials, which are generated as a result of interactions of elastic waves with spin waves in conditions of magnetoacoustic resonance [50], resemble, to a wide extent, polaritons in the excitonic domain of the spectrum and phonon polaritons [60].

Another typical example of the branching of a research direction is represented by the phenomenon of NMR, discovered in 1946 independently by Purcell, Torrey, and Pound [61] and by Bloch, Hansen, and Packard [62]. Because the local magnetic fields on the nuclei of magnetic (and in some cases, nonmagnetic) ions conditioned by the hyperfine interaction in ferro- and antiferromagnetic materials are two to three orders of magnitude higher than those for these systems in the paramagnetic phase, as soon as radio spectrometers with frequencies in the range of 10^2–10^3 MHz were built (13 years after the discovery of NMR in paramagnetic materials), an independent research direction of great importance—NMR in magneto-ordered crystals [63–65]—was developed. However, the new peculiarities of magneto-ordered phase have led to the discovery of new phenomena, such as the effect of amplification of NMR signals, electron–nuclear double ferromagnetic and electron–nuclear double antiferromagnetic resonances [66], a new type of electron–nuclear double resonance that is determined by a substantial change in the rate of nuclear transversal relaxation at parametric excitation of electronic spin waves in ferrites [67], and so on.

The further development of the methods themselves and the realization of substantial new ideas are important as well. A good illustration of this aspect of the process in radio spectroscopy development is represented by the sharp increase in information given by NMR methods in studying condensed matter by using multipulse sequences and the successful experimental realization of "coherent averaging" methods in spin spaces [68, 69].

Radio-frequency spectroscopic methods can be applied under specific conditions to study degenerate exciton states in crystals. Regarding their real application, magnetic microwave spectroscopy of the Frenkel excitons in molecular crystals has been developing for a long time. Meanwhile, the application of these methods to study the Wannier–Mott excitons in semiconductors is a topic of future research.

The lack of hyperfine structure in the paramagnetic resonance spectra of triplet excitons due to the effect of translational movement [70] allows us to firmly differentiate experimentally the exciton states in molecular crystals from the localized triplet excited states of molecules. The small radius of the Frenkel exciton and, accordingly, the narrow exciton band which differs insignificantly from the enlarged level, as well as the sufficiently large lifetime of metastable triplet states in molecular crystals (noticeably larger than the period of a microwave) mean that the study of EPR spectra of Frenkel excitons differs insignificantly from the results obtained for EPR spectra of paramagnetic centers. In this way, naturally, the investigation of EPR of triplet Frenkel excitons in molecular crystals actually began in the 1960s [58].

Things look completely different in the study of Wannier–Mott excitons in semiconductors by methods of radio spectroscopy. The first experimental works in this field were performed substantially later [71–74]. It is thus reasonable to elucidate the research possibilities of exciton states in semiconductors by means of radio and microwave spectroscopy at small and large levels of optical excitation in crystals.

During the last decades, many of the physical systems that we discuss in this book, as well as the magnetic resonance techniques that were developed to study them, have been investigated from a very different angle: they may be used as "quantum bits" or qubits, for storing and processing information. The field of quantum information (or, more generally quantum technology) was born from the realization that information is not only an abstract concept, but that it is intimately connected to its physical representation [75]. In particular, it was conjectured [76] and proved [77] that quantum mechanical systems can process information in a way that makes them qualitatively more powerful than classical systems. As a result, problems for which no efficient solution appears to exist for classical computers may be solved efficiently by information processing devices that use the laws of quantum mechanics [78, 79]. The basic building blocks of quantum information processing are called quantum logical gates, in analogy to the logical gate operations of classical computers.

The realization of this potential requires two main ingredients: a physical system for storing the quantum information and external controls for implementing the quantum logical gate operations. The basic elements for storing the information are quantum mechanical two-level systems: the qubits. Since spins $1/2$ are the only physical systems whose Hilbert space is exactly two dimensional, they represent ideal blueprints for qubits. Similarly, all required quantum gate operations can be implemented by pulses of resonant radio-frequency or microwave fields interacting with the spins, in combination with free precession of the spins under an internal Hamiltonian that couples them to each other. As a result, the basic principles of quantum information processing have all been demonstrated on the basis of physical systems and spectroscopic methods described in this book.

In Chaps. 1–7 we discuss Wannier–Mott excitons, free and localized biexcitons, paramagnetic centers and nuclei which interact with the high-density excitons, as well as the influence of paramagnetic centers of high concentration on low-density excitons. The aim of these investigations is to obtain, by means of radio spectroscopy methods, research data on the hyperfine interaction of nuclear spins with excitons for small levels of optical excitation in crystals, to elucidate the mechanisms of influence of free excitons and free and localized biexcitons on the magnetic properties of semiconductors at large levels of their optical excitation, and also to obtain the relations regarding the inverse influence of magneto-condensed systems of impurity centers on free excitons. The methods of radio and microwave spectroscopy then have a special significance in those cases when the exciton states cannot be studied by means of optical methods (e.g., optically forbidden transitions in the exciton states, or extremely low concentrations of excitons).

Chapter 1 reviews the individual and collective properties of excitons in semiconductors studied by means of optical spectroscopy. Special attention is given to some fundamental difficulties in experimentally supporting the theory of Bose–Einstein condensation of excitons by means of optical spectroscopy experiments. These difficulties can be overcome by using the methods of radio spectroscopy.

In Chap. 2 we present the research on exciton paramagnetic and exciton paraelectric resonances. The possibilities of generation by excitons of coherent magnons (in magnetic semiconductors) and coherent electromagnetic radiation in submillimeter and infrared diapasons are analyzed.

In Chap. 3 we study the acoustic resonance of excitons and biexcitons. The theory of phonon masers based on exciton transitions is developed.

Chapter 4 is devoted to double resonances. Electron–nuclear double resonance and electron–nuclear magnetoacoustic double resonance of paramagnetic centers, hole–nuclear double resonance of the biexcitons captured by isoelectronic traps in GaP:N crystals, transfer of radio-frequency coherence in the cycle of optical pumping of excitons and, thereby, excitonic optical radio-frequency double resonance are presented.

In Chap. 5 we study excitons and biexcitons by means of NMR spectroscopy. The effects of excitons and orthobiexcitons on the relaxation rate of nuclear spin are discussed. The Knight excitonic shift of NMR lines for a degenerate and nondegenerate exciton gas is studied. The possibility of expanding the multipulse methods of high-resolution NMR spectroscopy to the optical spectroscopy of excitons is suggested based on an example of a four-pulse sequence of WAHUHA (WAugh, HUber, HAeberlen) type.

In Chap. 6 we study the interaction of excitons with paramagnetic centers: exchange exciton scattering on deep paramagnetic centers, indirect exchange interactions between paramagnetic centers through excitons, giant spin splitting of the exciton band in the exchange field formed by paramagnetic centers of high concentration (first being partially spin-polarized by using an external magnetic field) and, thereby, anomalous high increases in magneto–optical effects in the exciton band.

Chapter 7 is devoted to the effects of strong saturation of exciton and spin transitions. Nonstationary exciton states in the field of an intense hypersonic wave, nonstationary spin states (for an arbitrary spin value) in a constant magnetic field and low-frequency magnetic field of high amplitude, and the time-reversal symmetry for systems having a quasi-energy spectrum are studied.

In Chap. 8 we give a brief overview of the basics of quantum information processing. Taking the quantum mechanical nature of the physical system into account allows one to build qualitatively more powerful computers than is possible with the already extremely powerful classical devices. The fundamental notions are quantum superposition and the parallel processing of superposition states by unitary control operations.

Chapter 9 discusses specific examples where spins in solids have been used to demonstrate important concepts of quantum information processing. This includes very different systems like semiconductor quantum dots, dielectric solids containing rare-earth ions, or defects, such as the nitrogen–vacancy center in diamond.

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