Local Lattice Information for GaN and ZnO relevant to Spintronics

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Abstract

ZnO and GaN have been studied with the perturbed angular correlation method (PAC) using the ^{100}*Pd-probe. Rutherford Backscattering Channeling (RBS-C) supports the PAC results. Annealing of the materials does not lead to unique probe environments, effectively excluding that a significant fraction of probes is integrated substitutionally.*

Keywords: Spintronics, ZnO, GaN, Perturbed Angular Correlations, Rutherford Backscattering

1. INTRODUCTION

For dilute magnetic semiconductors (DMS) based on GaN and ZnO with Curie temperatures above room temperature, theoretical work predicts promising and truly applicable properties. This may lead to suitable spintronics applications [1]. Specifically, doping with Co, Mn or Fe transition metals has been suggested as a possible way to create such DMS. However, real proof of ferromagnetic order in these semiconductors caused by isolated transition metal dopants has yet to be found. The perturbed angular correlation (PAC) probe ^{100}Pd/^{100}Rh is isoelectronic to Co and it is therefore a perfect tool to investigate how transition metals are incorporated into these compounds. An additional advantage of the ^{100}Pd/^{100}Rh probe is its strong sensitivity to magnetic fields.

In this work the ^{100}Pd/^{100}Rh probe has been introduced into the semiconductor samples via recoil ion implantation. Thus implantation-induced lattice defects and the annealing of the lattice can be studied. This is significant, since ion implantation is an important doping method for semiconductor materials .

This conference contribution gives a status report for the ongoing project. Preliminary results for ^{100}Pd/^{100}Rh in GaN and ZnO are reported and backed by Rutherford Backscattering Channeling measurements. In addition measurements investigating the influence of external magnetic fields on the probe have been performed.

2. EXPERIMENTAL DETAILS

ZnO single crystals samples with (0001) and (1010) orientation, freestanding GaN films and 6 µm thick GaN films on sapphire substrates were recoil-implanted with the ^{100}Pd/^{100}Rh probe as described in detail elsewhere [2]. Sample sizes were in the range from 2×2 mm^2 to 5×5 mm^2. The probe was produced using the fusion evaporation reaction ^{92}Zr(^{12}C, 4n)^{100}Pd at a beam energy of 70 MeV. After recoil implantation PAC spectroscopy [3,4] was performed at room temperature.

The samples were measured as-implanted, and following isochronal annealing for 10 min at increasing temperatures up to 1100 K. Ratio functions $R(t) = A_{22}G(t)$, which reflect the magnitude A_{22} and the time-dependence $G(t)$ of the angular correlation anisotropy, have been extracted from the data in the conventional way [3,4]. The extracted ratio functions are discussed below. In some PAC measurements an external magnetic field was applied with the sample housed in a specially designed ferromagnetic container.

For the RBS channeling measurements, a 10×5 mm^2 ZnO sample was implanted with ions of the stable ^{106}Pd isotope at a fluence of 3×10^{14}. One half of the sample was masked with a piece of silicon, so that the He-beam of the RBS-channeling measurement could be directed to sense both implanted and pristine regions on the same sample.

3. RESULTS

3.1 Annealing of ZnO and GaN

Figure 1 shows PAC ratio functions $A_{22}G(t)$ measured for ^{100}Pd/^{100}Rh implanted in ZnO and GaN after annealing at T_a = 600°C and 900°C (Fig. 1 a). In the measurement presented in Figure 1 b,c an external magnetic field of 0.48(5) T was applied. As a consequence of the ion implantation, the crystal lattice at probe sites is strongly disturbed. This results in the high damping of the ratio functions. This is caused by non-uniform probe environments with variations in distant defects or slightly different defect locations. The slight periodic modulations of the ratio functions are related to the quadrupole interaction of the probes with the electric field gradient at their site and can be fitted, considering the large damping. Fits are also shown in the figures. For ZnO the fits have been obtained by postulating two specific probe sites. The first probe site in the lattice corresponds to a slow frequency $\omega_{0,1}$, which might be caused by ^{100}Pd/^{100}Rh being integrated substitutionally on a lattice site. The probe fraction associated with this first site is of the order of 45 %. The second modulation with frequency $\omega_{0,2}$, as evidenced by the sharp drop of the data during the first 50 ns, is affect by much stronger damping. This second modulation may be interpreted as an ensemble of different types of local probe environments in the crystal that cannot be distinguished.

In the case of GaN no unique site can be identified in the data for the as-implanted sample. Furthermore annealing at T_a = 600 °C or 900 °C does not change the ratio function and its interpretation significantly. This implies that the probe is not integrated into the GaN lattice in a well-defined fashion.

For ZnO, the ratio function measured after annealing at 600 °C shows somewhat less damping compared to annealing at lower temperature. This may imply an onset of lattice recovery. Since the modulation frequency $\omega_{0,1}$ is affected by damping of the order of δ = 200 - 400 % it can only be defined to be within the range $\omega_{0,1}$ = 3 to 5 Mrad/s. The final annealing at 900°C appears to have cured the ZnO lattice in some places with a fraction of 45% of probes on the first probe site and a better definition of the modulation frequency with $\omega_{0,1}$ = 3.5(1) Mrad/s. This corresponds to a V_{zz} of the EFG of 10^{16} V/cm² which is in good agreement to measurements of other PAC probes (^{111}In [8]). However, the damping of δ = 64(5) % may still be considered high.

The lack of lattice recovery may be explained, if excessive lattice damage was caused by ^{12}C projectile scattering. However, Dogra *et al.* [8] showed that after production of ^{111}In/^{111}Cd and recoil-implantation of this probe into GaN and ZnO using the same set-up, under very similar experimental conditions, a complete annealing of the lattices is indeed possible with the ^{111}In/^{111}Cd probe being incorporated substitutionally.

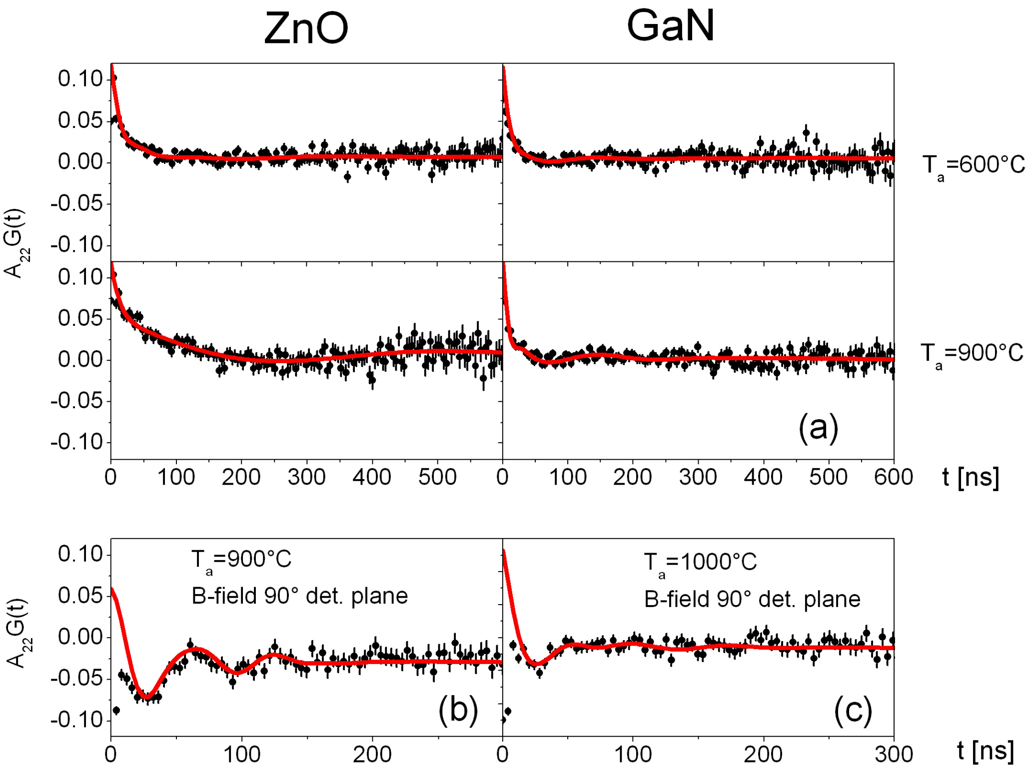

Figure 1 Measured PAC ratio functions for ZnO and GaN samples implanted with the ^{100}Pd/^{100}Rh probe. For details see text.

3.2 External magnetic field

The strong damping of the ratio functions in Fig. 1(a) might be indicative of the onset of internal magnetic orientation, since the presence of magnetic fields of different magnitude causes a distribution of interaction frequencies. Similar to non-unique probe environments, such a finite distribution strongly dampens any signal, which may arise from a large fraction of probes on substitutional lattice sites with unique environment. An external magnetic field can shift the mean of this finite distribution, so that the unique signal due to the field gradient may then be identified. This motivated an additional series of measurements with an external magnetic field of 0.5 T. The measured ratio functions after annealing at 900 °C and 1000 °C for ZnO and GaN, respectively, are shown in Figure 1(b,c).

The influence of the external magnetic field on the measurements can be clearly seen. For both, GaN and ZnO, the ratio function can now be described postulating the same two probe fractions discussed above. In the fitting it has been considered that the probes now experience a combined interaction, which involves, in addition to the electric field gradient, the external magnetic field.

3.3 RBS-channeling measurements

The un-implanted region of the ZnO sample shows after annealing at 900 °C a minimum yield of 3.7 %. The implanted part has a minimum yield of 4.7 % showing very good recovery of the implantation damage Fig. 2(a). Both yields are in the range of expected values of 2-4 % [9]. The annealed crystal is thus of very good quality. The analysis is made difficult by unknown surface impurities in the mass range of Pd. Results are, however, conclusive. Fig. 2(b) shows the respective Pd-peaks. About 40 % of the Pd is found to be on substitutional Zn sites, which agrees with the PAC measurement.

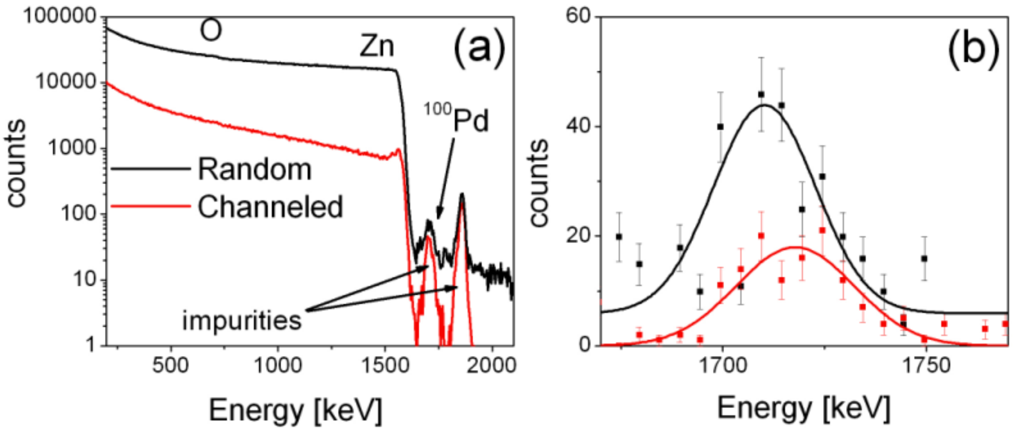

Figure 2 (a) RBS-Channeling measurements for ZnO implanted with ^{106}Pd. The Pd-peak is masked by heavy unknown impurities on the surface, which are also evident in the un-implanted region of the sample (not shown). (b) The Pd peak for random and channeled geometry. The curves are to guide the eye. Roughly 40 % of the ^{106}Pd is substitutional in the lattice.

4. CONCLUSIONS

Time-differential perturbed angular correlation spectroscopy has been performed on GaN and ZnO samples using the probe ^{100}Pd/^{100}Rh in an effort to ascertain how transition metals are incorporated into the lattice of these compounds. The results suggest that, in contrast to the probe ^{111}In/^{111}Cd, the ^{100}Pd/^{100}Rh probe is not incorporated in a unique fashion, rather tending to be in a multitude of disturbed local lattice environments. Results for ZnO show that after annealing at 900 °C the ZnO lattice recovers somewhat with a fraction of 45% of probes possibly substitutionally located on the Zn site. This is supported by RBS-channeling measurements. Additional PAC measurements with an external magnetic field have excluded that any unique signal is obscured by the possibility of internal magnetic ordering.

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