Regularities of elastic anisotropic strains caused by *T-H-P* influence on the structural transition and properties of magnetic semiconductors

P.I.Polyakov, S.S.Kucherenko*

Institute for Physics of Mining Processes, National Academy of Sciences of Ukraine, 72 R.Luxemburg St., 83114 Donetsk, Ukraine
*A.Galkin Donetsk Institute for Physics and Engineering, National Academy of Sciences of Ukraine, 72 R.Luxemburg St., 83114 Donetsk, Ukraine

A generalizing analysis of experimental results on resistivity, magnetostriction, and phase transitions has been performed for magnetic semiconductors, namely, $La_{0.7}Ca_{0.3}MnO_3$ polycrystals and $LaMnO_3$ single crystals exposed to temperature (T), magnetic field (H), and hydrostatic pressure (P). The magneto-,baro-, and baromagnetoresistive effects have been revealed, where their maxima temperatures, T_{PP}, have been found to be constant and to coincide with the metal-semiconductor structural phase transition (PT) temperature, T_{ms}. The "cooling" and "heating" effects of the magnetic field and pressure have been established, thus enabling to validate the regularities of $T_{ms}(H)$, $T_{ms}(P)$, and $H_g(T)$ variations. The correspondence between *T-H-P* effect (5.1 K ~ 2.42 kOe ~ 1 kbar) on the resistivity properties and *T-H* effect (5.2 K-2.5 kOe) on the magnetostriction properties of the magnetic semiconductors has been estimated. The sign alternation has been revealed in variations of properties and effects as well as regularities of competing influence of thermo-, baro-, and magneto-elastic anisotropically straining (EAS) stresses. The positions of critical lines $T_{ms}(H)$, $T_{ms}(P)$, $H_g(T)$ and points T_X, P_X, PP_X, P_X', $T_{PP} = T_{ms}$, T_C have been defined, their correspondences in resistivity and magnetostriction behaviors has been established. Basing on the variety of critical lines and points, the correspondence and sign alternation of *T-H-P* influence through the mechanism of EAS stresses have been substantiated.

Проведен обобщающий анализ экспериментальных результатов изменений резистивности, магнитострикции, фазовых переходов магнитных полупроводников: поликристаллического $La_{0.7}Ca_{0.3}MnO_3$ и монокристаллического $LaMnO_3$ под влиянием температуры (T), магнитного поля (H) и гидростатического давления (P). Выявлены магнито-, баро- и баромагниторезистивный эффекты, в которых определено постоянство температуры T_{PP} их максимумов, совпадающей с температурой T_{ms} структурного фазового перехода "металл-полупроводник". Установлены "охлаждающий" и "нагревающий" эффекты магнитного поля и давления, позволяющие обосновать закономерности изменения $T_{ms}(H)$, $T_{ms}(P)$ и $H_g(T)$. Получена оценка соответствий влияния *T-H-P* (5,1 K ~ 2,42 kOe ~ 1 kbar) на резистивные свойства, и влияния *T-H* (5,2 K-2,5 kOe) на магнитострикционные свойства магнитных полупроводников. Выявлена знакопеременность в изменениях свойств, эффектов и закономерности конкурирующего влияния термо-, баро- и магнитоупругих анизотропно деформирующих (УАД) напряжений. Определены положения критических линий $T_{ms}(H)$, $T_{ms}(P)$, $H_g(T)$) и точек T_X, P_X, PP_X, P_X', $T_{PP} = T_{ms}$, T_C. Установлены их соответствия на зависимостях изменений удельного сопротивления и магнитострикции. Из многообразия выделенных критических линий и точек установлено и дано обоснование значения оценок соответствий и знакопеременности влияния *T-H-P* через механизмы УАД напряжений.

A variety of interrelated properties and physical processes, correlation between electrical conductivity and magnetism in manganites are among interesting phenomena in solid state physics. Numerous works [1, 2] and reviews [3, 4] are dedicated thereto, but the interrelation between the magnetic, transport, and structure properties is not yet elucidated completely to date. Recently, a trend to study the resistivity under simultaneous influence of temperature (*T*), magnetic field (*H*) elevated hydrostatic and quasi-hydrostatic pressure is observed. Some significant results have been obtained, new effects and regularities have been established [5–12]. A generalizing analysis of changes in the phase transitions (PT), properties, revealed critical lines, points and regularities of their positions under *T-P-H* influence for $La_{0.7}Ca_{0.3}MnO_3$ polycrystals [8] and $LaMnO_3$ single crystals [13] is the main aim of this work.

We would like to note the consistency in study results obtained for polycrystals and films of magnetic semiconductors [9–12]. So, in [9], the baro- and baromagnetoresistive effects were first revealed, a correspondence of *T-H-P* influence on the polycrystalline sample resistivity was established, the role of the mechanism of EAS stresses was defined. The "cooling" and "heating" effects of pressure and magnetic field influence, their regularities in the change of $T_{ms}(P)$ and $T_{ms}(H)$ critical lines has been established [10]. A significant feature of T_{PP} temperature for magneto-, baro- and baromagnetoresistive peaks is its constancy and coincidence with phase transition temperature T_{ms}. Basing on the data concerning the phase transitions, resistivity, and magnetostriction behavior under *T-H-P* influence [11], a concept of sign alternation in properties and effects was introduced and substantiated. The correspondence of critical lines $H_g(T)$ and $T_{ms}(H)$ in magnetostriction and resistivity dependences was established, critical points P_X and T_X were defined. An advanced examination technique at high hydrostatic pressure, features of pressure sensors for 0 to 25 kbar range on the basis of magnetic semiconductor films was described in [12].

The analysis of previous studies [9–11] has shown a rather close correspondence of estimations for temperature, magnetic field, and pressure influence on the resistivity of polycrystalline manganite samples such as $La_{0.7}Mn_{1.3}O_3$ (8 K ~ 2 kOe ~ 1 kbar); $La_{0.9}Mn_{1.1}O_3$ (6.2 K ~ 2.7 kOe ~ 1 kbar); $La_{0.56}Ca_{0.24}Mn_{1.2}O_3$ (6.2 K ~ 2.37 kOe ~ 1 kbar); $La_{0.7}Ca_{0.3}MnO_3$ (5.1 K ~ 2.42 kOe ~ 1 kbar). This confirms the universality of our analysis method for bulk polycrystalline samples. As to dependences for magneto-, baro-, baromagnetoresistive effects (Fig. 1), it is to note the critical role of the thermoelastic expansion within a wide temperature range. The revealed critical lines $T_{ms}(H)$, $T_{ms}(P)$ (see Fig. 1a) are regularities of the phase transition displacement under magneto- and baro-induced EAS stresses, that differ by variance of elastic and magnetoelastic anisotropies. For $La_{0.7}Ca_{0.3}MnO_3$ sample, estimations show that the magnetic field intensity 2.42 kOe shifts the T_{ms} temperature by the same value as the applied hydrostatic pressure 1 kbar.

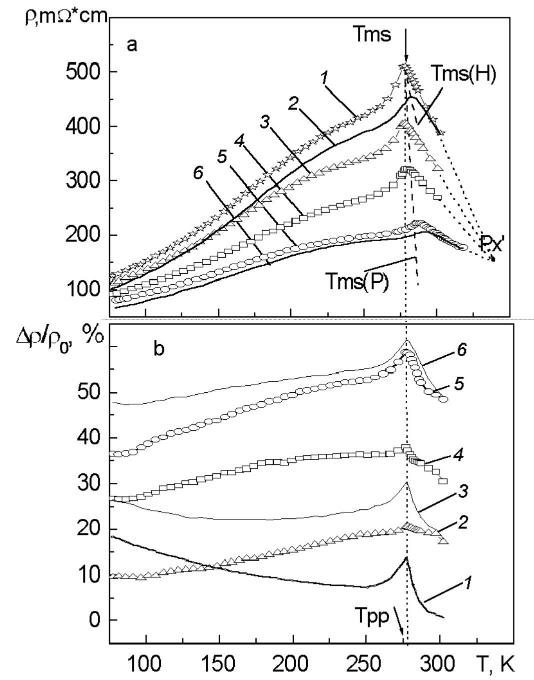

Fig.1. Temperature dependences of resistivity (a) and magneto-, baro-, baromagnetoresistive effects (b) of polycrystalline $La_{0.7}Ca_{0.3}O_3$ sample at magnetic field and hydrostatic pressure influence:(a) *1* – P=0, H=0; *2* – P=0, H=8 kOe; *3* – P=6 kbar, H=0; *4* – P=12 kbar, H=0; *5* – P=18 kbar, H=0; *6* – P=17 kbar, H=8 kOe; (b) *1* – P=18 kbar, H=8 kOe; *2* – P=18 kbar, H=0; *3* – P=12 kbar, H=0; *4* – P=6 kbar, H=8 kOe; *5* – P=6 kbar, H=0; *6* – P=0, H=8 kOe.

The peaks of baro-, magneto-, baromagnetoresistive effects (Fig. 1b) exhibit the same maximum temperature, T_{PP}, coincident with temperature of metal-semiconductor phase transition, T_{ms}, as it was observed in [10, 11]. It means that the main cause of the structural phase transitions is the thermo-induced EAS stress mechanism, while magneto- and baro-EAS stresses are the reasons for the "cooling" and "heating" effects. The revealed correspondence estimations [10, 11] of thermo-, baro-EAS stresses (8 K, 1 kbar) emphasize the role of the thermoelastic straining stress mechanism as the main reason of resistivity behavior formation under different magnetic fields and pressures. Having approximated resistivity behaviors at different P and H, we would mark the intersection point thereof as the critical point P_X' (Fig. 1a). The same construction is also used in [5–7].

Having noticed regularities of EAS stresses in resistivity properties, it is important to define their role in magnetic properties, that was made in part in [11]. Correspondence estimations of H and T influence (5.1 K, 2.5 kOe) on magnetostriction was shown there and correlation between $T_{ms}(H)$ and $H_g(T)$ was defined (Fig. 2), a critical line $H_g(T)$ and points $T_{ms} = T_{PP}$, T_C, P_X, T_X were revealed. These results are physically substantiated and allow to carry out the generalizing analysis, concerning a variety of magnetic semiconductors.

We would pay attention to the field dependences of $LaMnO_3$ magnetostriction (Fig. 2) which are the most informative in our opinion. The observed features in the non-linear range of magnetostriction behavior are caused by T and H competitive influence and can be explained through the EAS mechanism by introducing the sign alternation conception. A non-linear region of magnetostriction field is observed at low magnetic field and low fixed temperatures (Fig. 2). This is a result of competitive effect of fixed thermoelastic expansion and magnetoelastic strictions. As H increases, the priority passes to magnetoelastic striction and the linear dependence appears. At higher temperatures, the thermal EAS expansion has a priority comparatively to the magneto-EAS striction. A drastic increase of magnetostriction is observed, magnetic hysteresis and sign alternation are formed in magnetic properties.The sign alternation of T and H influence manifests itself as a variation of thermo- and magnetoelastic anisotropy affecting $H_g(T)$ hysteresis in the phase transition region. These anisotropies become equal to each other at the Curie temperature T_C. At competitive correspondence of simultaneous H and T influence, non-linear $H_g(T)$ dependence appears. A

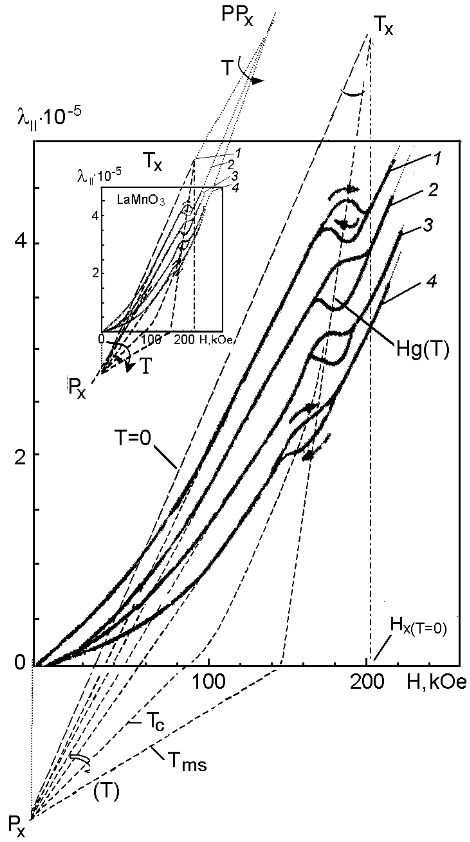

Fig.2. Field dependence of longitudinal magnetostriction of $LaMnO_3$ monocrystal [13]: *1*— 29 K, *2* — 77 K, *3* — 102 K, *4* — 130 K.

change of thermo- and magnetoelastic priority occurs there. The T_{PP} temperature remains constant.

As a whole, the study of thermo- and magnetoelastic influence reveals a strong correlation of magnetic and crystalline structures and the defining role of the EAS stress mechanism. The thermo- and magnetoelastic anisotropies cause the regularities of numerous anomalies taking place in magnetic semiconductors.

The experimental study of changes in properties can point to the existence of both critical points and lines. The ascertaining of the type, existence conditions, and characteristics helps in judging on the features of magnetization, magnetic susceptibility, heat capacity, resonance-frequency behavior, but it also can be a subject of independent investigation.

To date, there is no consistent analysis of critical phenomena being realized in the critical lines and points. Analytical study methods of the changes in parameters and properties, being the indications of critical points both in physically accessible and inaccessible regions form a missing link in the chain of scientific searches. The obvious signs of sufficient generality in the obtained results allow to fill up this deficiency and to carry out a comprehensive analysis in order to show true characteristics of thermodynamic and magnetic values in the general problem of studying of mechanisms implementing physical processes immediately at investigation of a diversity of critical lines and points. We have selected and shown methods of deriving the following critical lines and points in the phase transitions and resistivity behavior taking the dependences of resistivity change in magnetic semiconductors (Fig. 1a):

$T_{ms}(H)$, $T_{ms}(P)$ are dependences of the phase transition temperature on H and P, the difference thereof being defined by corresponding regularities of elastic and magnetoelastic anisotropy.

P_X' is the intersection point of resistivity versus temperature dependence under the influence of P and H. Note that the critical point P_X' is a starting point of counter-clockwise changes in properties under the influence of H and P.

$T_{ms} = T_{PP}$ is the phase transition temperature that coincides with the peaks of baro-, magneto- and baromagnetoresistive effects (Fig. 1a, b).

It should be noted that all the critical points are related to chemical composition of the compounds, the processing technology, and the symmetry features of structure.

The next result (Fig. 2) concerns the magnetostriction change under the influence of magnetic field and temperature:

$H_g(T)$ is the critical line in hysteresis field dependence corresponding to $T_{ms}(H)$.

P_X, PP_X are the intersection points for the approximated linear segments of field dependences of the magnetostriction before and after the phase transition; this is an interaction regularity between the changes in properties and the structure of the sample.

T_X is the intersection point for the dependence of phase transition $H_g(T)$ and the dependence of magnetostriction at $T = 0$ K.

T_C is the Curie temperature corresponding to $H_g(T)$ in the magnetostriction curve, that holds the structural phase transition fixed at the equality of thermo- and magnetoelastic anisotropy.

The analysis of the described critical lines and points in the systems studied allows to establish the role of the EAS stress mechanism. Let comparative investigations of obtained results in magnetic semiconductors be carrier out. First, let an attention be paid to the resistivity change (Fig. 1a, curve 1). The temperature variation is concerned with the priority of thermal EAS stresses, where the elastic anisotropy implements conductivity jump at the moment of the structural metal-semiconductor phase transition at the T_{ms} temperature and magnetic intensity $H = 0$. The influence of magnetic field manifests itself in properties and $T_{ms}(H)$ dependence via the "cooling" effect.

Let the magnetostriction change under the "heating" effect of magnetic field be considered (Fig. 2) where the priority of magneto-EAS stresses implements properties of magnetoelastic anisotropy at $T = 0$ in the critical point T_X. The subsequent influence of the thermal EAS stress competitive mechanism is displayed in shift of a hysteresis field in $H_g(T)$ dependence that is the regularity of difference in thermo-and magnetoelastic anisotropy.

Then, it is to consider the case of simultaneous H and T influence (Fig. 1a, Fig. 2) and note the linear and non-linear dependences $T_{ms}(H)$, $H_g(T)$. Keeping in mind the role of the EAS stress mechanism and correspondence estimations of T and H, it can be stated that the change of critical lines is the regularity of priority changes in T and H influence on phase transitions and properties. The influence of EAS stress mechanism is the sign-alternating one. It is important that the position of T_C is a consequence of the same sign alternation that results in the equality of elastic and magnetoelastic anisotropies. The investigation carried out allows to conclude that the T_{PP} constancy reveals the role of EAS stress in the nature of the colossal magnetoresistive effect. This means that the position of critical points T_X, $H_X(T = 0)$, P_X, PP_X and P_X' is a regularity defined by lattice features of the structure and its symmetry, but T-H-P influence brings to mechanism of thermo-, baro- and magnetoEAS stresses in properties, phase transitions, correspondences, sign alternation, effects, and anomalies.

The studies carried out using the analysis of experimental results allowed to establish the significant role of EAS stresses in formation and changing of the phase transitions and properties, and also to associate this mechanism with the composition and structure symmetry features of the materials.

Thus, taking into account the nature of T-H-P influence in magnetic semiconductors via the of EAS stress mechanism, the following results have been obtained. According to the resistivity and magnetostriction study, an approximate equivalence and conformity of *T-H-P* effect on resistive properties of polycrystalline manganites $La_{0.7}Mn_{1.3}O_3$ (8 K ~ 2.1 kOe ~ 1 kbar), and $La_{0.9}Mn_{1.1}O_3$ (6.2 K ~ 2.7 kOe ~ 1 kbar), $La_{0.56}Ca_{0.24}Mn_{1.2}O_3$ (6.2 K ~ 2.37 kOe ~ 1 kbar), presented in [9–11] has been established. The regularities of "cooling", "heating" effects, and the same temperature T_{PP} of baro-, magneto- and baromagnetoresistive peaks coincides with "metal-semiconductor" phase transition temperature T_{ms} have been observed. A regularity in $T_{ms}(H)$ and $T_{ms}(P)$ shifting has been defined, where magnetic field H = 2.42 kOe displaces the phase transition temperature by the same value as the pressure 1 kbar.

In $LaMnO_3$ single crystal, a conformity exists between the magnetic intensity 2.5 kOe and temperature of 5.2 K, that equally influence its magnetostriction. A regularity in change of the hysteresis parameter $H_g(T)$ under T and H influence in the phase transition region and the role of elastic and magnetoelastic anisotropies equality at Curie temperature T_C has been established. Correlations between $H_g(T)$ and $T_{ms}(H)$ have been found, the "heating" and "cooling" effects of magnetic field have been explained. The sign alternation in competitive influence of thermo- and magneto- EAS strictions has been established and explained. Correlation of the critical points T_X, P_X, PP_X, T_C, $H_X(T = 0)$ and lines $T_{ms}(H)$, $T_{ms}(P)$, $H_g(T)$ with the structure, resistive, magnetic properties and phase transitions via the of EAS stresses mechanisms of *T-H-P* influence has been established.

It can be stated without exaggeration that such a treatment of the experimental results shows the necessity and relevance of the counting for regularities of thermo-, magneto- EAS stresses forming and changing the properties and phase states. This allows to predict a participation of the same mechanisms in the long-studied effects of colossal magnetoresistance (CMR) and conductance in HTSC structures. To that end, it is necessary to carry out an analogous analysis for magnetic dielectrics properties that partially has been made in [14].

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Закономірності пружних анізотропних деформацій, обумовлених впливом *T-H-P* на структурний фазовий перехід та властивості магнітних напівпровідників

П.І.Поляков, С.С.Кучеренко

Виконано узагальнюючий аналіз експериментальних результатів змін резистивності, магнітострикції, фазових переходів магнітних напівпровідників: полікристалічного $La_{0.7}Ca_{0.3}MnO_3$ та монокристалічного $LaMnO_3$ під впливом температури (T), магнітного поля (H) та гідростатичного тиску (P). Виявлено магніто-, баро- та баромагніторезистивний ефекти, для яких визначено постійність температури T_{PP} їх максимумів, яка співпадає з температурою T_{ms} структурного фазового переходу "метал-напівпровідник". Виявлено "охолоджувальний" та "нагрівальний" ефекти магнітного поля та тиску, які уможливлюють обгрунтування закономірності змін $T_{ms}(H)$, $T_{ms}(P)$ та $H_g(T)$. Отримано оцінку відповідностей впливу T-H-P (5,1 K-2,42 kOe ~ 1 kbar) на резистивні властивості та впливу T-H (5,2 K-2,5 kOe) на магнітострикційні властивості магнітних напівпровідників. Виявлено знакозмінність у змінах властивостей, ефектів та закономірності конкурентного впливу термо-, баро- та магнітопружних анізотропно деформуючих (ПАД) напруг. Визначено положення критичних ліній $T_{ms}(H)$, $T_{ms}(P)$, $H_g(T)$ та точок T_X, P_X, PP_X, P_X', $T_{PP} = T_{ms}$, T_C. Встановлено їх відповідності на залежностях змін питомого опору та магнітострикції. З різноманіття виділених критичних ліній та точок встановлено та обгрунтовано значення оцінок відповідностей та знакозмінності впливу T-H-P через механізми ПАД напруг.