

ELECTRON SPIN RESONANCE IN SEMICONDUCTORS

ZBYŚLAW WILAMOWSKI^{1,2}, AGNIESZKA WOŁOŚ¹, HANKA PRZYBYLIŃSKA¹

¹Institute of Physics PAS, 02-668 Warsaw, Al. Lotników 32/46

²Faculty of Mathematics and Computer Science, Warmia and Mazury University in Olsztyn, Ul. Żołnierska 14, 10719 Olsztyn

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We present a review of possible applications of magnetic resonances in semiconductors. The Electron Spin Resonance (ESR) technique has been applied to investigations of free and bound electrons in classical 2D and 3D semiconductors as well as paramagnetic centers in diluted magnetic semiconductors (DMS). We demonstrate that the spin orbit coupling (Rashba coupling) determines both the anisotropy of the g-factor and the resonance line broadening, leading to specific effects, such as a tuning of the resonance field by electric current or excitation of ESR by *rf* electric field. In DMS Korringa broadening, Knight shift and giant spin splitting are discussed. In magnetically ordered materials the resonance method allows detailed evaluation of magnetic anisotropy, as demonstrated in case of ferromagnetic bulk (Ga,Mn)As and magnetic precipitates in (Ga,Fe)N.

INTRODUCTION

Semiconductor crystals are usually diamagnetic. Therefore, their lattices do not contribute to magnetic resonance signals allowing detailed investigations of paramagnetic centers. Electron Spin Resonance (ESR) is able to detect impurity and carrier spins of a very low concentration. Here we review the results of magnetic resonance studies of semiconductors and diluted magnetic semiconductors (DMS). We show that ESR data give important information on magnetic as well as electric properties.

MAGNETIC IMPURITIES IN SEMICONDUCTORS

Investigation of paramagnetic centers is the most typical goal of ESR studies. In semiconductors, however, ESR of free carriers and donor electrons can be also measured, giving some unique insight into spin orbit coupling and spin relaxation mechanisms. Below, we review the single ion anisotropy results determined for the Mn impurity in GaAs, Fe in GaN, and Mn in ZnO. In all these systems at a higher content of magnetic ions ferromagnetic order is observed.

ESR of paramagnetic ions

Mn²⁺ in GaAs. ESR gives a unique possibility to investigate the properties of paramagnetic centers. An investigation of Mn²⁺ in GaAs (Fedorych, Hankiewicz & Wilamowski, 2002) is an example of such a study. The resolved fine structure in insulating (Ga,Mn)As allows us to evaluate the crystal field parameters of the spin

Hamiltonian. On the other hand the exchange narrowing of the structure, related to the presence of free carriers, indicates long range exchange interaction.

The axial field parameter of single ions, D , is found to increase with Mn concentration, x , i.e., with the strain of (Ga,Mn)As layers. Extrapolation of D towards low Mn content shows, however, that the single ion anisotropy gives only a small contribution to the total magnetic anisotropy observed in ferromagnetic layers with greater Mn and hole concentrations. This indicates that the axial anisotropy of ferromagnetic (Ga,Mn)As layers originates from the strain induced anisotropy of the valence band.

The analysis of the EPR linewidth in strongly diluted (Ga,Mn)As shows that native defects at a concentration of $5 \cdot 10^{19} \text{ cm}^{-3}$, but not the Mn ions, are the main source of crystal field fluctuations. The study of magnetic resonances in (Ga,Mn)As allows us to distinguish phenomenologically different ranges of Mn and holes concentrations. For mole concentrations $x_{Mn} < 10^{-4}$ the low-temperature, MBE grown layers are insulating. Electrically, the Mn acceptors are fully compensated by native donors. No evidence of effective mass carriers is seen. Within the range $10^{-4} < x_{Mn} < 0.002$ the presence of carriers leads to exchange narrowing of a fraction of Mn centers. The fact that the length of the exchange interactions corresponds to the Bohr radius of the acceptor allows us to conclude that this concentration range corresponds to the Hubbard-Mott insulator. The insulator to metal transition takes place at $0.002 < x_{Mn}$. The ferromagnetic

phase occurs for $x_{Mn} \cong 0.015$ and the transition from the “weakly” to “strongly” anisotropic ferromagnet starts at $x_{Mn} \cong 0.04$.

Fe³⁺ in GaN. Electron spin resonance studies demonstrate the existence of Fe in the isolated 3d⁵ (Fe³⁺) state. Magnetic measurements reveal, apart from Curie paramagnetism due to Fe³⁺, a temperature independent contribution which was attributed tentatively to van Vleck paramagnetism of Fe in the 2+ state. We conclude that the Fe ions coexist in two charge states in the investigated samples. The fraction of Fe³⁺ ions was found to increase upon additional co-doping with Mg acceptors, both in ESR as well as in magnetization measurements (Przybylinska, Bonanni, Wolos, Kiecana, Sawicki, Dietl, Malissa, Simbrunner, Wegscheider, Sitter, Rumpf, Granitzer, Krenn & Jantsch, 2006).

The spin Hamiltonian parameters for the Fe³⁺ ion were determined by a least squares fit of the angular dependence of the ESR line positions for the magnetic field **B** rotated in the (11 $\bar{2}$ 0) plane with the spin Hamiltonian for an S=5/2 ion in C_{3v} symmetry:

$$H = \mu_B \mathbf{B} \mathbf{g} \mathbf{S} - \frac{2}{3} B_4 (O_4^0 + 20\sqrt{2} O_4^3) + B_2^0 O_2^0 + B_4^0 O_4^0. \quad (1)$$

Here the first term describes the Zeeman interaction, the second term is the cubic crystal field interaction, and the last two terms are second and fourth order trigonal crystal field interactions. The spin Hamiltonian parameters, especially the value of the second order trigonal parameter B_2^0 was found to vary slightly with sample thickness, which indicates that the layers are strained. Typical parameters for a 1000 nm thick layer are: $g_{\parallel}=2.009 \pm 0.002$, $g_{\perp}=2.005 \pm 0.002$, $|B_2^0| = 237.2 \pm 0.6$ G, $B_4^0 = 0.11 \pm 0.01$ G, and $B_4^0 = -0.8 \pm 0.1$ G (Bonanni, Kiecana, Simbrunner, Li, Sawicki, Wegscheider, Quast, Przybylinska, Navarro-Quezada, Jakiela, Wołos, Jantsch & Dietl, 2007).

In order to estimate the total concentration of substitutional Fe ions in the sample we analyzed the ESR line width of Fe³⁺. The line shape of Fe³⁺ was found to be excellently described by the derivative of the Gaussian error function. At very low Fe concentrations the linewidth is mainly governed by interactions with nuclear magnetic moments of the Ga and N isotopes, however, with increasing Fe concentration dipole-dipole interactions between iron ions (in both charge states) lead to line broadening. The broadening was found to increase linearly with increasing Fe doping (determined by SIMS) and saturate at a value corresponding to $1.8 \cdot 10^{20} \text{ cm}^{-3}$ Fe ions, *i.e.*, 0.4%. Excess iron was found to be incorporated in precipitates of different phases (Bonanni et al, 2007).

Er³⁺ in Si and GaN The high-resolution Fourier transform of the $^4I_{13/2}$ to $^4I_{15/2}$ photoluminescence (PL) transition of Er³⁺ near 1.5 μm , site-selective PL and PL excitation measurements have shown that in MOCVD grown GaN, even at very high doping levels (10^{20} to 10^{21} cm^{-3}), only one type of Er-centers exists. This conclusion has been confirmed by ESR measurements, where an axial symmetry Er³⁺ spectrum was observed with $g_{\parallel}=2.861$ and $g_{\perp}=7.645$ characteristic for substitutional Er ions at Ga sites (C_{3v} symmetry) (Palczewska, Wołos, Kamińska, Grzegory, Bockowski, Krukowski, Suski & Porowski, 2000). Angular dependence of the ESR did not point to the existence of lower symmetry centers (Makarova, Stachowicz, Glukhanyuk, Kozanecki, Ugolini, Lin, Jiang & Zavada, 2008). In contrast, in Er doped silicon numerous light emitting Er centers have been detected by high resolution PL investigations. In ESR experiments also a large number of paramagnetic Er centers, of trigonal and orthorhombic symmetry, were detected but they could not be ascribed to the dominant emitting centers. Preferential formation of one optically active Er center in sublimation MBE grown layers allowed us finally to perform high resolution magneto-optical experiments, which revealed that the symmetry of the center is orthorhombic (C_{2v}). Interestingly, both the ground and the lowest lying excited state of the Er ion turned out to possess only one non-vanishing g-tensor component, which explained why this center cannot be observed in an ESR experiment (Vinh, Przybylinska, Krasil'nik & Gregorkiewicz, 2003; Vinh, Przybylinska, Krasil'nik & Gregorkiewicz, 2004).

Exchange coupling of localized and carrier spins

The exchange coupling between spins of magnetic impurities, **S**, and carrier spins, σ ,

$$j \mathbf{S} \cdot \sigma \quad (2)$$

is the basic coupling which determines properties of DMS. Here j is the exchange integral, proportional to the density of the carrier spin at the impurity site and the exchange parameter $\{N_0 \alpha\}$. ESR investigations of the Knight and Overhauser shifts as well as Korringa scattering effects allowed us to evaluate $\{N_0 \alpha\}$.

Korringa relaxation in GaN:Mn. ESR shows that Korringa scattering, *i.e.*, scattering of the carrier spin on localized Mn spins is the dominant relaxation mechanism of Mn spins in highly *n*-type GaN:Mn bulk crystals (Wołos, Palczewska, Wilamowski, Kamińska, Twardowski, Bockowski, Grzegory & Porowski, 2003).

The temperature dependence of the spin relaxation time determines the effective *s*-*d* exchange constant between conducting electrons and Mn spins. For

GaN:Mn $\{N_0\alpha\} = 14 \text{ meV}$. Weak exchange interaction between Mn^{2+} and GaN conduction band electrons excludes carrier mediation as the origin of the reported high-temperature ferromagnetism in *n*-type (Ga,Mn)N. Thus, other possible explanations, such as indirect or double exchange mechanisms leading to ferromagnetism in homogeneous (Ga,Mn)N or, most probably, precipitating of Mn compounds have to be considered.

Specific Knight shift in ZnO. Originally the Knight shift stands for a change of NMR frequency of magnetic impurities due to exchange coupling with conducting electrons. The complementary effect, i.e., a change of the spin resonance frequency of conducting electrons is known as the Overhauser effect. Similar effects occur in DMS where *s-d* coupling, as defined by Eq. (2), affects the resonance condition as well for the Mn impurity (Knight shift) as for the carrier spins (Overhauser shift) which in DMS leads to a giant spin splitting. Michaluk (2010) was able to simultaneously monitor ESR of Mn impurities and donor electrons in *n*-type ZnO:Mn. Because of the very small spin concentration both shifts are very small, of the order of few Gauss, but due to very narrow lines they can be easily detected. The temperature dependence of the Overhauser shift reflects the Curie like behaviour of the magnetic susceptibility of Mn spins and allows us to determine the *s-d* coupling parameter $\{N_0\alpha\}$ in ZnO as equal to 20 meV.

The phenomenon of the Knight shift induced by donor bound electrons in semiconductors appears to be very specific. Because the spatial distribution of donor electron density is very inhomogeneous the Knight shifts of individual Mn spins may differ by orders of magnitude. Consequently, one has to treat the Knight shift as a random quantity with a special probability distribution. We postulated the logarithmic normal distribution which allowed us not only to explain the experimental data but also to find interesting new effects. In particular, we show that in insulating semiconductors: (i) the ESR line shape is asymmetric, (ii) the Knight shift is strongly reduced, (iii) the ESR line is narrowed – the peak to peak line width is much smaller than the fluctuations of the Korringa field, and finally, (iv) the integrated ESR amplitude of Mn spins is smaller than that corresponding to the real Mn content.

Giant spin splitting is the most characteristic property of DMS. When the concentration of magnetic ions is of the order of one percent then the low temperature *g*-factor of carrier spins is of the order of hundreds. Such a spin splitting is commonly measured by optical methods, e.g. as a splitting of the inter-band absorption lines. An interesting new effect has been found during high field, high frequency ESR measurements of a 2D electron gas in (Cd,Mn)Te layers (Teran, Potemski, Maude, Plantier, Hassan, Sachrajda, Wilamowski, Jaroszyński, Wojtowicz & Karczewski,

2003). Also in this case ESR of conducting electrons and Mn spins can be simultaneously monitored. At magnetic fields for which the excitation energies of free carriers and Mn spins are almost identical, an anomalously large Knight shift is observed. Our findings suggest the existence of a magnetic field induced ferromagnetic order in these structures, which is in agreement with recent theoretical predictions. The *s-d* exchange prevents the crossing of the Mn^{2+} and the electron spin related energy levels. Such an “anti-crossing” is a signature of the collective character of the spin excitations under these specific conditions.

FERROMAGNETIC SEMICONDUCTORS

When the concentrations of magnetic ions and carriers in DMS become high enough ferromagnetic ordering occurs. (Ga,Mn)As is a canonical example of a ferromagnetic semiconductor, with a Curie temperature above 170 K. Magnetic resonance is a powerful tool for the investigation of such materials. Below we present the evaluation of the magnetic anisotropy in (Ga,Mn)As and the explanation for the ferromagnetism observed in GaN:Fe, based on ferromagnetic resonance studies.

Magnetic anisotropy of ferromagnetic (Ga,Mn)As

Magnetic resonance studies allow us to distinguish paramagnetic, ferromagnetic and ferrimagnetic phases in (Ga,Mn)As (Fedorych, Wilamowski, Potemski, Byszewski & Sadowski, 2004a, Fedorych et al., 2004b). The results show that the transition from a ferromagnet to a ferrimagnet is correlated with the metal to insulator transition. The analysis of the dependence of the ferrimagnetic resonance on the direction of the applied magnetic field allows us to evaluate all important parameters describing the magnetic anisotropy. Moreover, the spin wave resonance spectra observed in the ferrimagnetic phase make the estimation of the magnitude and the distance dependence of exchange coupling possible. The experimentally evaluated long range exchange causes an effective averaging of the fluctuations of exchange interactions. As a consequence, in the semi-metallic phase both Mn and hole spin subsystems coherently precess forming a ferrimagnetic system. In the insulating phase fluctuations of the local exchange fields lead to a fast decoherence of the carrier spins and only localized Mn spins contribute to the ferromagnetic moment.

Ferromagnetic nanoprecipitates

Ferromagnetic MOCVD grown GaN films doped with about 1% Fe and unintentionally contaminated with oxygen were investigated by electron spin resonance. Simultaneously with the paramagnetic resonance of isolated Fe^{3+} on Ga sites numerous ferromagnetic

resonance (FMR) signals were detected. The narrow linewidths (below 50 G at the easy magnetization orientation) indicated a well-defined crystallographic structure. In all investigated samples the presence of spheroidal magnetite (Fe_3O_4) precipitates was detected. The fact that the easy and hard axes changed direction at 105 K and the resonance signal in x-band disappeared exactly below the Verwey transition temperature confirmed the identification unambiguously. Bcc iron particles with dimensions small enough to influence the magnetocrystalline anisotropy constant were also observed, but only in samples less contaminated with oxygen. Precipitates of both phases were arbitrarily oriented with respect to the GaN crystallographic axes. In addition, ferromagnetic resonance signals related to two distinctly different hexagonally ordered phases could be distinguished, both with the easy magnetization directions lying in the basal plane. The high uniaxial and small in-plane anisotropy constants of one of them are close to those observed in small diameter hematite ($\alpha\text{-Fe}_2\text{O}_3$) nanoparticles. The other hexagonal phase has a Curie temperature of about 360 K and is, yet, unidentified.

CONDUCTING ELECTRONS IN NON MAGNETIC SEMICONDUCTORS

The ESR spectra of conducting and donor electrons give detailed information on spin relaxation mechanisms. In particular, unique data on spin-orbit coupling can be obtained. Below we present results on conducting electrons ESR in “non magnetic” 2D silicon and GaN.

In addition to spin resonance spectra semiconducting samples placed in a microwave cavity of a standard ESR spectrometer show also spectral features related to electric conductivity, namely cyclotron and magnetoplasma resonances (CR). Simultaneous measurements of spin and electric properties give an exceptional possibility to study the influence of spin on the electric properties.

Electric properties

ESR and CR amplitude. Exploiting the spin resonance of two-dimensional electrons in SiGe/Si quantum wells we determine the carrier density dependence of the magnetic susceptibility χ_m . By combining ESR and transport experiments we have studied high-mobility 2D electron gas in Si/SiGe heterostructures on their way from the metallic to the insulating state. (Wilamowski, Sandersfeld, Jantsch, Többen & Schäffler, 2001). Magnetic susceptibility χ_m determines density of states at the Fermi level, $D(E_F)$.

We found that above $n_s \cong 1.5 \cdot 10^{11} \text{ cm}^{-2}$ $D(E_F)$ is independent on n_s as expected for an ideal 2D gas. It

means that 2D gas in that concentration range is reasonably well described by an effective Thomas-Fermi screening which leads to weak potential fluctuations. At lower n_s , however, $D(E_F)$ decreases with decrease of n_s showing that the potential fluctuations increase and diverge at a critical value of $n_s \cong 7 \cdot 10^{10} \text{ cm}^{-2}$. This process is driven by the mutual dependence of DOS, screening efficiency, and potential fluctuations in a self-consistent, self-amplifying way. Thus, even in our high-mobility modulation-doped samples scattering is obviously dominated by potential fluctuations and the loss of screening rather than by the $e-e$ interaction. Decrease of screening leads to the formation of puddles of mobile carriers with at least 1 μm diameter.

Magneto-plasma resonance. Edge magnetoplasma modes are studied in the two-dimensional electron gas confined at the GaN/ $\text{Al}_x\text{Ga}_{1-x}\text{N}$ interface using standard microwave resonance spectroscopy (Woloś, Jantsch, Dybko, Wilamowski, & Skierbiszewski, 2007). The position and shape of the resonance line are described by the theory for the dimension-dependent plasmon-cyclotron coupling and the Drude model of momentum relaxation. The analysis of the resonance line shape provides a contactless method for the determination of the sheet electron concentration and the mobility of the two-dimensional electron gas. In addition, we observe Shubnikov-de Haas oscillations using the same microwave resonance spectrometry. We compare values for the cyclotron and quantum mobilities obtained from the plasmon-cyclotron linewidth and the magnetic field dependence of the Shubnikov-de Haas signal, respectively.

Spin-orbit coupling – Rashba field

When the crystal lattice or a multilayer structure lacks inversion symmetry then itinerant electrons are affected by an effective spin-orbit field, the so called Rashba field. It occurs, e.g., in wurzite bulk crystals and in 2D structures of cubic semiconductors. The Rashba spin-orbit coupling leads to many specific effects and properties. It governs the spin relaxation and g -factor anisotropy, induces electric dipole transitions, and couples electric and magnetic properties in general. In particular, it couples the spin precession and Zitterbevegung motion of electrons leading to a spin dependent electric conductivity.

g -factor and line width of conducting electrons ESR. In spin resonance studies of two-dimensional conduction electrons in a modulation doped SiGe/Si/SiGe quantum well structure we find a 2D anisotropy of both the line broadening and the g factor (Wilamowski, Jantsch, Malissa & Rösler, 2002)). They can be explained consistently employing the Rashba term:

$$H_{BR} = \alpha(\mathbf{k} \times \boldsymbol{\sigma}) \cdot \mathbf{n}, \quad (3)$$

where \mathbf{k} and $\boldsymbol{\sigma}$ are electron momentum k-vector and spin, \mathbf{n} is a normal vector indicating a direction effective electric field, and α is a material parameter describing efficiency of Rashba coupling. The Rashba term (3) turns out here to be the dominant coupling between electron orbital motion and spin.

We have shown that ESR measurements provide a very sensitive tool to evaluate the Rashba constant, α . The Rashba field is the origin of the observed anisotropies of the g factor and of the ESR linewidth. Both of them increase with increasing carrier density. Extrapolating the mean g value for low carrier density we find the subband edge g factor of 2D electrons in Si, $g = 2.00073$. For perpendicular field the Dyakonov-Perel linewidth broadening originating from Rashba fields vanishes and the ESR is easiest to observe for that orientation. We obtain a BR parameter of $\alpha = 0.55 \times 10^{-12} \text{ eV} \cdot \text{cm}$ - three orders of magnitude smaller than in quantum well structures based on III-V semiconductors, consistent with the much smaller spin-orbit coupling in Si. The weak spin-orbit coupling in Si structures means that spin relaxation is also very slow.

The effect of cyclotron motion on spin relaxation.

We investigate the spin relaxation of two-dimensional electrons in a Si/SiGe quantum well by means of electron spin resonance. The longitudinal spin relaxation time, T_1 , is determined from the saturation of the ESR signal (Wilamowski & Jantsch, 2004). Simultaneous observation of cyclotron resonance allows us to evaluate the influence of momentum scattering on spin relaxation. We identify thus a dominant contribution due to the Dyakonov-Perel (DP) mechanism which is expected to be more efficient for slow momentum-relaxation while for low mobility the Elliott-Yaffet mechanism dominates. The observed relaxation times of microseconds can be explained, however, only by an additional motional narrowing due to modulation of the spin-orbit coupling caused by the cyclotron motion. The latter is evidenced by the observed dependence of spin relaxation on the direction of applied magnetic field which changes the cyclotron frequency of the 2D electrons. Evaluation of the DP mechanism shows that T_1^{-1} for high electron mobility can be effectively reduced by an external magnetic field.

Rashba field caused by electric current

ESR tuned by dc electric field. We investigate the Zeeman splitting of the two-dimensional electron gas in an asymmetric silicon quantum well, performing ESR experiments. Applying a small dc electric current we observe a shift in the resonance field due to the additional current-induced Bychkov-Rashba type of spin-orbit field (Wilamowski, Malissa, Schäffler &

Jantsch, 2007). These data demonstrate that a dc current allows one to tune the ESR signal. Important is that the tuning can be addressed locally, e.g., to a nanowire without heating the rest of a sample in contrast to methods employing a resonator.

Current induced ESR (CI ESR). We also show that a high frequency current may induce electric dipole spin resonance very efficiently (Wilamowski, Ungier & Jantsch, 2008). In asymmetric quantum wells an electric current induces an effective magnetic field acting on conduction electrons due to Rashba coupling. A high-frequency electric current can thus induce spin precession. We present and analyze a model for this current-induced electron spin resonance. In the low-frequency range, in high-mobility two-dimensional layers, CI ESR is the dominant mechanism of spin excitation. In the high-frequency limit, when the displacement current dominates, the drift current and momentum dissipation become irrelevant. There the CI ESR becomes equivalent to the well-known electric-dipole spin resonance. We show that in both limits the line shape of the power absorption spectra is described by the imaginary component of the dynamic magnetic susceptibility, $\chi''(\omega)$, in contrast to experiment, indicative of another mechanism.

In high-mobility systems, ESR excitation by current is much more efficient than by magnetic-dipole transitions. CI ESR is a form of ESR induced by electric-dipole transitions where damping of the electron motion leads to a reduction of the transition matrix element of electric dipole ESR by a factor $\omega\tau_p$. Here τ_p is momentum relaxation time and ω is the frequency. We discuss the efficiency of the resonance excitation by an rf electric field and the resulting Rabi frequency, and we evaluate one of the channels for the energy transfer. The strong enhancement of the microwave magnetic field will be most pronounced at low temperatures where the mobility of the 2D electrons is the highest. Detailed analysis of the experimental data shows that, in fact, there are two parallel mechanisms of the resonant power absorption. In addition to the discussed above CI ESR, due to a dependence of the electron velocity on the direction of electron spin, an excitation of the spin precession, e.g. by electric current, causes an oscillatory motion of electron. This oscillatory motion in rf electric field leads to additional power absorption, the spin dependent Joule heating. This absorption occurs at the ESR condition only but in contrast to CI ESR it can be as well positive as negative. It can enhance or decrease the Joule heat. We show that the Joule heat contribution to the ESR signal is described by a linear combination of real, $\chi'(\omega)$, and imaginary part, $\chi''(\omega)$, of magnetic susceptibility. Because both channels, CI ESR and the spin dependent Joule heat are of similar magnitude the resulting line shape of ESR induced by electric field is

asymmetric with the asymmetry ratio dependent on $\omega\tau_p$.

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