Spin electronics

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I. INTRODUCTION

Spintronics devices are expected to combine the speed and complexity of modern semiconductor elements with the storage and switching capabilities of the magnetic ones. The idea of using spins of semiconductor free carriers in the electronic devices requires, however, further comprehensive and systematic studies. An essential progress has to be made in synthesis of new spintronics materials and low-dimensional layer structures from semiconductor and magnetic materials. A development of new research methods for the analysis of such structures is also required.

In the project titled "Spin electronics" financed by Polish Science Foundation is realized at the Institute of Physics Polish Academy of Sciences in Warsaw (IF PAN), at Institute of Experimental Physics of Warsaw University (IFD UW) and at the Institute of Molecular Physics Polish Academy of Sciences (IFM PAN) and at the Adam Mickiewicz University (UAM) in Poznań. The part of researches is realized also at the Warsaw Technical University (PW), the Institute of Electron Technology (ITE) and the Institute of Technology of Electronic Materials (ITME) in Warsaw. We realize complex studies aimed at optimization of classical spintronics structures based on metallic ferromagnet, as well as, at the search of substantially new materials and spintronics structures from semiconductor compounds. We also plan to improve the technology of diluted magnetic semiconductors, already well established at the IF PAN and the IFD UW, to the stage that would enable the construction of a prototype of a spintronics device.

The development of spintronics shows that an application of a semiconducting ferromagnetic material could solve many spintronics problems and to take a real advantage of spin freedom degree in future electronics. Unfortunately, there is no natural ferromagnet with semiconductor attributes. Neither the properties of metallic, nor those insulating ferromagnets can be modified by light or by applied voltages. Therefore, preparation of an artificial magnetic semiconductor is an important challenge for the modern materials science. This is one of the most important target of our investigations.

II. SPINTRONIC STRUCTURES

II.1. Giant magneto resistant (GMR) and inter layer coupling

The GMR and tunneling magneto resistance (TMR) effects in metallic ferromagnetic layers separated by paramagnetic metal or insulating spacer are well known for more then decade. For many years they are studied in the IFM PAN and the UAM in Poznań. The aim of the "Spin electronics" project is to test new type of metallic GMR and TMR structures, to build similar structures based on semiconductor layer and to explain the mechanisms of spin dependent electron tunneling and interlayer coupling via insulating or semiconductor spacer. Semiconductor structures are also investigated in the IF PAN in Warsaw.

In particular, magnetic properties of Co/metal/Co, Fe/Si and Co/Si multilayers [1], spin injection through Fe/InAs interface [2], electron tunneling in planar double junction with

ferromagnetic barriers [3], origin of magneto-resistance in Ca: YIG [4], tunnel magneto-resistance in planar junctions with ferromagnetic barriers [5], and problem of spin currents *via* the domain walls [6] are studied.

The problem of interlayer coupling, the basic problem of GMR and TMR effects, is studied in various systems, as well in ferromagnetic metal as in antiferromagnetic semiconductor layered structures. *E.g.*, the exchange coupling between ferromagnetic layer is monitored in Fe/Si_xFe_{1-x} multi-layers [7, 8] and between antiferromagnetic layers via semiconductor spacer. Experimental measurements are supported by theoretical modeling [9,10]. The system of $Ga_{1-x}Mn_xAs/GaAs$ is of a special interest [11]. First of all $Ga_{1-x}Mn_xAs$ is a ferromagnet and both components are semiconductors with a strong spin polarization. The observed fact that very narrow $Ga_{1-x}Mn_xAs$ layers are characterized by a higher critical temperature is another intriguing property of such a structure.

Only a weak GMR effect in metal/semiconductor structures has been found. The difference in the rersistivity of two sub-systems is the main reason of the weak dependence of resistance of metal/semiconductor structure on spin polarization. Therefore we undertook investigation of spin dependent transport in structures made of semiconductors only. A wide class of spintronics materials, structures and effects are investigated. From a simple magneto-resistance in diamagnetic [12] and ferromagnetic semiconductors, via investigation of spin alignment of electrons in diluted magnetic semiconductors (DMS) and DMS/semiconductor structures [13]. At present many various experimental methods are used for investigation of spin dependent effect. In addition to magneto-resistance and electrically detected magnetic resonance, which reflects the dependence of electrical conductivity on spin polarization, we apply a spin dependent optical methods. The circularly polarized photo luminescence (PL) allows to measure the spin polarization and spin diffusion within the layered structures. The time dependent PL and combine magneto-optical resonances, allow us to investigate the complex problem of spin dynamics. The most of these data are still unpublished.

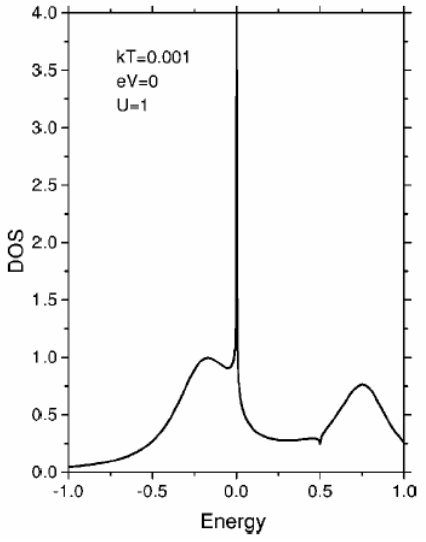

Fig. 1. DOS in equilibrium situation. The two broad bands are splitted by the correlation energy *U*. The narrow peak at the Fermi level is the Kondo peak [16].

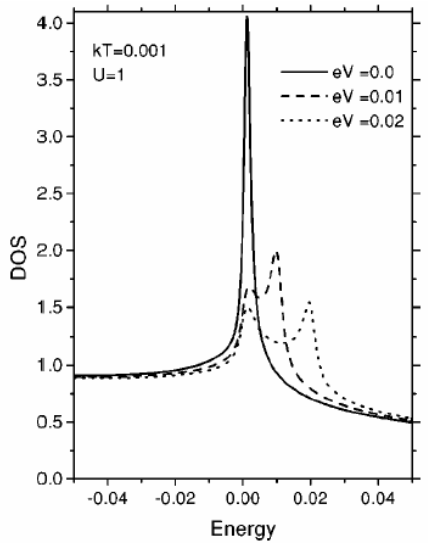

Fig 2. Splitting of the Kondo peak in DOS with increasing bias shown for three values of the applied voltage [16].

II.2. Single electron structures

Theory of spin related effects in mesoscopic systems are studied at the UAM and the IFM PAN in Poznań. Generally the spin polarized transport through quantum dot coupled to magnetic electrodes is analyzed. The variety of effects come from the spin splitting of electron states in the central dot due to the presence of ferromagnetic electrodes. The splitting depends on the direction of electrode magnetization, but can be modified by an external magnetic field and by applied voltages. The analysis requires simultaneous considerations of the Coulomb blockade, the Kondo effect, and spin fluctuations [14-17].

Calculation has been done for structures with ferromagnetic electrodes as well for metallic central island as for a quantum dot. The spin dependent current and its spectral function are analyzed. It has been shown that for metallic island the Coulomb blockade effect dominates the electron transport but, so called, second order effect rules the current voltage characteristics. In particular the splitting of the resonance peak reflects the spin accumulation at the island, caused by spin dependent tunneling by the barrier (see Figs 1 and 2). In the system with quantum dot two new effect has been found. The first, it is an anomaly of conductance which occurs for anti-parallel configuration of electrode magnetization. It is seen as a narrow maximum at zero voltage. The second effect results from the spin asymmetry in elastic and inelastic scattering co-tunneling processes. It leads to different magneto tunneling resistance for even and odd number of electrons an the quantum dot.

Spin effects in single electron objects are experimentally investigated (at the IF PAN and the IFD UW) in two dimensional semiconductor structures, where the initial spin structure of exciton can be controlled by the circular polarization of excitation light while the final spin structure by circular polarization of photoluminescence. Comparison of the initial and the final states allows us to conclude about coupling of exciton to another electron of spin, *i.e.*, formation of trion, and about spin diffusion and spin relaxation. The most of these effect are investigated in diluted magnetic semiconductors where the spin splitting energy is strongly enhanced.

III. SPINTRONIC MATERIALS

The physics of DMS has been investigated in Poland for few last decades. The giant spin splitting of conduction bands, caused by the magnetization of magnetic impurities and mediated by sp-d exchange, is an essential attribute of DMS. Such splitting allows selective spin manipulation in excitons or in quantum dots. Moreover, DMS are the most effective spin injectors in spintronics devices. Unfortunately, in paramagnetic DMS, external magnetic field is required for magnetization of local spins, and this limits possible applicability of DMS. On the other hand, increased carrier concentration allows inducing an exchange (Zener-like) field which causes magnetization of the local spins. The occurrence of a ferromagnetic phase generated by such positive feedback has been evidenced in many materials. Unfortunately, non of ferromagnetic semiconductor is characterized by the Curie point above the room temperature. In II-VI Mn based DMS, the high carrier concentration requires strong, additional doping and the typical T_c is of the order of few Kelvin. The temperature of 170 K has been found for $Ga_{1-x}Mn_xAs$. Now this is the highest Curie temperature observed, at least, this is the highest critical temperature of ferromagnetic semiconductor, where the ferromagnetism is related to semiconductor volume and the mechanism of ferromagnetism is understand.

Nowadays our investigations are focused in three directions.

- Two dimensional structures build from insulating ferromagnet and semiconductor layers. When the layer thickness is small the total superlattice has some attributes of magnets as well as semiconductors [10]. We investigate systems based on lead compounds. Here EuTe is an antiferromagnet, while EuS is a ferromagnet. Both of them can be grown as super-lattices with PbTe, which is a narrow gap semiconductor. The doping of EuTe by Gd changes this material from insulating antiferromagnet to semi-metallic ferromagnet.
- Explanation of the details of the nature of magnetic order in $Ga_{1-x}Mn_xAs$. This is a canonical diluted magnetic ferromagnet. Magnetic order in $Ga_{1-x}Mn_xAs$ has been well evidenced, but many of its properties differ from the properties of classical magnets and they have to be investigated.
- Search for another chemical system which could simultaneously show semiconductor and magnetic properties. Our investigations focus on DMS of group III-V and II-VI with various transition metal elements. The most advanced studies was done for the nature of Mn impurity in GaN, which had been expected to be the best candidate for room temperature semiconducting ferromagnet.

III.1 Magnetic orders in $Ga_{1-x}Mn_xAs$

Mn ion in the GaAs lattice introduces a large local spin, S_{Mn}=5/2, and simultaneously plays the role of acceptor. Consequently, the high concentration of holes allows the ferromagnetism to occur. The magnetic order in semiconducting $Ga_xMn_{1-x}As$ is already well determined and the theoretical models explain well its origin, and also allow satisfactory estimation of the critical temperature. On the other hand, the details of the spin structure, the role of fluctuations, the puzzle magnetic anisotropy and the peculiar spectrum of spin waves are still under debate.

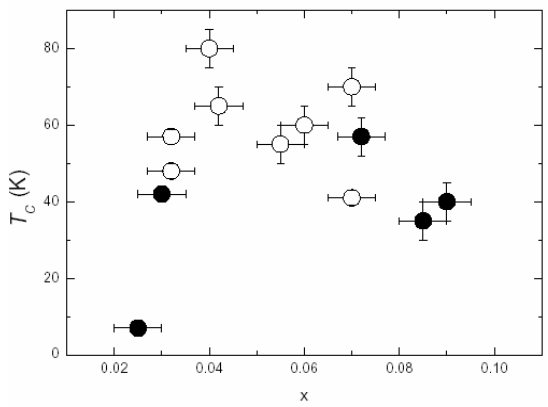

Fig. 3. The dependence of the critical temperature, evaluated from magnetic resonance studies, on Mn concentration, *x*. Full and open circles correspond to ferromagnetic and ferrimagnetic samples respectively [19].

Our magnetic resonance studies allow distinguishing two different magnetic structures of $Ga_{1-x}Mn_xAs$ [18,19]. Metal-to-insulator transition is accompanied by a principal change of magnetic resonance spectra (see Fig. 3). The single isotropic line of ferromagnetically ordered Mn spins changes to a very complex, strongly anisotropic spectrum. We interpret this change to a transition from ferromagnetic to ferrimagnetic phase, where both spin sub-systems are magnetically correlated. The observed resonance frequency and magnetic anisotropy can be well described using the formalism of ferrimagnetic resonance [20]. In that sense, one can conclude that in metallic phase the $Ga_xMn_{1-x}As$ constitute a ferrimagnetic system.

The complex structure corresponds to spin wave (SW) resonance (see inset in Fig. 4). Just the fact alone, that SW resonance is observed, is intriguing because it implies a long mean free path of SW, which can occurs in a very uniform magnetic medium only. Also the observed dispersion of SW (see Fig 4) and its angular dependence is very different from the classical expectations. We are able to explain these peculiarities by taking into considerations the strong momentum scattering of holes. Therefore, the latter ones cannot be treated as a plane waves but rather as partially localized carriers. The localization affects neither Zener field nor

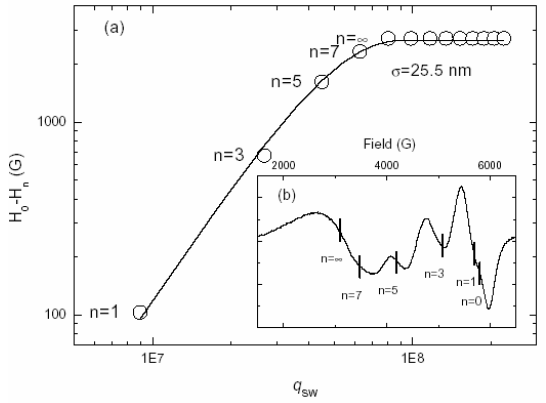

Fig. 4. (a) Dispersion dependence of spin waves in $Ga_{1-x}Mn_xAs$. (b) 9.44 GHz resonance spectra obtained at $T = 2K$ in normal configuration. A complex wave structure corresponds to the SW resonance. Numbers indicate the positions of the consecutive spin waves.

mean Mn-Mn exchange field. In contrast, the mean range of Mn-Mn exchange for localized carriers corresponds with the localization range and it can be much longer if compared to the range of RKKY coupling. The long exchange range can be directly evaluated from the observed dispersion of SW and allows the explanation of the long mean free path of SW. The experimentally estimated number of coupled Mn spins is of the order of 10^4-10^5. The resulting exchange field, responsible for the propagation of SW, is very well averaged.

The well averaged alloy disorder in semi-metallic $Ga_{1-x}Mn_xAs$ contrasts with the fluctuations in insulating crystals. In that case, local spins are coupled via a double-exchange mediated by effective mass holes occupying acceptor states [21, 22]. The characteristic length of interaction is that of the Bohr radius order, *i.e.*, of the order of mean Mn-Mn distance. The alloy disorder, namely fluctuations of exchange and anisotropy fields seen by hole spins, does not allow any coherent precession of hole spins. As a consequence, only the Mn spins can form a macroscopic, ferromagnetic moment, while the role of hole spins is reduced to mediation of Mn-Mn interaction.

Two different types of magnetic order in different electrical phases are described by two different types of theoretical models. The ferromagnetic order is well modeled by calculations which takes into consideration a double exchange and fluctuations of the local charge density [21, 22]. Whereas in ferrimagnetic, semimetallic $Ga_{1-x}Mn_xAs$ the energy scale is well estimated by mean field Zener/RKKY model where the charge distribution is approximated by an uniform medium (Dietl, 2001). This simple mean field model well predicts the critical temperature, but the description of the details of spin wave dispersion requires a small modification of Zener model. A finite carrier localization length, corresponding to the range of exchange interaction, and a finite range of a coherent spin precession must be taken into consideration. The fact that the range of coherent precession is shorter than the range of exchange coupling results in the softness of spin waves. In diluted $Ga_{1-x}Mn_xAs$ alloys the energy of spin wave is by two orders of magnitude smaller than in the regular ferromagnet with a similar Curie temperature. The spin wave softness leads to a specific dependence of material magnetization on the temperature and the applied magnetic field.

The native defects in $Ga_{1-x}Mn_xAs$ is another important issue. It was shown that the interstitial Mn is of a crucial importance [23, 24]. First of all, it plays the role of a double donor, consequently, it reduces the hole concentration and Curie temperature. Moreover, the indirect coupling of an interstitial to a substitutional Mn is of an antiferromagnetic character thus the total magnetic moment is accordingly reduced. The occurrence of interstitial Mn ions, and a strong increase of their concentration with an increase of the total Mn concentration, is considered as the main reason limiting the high Curie temperature.

We find out that the sample annealing is an effective tool for a reduction of the concentration of Mn interstitial. Systematic studies of annealing of $Ga_{1-x}Mn_xAs$ layers of different thickness at various temperatures allow to find optimal conditions and to evaluate the height of the barrier for diffusion of interstitial Mn ions. The experimentally evaluated height well fit to the value obtained from *ab initio* calculations [24].

III.2. Properties of $Ga_{1-x}Mn_xN$

We applied an effort to investigated $Ga_{1-x}Mn_xN$ [25-27]. $Ga_{1-x}Mn_xN$ was treated as a very promising high temperature ferromagnet. It represents a system similar to $Ga_{1-x}Mn_xAs$, but, due to a smaller lattice constant, the p-d exchange is expected to be greater. Consequently, the Curie temperature, which scales with the square of p-d exchange, was expected to be considerably greater. Regrettably, nobody has grown crystals of p-type semi-metallic $Ga_{1-x}Mn_xN$.

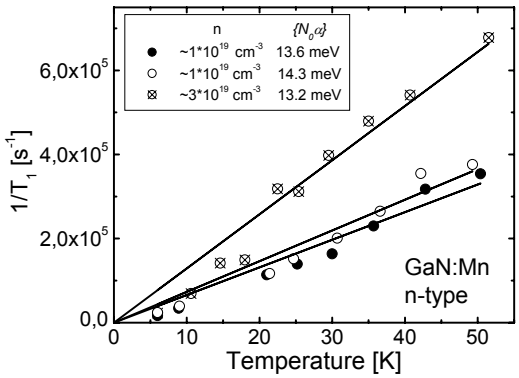

Fig. 5. Temperature dependence of spin relaxation rate of Mn spins for n-type GaN:Mn samples with different free electron concentrations. Linear dependence $1/T_1$ on the temperature is characteristic for Korringa spin relaxation mechanism [27].

Because of very high melting temperature the technology of GaN based compounds is very difficult. It is possible to obtain semi-metallic n-type crystal, but s-d exchange was found to be very small [28]. The exchange coupling was evaluated from the Korringa broadening of Mn resonance, *i.e.* the broadening of electron paramagnetic resonance (EPR) line due to scattering of carrier spin on Mn spin. The temperature slop (see Fig. 5) of the line width allows for a direct estimation of s-d exchange coupling.

The ionization energy of magnetic impurity, *i.e.*, the position of impurity level in respect to the valence and the conduction bands, is the basic parameter which rules the character of impurity and determines the electric properties of the alloy. In particular, the $Mn^{2+/3+}$ level in GaAs, corresponding to ionization of $3d^5$ shell, is degenerated with the valence band. Consequently, $3d^5$ plus an effective mass hole is the ground state configuration of Mn impurity in GaAs. It means that the holes can occupy Mn acceptor or valence band states

(when Mn concentration is big enough to induce insulator-to-metal transition). In both cases, effective mass holes can mediate Mn-Mn exchange and to form a magnetic order.

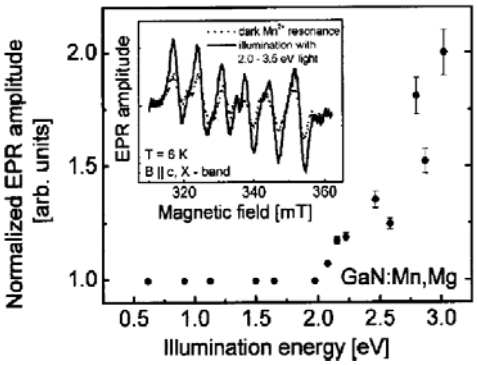

Fig. 6. A photo – EPR experiment performed for GaN:Mn,Mg sample. Spectral dependence of the photo – stimulated Mn^{2}+(d^5) amplitude is shown. Inset shows the difference of EPR signal of Mn^{2+} dark (dotted line) and illuminated sample [28].

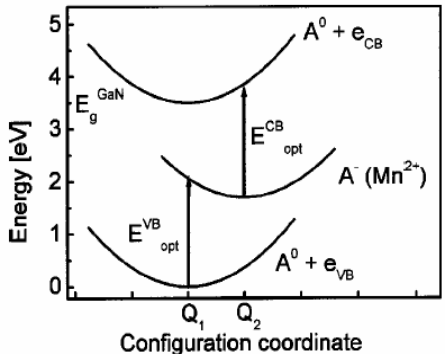

Fig. 7. Configuration-coordinate diagram showing Mn-related photo-ionization transitions in GaN [28].

To define the position of Mn level in GaN we done complex studies. Photo-EPR measurements show that the concentration of Mn^{2+} ions can be modified by sample illumination. As it is shown in the Fig. 6 an illumination increases concentration of Mn^{2+} showing that a hole from the valence band can be optically transferred to Mn 3d shall. The photo-ionization edge occurs at energy of 2 eV. The corresponding energy diagram is shown in Fig. 7. The studies optical absorption which shows excitation of $3d^4$ configuration and magnetization anisotropy, which is characteristic for Jahn-Teller state, prove that $3d^4$ is the main configuration in GaN and that the corresponding impurity level is located in the vicinity of the center of energy gap.

As a consequence, for strongly doped GaN the Fermi level is pinned to the Mn level, Mn occurs in two valance states and there are no carriers present which could mediate the Mn-Mn exchange. The system remains semi-insulating and paramagnetic [29].

III.3. Search for a room temperature ferromagnetic semiconductor.

Searching for good material for the room temperature ferromagnetic semiconductor we tested a wide class of II-VI and III-V based DMS. Within the III-V compounds with Mn the $Ga_{1-x}Mn_xAs$ seems to be the best one. In the wide gap semiconductors the character of Mn impurity is similar to that in GaN (see diagram in Fig. 8). In narrow gap materials the Mn level is degenerate with the valence band, p-type semi-metallic crystal can be obtained, but the crystal quality is bad and the Curie is low.

Similar situation, *i.e.*, location of the energy level of transition metal impurities within the energy gap, occurs in the most of investigated semiconducting compounds.

There is still a hope that a high temperature ferromagnetic order can be found in $Ga_{1-x}Fe_xN$ or in Zn_{1-x} Mn_xO systems. In these alloys the magnetic impurity are expected to be in $3d^5$ configuration and characterized by S=5/2 spin. These systems differs, however, from $Ga_{1-x}Mn_xN$ because in these case magnetic impurities are iso-electronic, *i.e.*, they do not introduce carriers. As a consequence, carriers, which are needed for mediation of exchange, must be introduced by an extra doping. We expect that p-doped systems can be either of

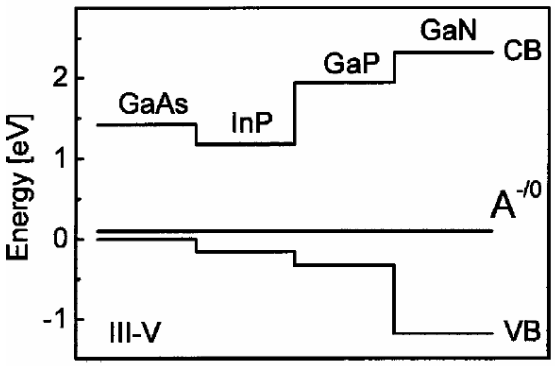

Fig. 8. Localization of Mn acceptor level in III-V semiconductors. In wide gap compounds the impurity level is located within the energy gap avoiding a metallic character of Mn doped crystals [28].

semimetallic type or that holes can be bound at iso-electronic impurities by exchange interaction, forming a specific impurity band. Unfortunately, simultaneous doping by acceptors and magnetic impurities is still a difficult technological challenge.

IV. SPIN RELAXATION

The spin relaxation is the basic problem of spintronics. In many cases a long spin memory is a necessary condition for spintronics devices. The spin memory time must be long enough to provide the necessary time for spin manipulation. However, in some cases when fast spintronics devices are build, a force acting on spin must be strong enough to provide a fast spin manipulation. In contrast, sometimes a fast spin relaxation is required. One has to underline that forces which allow the spin manipulation simultaneously lead to a fast spin relaxation [12, 30, 31]. Because of that complexity the detail knowledge of spin interaction and spin dynamics, including the spin relaxation mechanisms, are crucial.

Generally, the problem is very complex. One cannot expect any general rule. Various details must be separately analyzed. One cannot simultaneously discuss the spin memory of ferromagnetic objects and single electron. Also the relaxation rate of electron spin in diamagnetic semiconductor and DMS are ruled by different mechanisms and they differ by order of magnitudes. Even the relaxation rate of electron spin in different semiconductors can differ by few orders of magnitude. In particular, the spin relaxation rate of electrons in Si is by 3 or 4 orders of magnitude slower as compare to holes GaAs [12]. On the other hand, the spin relaxation in DMS, where the mechanism of spin flip scattering on paramagnetic impurities dominates, the spin relaxation rate is by another few orders of magnitude faster [32].

The spin relaxation problem is investigated by different methods in various systems. The time resolve photoluminescence, pump-probe measurements, magnetic resonances and magneto optical combined resonances are applied. But there are a few cases where spin relaxation can be quantitatively described. In spintronics material we understand the spin relaxation mechanisms for carriers in paramagnetic DMS [28] and in purely diamagnetic semiconductors [12].

As it was already mentioned above, in DMS the spin relaxation of carriers is ruled by the Korringa mechanism (see Fig. 5). In the case of degenerate statistic in semimetals, the Korringa relaxation depends on the square of the sp-d exchange coupling and is proportional to temperature [28]. The description becomes more complex for non degenerate statistic for low carrier concentration and much more complex for ferromagnetic DMS [19]

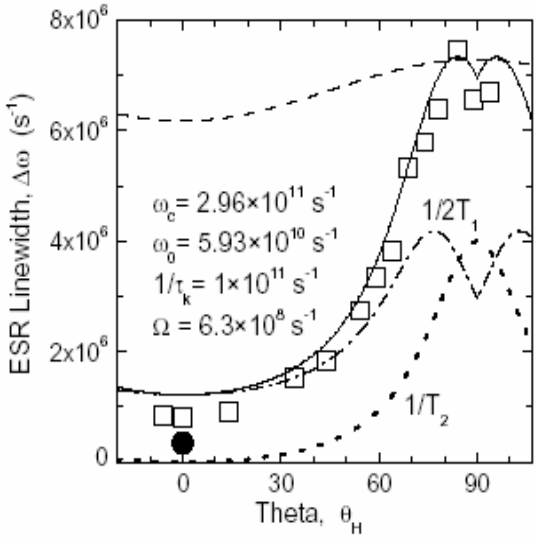

Fig. 9. Angular dependence of the ESR line width (squares) of high mobility 2D electrons in Si quantum well. θ is the angle between the growth direction and the direction of the applied field. The full dot shows the contribution of the longitudinal relaxation to the linewidth, $1/2T_1$. Dashed line: the anisotropy predicted by Dyakonov-Perel mechanism when the modulation of BR field caused by cyclotron motion is neglected. The solid, dash-doted and dotted lines correspond to the model described in Ref. [12].

The high mobility 2D electron gas in Si quantum well is the another case where spin relaxation mechanisms are quantitatively explained [12]. In that case, the specific spin-orbit coupling, so called Bychkov-Rashba (BR) and Dresselhaus terms, are responsible for spin relaxation. In 2D Si the BR effect dominates. According to D'yakonov-Perel the spin relaxation is proportional to square of the BR field and to characteristic modulation time. Spin resonance measurements allow us to evaluate the BR field and to explain the longitudinal as well as the transverse relaxation rates. The same value of BR field allows for explanation of g-factor anisotropy. We show that in the case of a high electron mobility, when momentum relaxation is slow, the modulation caused by the cyclotron motion has to be additionally considered. The observed anisotropy of spin relaxation, and in particular of spin resonance line width, is finger print of the effect cyclotron motion on spin relaxation. The anisotropy shown in the Fig. 9 reflects the anisotropy of the cyclotron frequency of 2D electrons.

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