

## Weak localization in ZnO:Ga and ZnO:Al thin films

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**Abstract.** The temperature dependences of the resistivity, magnetoresistance and the Hall effect of ZnO thin films doped with Ga and Al were investigated at low temperatures. According to obtained experimental data the dimensionality of the investigated films with respect to the weak localization theory changes from 2D to 3D with an increase of magnetic field and temperature. To describe the observed negative magnetoresistance under these conditions we derived a new expression for the weak localization correction to the conductivity. It was found that the obtained expression describes magnetoresistance of investigated films much better than all known expressions for negative magnetoresistance related to weak localization. The values of the electron diffusion length during the phase relaxation time of wave function were obtained by fitting of experimental magnetoresistance with derived expression. Obtained values are consistent with the applied approach.

### Introduction

The theory of quantum correction to conductivity (QC) was developed to explain experimentally observed peculiarities in the low temperature behavior of the resistivity and negative magnetoresistance (NMR) in disordered conductors [1-3]. Typically NMR in such systems is described by correction to the conductivity due to weak localization (WL). The expressions for corrections to conductivity due to WL for two-dimensional (2D), quasi two-dimensional (quasi-2D) and three-dimensional (3D) systems and corresponding magnetoresistance are known and widely used [4-5]. But there are only few publications devoted to the analysis of weak localization phenomena for the systems with dimensionality intermediate between purely 3D and quasi 2D [6].

The dimensionality of electron system with respect to WL theory in absence of magnetic field is determined by the ratio of diffusion length for the phase relaxation time of wave function ( $L_\varphi = \sqrt{D\tau_\varphi}$ ) to the least size of the sample ( $d$ ) [4-5]. In the presence of magnetic field the value of magnetic length ( $l_B = \sqrt{\hbar/eB}$ ) also should be taken into account [4-5]. The magnetic length decreases with an increase of magnetic field. If  $L_\varphi \gg d$  but  $l_B \ll d$  the electron system will behave as 3D system with respect to the WL theory. Therefore expressions for quasi-2D or 3D electron system will not describe

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the magnetoconductivity of the system in the range of magnetic fields where dimensionality transition caused by the decrease of  $l_B$  takes place. The accurate consideration of weak localization in the film should account the boundary effects as shown in Ref. [6]. The author of this ref. showed that the WL correction to the conductivity of semi-infinite samples and films always comprises 2D and 3D contribution. The formula for the magnetoconductivity of thick films related to WL and accounting both contributions was derived in Ref. [6]. However the expression for magnetoconductivity related to weak localization for the case of 2D-3D crossover is absent in available literature.

This research is devoted to the experimental and theoretical study of the weak localization phenomenon in heavily doped ZnO films with a crossover from quasi 2D to 3D induced by temperature or magnetic field.

### Experimental

Investigated films were synthesized by the oxygen-assisted chemical vapor deposition method (MOCVD) and magnetron sputtering technique (MS). Films were deposited on monocrystalline substrates of R-sapphire, Yttrium stabilized zirconia (YSZ) and on glass. The films differed in thickness and donor impurity concentration. Some characteristics and deposition parameters of studied films are listed in Table 1.

**Table 1.** Deposition parameters of studied films

Film	Deposition technique	Substrate	T <sub>deposition</sub> (°C)	Thickness (nm)	Concentration impurity
R1	MOCVD	R-sapphire	600	60	7.2 at% (Ga)
Y1	MOCVD	Zr <sub>2</sub> O <sub>3</sub> (Y <sub>2</sub> O <sub>3</sub> )	600	90	6.8 at% (Ga)
Al	MS	Glass	70	100 <sup>b</sup>	2 wt% (Al)

According to the X-ray diffraction data all films deposited by oxygen-assisted MOCVD were epitaxial with orientation determined by substrate. The details on synthesis and structure analysis of films could be found in Ref. [7-8].

Resistivity and Hall effect of the films have been measured by the standard four probe method. Samples were made rectangular form with typical dimensions 8×3 mm. Plates of electrical contacts were made by evaporation of Au and copper wires were soldered to the plates. For the measurements samples were mounted in the cryostat where the temperature was varied from 4.2 to 293 K. The Hall effect and magnetoresistance were measured in magnetic fields up to 6 T using a superconducting solenoid.

### Results and discussion

Temperature dependencies of resistivity for studied films are shown in figure 1. Resistivity of ZnO:Ga films as well as ZnO:Al film is a nearly temperature independent except a slight increase at helium temperatures. Such behavior of resistivity is specific for band electron transport in degenerated semiconductors and disordered metals.

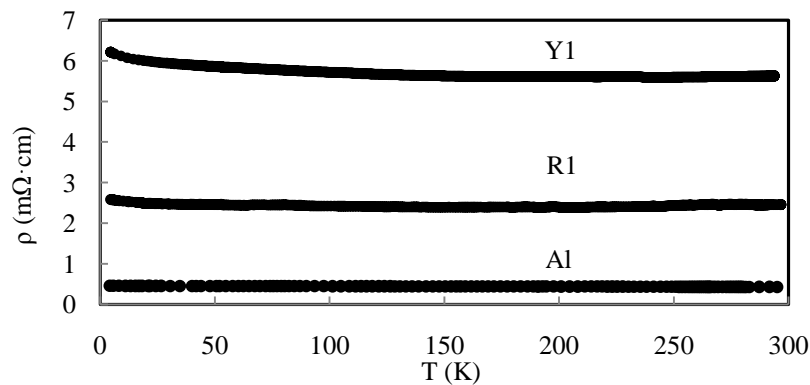
Hall resistivity of all investigated films depends linearly on magnetic field in the measured range of the magnetic fields. The Hall effect data at 4.2 K enabled us to determine concentration ( $n$ ) and mobility ( $\mu$ ) of electrons in films. The results are presented in table 2.

All films exhibit NMR. We used the values of  $n$  and  $\mu$  to verify the fulfillment of Ioffe-Regel criterion in studied films:  $k_F l \gg 1$ , where  $k_F$  is the Fermi wave vector of electrons,  $l$  is the electron mean free path. The criterion is satisfied for all investigated films therefore we can use the theory of QC for analysis of observed NMR.

<sup>b</sup> The thickness was estimated from the conditions of film deposition

We tried to approximate observed magnetoconductivity by the expressions for WL correction to the conductivity for quasi-2D, 3D electron system and by expression (1) for thick films derived in Ref. [6]

$$\Delta\sigma(B) = d \cdot \Delta\sigma_{3D}(B) + \frac{1}{2} \Delta\sigma_{2D}(B) \quad (1)$$



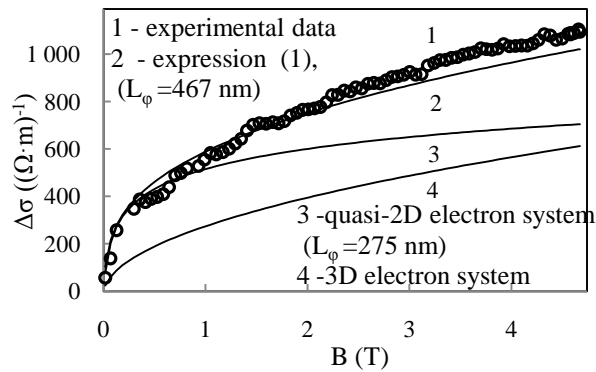
**Figure 1.** Temperature dependences of resistivity of ZnO:Ga and ZnO:Al films

**Table 2.** Calculated characteristics of investigated films at 4.2K

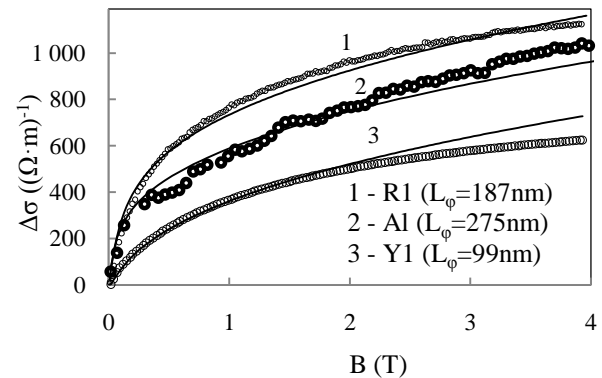
Film	$n, 10^{19} \text{ (cm}^{-3}\text{)}$	$\mu \text{ (cm}^2\text{/(V·sec))}$	$E_F \text{ (meV)}$	$l, \text{ nm}$	$k_F l$
R1	6.7	38	240	3.3	3.9
Y1	5.6	18	210	1.6	2.1
Al	140	9.8	1650	2.2	7.7

$\Delta\sigma_{3D}, \Delta\sigma_{2D}$  are WL correction to conductivity for quasi-2D, 3D electron system correspondingly. The expressions for  $\sigma_{3D}, \sigma_{2D}$  were taken from Ref. [9]. The ratio of parameter  $L_\phi$  to the least size of the sample ( $d$ ) determines the dimensionality of the system with respect to the WL theory. If  $d < \lambda < L_\phi$ , ( $\lambda$  is de Broglie wavelength), then electron system is considered to be 2D with respect to the WL theory, if  $\lambda < d < L_\phi$ , then it is quasi-2D and in case of  $d > L_\phi$  it is 3D [4-5]. The parameter  $L_\phi$  was determined by fitting of experimental MR data. The example of approximation of magnetoconductivity of the investigated film by expressions for WL correction to conductivity of quasi-2D, 3D electron system and expression for thick films (1) is shown in figure 2. The expression for quasi-2D system satisfactory approximates experimental data in low magnetic fields up to 0.7 T. Resulted value of  $L_\phi = 275 \text{ nm}$  exceeds the thickness of the film  $d = 100 \text{ nm}$  and thus satisfied to the applied quasi-2D WL theory. The calculated magnetoconductivity for 3D system goes lower than the experimental curve for all reasonable values of  $L_\phi$ . Expression (1) satisfactory fits the experimental data, however resulted value of  $L_\phi = 467 \text{ nm}$  is not consistent with applied theory, because obtained value of  $L_\phi$  exceeds the thickness of the film (100nm), while the expression (1) is valid for  $d > L_\phi$  [6].

The calculated magnetoconductivity of quasi-2D electron system deviates from the experimental dependence in high magnetic fields. The deviation of the theoretical dependence from the experimental one points to the magnetic field induced 2D-3D crossover of the dimensionality of electron system with respect to WL theory. To describe observed NMR in the whole range of magnetic fields we derived an expression for magnetoresistance which is valid for the arbitrary film thickness within diffusion limit.



**Figure 2.** Experimental data (symbols) and calculated curves (line) for Al film at 4.2K



**Figure 3.** Magnetoconductivity of films experimental data (symbols) and fitting by (5) (line)

During this procedure we followed conventional approach developed in Ref. [4]. Namely the equation (2) for Cooperon  $C(r, r, \omega)$  was solved with boundary conditions (3) :

$$(-i\omega + D(-i\vec{\nabla} - 2e/\hbar \vec{A})^2 + \tau_\varphi^{-1})C(\mathbf{r}, \mathbf{r}', \omega) = \frac{\delta(\mathbf{r}-\mathbf{r}')}{\tau} \quad (2)$$

$$(\nabla_z - 2ie/\hbar A_z)|_{z=0}C = 0 \quad (3)$$

$$(\nabla_z - 2ie/\hbar A_z)|_{z=d}C = 0$$

The expression for QC and Cooperon are related as following [4]

$$\delta\sigma(\mathbf{r}, \omega) = \frac{-2e^2 D}{\pi^2 \hbar} \tau C(\mathbf{r}, \mathbf{r}, \omega)$$

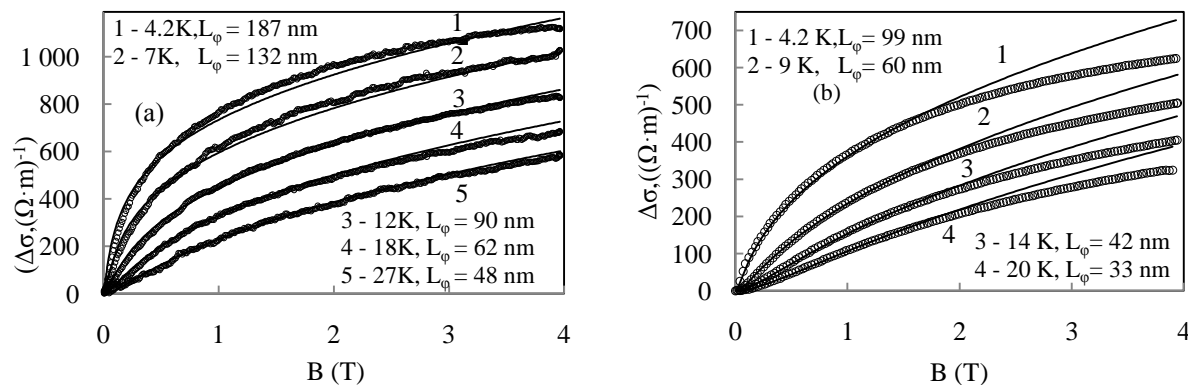
where  $\vec{A}$  is a magnetic vector potential,  $\omega$  is an external field frequency,  $D$  is an electron diffusion coefficient,  $\tau$  is electron momentum relaxation time. Using mathematical technique described in [10] we found the expression for magnetoconductivity at  $\omega=0$ :

$$\Delta\sigma(B) = \frac{e^2}{2\pi^2 \hbar} \sum_m \left[ \ln x_m + \Psi\left(\frac{1}{x_m} + \frac{1}{2}\right) \right] \quad (5)$$

$$\text{where } x_m = \frac{\omega_c}{\tau_\varphi^{-1} + D(\frac{\pi m}{d})^2}, \quad \omega_c = \frac{4eBD}{\hbar}, \quad m=0,1,\dots$$

Approximation of experimental magnetoconductivity measured at 4.2 K by formula (5) for investigated films and corresponding values of the fitting parameter  $L_\varphi$  are shown in figure 3. The expression (5) describes the experimental data better than all other expressions for magnetoconductivity related to WL. In contrast to the expression (1) the formula (5) could be applied for the analysis of films of arbitrary thickness and resulted values of  $L_\varphi$  are consistent with applied theory. We utilized expression (5) to analyze observed NMR of films R1 and Y1 at different temperatures (figure 4).

Values of  $L_\varphi$  are listed in figure 4. Formula (5) satisfactory approximates magnetoconductivity of the films at helium temperatures as well as at higher temperatures. With increase of the temperature the value of diffusion length  $L_\varphi$  decreases. This is consistent with the majority of the experimental data and theoretical predictions [3, 4].



**Figure 4.** Magnetoconductivity at different temperatures (a) – R1 film,  $d_{film}=90$  nm, (b) – Y1 film,  $d_{film}=60$  nm experimental data – scatter, approximation by (5) – solid line

The decrease of the value of  $L_\phi$  is accompanied by the change of the dimensionality of electron system and at about  $T=20$  K the electron system becomes effectively 3D in respect to the WL theory. The deviation of calculated curve from experimental data for Y1 film at high magnetic field in figure 4 (b) has systematical character and most probably associated with the electron-electron interaction.

### Summary

Temperature dependence of resistivity, magnetoresistance and Hall effect were studied for thin ZnO:Ga and ZnO:Al films. All investigated films exhibited negative magnetoresistance at low temperatures explained by quantum corrections to the conductivity. Analysis of the experimental results points to the magnetic field and temperature induced 2D-3D crossover of the dimensionality of electron system with respect to the WL theory. An expression for magnetoresistance caused by weak localization valid for arbitrary film thickness was derived. The obtained expression satisfactorily describes observed NMR. Obtained values of the electron diffusion length during the phase relaxation time of electron wave function are consistent with the theory.

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