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# TECHNIQUE OF OPTICALLY DETECTED MAGNETIC AND CYCLOTRON RESONANCES – BASICS AND APPLICATIONS

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**Optical detection of magnetic and cyclotron resonances enables not only to study magnetic and cyclotron resonances by observation of light emission modifications, but also allows identification of channels of radiative and nonradiative recombination in a given material. In the present review we shortly describe applications of the two optically detected resonance methods for studies of magneto-optical effects in diluted magnetic semiconductors.**

## INTRODUCTION

## INTRODUCTION TO THE ODMR

Principles of **O**ptically **D**etected **M**agnetic **R**esonance (ODMR) were proposed by J. Brossel and A. Kastler in 1949 for triplet-singlet spin configuration of the excited and ground states, often found for organic materials. Due to spin selection rules such system stays long enough in the excited triplet state to measurably repopulate its spin states (split by an external magnetic field) at magnetic resonance (MR) conditions. Thus, the MR of the molecule in the excited triplet state can be detected via a change in photoluminescence (PL) polarization.

The ODMR method was then successfully used for investigations of the origin of donor-acceptor pair (DAP) transitions in semiconductors. Information on these ODMR works can be found in the reference (Cavenett, 1981).

For DAP transitions rates of optical transitions can be increased by flipping spin of either donor or acceptor, for pairs with an initial parallel spin orientation. For such pairs electric dipole transitions are forbidden by spin selection rules. Spin flip allows radiative decay for such pairs, thus intensity of DAP PL is increased at the MR of either donor or acceptor. Importantly observation of these two resonances identifies not only the nature of the PL transition, but also identifies centers active in a given DAP PL process. This is the unique property of the ODMR.

Bound excitonic transitions (e.g. for excitons bound at isoelectronic centers and neutral complexes (Chen *et al.*, 1988)) and deep centers of nonradiative recombination were also studied with the ODMR (Killoran *et al.*, 1982). In the present work we shortly review application of the ODMR to the investigations of diluted magnetic semiconductor (DMS) materials.

Further details on the ODMR use for the studies of DMS samples can be found in recent papers (Godlewski, 2002; Godlewski, 2006a; Godlewski, 2006b).

## INTRODUCTION TO THE ODCR

The first cyclotron resonance (CR) experiment was performed by Dresselhaus, Kip and Kittel in 1953 (see (Godlewski, Chen & Monemar, 1994) and references given there). To reduce scattering efficiency experiments were performed at low temperature, but this resulted in carriers freeze out. Thus, Dexter, Zeiger and Lax introduced photo-excitation to introduce the sufficient concentration of free carriers (see (Godlewski, Chen & Monemar, 1994) and references given there). This led to a further modification of the CR method by Lax, who introduced a synchronous detection in phase with on-off modulated light excitation (see (Godlewski, Chen & Monemar, 1994) and references given there). The next obvious step was introduction of the Optical Detection of CR (ODCR) by Baranov et al. (Baranov *et al.*, 1977) and Romestein and Weisbuch (Romestein & Weisbuch, 1980).

Considering how complicated and expensive set ups are used in the ODCR investigations an obvious question is what extra information we obtain from such studies. The fact of simultaneous observation of electron and hole CR signals (detected via different PL transitions (as demonstrated in (Weman, Godlewski & Monemar, 1988)) only partly answers this question.

In our studies we demonstrated the important role of impact ionization processes in optical detection of the CR signals (Godlewski *et al.*, 1995). We further showed that ODCR can be used as an advanced spectroscopic method (see (Godlewski, Chen & Monemar, 1994) and examples given there). At the CR we could change formation rates of bound excitons, trapping rates of free carriers, or impact ionize shallow centers and excitons (Godlewski, Chen & Monemar, 1994). This allows for identification of channels of radiative recombination, evolution of strength of localization processes in quantum well structures (Godlewski *et al.*, 1995; Godlewski, 2000), and studies of carriers and spin relaxation processes in Diluted Magnetic Semiconductor (DMS) materials (Godlewski, 2000), as described in the reviews on the ODCR (Godlewski, Chen & Monemar, 1994; Godlewski, 2000).

## EQUIPMENT

Techniques of the ODMR and ODCR are developed by us in the laboratory of Magneto-optics in the Institute of Physics Polish Academy of Sciences in Warsaw, Poland. In Fig. 1 we show schematic diagram of the 60 GHz system used by us. Q-band system is also used by us for some ODMR/ODCR investigations.

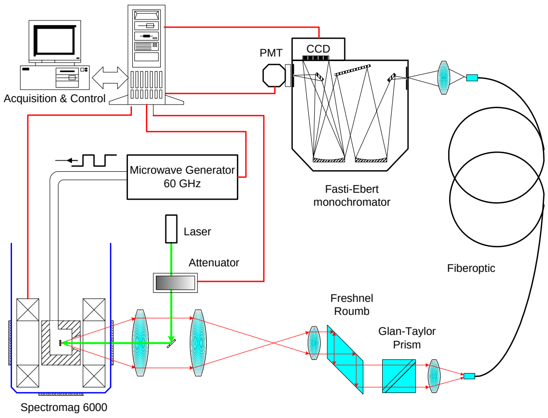

Fig. 1. Diagram of the 60 GHz ODMR/ODCR system.

A core of the system is Spectromag 6000 split-coil superconductive magnet of the Oxford Instruments with magnetic field up to 7.5 T and with four windows allowing an optical access.

We use set of different gas and solid state lasers for the photo-excitation. Samples are mounted in a microwave cavity with an optical access allowing PL excitation. PL is collected using set of lenses and transferred by fibers to a double grating monochromator.

Signal is detected with either CCD camera or photon-counting photomultiplier. In the ODMR/ODCR study PL changes induced by MR or CR transitions are detected in phase with on-off modulated microwave power.

Fresnel rhomb and Glan-Taylor prism allow studies of PL polarization. Magnetic field steps, microwaves modulation and PL detection are controlled by a computer.

Further modification of the system allow us measurements of time evolution of the PL changes (time-resolved ODMR) and spatial resolution of the PL modifications and magnetic/cyclotron resonances.

## MECHANISMS OF THE ODMR/ODCR DETECTION SPECIFIC FOR THE DMS SYSTEMS

In early works on DMS samples (see (Gisbergen *et al.*, 1993) and references given there) spin cross-relaxation processes were identified as responsible for the ODMR detection.

Most of these experiments were performed for DMS samples containing Mn ions. In such cases the Mn2+ MR is detected optically via an increase in intensity of DAP PL processes (see (Gisbergen *et al.*, 1993) and references given there). A new situation we found for ZnMnS samples for which the $Mn^{2+}$ MR, by spin flip between two adjacent Mn ions, resulted in an increase in intensity of the $^4T_1 - {}^6A_1$ intra-shell transition of the excited $Mn^{2+}$ ions (Godlewski *et al.*, 2002; Godlewski, 2004 *et al.*; Godlewski, 2006a; Godlewski, Yatsunenko & Ivanov, 2006b;). The process was identified by measuring of the so-called ODMR-PL spectrum. In the ODMR-PL experiment a spectral response of the MR signal is measured, which allows a direct relation between a given MR signal (and thus identified center) and process of light emission.

For CdMnTe (Godlewski *et al.*, 2002; Godlewski, 2006a; Godlewski, Yatsunenko & Ivanov, 2006b) and ZnMnSe (Ivanov *et al.*, 2008) we demonstrated another important mechanism of the ODMR detection. Due to very long $T_1$ spin-lattice relaxation times for $Mn^{2+}$ ions sample magnetization can be macroscopically quenched at the MR conditions. Quenching of samples magnetization result in changes of the spectral position of excitonic transitions. The so-induced shifts (up in energy) of excitonic transitions measure the magnitude of the magnetization quenching (Godlewski *et al.*, 2002; Godlewski, 2006a; Godlewski, Yatsunenko & Ivanov, 2006b; Ivanov *et al.*, 2007; Ivanov *et al.*, 2008).

Such shifts we observed only at very low excitation density. Once free carriers concentration increases very efficient Mn-free carriers spin flip process shortens considerably spin relaxation times (Godlewski, 2002; Godlewski, 2006a; Godlewski, Yatsunenko & Ivanov, 2006b).

The latter could be directly proved in the developed by us modification of the ODMR technique – in time-resolved ODMR study (Ivanov *et al.*, 2007; Ivanov *et al.*, 2008). At low excitation density the measured time of magnetization recovery (after the MR transition) is equal to the $T_1$ spin-lattice relaxation time. Once the excitation density increases this time shortens considerably, indicating a very high efficiency of Mn-free carriers spin flip interactions

(Ivanov *et al.*, 2007; Ivanov *et al.*, 2008). At such condition the MR signal can not be detected as the magnetization quenching. For further details on time-resolved ODMR see our recent reviews (Godlewski *et al.*, 2002; Godlewski, 2006a; Godlewski, Yatsunenko & Ivanov, 2006b).

High efficiency of Mn-free carriers spin flip interactions allowed us to explain the origin of shortening of $Mn^{2+}$ PL decay observed in nanoparticles (see (Godlewski *et al.*, 2004) and references given there). Also the ODCR investigations can yield important information on spin interactions in DMS samples. The application of the ODCR to the investigations of DMS samples is described in details in the reference (Godlewski, 2000) and thus will not be discussed here.

## CONCLUSIONS

Two techniques of optically detected resonances (ODMR and ODCR) are used for investigations of recombination channels in semiconductors. As compared to conventional MR, CR and PL methods the ODMR and ODCR yield additional information on nature of recombination processes and allow the direct identification of centers active in these processes. For DMS samples these two techniques allow the direct identification of various spin flip interactions and measurements of spin relaxation times

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