

Determination of the Diamagnetic and Paramagnetic Impurity Magnetic Susceptibility in Ge:As near the Metal Insulator phase Transition

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Abstract. Low-temperature magnetic susceptibility of heavily doped Ge: As samples has been investigated by methods SQUID magnetometry and ESR spectroscopy near the metal-insulator phase transition. Paramagnetic component of the impurity magnetic susceptibility was investigated by ESR previously. Using both techniques make possible to determined the diamagnetic component of impurity susceptibility. The value of the impurity diamagnetic susceptibility equals to $5 \times 10^{-8} \text{ cm}^3/\text{g}$ and corresponds to the localization radius of the As donor-electron near the metal-insulator phase transition.

1. Introduction

The magnetic susceptibility is a dimensionless proportionality constant that indicates the degree of magnetization of a material in response to an applied magnetic field. The full magnetic susceptibility χ of a doped semiconductor is comprised of diamagnetic magnetic susceptibility of the lattice χ_{LD} and magnetic susceptibility defined by impurities χ_I (impurity paramagnetic susceptibility χ_{IP} and impurity diamagnetic susceptibility χ_{ID}):

$$\chi = \chi_{LD} + \chi_I = \chi_{LD} + \chi_{IP} + \chi_{ID}. \quad (1)$$

Small impurities with one unpaired electron or hole result in temperature-dependent paramagnetic contribution to magnetic susceptibility of semiconductor χ_{IP} . These electrons, moving on their orbits in impurities at low temperatures, also make their diamagnetic contribution χ_{ID} to the full magnetic susceptibility.

SQUID method (Superconducting Quantum Interference Device) is usually used to measure the magnetic susceptibility. SQUID magnetometer measures the full magnetic susceptibility where the biggest contribution is made by lattice susceptibility χ_{LD} .

The method of electron spin resonance (ESR) was used earlier for the similar measurements [1]. It enables to define only paramagnetic component of impurity susceptibility χ_{IP} . A strong influence of spin the interaction near the metal-insulator phase transition (MI transition) in Ge:As [2-4] on



paramagnetic susceptibility has been established. Electron location radius is separated closed to MI transition, what should result in increased value χ_{LD} .

Using both SQUID and ESR methods we would like to define behavior of the impurity diamagnetic susceptibility χ_{ID} and influence of spin interaction on it.

Lattice diamagnetic susceptibility of the substance is defined as follows:

$$\chi_{LD} \approx - (Ne^2/6mc^2) \sum_k^Z \langle r_k^2 \rangle = -2.83 \times 10^{10} \sum_k^Z \langle r_k^2 \rangle, \quad (2)$$

where $N = 6 \times 10^{23}$ – Avogadro constant, e – electron charge, m – electron weight, c – speed of light, $\langle r_k^2 \rangle \approx 10^{-16} \text{ cm}^2$ – averaged square of electron orbit, rotating around one of the lattice atoms. For Ge $\chi_{LD} = -8 \times 10^{-7} \text{ cm}^3/\text{g}$.

Based on experimental data [5] an empiric formula was composed for temperature dependence within range from helium to room temperatures:

$$\chi_{LD} = (-5.9 \times 10^{-7} + 8.9 \times 10^{-11} T) \text{ cm}^3/\text{g}. \quad (3)$$

The component, which is provide the temperature correction, is much less than constant part of the magnetic susceptibility. Lattice magnetic susceptibility practically does not depend on temperature.

Paramagnetic susceptibility χ_{IP} in case of no spin interaction (temperature area is above 1 K) follows Curie's law:

$$\chi_{IP} = n_s \mu_B^2 p_{ef}^2 / 3kT, \quad (4)$$

where n_s – spin concentration, μ_B – Bohr magneton, p_{ef} – effective magnetic moment on atom, k – Boltzmann constant. For samples being studied (close to metal-insulator phase transition) concentration of impurities $n_s \sim 10^{17} \text{ cm}^{-3}$, at $T = 2 \text{ K}$, $k = 1.38 \times 10^{-16} \text{ erg/K}$ and $\mu_B = 9.27 \times 10^{-21} \text{ erg/Gs}$, $p_{ef} = 1$ we shall have $\chi_{IP} \sim 10^{-8} \text{ cm}^3/\text{g}$. Adduced estimate is extreme from the top, and in case of antiferromagnetic alignment the value of paramagnetic susceptibility is reduced.

Diamagnetic susceptibility of electrons localized on impurities:

$$\chi_{ID} = - (n_s e^2 / 6m^* c^2) \langle r_B^2 \rangle, \quad (5)$$

where m^* - effective electron weight of state density, r_B – Bohr radius of electron orbit for impurities. At $n_s = 10^{17} \text{ cm}^{-3}$ and $r_B = 5 \times 10^{-7} \text{ cm}$ we shall have $\chi_{ID} \approx 7 \times 10^{-9} \text{ cm}^3/\text{g}$.

2. Samples and method used to measure magnetic susceptibility

2.1. Samples

Magnetic susceptibility was measured on a series of samples Ge:As, which had been used before in papers [2-4]. Phased reduction of electron concentration compared to original value for a series of samples $\sim 3.6 \times 10^{17} \text{ cm}^{-3}$ was performed on account of introducing compensation impurity Ga during transmutation neutron doping. Sample parameters are given in Table 1.

Table 1. Samples

Sample No.	Electron concentration $n_H, 10^{17} \text{ cm}^{-3}$	Potassium concentration $N_{As}, 10^{17} \text{ cm}^{-3}$	Compensation $K = N_{Ga} / N_{As}$
1	3.49	3.63	0.04
2	3.25	3.76	0.14
3	2.35	4.24	0.44
4	1.91	4.48	0.57

2.2. ESR method

The impurity paramagnetic susceptibility χ_{IP} was measured by ESR method. ESR spectrometer E-112 “VARIAN” with cryostat ESR-910 “OXFORD INSTRUMENTS” and digital registration system was used. This spectrometer registered derivative of resonance absorption line. Magnetic susceptibility is proportional to first integral of absorption line [3]. To define absolute value of impurity paramagnetic susceptibility χ_{IP} double integrated signal from the sample I_S was compared with the second integral from reference sample I_{st} . The spin concentration of the reference sample by “VARIAN” is equal $2.58 \times 10^{15} \text{ cm}^{-2}$, $\chi_{st} = 1.8 \times 10^{-10} \text{ emu}$ at $T = 300 \text{ K}$. Impurity paramagnetic susceptibility of the sample was defined by formula:

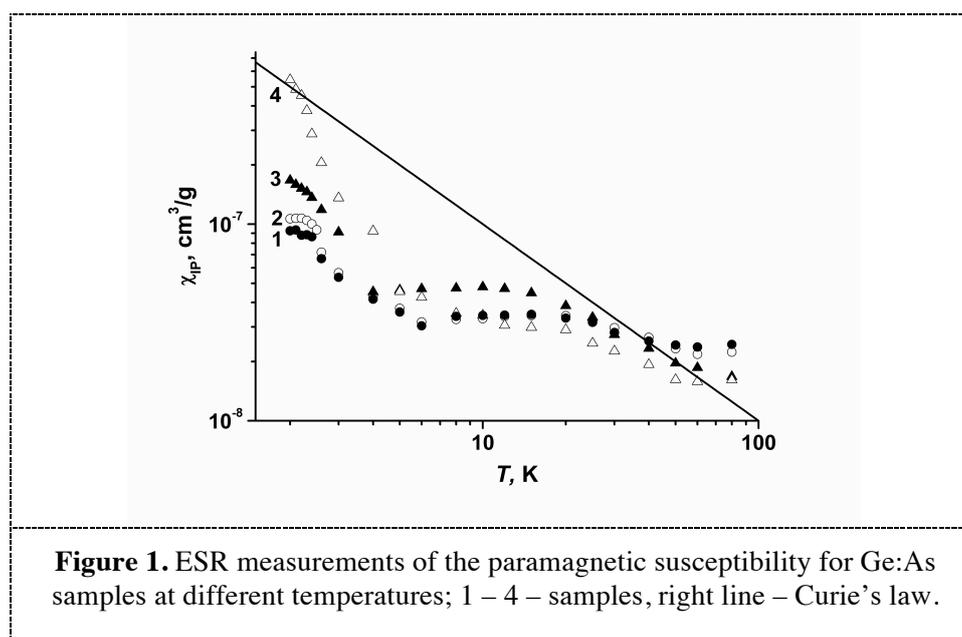
$$\chi_{IP} = 1.8 \times 10^{-10} I_S / I_{st} \quad (6)$$

2.3. SQUID method

MPMS-XL-1 device operating within temperature range 1.7 – 100 K and magnetic fields up to 10 kOe was used to measure magnetic susceptibility with the help of SQUID method.

3. Experimental results

Temperature dependences of the impurity paramagnetic susceptibility χ_{IP} obtained for these samples from ESR signal analysis are given in figure 1. It also shows $\chi_{IP} \sim 1/T$ dependence, meeting Curie’s law, for comparison.



The figure shows that all curves have similar characteristic features. At temperatures $50 \geq T \geq 20$ K $\chi_{IP}(T)$ dependence close to Curie's law is seen. At temperatures $20 \geq T \geq 6$ K $\chi_{IP}(T)$ dependence saturated. Such unusual behavior could be explained by occurrence of antiferromagnetic spin interaction.

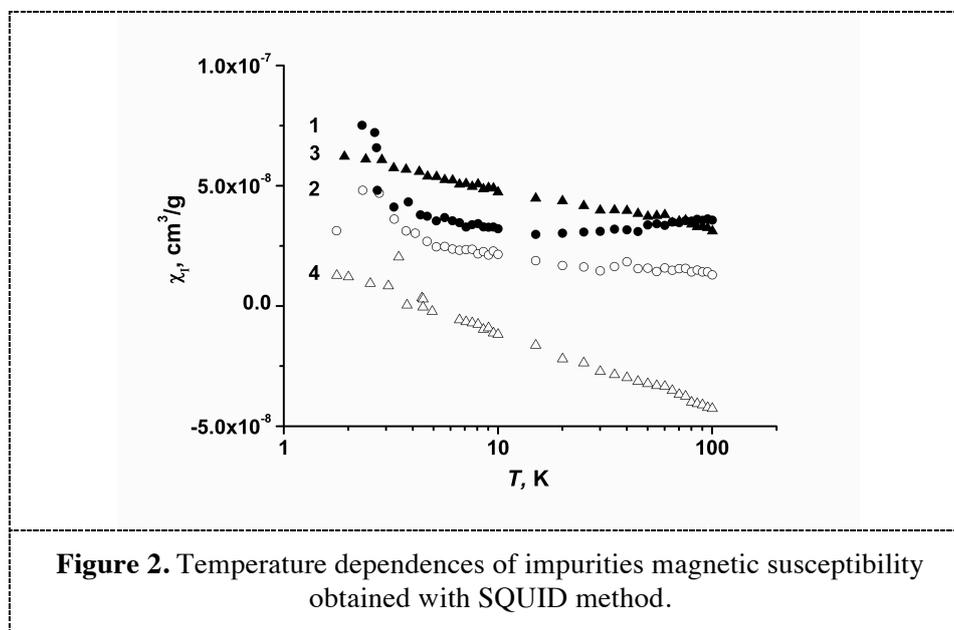
At low temperatures $T \leq T^*$, where $T^* \approx 3 \div 6$ K, impurity paramagnetic susceptibility increases sharply (stronger than the one stipulated by Curie's law).

We explain such behavior by transition from antiferromagnetic to ferromagnetic spin coupling at low temperatures [2,6].

Major contribution to SQUID is made by the lattice diamagnetic magnetic susceptibility, and to separate the impurity magnetic susceptibility χ_I from values χ measured by SQUID method, lattice susceptibility should be deducted.

$$\chi_I = \chi - \chi_{LD}. \quad (7)$$

Obtained values of the impurity magnetic susceptibility χ_I for the samples are given in figure.2.

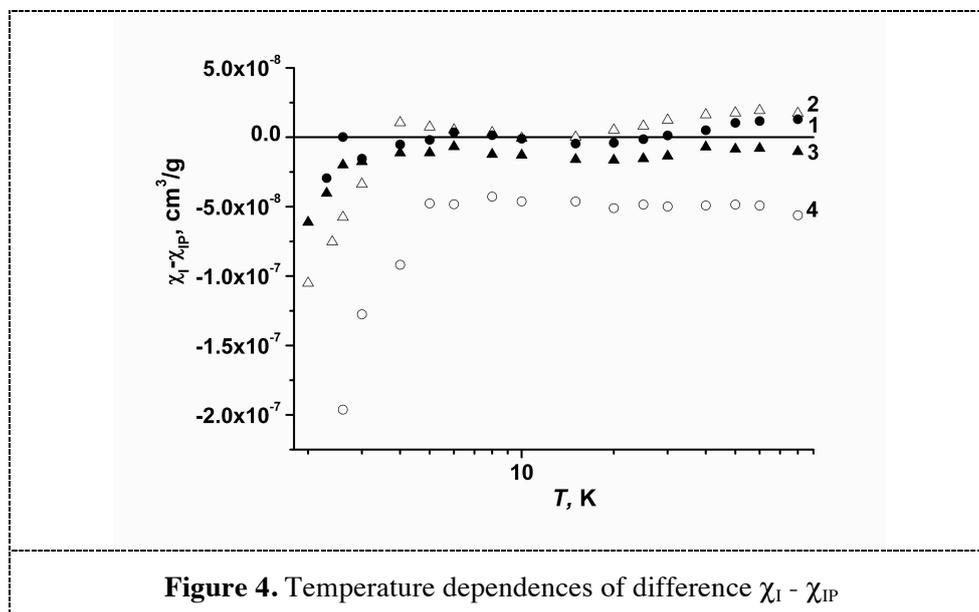
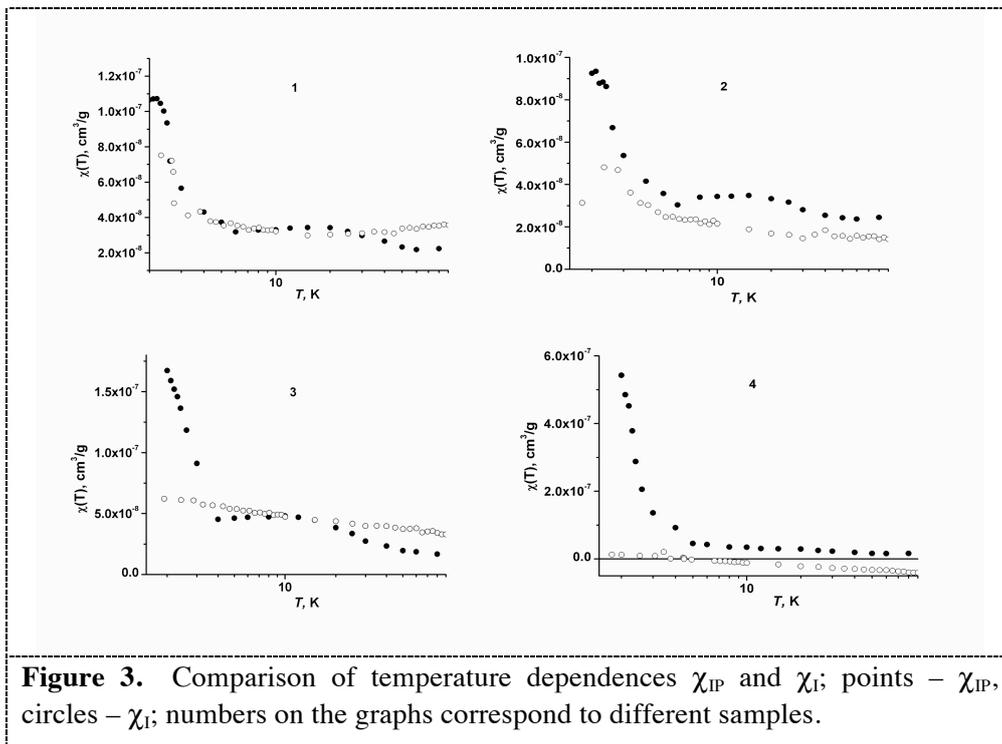


Temperature dependences χ_{IP} and χ_I for four samples are shown in figure 3. The major difference is that χ_{IP} increases at low temperatures stronger than χ_I .

Let's define χ_{ID} as follows:

$$\chi_{ID} = \chi_I - \chi_{IP}. \quad (8)$$

Figure 4 shows obtained results for the samples. It can be seen that at $T \geq T^*$, the temperature dependence of difference $\chi_I - \chi_{IP}$, is weak. But the difference $\chi_I - \chi_{IP}$ is sharply reduced at lower temperatures $T \leq (3 - 5)$ K.



4. Discussion

The value of the impurity diamagnetic susceptibility χ_{ID} should be negative at temperatures higher than T^* . It could be seen from figure 4 that for the samples 1, 2 value $\chi_I - \chi_{IP}$ is positive at $T \geq T^*$. It could be explained by random ingress of “dirt” to SQUID measuring device.

Expected low values of difference $\chi_I - \chi_{IP}$ were registered for samples 1, 2. Average value $\chi_I - \chi_{IP} = 5 \times 10^{-8} \text{ cm}^3/\text{g}$ is the sought value of the impurity diamagnetic susceptibility χ_{ID} .

Based on it and using formula for χ_{ID} and average sample value $n_s = 2 \times 10^{17} \text{ cm}^{-3}$, let's estimate radius of electron location in impurity As.

$$\langle r \rangle = (6|\chi_{ID}| m^* c^2 / n_s e^2)^{1/2} \approx 10^{-6} \text{ cm}. \quad (9)$$

This value exceeds Bohr radius of small impurity As in low-alloy Ge almost twice. Obtained result corresponds to approximation of Ge:As system to metal-insulator phase transition, which is accompanied by increased radius of location.

The measurements of the temperature dependences obtained with two methods are different at low temperature. In higher compensated samples ESR measurements show a sharp increase of $\chi_{IP}(T)$ at $T \leq T^*$. In SQUID measurements this effect is not witnessed.

We tried to explain such difference by increased radius of electron orbits at their parallel orientation, similar to the structure of hydrogen molecule. It is formed only in case of anti-parallel oriented spins. At parallel spin orientation molecule is not formed, in our case at transition from anti-parallel coupling to parallel one, electrons start moving on orbits with a great radius as a result of repulsion.

Thus, changes in the exchange interaction leads not only to a transition from the antiferromagnetic to ferromagnetic ordering in impurity spins, but also to increase the value of the diamagnetic susceptibility due to the increase of the electron orbits.

5. Conclusions

Combined application of ESR and SQUID methods enables dividing contributions of the spin (paramagnetic) susceptibility and orbit (diamagnetic) susceptibility on full magnetic susceptibility of impurities.

In the area of low temperatures transition from anti-parallel to parallel coupling of localized spins results in increased radius of electron orbit, which leads to increased impurity diamagnetic susceptibility.

References

- [1] Ch. P. Poole, Jr., *Electron Spin Resonance: A Comprehensive Treatise on Experimental Techniques* (John Wiley and Sons, New York, 1967; Mir, Moscow, 1970).
- [2] A. I. Veinger, A. G. Zabrodskii, T. V. Tisnek, S. I. Goloshchapov, P. V. Semenikhin, *AIP: Conference Proceedings*. Vol. 1610, 119, (2013)
- [3] A. I. Veinger, A. G. Zabrodskii, T. L. Makarova, T. V. Tisnek, S. I. Goloshchapov, P. V. Semenikhin, *AIP: Conference Proceedings*. Vol. 1610, 129, (2013)
- [4] A.I. Veinger, A.G. Zabrodskii, T.L.Makarova, T.V. Tisnek, S.I. Goloshchapov and P.V.Semenikhin. *JhETP*, 116, 796, (2013)
- [5] D.K. Stevens, J.W. Cleland, J.H. Crawford, H.C. Schweinler. *Phys.Rev.* 100, (1955), 1084.
- [6] H.Kamimura. In book "Crystalline Semiconducting Materials and Devices". Eds by P.N.Butcher, N.H.March M.Tosi, (Plenum, 1986).