PHYSICAL REVIEW B VOLUME 56, NUMBER 8 15 AUGUST 1997-II

Electron spin resonance on a two-dimensional electron gas

N. Nestle
MPI für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

G. Denninger, M. Vidal, and C. Weinzierl
Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany

K. Brunner, K. Eberl, and K. von Klitzing
MPI für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

(Received 7 May 1997)

Electron-spin-resonance (ESR) data obtained from modulation-doped Si/$Si_{1-y}C_y$ heterostructures are reported. The observed signals show that the small number of electron spins in these two-dimensional structures is still sufficient to detect ESR directly, and we have strong indications that the prominent line in the spectra is due to conduction electrons confined in the $Si_{1-y}C_y$ layers, forming a two-dimensional electron gas. We compare our results to the case of confined electrons in III-V heterostructures and quantum wells, where only indirectly detected ESR experiments have been reported so far. [S0163-1829(97)50732-6]

I. INTRODUCTION

Two-dimensional electron gas (2DEG) systems are of great interest in semiconductor physics and in technology. The confinement of the electrons in one dimension leads to interesting quantization effects and can be used for the construction of highly efficient electronic and optoelectronic devices.

Direct electron-spin-resonance (ESR) investigations of the quantized electrons are complementary to transport methods and optical experiments. ESR experiments probe directly the equilibrium distribution as well as the chemical environment of the electrons. Both static properties (e.g., intensity as a quantitative measure of the electron density) and dynamic properties (e.g., spin relaxation) are accessible. The method requires only moderate magnetic fields and the experiments can be performed in semiconductors with direct and indirect band gap. Since in two-dimensional electron gases the highly conducting layers are very thin, the problems with the skin effect usually encountered in ESR experiments on conducting samples should be small or absent.

In this contribution we present results on a 2D electron system, where layers of $Si_{1-y}C_y$ (the carbon concentration y is in the 0.005–0.04 range) are grown by molecular-beam epitaxy (MBE) between Si buffer layers and form a quantum well for the electrons.[1,2] The $Si_{1-y}C_y$ material system has become available only recently[2–5] and it is particularly suited for direct ESR investigations, as shown in this contribution. By direct ESR we mean detection of the absorption via magnetic dipole transitions in a conventional continuous-wave ESR spectrometer. Si/$Si_{1-y}C_y$ is not just an interesting system for doing direct ESR experiments but also a very promising material for various electronic quantum device applications. Especially important aspects are its good compatibility with standard Si-based VLSI technology and its good chemical and thermal stability.

Before we proceed to our experimental results we will give a short review of indirect ESR investigations in the III-V systems like GaAs/Ga_xAl_{1-x}As and address the specific problems for direct ESR in these systems.

II. REVIEW OF EXPERIMENTS IN 2DEG's

The main obstacle to ESR experiments in typical 2D samples is the rather low number of spins. These sensitivity problems were overcome by using optical detection methods for ESR, known from work on bulk semiconductors. The experimental methods are reviewed in Ref. 6. In Ref. 7 the optical detection method was extended to the detection of ESR in GaAs/Ga_xAl_{1-x}As heterostructures. The ESR line of the conduction electrons had a width of ≈30 mT (full width at half maximum). A drawback consists in the fact that the method is more or less restricted to direct semiconductors. Furthermore, the method works best with p-type material, since the total electron spin polarization in the conduction band is essential and only optically excited electrons are spin polarized in excess to the thermal polarization.

Electrical detection via resonant changes in the quantum Hall transport behavior was successfully used to detect the ESR (Ref. 8) and even NMR:[9] due to the spin splitting of the Landau levels, spin gaps are opened in the energy spectrum with vanishing conductivity if the Fermi energy is located in such a gap. Under this condition, spin-flip transitions lead to a relatively strong photoconductive signal.

Direct ESR detection in a dedicated resonator was reported on a 2D inversion layer in a Si metal-oxide-semiconductor field-effect transistor structure.[10] However, in addition to being experimentally quite difficult, these results were heavily disputed afterwards.[11,12] In a detection scheme via spin-dependent transport,[13] resonant changes in the conductivity of a 2DEG in a 2D Si inversion layer due to spin-dependent scattering on neutral impurities were measured.

The most popular 2DEG's are made from the III-V systems. A major obstacle for direct ESR on these 2DEG's is

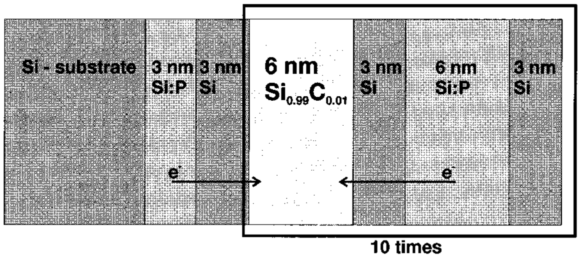

FIG. 1. Modulation-doped $Si/Si_{1-y}C_y$ multi-quantum-well sample used in most experiments (sample 3).

the hyperfine interaction of the electrons with the nuclei: In these materials, there are NMR-active nuclei with appreciable magnetic moments and high natural abundance (e.g., ^{31}P, ^{75}As, ^{27}Al, ^{69}Ga, ^{71}Ga, ^{115}In). Furthermore, there is a rather strong hyperfine interaction between electrons and nuclei, due to the high Z. The resulting hyperfine splitting already leads to an inhomogeneous line broadening on the order of 20 mT (see, e.g., Ref. 7). The situation is aggravated by dynamic polarization effects of the nuclei: At low temperatures (where one has the best thermal polarization of the electrons), the nuclear relaxation times in III-V semiconductors are very long (several minutes or even hours). Upon excitation of electrons, polarization transfer to the nuclear system sets in and dynamically polarizes the nuclei.[14] The field produced by the polarized nuclei is of the order of several 100 mT. Therefore, the position of the line will be shifted drastically during the buildup of the nuclear polarization. Furthermore, the efficiency of the magnetization transfer (or of the nuclear relaxation) is often different at different points in the sample. This leads to a further inhomogenous broadening of the linewidth. Even when suppressing these nuclear polarization phenomena by NMR saturation of the nuclei, one still is left with the inhomogeneous broadening due to the hyperfine interaction.

Another problem is that the g factors in some III-V materials (e.g., $g=-0.44$ in GaAs and $g\approx-51$ in InSb) are outside the field range of standard ESR spectrometers. Furthermore, the large deviations from the free-electron g factor of 2.002 319 in these semiconductors depend on the band gap, on the spin-orbit coupling, and on the crystal structure. Fluctuations of the g factor occur in heterointerfaces (''g strain''). This effect will further increase the linewidth and therefore decrease the sensitivity in ESR experiments.

III. EXPERIMENTAL RESULTS AND DISCUSSION

In $Si_{1-y}C_y$ heterostructures and quantum wells at least some of the problems for direct ESR are not present: The natural abundance of nuclei with nuclear spin is low (^{29}Si, 4.67%, ^{13}C, 1.11%) and the hyperfine interaction is small (low Z). Due to the rather small spin-orbit interaction of the electrons in Si, the g factor of the conduction electrons in Si and in $Si_{1-y}C_y$ is near the free-electron value. Thus, there is no expected broadening due to g strain.

$Si/Si_{1-y}C_y$ heterostructures containing 2DEG's have been successfully manufactured by MBE growth techniques only recently.[1,2,5] Due to strong tensile biaxial strain in the $Si_{1-y}C_y$ layers (typical C content between 1% and 3%), a type-I quantum well is formed in the $Si_{1-y}C_y$ layer. While the potential well for the holes is quite shallow, the electron band offset is of the order of 50 meV for every percent of C alloyed into the Si.[2]

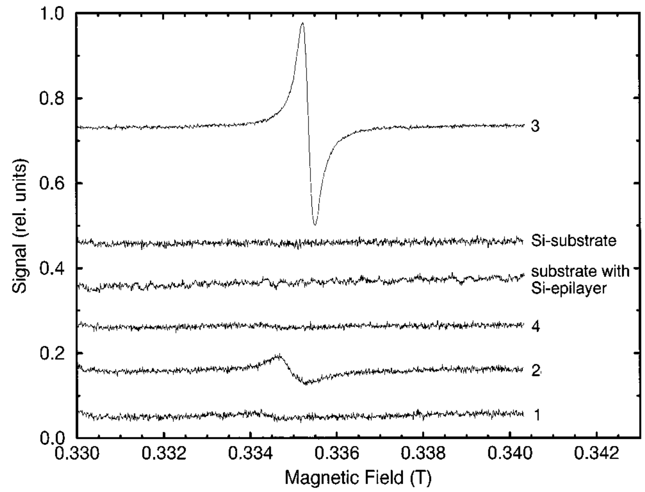

FIG. 2. ESR spectra of 2DEG samples and reference samples. Sample 3, 10 quantum wells, see Fig. 1; Sample 2, single quantum well; Sample 4, 30 quantum wells, no doping; Sample 1, doping layer, no quantum well.

Presently there is still no reliable protocol for making good Ohmic contacts to $Si/Si_{1-y}C_y$ 2DEG's and the electron mobilities achieved so far are not yet optimal.[5] Contactless characterization methods such as ESR are useful in this case.

The sample used in our investigation is depicted in Fig. 1. The quantum well is formed in the 6-nm-thick $Si_{0.99}C_{0.01}$ section, sandwiched between two 3-nm Si buffer layers. The electrons are supplied by the Si:P layers doped with P (4×10^{18} cm^{-3} doping concentration, 6 nm thick). This modulation doped structure is repeated 10 times in our sample 3 and capped with a Si protection layer. In addition, we investigated several control samples: the pure substrate, the substrate with MBE-grown Si epilayer, a nominally undoped $Si/Si_{1-y}C_y$ multiple-quantum-well sample with 30 layers (sample 4) and a Si epilayer with the same doping as one of our 2DEG samples, but without the $Si/Si_{1-y}C_y$ quantum-well layer (sample 1). In addition, a sample with a single quantum well and a similar doping concentration (sample 2) was investigated, but the signal/noise ratio was much lower than in sample 3.

The experiments were performed with a Bruker X-band spectrometer (9.34 GHz) using a standard rectangular resonator (TE102 mode, Bruker ER4102ST) and a helium-flow cryostat (Oxford 900). Microwave power up to 100 mW was available, and the temperature range was 300 K down to 5 K. The results of the measurements at 5 K (where the sensitivity is the highest) are depicted in Fig. 2. One can clearly see from the results of the multiple-quantum-well sample (3) and of the single-quantum-well sample (2) that there is a narrow ($\approx$0.23 mT half-width at half maximum) line with Lorentzian line shape in the samples with both doping layers and $Si/Si_{1-y}C_y$ layers. As usual in ESR spectroscopy, the measurements are presented as the first derivative of the signal. In all the other samples such a signal is not observed. This is a first indication that the narrow line belongs to the 2DEG.

The g factor of 1.9993 ± 0.0001 [calibrated to the conduction electron g factor of Li metal particles in LiF (Ref. 18)] is near the values obtained for conduction electrons in Si.[15,16,17]

A closer inspection of the observed lines in the various samples reveals several other details: one can obtain a better fit to the single line signal by including an additional weak, broader ($\approx$0.3 mT) line at a slightly lower g factor. The separation of the two signals is only $\approx$0.1 mT and thus below the linewidth. At low doping concentrations and at low temperatures, one can observe satellite lines at $\pm$2.1 mT symmetrical to the main line, due to the hyperfine interaction at the ^{31}P (spectra are not shown in Fig. 2). These satellite lines suggest that not all P donors are ionized under these conditions (ionization energy of P in Si: 45 meV). From various ESR studies in P-doped Si (see, e.g., Ref. 16 and the references therein) it is known that the g factors of the donors are very close to those of the free conduction electrons. A reliable separation on the field axis is not possible at the signal/noise ratio and linewidth conditions in our experiments at 9.34 GHz.

In order to further clarify the origin of the main ESR line, we measured its temperature and saturation behavior. The most important results are the following. The intensity versus temperature corresponds only very roughly to a Curie law, and a closer look reveals systematic deviations. In Fig. 3 we plot the ESR intensity (obtained via a careful fitting procedure of the line to a Lorentzian) versus $1/T$. A Curie-type behavior would be expected for isolated electrons (e.g., electrons on the P donors) or for electrons with only a small exchange interaction. The intensity would scale with $1/T$ in this case. On the other hand, for a Fermi system with a high Fermi energy $E_F \gg kT$ we expect a constant intensity independent of T (Pauli susceptibility).

The data in Fig. 3 can be quantitatively explained by a linear superposition of the contribution of a Fermi system with a rather low Fermi energy (5 meV to 10 meV) and a Curie contribution. In addition, we include a temperature-dependent density of the electrons in the quantum well. The Pauli part of the susceptibility is due to the electrons in the quantum well. The density of states for 2D electrons is $n(E)=M_x m^*/(\pi\hbar^2)$, if only the lowest subband in the quantum well is occupied. M_x is the number of equivalent valleys of the conduction-band minima. The Fermi energy E_F is proportional to the density n_{2D} with $E_F=(\pi\hbar^2 n_{2D})/(M_x m^*)$. We use $m^*=0.19m_0$ and $M_x=2$, since only the two valleys in the growth direction are lowest in energy. The corresponding density of states is 1.587×10^{14} eV^{-1} cm^{-2}. The density of donor atoms is 2.4×10^{12} cm^{-2} and if all the donors are ionized and occupy the quantum well, a Fermi energy of 15.12 meV would result.

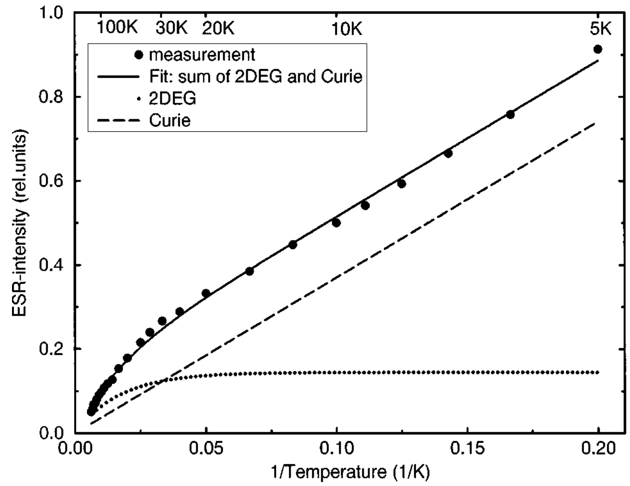

FIG. 3. Intensity of the ESR signal (sample 3) versus $1/T$. Solid line: fit to the data according to Eq. (3). The individual Pauli-type (dotted) and Curie-type (dashed) contributions are shown separately.

The magnetization of the 2D electrons (and thus the ESR intensity) can be calculated under the assumptions $g\mu_B B \ll E_F$ and $g\mu_B B \ll kT$, where μ_B is the Bohr magneton, B the external magnetic field, and k the Boltzmann constant. Both conditions are satisfied in our case, since at $B\approx$ 0.35 T we have $g\mu_B B=40.6$ μeV, which is small compared to E_F and kT even at low temperatures. However, due to the rather low Fermi energy, the temperature variation of the magnetization is noticeable at higher temperatures and has to be taken into account.

We proceed by calculating the temperature dependence of the electrochemical potential $\mu(T)$:

$$\mu(T)=kT\ln\left[\exp\left(\frac{E_F}{kT}\right)-1\right], \tag{1}$$

and then solving for the magnetization,

$$M_{2D}(T)=\frac{M_x m^*}{2\pi\hbar^2}(g\mu_B)\int_0^\infty\left[\frac{1}{1+\exp\left(\frac{E-\mu(T)-g\mu_B B/2}{kT}\right)}-\frac{1}{1+\exp\left(\frac{E-\mu(T)+g\mu_B B/2}{kT}\right)}\right]dE. \tag{2}$$

The solution of Eq. (2) is:

$$M_{2D}(T)=\frac{M_x m^*}{2\pi\hbar^2}(g\mu_B)\left\{kT\ln\left[\exp\left(\frac{E_F}{kT}\right)-1+\exp\left(\frac{-g\mu_B B}{2kT}\right)\right]\right.$$
$$\left.-kT\ln\left[\exp\left(\frac{E_F}{kT}\right)-1+\exp\left(\frac{g\mu_B B}{2kT}\right)\right]+g\mu_B B\right\}. \tag{3}$$

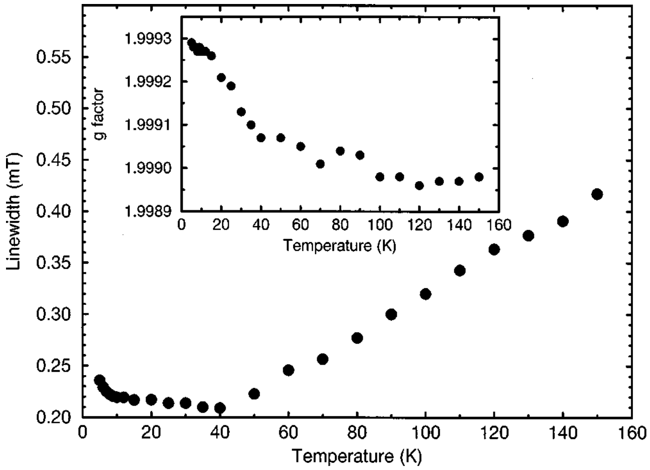

FIG. 4. Temperature dependence of the g factor and the linewidth (sample 3).

The solid line in Fig. 3 is a fit to the data points of a linear superposition of $M_{2D}(T)$ from Eq. (3) and a Curie-type behavior $\propto 1/T$. Both individual contributions are shown in Fig. 3 and we arrive at a $E_F = (10 \pm 3)$ meV from the fit of the intensity versus T. This is consistent with the result we expect from the density of electrons: with the calculated Fermi energy of 15.12 meV from the donor density, we conclude that $(66 \pm 20)\%$ of the electrons are in the quantum well at low temperatures. A consistent conclusion can be drawn from the individual contributions $n_{\mathrm{Pauli}}/n_{\mathrm{Curie}}$. If we interpret the Pauli contribution as arising from the electrons in the quantum well and the Curie contribution as arising from electrons localized at the donor atoms, we conclude quantitatively that about 90% of the electrons are in the quantum well at low temperatures. Considering the experimental error of $\approx$30% in the determination of E_F from the intensity data, this agreement is good.

We have measured the linewidth and the g factor as a function of temperature, and the results are depicted in Fig. 4. The linewidth slightly decreases from 0.25 mT at 4.2 K to 0.21 mT at 40 K, followed by a steep increase for temperatures above 40 K. Above 150 K, the ESR line is no longer resolved due to both the line broadening and the decrease in intensity. The g factor shows a small but systematic monotonic decrease with temperature. We note that the variation of the linewidth with the temperature could be explained by the combined influence of motional narrowing and the spin flips by phonons at higher temperature. The temperature variation of the g factor could result from the temperature dependence of the band gap, but no quantitative model is available at present to our knowledge. Up to power levels of 100 mW we have seen no indication of saturation effects either in the intensity or in the linewidth. This could be expected for conduction electrons with $T_1 \approx T_2$.

IV. CONCLUSION

In conclusion, we have presented ESR results from $Si/Si_{1-y}C_y$ samples, which can be interpreted as resulting in part from the electrons in the quantum well. These results show that there is no inherent sensitivity problem for ESR experiments on 2DEG's, provided that the linewidth is sufficiently narrow. Similar investigations can be envisaged for suitable systems such as II-VI semiconductors, where the natural abundance of nuclei with magnetic moments is low as well. Our experiments show that with quantitative relative values for the intensity versus T one can use ESR to distinguish between different contributions of the magnetization and thus single out the 2DEG part. Since the method requires no contacts, it could allow a rapid characterization of such samples. Extensions to higher frequency (34 GHz, 94 GHz) are under way and should help to disentangle the different magnetization contributions by their g-factor differences.

ACKNOWLEDGMENTS

We thank T. Ruf for critical reading of the manuscript. C.W. acknowledges financial support from ''Schwerpunkt Magnetische Resonanz in Metallclustern.''

[1] K. Brunner, K. Eberl, and W. Winter, Phys. Rev. Lett. **76**, 303 (1996).
[2] K. Brunner, W. Winter, and K. Eberl, Phys. Bl. **52**, 1237 (1996).
[3] S.S. Iyer, K. Eberl, M.S. Goorsky, F.K. LeGoues, J.C. Tsang, and F. Cardone, Appl. Phys. Lett. **60**, 356 (1992).
[4] K. Eberl, S.S. Iyer, S. Zollner, J.C. Tsang, and F.K. LeGoues, Appl. Phys. Lett. **60**, 3033 (1992).
[5] W. Faschinger, S. Zerlauth, G. Bauer, and L. Palmetshofer, Appl. Phys. Lett. **67**, 3933 (1995).
[6] *Optical Orientation*, edited by F. Meier and B.P. Zhakarchenya (North-Holland, Amsterdam, 1984).
[7] M. Krapf, G. Denninger, H. Pascher, G. Weimann, and W. Schlapp, Solid State Commun. **74**, 1141 (1990).
[8] D. Stein, K. von Klitzing, and G. Weimann, Phys. Rev. Lett. **51**, 130 (1983).
[9] M. Dobers, K. von Klitzing, and G. Weimann, Phys. Rev. B **38**, 5453 (1988).
[10] W.J. Wallace and R.H. Silsbee, Phys. Rev. B **44**, 12 964 (1991).
[11] A. Stesmans, Phys. Rev. B **47**, 13 906 (1993).
[12] R.H. Silsbee and W.J. Wallace, Phys. Rev. B **47**, 13 909 (1993).
[13] R.N. Ghosh and R.H. Silsbee, Phys. Rev. B **46**, 12 508 (1992).
[14] M. Krapf, G. Denninger, H. Pascher, G. Weimann, and W. Schlapp, Solid State Commun. **78**, 459 (1991).
[15] G. Feher, Phys. Rev. **114**, 1219 (1959).
[16] V. Dyakonov and G. Denninger, Phys. Rev. B **46**, 5008 (1992).
[17] A.M. Portis, F. Kip, C. Kittel, and W.H. Brattain, Phys. Rev. **90**, 988 (1953).
[18] A. Stesmans and G. van Gorp, Rev. Sci. Instrum. **60**, 2949 (1989).