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Influence of Free Charge Carriers on EPR Parameters of Gd^{3+} Centers in the $Pb_{1-x}Ag_xS$ and $Pb_{1-x}Cu_xS$ Semiconductors

V. A. Ulanov[1,2], **A. M. Sinicin**[2], and **R. R. Zainullin**[1,2]

[1] Zavoisky Physical-Technical Institute, Kazan 420029, Russian Federation,
ulvlad@inbox.ru
[2] Kazan State Power Engineering University, Kazan 420066, Russian Federation

PbS (galena) has the rock salt structure and belongs to family of the lead chalcogenide narrow gap semiconductors (PbS, PbTe, and PbSe). Due to unusual physical properties these materials have important applications in thermoelectric and infrared devices. To modify the physical properties of the lead chalcogenides, usually they prepare solid solutions, mixing the PbS, PbTe, and PbSe in various proportions. For such purpose the various impurity and native defects in volumes of the lead chalcogenides are created too. Prevalent native defects in PbS crystal are sulfur vacancies. Each of them provides two free electrons. So, the sulfur vacancies in galena can be considered as donors of free electrons. Doping PbS by an acceptor impurity one can get a semiconductor with hole type conductivity.

The goal of the present work was to study, using the EPR method, the effects that may be realized by doping the galena with Cu and Ag, transition group elements. As in various compounds these elements frequently are found in monovalent states, in galena they can be considered as acceptor impurities. A few Gd^{3+} paramagnetic centers were created in volumes of the $Pb_{1-x}Cu_xS$ and $Pb_{1-x}Ag_xS$ crystals to get some information about lattice deformations and characteristics of free charge carriers (electrons or holes) present in the crystals with concentration depending on quantities of Cu and Ag. The crystals under study were all grown using the Bridgeman method. Copper, silver, and gadolinium

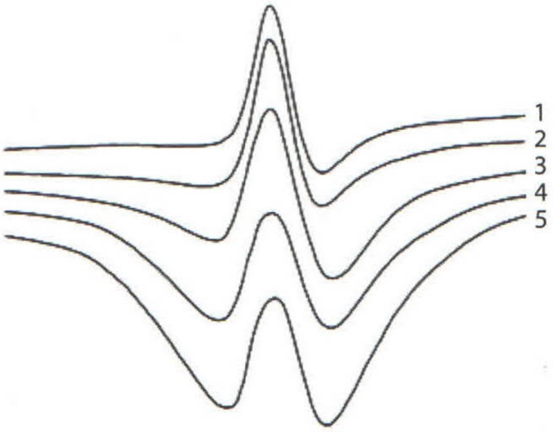

Fig. 1. Dependence of EPR line shape of the low field fine structure component of the EPR spectrum of Gd^{3+} on microwave power observed at $T = 4.2$ K ($B_0 \parallel <001>$)

were introduced into the galena in stoichiometric proportions. In the crystals thus grown free carrier concentrations were measured by Hall method using the formula $c = 1/eR$ ($c = n, p$). In these measurements it was found that type of conductivity of $Pb_{1-x}Ag_xS$ crystals was inverted from n to p at Ag concentration about $x = 0.0045$. But, no inversion of the type of conductivity was found for $Pb_{1-x}Cu_xS$ in the range $0 \leq x \leq 0.015$. As it was found, Gd^{3+} formed in the $Pb_{1-x}Cu_xS$ and $Pb_{1-x}Ag_xS$ crystals the paramagnetic centers with cubic symmetry. The X-band EPR spectra of these centers could be described by well-known spin-Hamiltonian

$$H_s = \beta_e gH \cdot S + (1/60)b_4(O_4^0 + 5O_4^4) + (1/1260)b_6(O_6^0 - 21O_6^4)\,.$$

It was found too that the g, b_4, and b_6 parameters of the spin-Hamiltonian are dependent on temperature and free carrier concentration. Comparing g-factors for PbS:Gd^{3+} and PbTe:Gd^{3+} one could find that g-factor of Gd^{3+} was not sensitive to lattice distortions induced by Ag and Cu impurities, while sensitivity of the crystal field parameters b_4 and b_6 to distortions could be observable. In the PbS:Gd^{+} crystal the parameters determined at 4.2 K were following: $g = 1.9901 \pm 0.0002$; $b_4 = 59.37 \pm 0.05$; $b_6 = -0.18 \pm 0.05$ (b_4 and b_6 in MHz). For $T = 77$ K these parameters were: $g = 1.9922 \pm 0.0002$; $b_4 = 57.92 \pm 0.05$; $b_6 = -0.32 \pm 0.05$. Results of g-factor dependences on the concentration and type of conductivity of the $Pb_{1-x}Cu_xS$ and $Pb_{1-x}Ag_xS$ samples studied in this work show that these ones are qualitatively the same as for Mn^{2+} centers in PbTe [1], but much less pronounced. Main result of the present work is an unusual dependence of the EPR line shapes of Gd^{3+} centers on microwave power P_r acting in the resonator of the spectrometer (Fig. 1). In this figure the fragment 1 corresponds to $P_r = 0.02$ mW, and fragment 5 to $P_r = 25$ mW. The same result was observed for other six EPR lines at $T = 4.2$ K.

From Fig. 1 one can see that at low microwave powers the shapes of the EPR lines have a Dysonian form, but, at higher powers these lines acquire a new form which cannot be described by Dyson's theory.

1. Story T., Swuste C.H.W., Egenkamp P.J.T, Swagten H.J.M., de Jonge W.J.M.: Phys. Rev. Lett. **77**, 2802 (1996)

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АО “Информационно-издательский центр” · Казань, ул. Чехова 28, телефон +7 (843) 236 94 26