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ELECTRONIC PROCESSES IN ACOUSTORESISTIVE SENSORS

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Abstract

The analysis of electronic processes in acoustoresistive sensors is presented in the paper. The change of electrical resistance under the action of the high frequency acoustic wave (AW) is called an acoustoresistive effect (ARE). In difference of the acoustoelectric effect the phenomena of ARE is not galvanic. The ARE is observed in semiconductors: Ge, Si, CdS, CdSe etc. and in layered structures: piezoelectric and semiconductor. The analysis of experimental data is based on the crystal and its energetic levels modulation; the reason of this phenomena is piezoeffect and deformation potential. In the paper the main attention is given to piezosemiconductors and to the layered piezoelectric-semiconductor structures. Some basic mechanisms of acoustoresistive effect are presented. The acoustoresistive effect in the high photosensitive semiconductors is accounted. In these materials the ARE is determined(caused) by modulation of the trapping and recombination levels in the band gap. The model with one recombination level was discussed in works of Y. V. Gulyaev et theoretically. and negative ARE is provided. The ARE is caused by the charge carriers ejection induced by AW. The strong positive ARE, as provided in model with two recombination levels, is observed experimentally. In this case the ARE is caused by the acoustic damping of photoconductivity. The strongest ARE is observed in layered structures with thin films where the resistance of film under the action of AW changes twice. In the layered structures the reason of the ARE is the change of (1) intercrystalline barriers and (2) surface electrokinetic process conditions by the propagating AW. Both the positive and the negative ARE is observed. The new type sensors on the base of acoustoresistive effect are created.

Key words: gas sensors, acoustoelectric interaction, acoustoresistive effect, the modulation of energy levels.

Аннотация

ЭЛЕКТРОННЫЕ ПРОЦЕССЫ В СЕНСОРАХ НА ОСНОВЕ АКУСТОРЕЗИСТИВНОГО ЭФФЕКТА

Р. Гирюнене, Эв.-Л. Гаршка

В последнее время созданы ряд сенсоров, особенно газовых, с использованием акусторезистивного эффекта (АРЭ). В статье обсуждаются различные физические механизмы, приводящие к изменению электрического сопротивления вещества в присутствии высокочастотной акустоэлектрической волны. В более популярных сенсорах данное явление используется через появление вторичных эффектов. Рассмотрено проявление АРЭ в

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объеме и на поверхности вещества. В полупроводниковых кристаллах при модуляции электронных уровней волной возникает освобождение, “выталкивание” носителей тока из уровней захвата. При более сложной модели электронных уровней может проявляться задержка носителей на этих уровнях. Отмечается, что значительное изменение сопротивления возникает в сенсибилизированных кристаллах фотопроводников вследствие акустического гашения фотопроводимости. В некоторой группе кристаллов с “одним” уровнем возникает уменьшение сопротивления кристалла-отрицательный АРЭ; во второй группе — с “двумя” уровнями — возникает значительная положительная акусторезистивность.

В настоящее время больший интерес представляют сенсоры с применением явлений в тонких слоях или на поверхности. Картина акусторезистивности здесь более сложная. Реализовать АРЭ на основе выше рассмотренных явлений взаимодействия через уровни захвата трудно из-за очень развитой системы уровней. Выяснено, что в поликристаллических тонких слоях превалирует АРЭ вследствие нелинейного возмущения барьерной электропроводности при акустической модуляции высоты барьеров. Нелинейность вольтамперной характеристики приводит к постоянной добавке к концентрации туннелирующих носителей, определяемой глубиной модуляции, т.е. интенсивностью волны. В такой модели проявляется отрицательная акусторезистивность.

Другая модель АРЭ на поверхности основана нарушением адсорбционного равновесия с окружающей газовой средой при модулированном поверхностном потенциале. Исследования показали, что для изготовления сенсоров весьма подходящими являются слои касситерита (SnO_2), в которых возможна реализация как положительной, так и отрицательной акусторезистивности; данное обстоятельство определяется созданием поверхности или донорного, или акцепторного характера при адсорбции газа различного химического состава.

Ключевые слова: сенсоры газов, акусторезистивный эффект, модуляция электронных уровней.

Introduction

Recently are created a lot of sensors, especially gas sensors, which based on acoustoresistive effect (ARE) [1,2] and various phototransducers. The acoustoresistive effect (ARE), discovered nearly thirty years ago, it has revived in sensors and it is still discovered in new materials such as gallium nitride [3]. The purpose of this paper is to analyze the basic mechanisms of the ARE and discourse its experimental investigations.

Grouping of the carriers into the bunches of inhomogeneous concentration propagating with a given velocity in the direction of the wave implies perturbations of processes of carrier capture by impurity levels, generation (both majority and minority charge curriers) and recombination. The capture in the acoustoelectric interaction is accounted for by introducing the capture factor f, which appears to be nonlinear depending not only on characteristic relaxation times, but also on the wave intensity.

Considering acoustoelectric interaction in electrically inhomogeneous medium with potential barriers, one more variety of the medium resistance change owing to perturbation of barriers by a field of a wave is observed. It is especially precisely exhibited in layered structures: piezoelectric and, for example, polycrystalline semiconducting layer.

The existence of acoustoelectric interaction in some crystals, especially in piezosemiconductors,

and in various layered structures creates premises by their strong acoustosensitivity. The acoustosensitivity is similar to the photosensitivity, however it is stipulated by completely other reasons. The acoustoresistive effect alongside with piezoresistive and strain sensitivity resistive effects envelops the whole class of related phenomenon: the ARE is possible to refer to a variety of the mentioned phenomena at high frequencies. The theory of the resistance variation under the action of AW is developed in works of Y. V. Gulyaev et. al.[4,5]. The various mechanisms of a possible conductivity variation (They it called as effect of acoustoconductivity[6,7]) are considered, and it is shown, that the greatest value ΔR/R can be reached in the semiconductor with traps, where it can achieve several per cent owing to "eject out"(force out) of carriers from impurity centers at grouping.

The acoustoelectric interaction at not so high frequencies is precisely described by the phenomenal approach not attracting a quantum nature of an acoustic wave. However, underlining the initial reason of the ARE as the interaction of a simple harmonic wave (stream of phonons) with others elementary excitation of the medium including charge carriers, proceeding from a generality of exposition of all frequency band (ultrasound, hypersound), was called an electron — phonon ARE.

From the beginning the problem of a possible change of electrical conductivity of the crystal was interesting for many of investigators. The researches were subdivided into investigation of customary multivalley semiconductors, in basic germanium and silicon, in which AEI is called by a deformation potential, and piezosemiconductors , in basic cadmium sulphide, in which AEI is stipulated by piezoactivity of the medium.

For the sake of a historical validity it is necessary to specify, that the first who have tried theoretically to describe the change of crystal resistance there were authors of the work [8], where the change of resistance is interpreted as influence of a deformation potential, and as piezoeffect; the expressions of a relative modification of resistance $\Delta R/R$ at piezoelectric interaction $(\Delta R/R)_{\varphi}$, at the deformation potential $(\Delta R/R)_{\chi}$, and also owing to the variation of concentration of charge carriers $(\Delta R/R)_{n}$, and also variation of their mobility $(\Delta R/R)_{\mu}$ are presented . Two reasons, under our judgment, are enveloped by essence of an appearance and consequently there is not absolutely clear a problem on the contribution of first two reasons. Here we shall not develop the further controversy in this direction, we shall not present obtained expressions owing to them complication.

It would be desirable to mention work [9], in which acoustic wave propagates along the piezoactive axes of crystal and external electric field E_1 is applied at the same direction the component of electric current density $I=I_0+I_{ae}+I_{\Delta\sigma}$., taking place in this direction are considered, here I_0 — ohmic current density, I_{ae} -acoustoelectric current density, $I_{\Delta\sigma}$ — current density owing to a modification of electrical conductivity of a medium because of AEI.

Our experimental research completely confirmed analytical results [10]. However, it is necessary to direct attention that such ARE is revealed in most perfect crystals from the point of acoustoelectric interaction, that is the electronic absorption factor completely corresponds to its theoretical calculated magnitude.

In consideration of acoustoelectric current the strong ARE in some CdS crystals we observed [11]. The strong ARE links with a high photosensitivity of the crystals. These results are explained by acoustic quenching of photoconductivity caused by modulation of electronic levels and by change of their role during the period of AW.

1. Acoustic quenching of photoconductivity

As AW propagates in crystal the modulation of energy band gap, bottom of conductivity band and ceiling of valence band occurs. In the case of crystal with strong piezoelectric properties the influence of piezoeffect predominates over a deformation potential, and the modulation of edges of a band gap

occurs with an identical phase, and the depth of modulation ΔW is determined by a strain and operating piezoconstant ε_{ik}, then $\Delta W = qe_{ik}u_0 / \varepsilon_{ik}\varepsilon_0$, q is elementary charge, ε_0 is dielectric constant.

At AW intensity of the order W / cm^2 the depth of modulation, for example, for crystals of cadmium sulphide can reach the order an electronvolt. Where as the displacement is the function of the coordinate and time, it causes a heterogeneity of energy state of crystal. At the presence of electron gas its thermodynamic equilibrium is breaked, the areas with the increased and reduced pressure of electron gas are created. At screening by free carriers of interior fields and their grouping the modulation of quasi-Fermi level occurs, and its non-equilibrium part can be shown as a charge density wave. Then a modulated quasi-Fermi level is

$$W_{F'} = W_F + \Delta W_F = W_F + \Delta W_{F_0} \exp\ i(kx - \omega\ t),$$

here W_F is the Fermi energy. The value ΔW_{F_0} is determined by the non-equilibrium concentration, which is equal to the local concentration of grouped carriers.

Thus in a semiconductor with impurity levels (by analogy to sensitization of a photoconductor by light) "the acoustic" sensitization is possible. Naturally, acoustic sensitization is the peculiar harmonic (in linear case) spatial-temporal inhomogeneity. On the base of the acoustic sensitization it is possible to explain a series of developments of acoustoresistive effect in bulk and on a surface. It can be illustrated on the base of photosensitive crystal with a developed system of impurity levels in a forbidden band gap. Let here are available both small-sized (traps), and deep (recombination) levels. As AW propagates in the crystal and grouping of the curriers occurs, the demarcation level W_D , defining character of levels, are modulated too. The depth of its modulation corresponds to the modulation depth of $W_{F'}$, and the phase is turned (biased) on an integer π. If to suppose, that in conditions of acoustic modulation the previous capture cross-sections for electrons S_n and holes S_p are saved, then the demarcation level W_D is mirror map W_F concerning a middle of zone W_i (fig. 1).

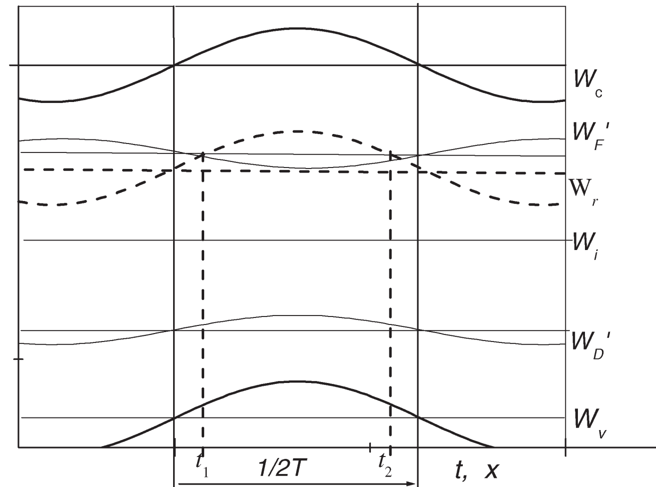

Fig. 1. Modulation of power levels by an acoustic wave inphoto sensing piezoelectric crystal.

That's why the interval between W_F and W_D, defining basic mass of recombination levels, is modulated from the point of view of impurity levels system in a forbidden band. The solution of a set of equations $\partial\ n / \partial\ t$ and $\partial n_t / \partial t$ for these conditions is difficult, therefore we shall limit by only qualitative consideration. Let's consider two cases: with one recombination level W_r (fig. 1) and with two W_{r1} and W_{r2} (fig. 2). Let in an initial moment of consideration quasi Fermi level $W'_{F'}$ is closer to the conductivity band bottom, that corresponds to the increased local concentration of grouped carriers.

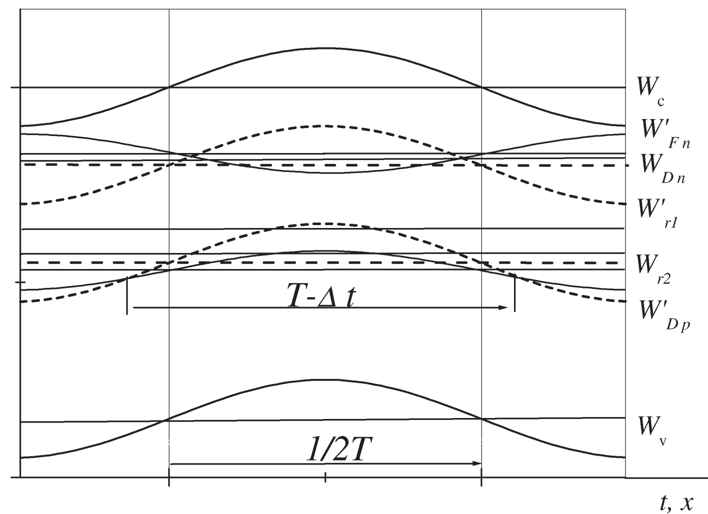

Fig. 2. Modulation of a two-level structure by an acoustic wave.

At transition to area with reduced concentration quasi Fermi level $W_F^{/}$ is lowered, and demarcation level $W'_{D'}$ is increased. The interval $W'_F - W'_\Delta$ is narrowed down, the level W_r goes out far from the bounds of this interval. The holes life time is $\tau_p = 1/N_r v_T S_p$, and at enough large concentration p_r, and also n_r, the electrons life time $\tau_n = 1/p_r v_T S_n$ is increased, as the probability of a recombination through this level decreases:

$$1/\tau \sim \exp\left(-W_r/kT + W_{F'}/kT\right).$$

The average concentration of free carriers in zone is increased. Thus, we obtain an experimental outcome coincided with analytical data by Y. V. Gulyaev et. al.[4,5].

The more complicated picture is observed for photosensitive crystal possessing two recombination levels. Lets the part of impurities is compensated. In such crystal the levels for electrons W_{F_n} ,W_{D_n} and for holes W_{F_p} ,W_{D_p} are various; for everyone W_r exists two demarcation levels. It is possible to accept, that the intervals $W_{F_n} - W_{D_n}$ and $W_{F_p} - W_{D_p}$ are approximately equal among themselves and are modulated in the dependence on local concentration n and p:

$$W_{F_n} - W_{D_n} = W_{F_n} - W_{F_n} + kT\ln(nS_n/pS_p) = W_{F_p} - W_{D_p}.$$

The interval of recombination levels $W'_{D_n} - W'_{D_p}$ becomes modulated too. Lets to the levels W_{r1} and W_{r2} there correspond concentrations N_{r1} and N_{r2}, and N_{r2} even on an order are higher N_{r1}, the cuts $S_{n2} << S_{n1} = S_{p1} = S_{p2}$. Let without of AW levels W_{r2} are located close to the level W_{D_p} and are closer to a middle of a forbidden band gap; such location can be chosen by illumination. In these conditions the recombination occurs, in basic, through W_{r2}, as $n_{r2} << n_{r1}$. In the case of modulated levels (fig. 2) W'_F and $W_D^{/}$ (in a fig. 2 modulations W_{F_p} and W_{D_n} is not represented, $\Delta W_{F_p} < \Delta W_{F_n}$) the role of centers W_{r2} varies: a defined time interval T — Δt centers W_{r2} appear as sensitization centers, but in remaining time they fall outside the limits W_D and play a role of usual trapping centers for holes. If $\tau < T$, at this time the filling of centers W_{r2} by holes falls down, $p'_{r2} < p_{r2}$. The filling of centers W_{r1} by holes is increased. The resettlement holes from W_{r2} on W_{r1} (through band W_V) reduces in increase of a role of a recombination through W_{r1}: the process reduce sensitization and decrease the majority carriers τ_n life time and increase the photoresistivity.

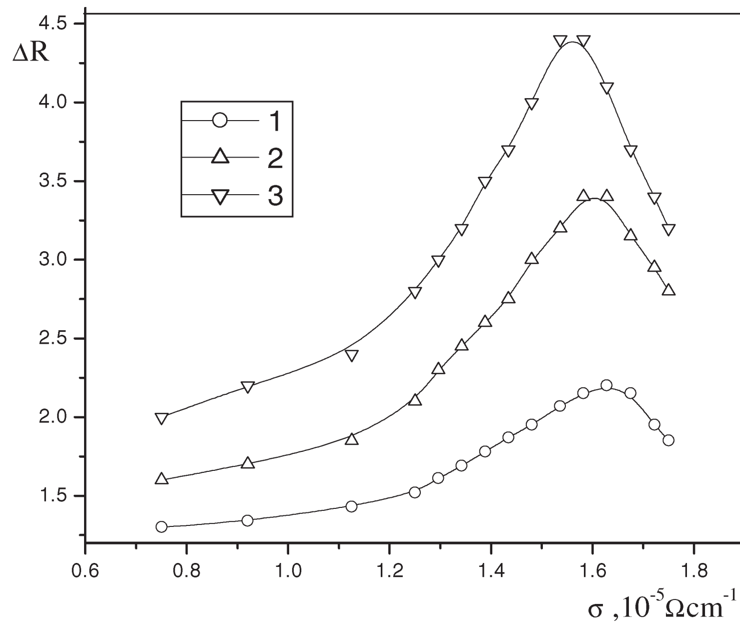

Fig. 3. The dependence of acoustoresistivity on illumination.

It was repeatedly confirmed experimentally; in such crystals we observed the strong acoustosensitivity. The presented model is illustrated by the dependences of resistance modification on illumination of CdS or CdSe crystals, when the regulating of a position of non-equilibrium Fermi level realizes greatest acoustosensitivity: curve ΔR (fig. 3) have an expressed extremum. The model becomes some complicated by the account of trapping levels. The significant role in a release of traps can be played by tunneling through a barrier; when the equilibrium carriers almost are absent, and the bending of band is the greatest, AW can exhibit itself similarly to a long wavelength illumination. At grouping, as repeatedly was shown, the traps are liberated too .

Both these appearances direct the increases of concentration both majority (from donor), and minority (from acceptor) carriers. But the increase of minority carriers in two-center model sharply intensifies the recombination rate. Acoustoexitation of impurity we observed experimentally in the darkened out CdS or CdSe crystals. The complicated pulse — transitional characteristic of a resistance (fig. 4) was observed. In its initial part the fast fall of a resistance with duration time of order of μs was observed. Further resistance begins to increase, reaching a fixed value. Here relaxation time — some tens of μs. It can be explained by acoustoexitation of minority carriers, amplification of recombination rate with switching of a channel on one faster and decrease of basic carriers life time. In this experiment even weak illumination of crystal by long wavelength light cut-off the curve peaks in direction of resistance decrease.

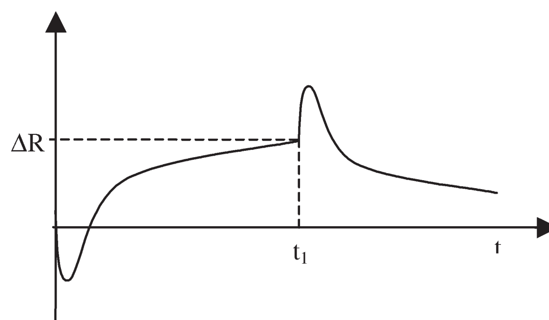

Fig. 4. The resistance relaxation at acoustoexitation of the impurities

Basically similar situation is realized on a surface. Thus the transversal component of acousto-emf creates a homogeneous charge carrier depletion or enrichment of a surface, displacing a surface Fermi level. The non-equilibrium bending of quasi Fermi level near the surface breaks an adsorption equilibrium and interchanging between a surface and environment. In the mentioned crystals on a surface the slow traps predominate and the equilibrium is reached slowly. The acoustoelectroadsorption effect is determined by conditions of acoustoelectric interaction on a surface. The considerable modification of a resistance was revealed by us in [12] for strongly photosensitive cadmium sulphide crystals, and later for cadmium selenide. It has appeared, that in such ARE a basic role is played by perturbation of recombination processes in sensitizing photoconductors.

2. Acoustoresistivity of thin films

We experimentally realized the significant ARE in a monolithic layered structure of piezoelectric — lithium niobate and a photo semiconductor — cadmium sulphide [12]. The layer cadmium sulphide were made on the special process engineering, had photosensitivity and was of polycrystalline structure (small-sized monocrystalline grains oriented chaotically). In such layers the nonlinear electrical conductivity in a medium with an infinite amount of heterogeneities of potential barriers between crystallites was assumed. Taking into account such the intercrystalline nonlinearity is possible to assume by ARE existence, based on perturbation of potential barriers by an electrical field of the wave. Really, the high-frequency electric field of acoustic wave in piezoelectric modulates an intercrystalline field, that is, locally modulates a height of a barrier.

If to present the layer as an infinite circuit of equivalent resistors connected sequentially, in the areas of increase of barriers the value of these resistors at action of a high-frequency electric field of the wave remain the large and almost not influences the initial value of a resistance of the layer; in areas where the barrier is decreases the injection of not basic carriers intensifies and value of resistors considerably falls down. The high-frequency field stimulates the current : combined operation of an exterior dc field and high-frequency field of the wave calls transition of an enlarged density electric current through the complex of potential barriers in polycrystal during passage of acoustic impulse. Short (on a comparison with early discussed) the relaxation time of the current indicates on the simple injection mechanism. It is known that it is possible to reduce influence of barriers by illumination on passage of a current in a polycrystalline sample. In the considered mechanism ARE, increasing layer conductivity by illumination it is possible to reduce a change of $\Delta R/R$.

At such consideration it is necessary to take into account, that in photoconductive cadmium sulphide in addition electrons a defined role play not basic carriers — holes, especially at small conductivity. The holes concentration can be close to electrons concentration. Taking into account the holes in barrier area the current through barriers increases. Probably, to recombine the carriers have no time (radiation by crystal it isn't marked). Further both majority and minority carriers participate in transfer of the current on the crystal.

In a fig. 5 the basic results of ARE in a structure with a polycrystalline film CdS are presented; the similar results were obtained for CdSe film in work [13]. The significant negative modification of a resistance ΔR is marked. On electrodes AB observed and acoustoelectric voltage U_{ae}; we carefully investigated its influence to summarized voltage. The voltage U_{ae} is proportional to the intensity W of a wave, $W \sim U_t^2$. The voltage U_{ae} is dependent on an electronic absorption coefficient α_e, which has an extremum in the range 10^6 Ω of a resistance (fig. 5, curve 3), that is, the value U_{ae} was not constants in all resistance range of the layer. The voltage U_{ae} contribution is not constants at representation of data in a scale U_t. According to experimental results the contribution of the voltage U_{ae} to total voltage drop ΔU across the resistance R_H is unimportant and it is much lower than

voltage U_{ARE}, $U_{ae} << U_{ARE}$. We had not separate it from total voltage *ΔU*. We have investigated *ΔR/R* across the wave propagation direction. The dependence *ΔR/R* on an initial resistance *R* is shown in fig. 6.

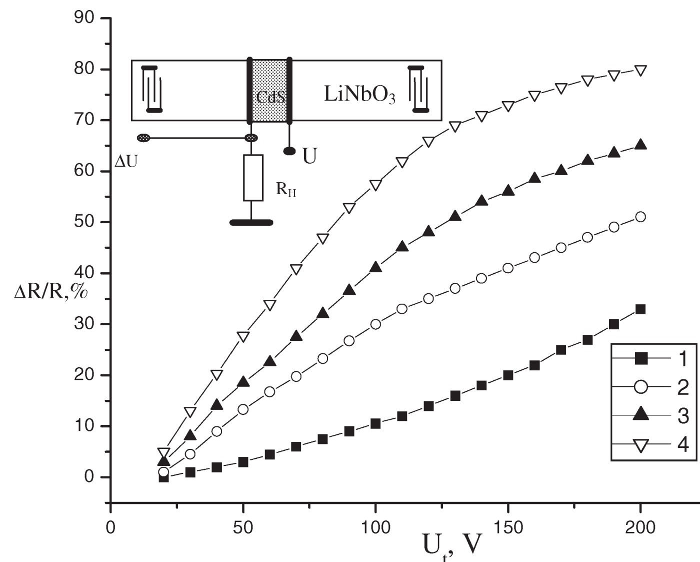

Fig. 5 Scheme of experiment for the definition longitudinal ARE and the dependence of a relative resistance *ΔR/R* on voltage U_S on an entering SAW transducer at measuring voltage on electrodes AB U=100 V and different initial resistances of a film CdS *R*, Ω: 1 — 10^8, 2 –$1 \cdot 10^7$, 3 — $1 \cdot 10^6$, 4 — $6 \cdot 10^5$.

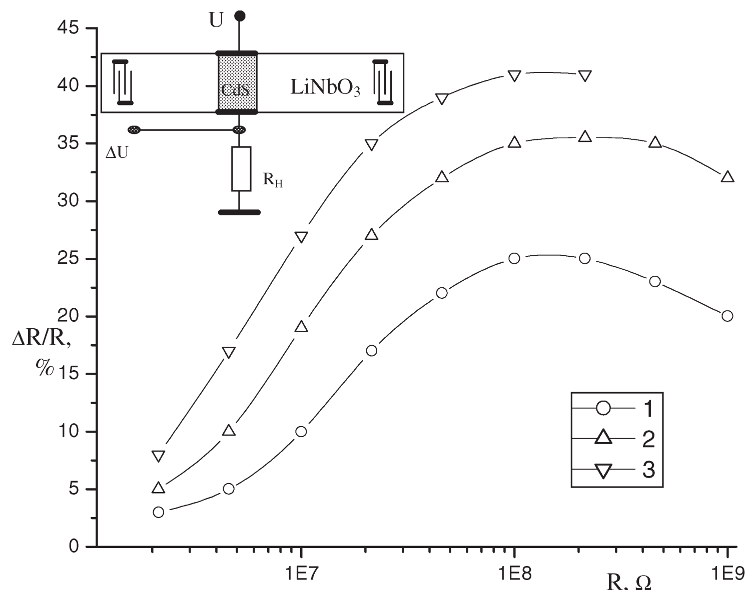

Fig. 6. Scheme of experiment for a research transversal ARE and the dependence of a relative resistance *ΔR/R* on an initial resistance R of a film CdS (b) at different measuring voltages on electrodes AB. *U*, V: 1 — 50, 2 — 200, 3 — 400.

The resistance *R* was controlled by optical illumination by white light. According to data of Fig. 6 it is visible, that the photo carriers concentration increase leads to a significant decrease of the *ΔR* : at large conductivities the ARE weakens. Also we conclude that at the low conductivity the barrier mechanism of ARE predominates. For the benefit of the barrier mechanism also indicate the data $\Delta R/R=f(U_t)$ (fig. 5): the increase of voltage *U* between electrodes AB, both in longitudinal, and in transversal directions, homogenizes a film smoothing barriers.

In this films, and also on a surface of CdS monocrystals the observation of positive ARE is possible; in all cases the sign ARE did not depend on direction of propagation of a wave.

3. The influence of surface electrokinetic processes on are

Taking certain surface electrokinetic processes and, also, interaction of a surface with a various gas environment into account, it appears to possible deriving ARE of both signs on the same substance.

In semiconductive polycrystalline films of tin oxide both the positive and the negative ARE is observed, when this film serves as a catalyst: if electron-acceptor heterogeneous reaction take place on the surface of the film the positive AR effect it was observed and in the case of electron-donor heterogeneous reaction the negative ARE [14]. In fig. 7 is shown such case. ARE effect in this films is used in sensors. The sensing activity is based on the SAW acoustoelectric interaction in semiconductor and the change of acoustoresistivity and longitudinal acoustoelectric current measurements. The main element of the sensor is the tin oxide film / lithium niobate layered structure. The structure is placed in a chamber, which can be filled with various gases or vented out. As a rule, semiconductor- type gas sensors have been fabricated mainly by sintering tin oxide powder [15,16]. In our case the tin oxide film on the surface of lithium niobate was formed by acoustochemical oxidation of metal tin film in the oxygen atmosphere using the method created by us [17].

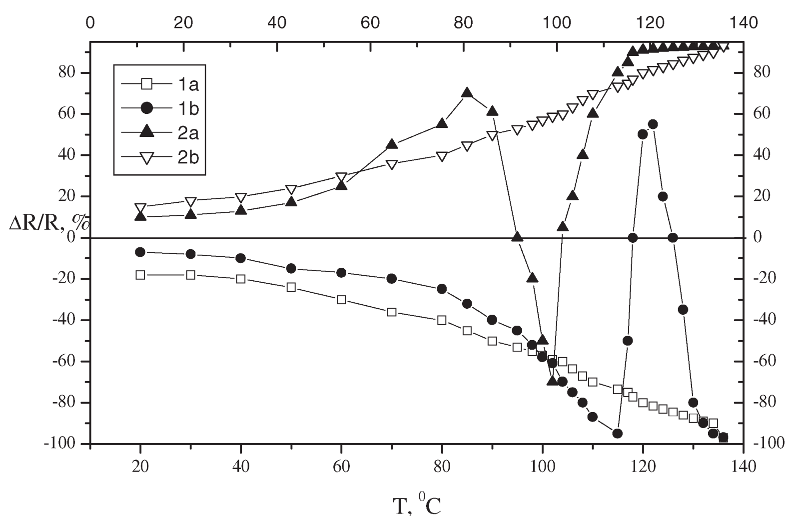

Fig. 7. Acoustoresistivity as a function of operating temperature in oxygen atmosphere (curves 1a, 1b) and in air with hydrogen (hydrogen concentration 1000ppm, curves 2a, 2b).

Using such method, it easy to control the properties of the film. Polycrystalline tin oxide film made by this method distinguishes in acoustoelectric interaction. An enormous acoustoresistive effect is observed in it too, and the film sensitively reacts to exterior influence including an ambient gas.

Tin oxide is the n-type semiconductor. The conduction electrons of tin oxide play a major role in gas sensing, since the concentration of conduction electrons changes as the surface of polycrystalline tin oxide is exposed to the gas to be detected. Some gases are electron-accepting particles (For example O_2) and are decreasing the conductivity, and some — electron-donoring (For example H_2) and increasing the conductivity. For the polycrystalline film, the neck size connecting adjacent grains is also an important factor in determining the gas sensitivity. The neck size not simply can be controlled; since it can be changed by various experimental conditions such as the parameters of continuous mode SAW, the oxidation temperature and acoustoelectrochemical transition time. Polycrystalline thin films also seem suitable for gas sensing, because they have a large surface — to — volume ratio and good

possibilities of fast adsorption-desorption processes on their surfaces. Furthermore, the use of SAW in continuous mode increases the adsorption ability of the surface [18]. SAW propagating in the structure changes the parameters of surface states and creates new states [19] and it can change the adsorption equilibrium between solid and gas phase. SAW during wave period induces the additional charge $\delta\ Q$ [20]

$$\delta Q = \frac{\varepsilon e L_D |E_s|^2}{24\pi T},$$

so the relative surface adsorption ability will be:

$$\frac{\Delta N}{N} = (1-\eta_0)(\pm\kappa|E_s|^2 - 1 + \sqrt{\kappa^2|E_s|^2 + 1}),$$

where $\kappa = \frac{\varepsilon e L_D}{48\pi TSN_0(1-\eta_0)}$; *e* is the electronic charge, *e* is the dielectric constant of semiconductor, $|E_s|^2$ is the SAW electric field on the semiconductor surface, *T* is the temperature in energy units, *S* is the area of the film surface, $N = N_0(T, P)$ is the equilibrium concentration of adsorbed particles on the surface, L_D is the Debye screening length. The sign "+" corresponds donor and "-" corresponds acceptor particles adsorption.

Conclusion

The influence of acoustic waves in material electrical resistance is determined by some physical mechanisms of interaction. The strong ARE reveals in thin films and layers. Both positive and negative ARE have been revealed by controlling the population of surface levels. In presented paper the ARE of p-n junction and others inhomogeneous(heterogeneous) structures is not discussed.

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