

Geometry of chaos – the Bernoulli equation for ternary and quaternary alloys

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Abstract: To understand and interpret the experimental data on the phonon spectra of the solid solutions it is necessary to describe mathematically the non-regular distribution of atoms in its lattices. It occurs that such description is possible in case of the strongly chaotically (stochastically) homogenous distribution what require a very great number of atoms and a very carefully mixed alloys. These conditions are fulfilled generally in case of the high quality homogenous semiconductor solid solutions of the III-V and II-VI semiconductor compounds. In this case we can use the Bernoulli equation describing a probability to occur a one from n equivalent events what can be apply to the probability to find one from n configurations in the solid solution lattice. The results described in this paper for ternary HgCdTe and quaternary HgZnCdTe can give affirmative answer on the question: whether geometry of chaos e.g. the Bernoulli equation is enough to describe the observed phonon spectra.

Keywords: Chaotic modeling, Phonon spectra, Solid solutions, Chaotic simulation.

1. Introduction

The role of alloys of semiconductors in electronics and optoelectronics is constantly increasing; more and more devices (lasers, convertors, detectors, memory elements etc.) are based on semiconductor solid solutions – another name for alloys. A homogenous high quality solid solution with the substitution of a cation or anion (we consider here mainly the semiconductor compounds III-V and II-VI) should be characterized by the random distribution of atoms in a lattice. However, we cannot call this a crystalline lattice because it does not possess one very important feature of crystals – long-acted ordering. From the point of view of electronic structuring it means some fluctuations of periodic potential exist in relation to different atoms (cation or anion) in the structurally reproducible basic cell in the lattice – a tetrahedron in the case of a zinc-blend structure and wurzit structure. Experiments have shown that electrons having a long-length wave function (belonging to the minimum in the center of the Brilluoin zone) are practically insensitive to these fluctuations and a k -vector can be attributed to electrons as a “good” quantum number.

A different situation occurs, in principal, in the case of the phonon spectra in mixed crystals. The atom masses (cation or anion) in a structurally reproducible cell are not repeated for different cells in the lattice because of the chaotic (random) distribution of the “foreign” atoms. This means that



definitively different vibration frequencies of the dipole pairs presented in cells exist in the case of optical oscillations. Thus, the phonon spectrum of alloys behaves ambivalently: usually the optical phonon branches split into several modes proper to the mass of the components of the mixed crystal lattice; although research contributions about one-mode behavior exist also [1]. The principally important question is: whether these vibrations of different dipole pairs are connected in the alloy lattice and form a running wave (phonons) with dispersion relations or, to the contrary: they are disseminated on a great number of local modes? This question is connected with another problem concerning the local structure of solid solutions: that which can be described by the random distribution of atoms – the geometry of chaos – or is this geometrical factor not sufficient and it is necessary to add a thermodynamic factor?

In this paper we attempt to generalise the experimental results obtained from the ternary $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ and quaternary $\text{Zn}_y\text{Cd}_x\text{Hg}_{1-x-y}\text{Te}$ far-infrared spectra (FIR-spectra) using synchrotron radiation as source in the far-infrared region. Their interpretation based on the Bernoulli equation can, in part, respond to the formulated above questions concerning the phonon spectra of these materials.

2. The Model

To understand and interpret the experimental data on the phonon spectra of the solid solutions it is necessary to describe mathematically the non-regular distribution of atoms in its lattices. It occurs that such description is possible in case of the strongly chaotically (stochastically) homogenous distribution what require a very great number of atoms and a very carefully mixed alloys. These conditions are fulfilled generally in case of the high quality homogenous semiconductor solid solutions of the III-V and II-VI semiconductor compounds. In this case we can use the Bernoulli equation [2] describing a probability to occur a one from n equivalent events what can be apply to the probability to find one from n configurations in the solid solution lattice.

The crystalline structure of the most III-V and II-VI compounds (possessed zinc-blend or wurzit structure as was mentioned above) is characterized by basic cell – tetrahedron – each with a central ion surrounded in the first coordination shell by four nearest neighbours (NN) at the vertices. In a $A_{1-x}B_xZ$ ternary solid solution with substitution of the cation A by cation B, different tetrahedron configurations T_n (n is the number of B-atoms in the tetrahedron) coexist simultaneously: 2 strictly-binary ones corresponding to the AZ and the BZ compounds, whose lattices are characterized by the tetrahedron units T0 and T4 (configurations), respectively and 3 strictly-ternary ones actually characterized by the configurations T1, T2 and T3. The similar configurations exist in a AY_yZ_{1-y} solid solution where anions Z are substituted by anions Y – the tetrahedra will be looked at similarly because in zinc-blend lattice we can represent a basic unit as tetrahedron in two versions: centred by anion and surrounded by four cations or oppositely: four anions surrounded cation in centre.

The probability to find the T_n configuration in ideal lattice of the $A_{1-x}B_xZ$ or AY_yZ_{1-y} ternary solid solution can be find using the Bernoulli polynomial [2]:

$$P_n(x) = \binom{4}{n} (1-x)^{4-n} x^n \quad (1)$$

where $\binom{4}{n} = \frac{4!}{n!(4-n)!}$ is the number of combinations with n elements in the fourth set:

$$\binom{4}{0} = \binom{4}{4} = 1, \binom{4}{1} = \binom{4}{3} = 4, \binom{4}{2} = 6,$$

x is a mol composition of BZ compound in the solid solution what is equal to the ratio of the B-Z ion pairs per whole number of ion pairs in lattice.

It is obvious that probability $P_n(x)$ must be function of composition x because increasing of x means increasing of the B-atoms number in lattice what leads to increasing of the tetrahedron's number with high value of n (not higher then 4). The sum of probabilities to find all configurations in lattice of alloy with composition x must be equal to 1:

$$\sum_n^4 P_n(x) = 1 \quad (2)$$

The probabilities to find a some of cation A or B respectively are:

$$P_n^A(x) = \frac{4-n}{4} \binom{4}{n} (1-x)^{4-n} x^n \quad (3)$$

$$P_n^B(x) = \frac{n}{4} \binom{4}{n} (1-x)^{4-n} x^n \quad (4)$$

It is easy to determine that

$$\sum_n^4 P_n^A(x) = 1 - x \quad (5)$$

$$\sum_n^4 P_n^B(x) = x \quad (6)$$

It is necessary to note that (3) and (4) are simultaneously the probabilities to find in the solid solution lattice the ion pairs A-Z and B-Z.

The oscillator strength of the vibrational mode generated by a A-Z-dipole in the T_n configuration is [3]:

$$S_n^{A-Z}(x) = f_{AZ} N_0 P_n^A(x) \quad (7)$$

where f_{AZ} is the oscillator strength of the single dipole A-Z-pair, N_0 is number of dipole pairs A-Z in the solid solution crystal, probability $P_n^A(x)$ is determined by (3).

It is important to remember that three assumption are introduced in this consideration:

- 1) the role of defects is negligible;
- 2) the alloy lattice is ideally homogenous and a random distribution of atoms in lattice takes place (stochastic homogeneity);
- 3) the oscillator strengths of the single dipole pairs for different configurations T_n are the same e.g. f_{AZ} or f_{BZ} depends not on index n .

If these conditions are fulfilled, the oscillator sum rule

$$\sum_n S_n^{A-Z}(x) = \sum_n f_{AZ} N_0 P_n^A(x) = f_{AZ} N_0 \sum_n P_n^A(x) = f_{AZ} N_0 (1-x) \quad (8)$$

has to be satisfied.

Similarly for $B-Z$ dipole pairs:

$$S_n^{B-Z}(x) = f_{BZ} N_0 P_n^B(x) \quad (9)$$

and oscillator sum rule

$$\sum_n S_n^{B-Z}(x) = \sum_n f_{BZ} N_0 P_n^B(x) = f_{BZ} N_0 \sum_n P_n^B(x) = f_{BZ} N_0 x \quad (10)$$

The experimental $\text{Im } \varepsilon(\omega)$ -curves (obtained by Kramers-Kronig transformation from experimentally measured $R(\omega)$ -curve [4]) enable us to find the S_i values, to identify that with certain S_n^{A-Z} or S_n^{B-Z} and to verify the sums (9) or (10) what means the proportionality of the oscillator sum to the contain of the each component in alloy ($N_0 x$ is equal to molar percent of the BZ component and $N_0(1-x)$ – to the molar percent of the component AZ).

We consider here also the four-component solid solution $A_x B_y C_{1-x-y} Z$ with three kinds of cations A , B and C and with the same anion Z . The lattice of quaternary alloy contents 15 basic units (tetrahedra): three binary AZ , BZ , CZ and nine strictly ternary ABZ , ACZ , BCZ . If quaternary alloy have x mol part of AZ compound and y mol part of BZ we can determine the probability to find in lattice the tetrahedron T_{nm} with n A-cations and m B-cations. This probability is equal to:

$$P_{n,m}(x, y) = \binom{4-n}{m} \binom{4}{n} (1-x-y)^{4-n-m} x^n y^m \quad (11)$$

where matrixes $\binom{4-n}{m}$ and $\binom{4}{n}$ are the same number of combinations as in

Eqn. (1).

The correspond probabilities to find particular cations in lattice are:

$$P_n^A(x) = \frac{n}{4} \binom{4-n}{m} \binom{4}{n} x^n y^m (1-x)^{4-n-m}, \quad (12)$$

for cation A ;

$$P_n^B(x) = \frac{m}{4} \binom{4-n}{m} \binom{4}{n} x^n y^m (1-x)^{4-n-m}, \quad (13)$$

for cation B , and

$$P_n^C(x) = \frac{4-n-m}{4} \binom{4-n}{m} \binom{4}{n} x^n y^m (1-x)^{4-n-m} \quad (14)$$

for cation C .

By this way the Eqns. (3,4,7,8, 10, 12 – 14) [5] represent the complete description of the random atom distribution in the ternary $A_xB_{1-x}Z$ and quaternary $A_xB_yC_{1-x-y}Z$ solid solutions with substitution of cations. The four-component solid solution $A_xB_yC_{1-x-y}Z$ in ideally random case described by relations (11) and (12-14) consists from fifteen structural units – tetrahedra – which can generate 66 optically active phonon (vibrational) modes. These number of modes arose by next way: three strictly binary tetrahedra generate three vibrational modes AZ-like, BZ-like and CZ-like, nine (3x3) strictly ternary tetrahedra generate $9 \times 6 = 54$ vibrational modes and three strictly quaternary tetrahedra generate $3 \times 3 = 9$ vibrational modes: in sum 66 vibrational modes. In practice the most of these modes are degenerated (have the same frequencies): for example, the AZ-like modes generated in tetrahedra ABZ could have the same frequencies as AZ-like modes in tetrahedra ACZ. The same concerns the BZ-like and CZ-like modes. By this way the number of distinguished modes should be 30.

The expression for the oscillator strengths are similar as for ternary alloys:

$$S_{n,m}^{A-Z}(x,y) = f_{AZ} N_0 P_{n,m}^A(x,y) \quad (15)$$

where probability $P_{n,m}^A(x,y)$ is determined by (14). Similarly are given the expression for $S_{n,m}^{B-Z}$ and $S_{n,m}^{C-Z}$. The corresponding oscillator sum rule are:

$$\sum S_{n,m}^{A-Z} = N_0 f_{AZ} (1-x-y) \quad (16)$$

$$\sum S_{n,m}^{B-Z} = N_0 f_{BZ} x \quad (17)$$

$$\sum S_{n,m}^{C-Z} = N_0 f_{CZ} y \quad (18)$$

The role of these oscillator sum rules (16-18) would be the same as in ternary alloys (8,10) but in the practice it is more difficult to relies the verification of the random distribution of atoms because the number of theoretically possible modes is very large and this factor prevent calculation of the oscillator sum rule.

3. Experimental results and interpretation

Here will be presented the results on FIR-spectra for ternary and quaternary alloys in order to illustrate application of presented above model for interpretation these experimental spectra. The optical reflection spectra in the region from 10 to 10 000 cm^{-1} where the phonon frequency values (100 – 400 cm^{-1}) of semiconductor's compounds are located, were measured in the wide temperature interval and composition regions. Experiments were performed at the DAFNE-light laboratory at Frascati (Italy) using the experimental set-up described in paper [6].

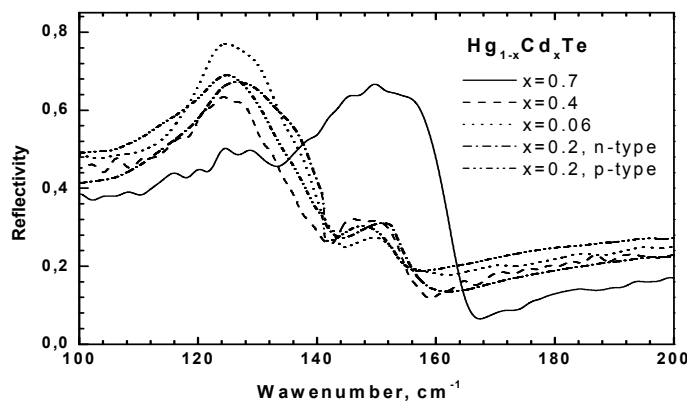


Fig.1. Reflectivity spectra for $Hg_{1-x}Cd_xTe$ (x is changing from 0.06 to 0.7) obtained at 300K .

The high-resolution reflection FIR-spectra obtained for the ternary $Hg_{1-x}Cd_xTe$ for compositions from $x=0.06$ to $x=0.7$ at a temperature 300K and in the spectral range from 100 cm^{-1} to 200 cm^{-1} are shown in Fig.1 as reflectivity curves $R(\omega)$. Two bands which shift weakly with the composition, are observed: first one around $118\text{-}128\text{ cm}^{-1}$ and second one around $145\text{-}155\text{ cm}^{-1}$. The amplitude of first band increases when the content of $HgTe$ increases and amplitude of second band increases when the $CdTe$ content increases. The first band corresponding to the $HgTe$ -like sub-band and second sub-band is the $CdTe$ -like one. This type of a reflectance spectrum again shows according to previous works [7,8,9] a two-mode behavior of the optical phonons in the $Hg_{1-x}Cd_xTe$ alloys from one hand. From other one, the subtle structure of both sub-bands is clearly observed too what undoubtedly indicates on multi-mode character of the phonon spectra (above two modes). Authors [10] interpreted these subtle structure in frame of the random model but they limited consideration of the FIR-spectra in the spectral region $118 - 160\text{ cm}^{-1}$. Whereas, in the region $90 - 116\text{ cm}^{-1}$ are observed additional lines (we can call them as Additional Phonon Modes (APM) whereas the main sub-bands we can call as Canonical Phonon Modes (CPM)), registered earlier in the above mentioned References.

Optical reflectivity from surface of $Zn_xCd_yHg_{1-x-y}Te$. The measurements of reflectivity were performed in temperature region from 30 K to 300 K. In Fig. 2 are presented reflectivity FIR-spectra obtained for composition $x=0.05$, $y=0.23$ for three temperatures: 30 K, 100K and 300 K. These curves are similar to typical reflection spectra but these curves have a much richer structure of spectra, as were observed for ternary alloys. It is seen three main bands at 130 cm^{-1} , 160 cm^{-1} and 180 cm^{-1} which can be point out in the reflective spectra. However, each of these sub-bands has additional subtle

structures, which point to the superposition of a greater number of lines. With increasing of temperature from 30 K to 300 K the subtle structure of observed sub-bands becomes more smooth.

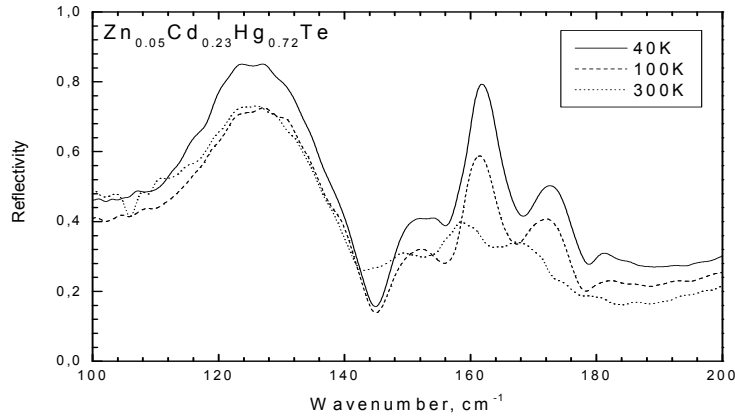


Fig.2. Reflectivity Spectra of $Zn_{0.05}Cd_{0.23}Hg_{0.72}Te$.

The lines corresponding to phonon modes are clearly observed on the $Im[\varepsilon(\omega)]$ curves calculated by Kramers-Kronig analyses from the experimental FIR reflectivity curves $R(\omega)$. In an attempt to treat the measured reflection spectra $R(\omega, \nu)$ of semiconductor materials (such as MCT with a considerable concentration of free carriers) in a reproducible way, as well as minimizing uncertainties due to subjective manual intervention, we have strictly abided by the rough procedure described in [4]. In Fig. 3 and 4 are shown $Im[\varepsilon(\omega)]$ -curves for p - $Hg_{0.8}Cd_{0.2}Te$ obtained for temperature 300 K and for $Zn_{0.05}Cd_{0.23}Hg_{0.72}Te$, respectively. The dispersion analysis of the CPMs and APMs was performed by approximating the $Im[\varepsilon(\omega, T)]$ curves by the Lorentzian sum

$$Im \varepsilon(\omega) = \sum_{i=1}^k \frac{S_i \gamma_i \omega}{(\omega_{TOi}^2 - \omega^2)^2 + \omega^2 \gamma_i^2} \quad (22)$$

where S_i , ω_{TOi} and γ_i are the oscillator strength, frequency and damping parameter of the i -phonon mode, respectively.

The results of spectral analysis for p - $Hg_{0.8}Cd_{0.2}Te$ are presented in Fig.3 and for n - $Hg_{0.8}Cd_{0.2}Te$ in Fig. 4. Parameters of Lorentzian's oscillators used for fitting the $Im[\varepsilon(\omega, T)]$ -curves are shown in Table 1.

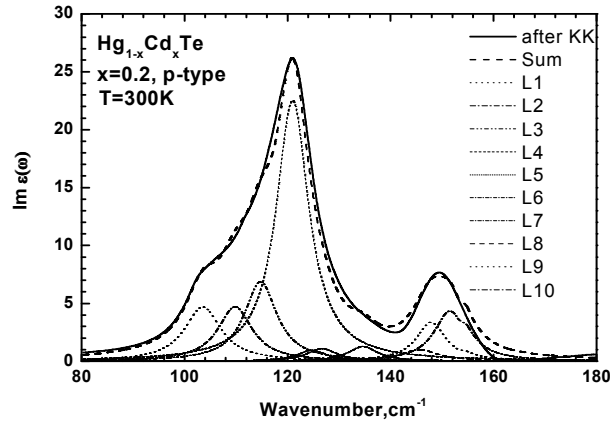


Fig.3. Imaginary part of the dielectric function of $Hg_{0.8}Cd_{0.2}Te$ p-type in the temperature 300K.

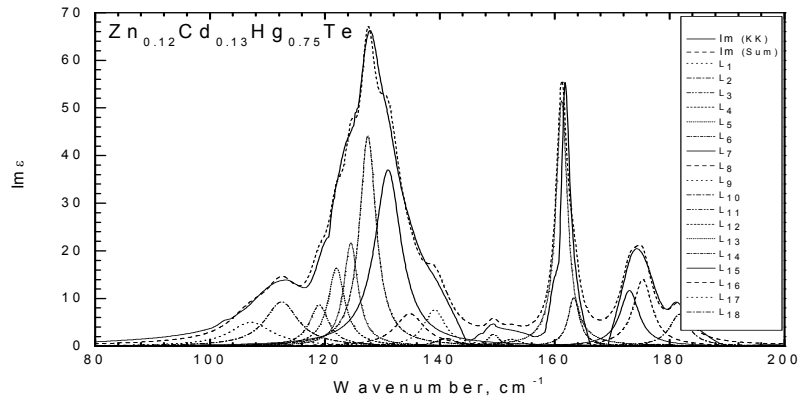


Fig. 4. Spectral analysis for sample $Zn_{0.12}Cd_{0.13}Hg_{0.75}Te$.

To identify observed lines, the probability to find the atoms Hg and Cd in the particular tetrahedra T_n (n is number of the Cd-atoms in tetrahedral) in the $Hg_{0.8}Cd_{0.2}Te$ lattice should be taken into account using formulas (3) and (4). If $x=0.2$ the values of $P_n^{Hg}(x)$ for different n are equal to: 0.410 ($n=4$), 0.307 ($n=2$), 0.077 ($n=3$) and 0.006 ($n=4$) while the $P_n^{Cd}(x)$ values are: 0.102 ($n=1$), 0.077 ($n=2$), 0.192 ($n=3$) and 0.002 ($n=4$).

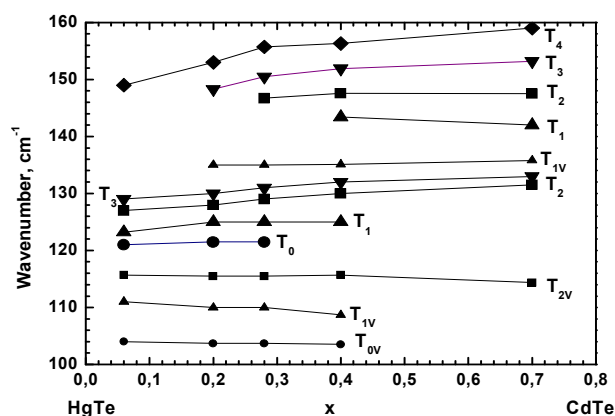
Table 1. Parameters of Lorentzian's oscillators used for fitting the $Im[\varepsilon(\omega, T)]$ -curves of the p - $Hg_{0.8}Cd_{0.2}Te$ for the temperature 300K.

Number of Line	S	ω	γ	ΣS_{HgTe}		ΣS_{CdTe}	
				CPM	AVM	CPM	AVM
1	4200	103.7	8.6		11400		
2	4200	110.0	8.1				
3	6100	115.0	7.7				
4	21100	121.3	7.7	31300			
5	830	125.0	7.0				
6	1000	127.0	7.0				
8	1200	135.0	7.0				1000
9	1300	146.1	8.9			9690	
10	3500	148.3	7.0				
11	4890	152.0	7.4				

At $T=30$ K all HgTe-like CPMs oscillate at the same frequency because tetrahedra with different number n are not deformed and we observed a degeneration of vibrational modes (Hg-Te and Cd-Te bonds have the same length). If $T=300$ K the splitting of the mode frequency takes place: the most strong line at 122.6 cm^{-1} should be generated by Hg-Te dipoles in the T_0 tetrahedron while the line at 125.0 cm^{-1} – by this dipoles in the T_1 one and very small line at 128.6 – in the T_2 . So, the frequency consequence takes place for HgTe-like modes: ${}^{HgTe}\omega_0 < {}^{HgTe}\omega_1 < {}^{HgTe}\omega_2 < {}^{HgTe}\omega_3$ according with work of Kozlyev et al. [10]. Analogical analyses for CdTe-like modes shown that the line at 151.5 cm^{-1} is generated by Cd-Te dipoles in T_1 tetrahedron and the line at 147.3 cm^{-1} – by the same dipoles in T_2 one. The frequency consequence for CdTe-like modes is: ${}^{CdTe}\omega_1 > {}^{CdTe}\omega_2 > {}^{CdTe}\omega_3 > {}^{CdTe}\omega_4$ what agree with the data of work [10] also. It allow to find to what basic cells (tetrahedra) belongs each observed vibrational mode generating by Hg-Te and Cd-Te dipoles: corresponding tetrahedra are shown in Fig. 5 for CPM (T_n) as well as for APM (T_m).

The general description of the phonon spectra is based on Fig. 5 where the composition dependences of the mode frequencies at the room temperature are present. In the MCT lattice there are two positions for Hg-atoms: stable one (Hg^I) and metastable one (Hg^{II}) [4]. If temperature increase, the number of Hg-atoms occupied the meta-stable positions (Hg^{II}) increases also and the deformation of crystal lattice rises, respectively. The last factor can cause the removing of degeneracy of the HgTe-like CPMs in n - $Hg_{0.8}Cd_{0.2}Te$ when the temperature increases over the 100 K: the AVMs appear simultaneously, too. Indeed, the AVM at 112 cm^{-1} (beside very weak from 30 K at 108 cm^{-1}) take place after 100 K in n-type $Hg_{0.8}Cd_{0.2}Te$ and after 200 K appear additionally one

AVM at $115-116\text{ cm}^{-1}$. The presence of Hg^{II} in a tetrahedron leads to the stretching of bonds which in its turn causes the shift of the $Hg-Te$ oscillation frequency towards smaller frequencies. This effect can occur in three kinds of tetrahedra: 1) containing 3 Hg -atoms in stable position (Hg^I) and one Hg^{II} ; 2) containing two Hg^I , one Cd -atom and one Hg^{II} ; 3) containing one Hg^I , two Cd -atoms and one Hg^{II} . The frequencies of $Hg-Te$ oscillations in these tetrahedra should be arranged in the next sequence: the lowest frequency corresponds to the $Hg-Te$ oscillations in the tetrahedron of first type and most higher corresponds to the oscillations in the tetrahedron of third type.



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Fig.5. The composition dependencies of the phonon mode frequencies for $p\text{-Hg}_{1-x}\text{Cd}_x\text{Te}$, at temperature 300K.

The lines in the range of $135-137\text{ cm}^{-1}$ are generated as could be assumed, by the oscillation of $Cd-Te$ pair in the tetrahedra containing two Hg^I , one Cd -atom and one Hg^{II} .

Therefore, the Figure 5 enable us to assume that the phonon spectra in *MCT* are reproduced in two versions: first one is realized in the lattice consisted only from the Hg^I -atoms (that are *CPM*) and second one occurs in the lattice included the Hg^{II} -atoms too (that are *APM*).

In Fig. 6 are shown the values of the oscillator strengths sums (OSS) for the $Hg-Te$ dipoles and for $Cd-Te$ dipoles for each samples investigated. The data are presented in two way: i) only OSS for *CPM* are included (open circles and squares), for $x=0.2$ there are two open circles because first one (upper open circle) is regarded to $n\text{-Cd}_{0.2}\text{Hg}_{0.8}\text{Te}$ and lower open circle – to $p\text{-Cd}_{0.2}\text{Hg}_{0.8}\text{Te}$; ii) in the OSS are included the *APM* OSS also (filled circles and squares). That enable us to obtain the dependencies of the OSS on composition. As follow from Fig. 6 if the oscillator strengths of *APM* are included in the sum of the oscillator strengths for the modes generated by $Hg-Te$ dipoles as well as $Cd-Te$ dipoles the OSS are proportional to the contain of correspond compound: to the

x in case of Cd-Te dipoles and to the $1-x$ in case of Hg-Te ones. As was mentioned above (see Eqns. (8) and (10)) these dependences are considered as a criterion of applying the Bernoulli equation to the phonon spectra interpretation of the ternary solid solutions.

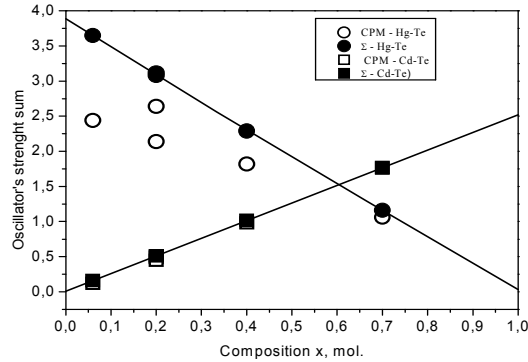


Fig. 6. The oscillator strength sum dependence on composition for the HgCdTe alloys

By this way, the same model developed for the quaternary alloys enable us successfully explain observed structure of the sub-bands for the $Zn_xCd_yHg_{1-x-y}Te$ alloys [11].

Table 2. The oscillator strength sum for Zn-Te dipoles in measured samples of $Zn_xCd_yHg_{1-x-y}Te$.

x, mol	OSS for Zn-Te
0.02	0.065
0.05	0.197
0.07	0.211
0.12	0.769
0.18	0.907

The next step is calculation of the OSS for certain dipole pairs. It is reason to consider the dependence on composition of the OSS for Zn-Te dipoles (the ZnTe contain is changed from 0.05 to 0.18). In Table 2 are shown calculated OSS for this dipoles. From Table 2 follow that really OSS for Zn-Te dipoles approximately is proportional to contain x of ZnTe in the $Zn_xCd_yHg_{1-x-y}Te$ alloys. Therefore, this important consequence of the random model (Eqn.17) is fulfilled.

4. Conclusions

The results described above for HgCdTe and HgZnCdTe can give affirmative answer on the question: whether geometry of chaos e.g. the Bernoulli equation is enough to describe the observed phonon spectra of ternary and quaternary alloys. The main criterion of the random distribution of atoms in the solid solution lattice – proportionality of the oscillator strength sum for the

each dipole pair to the content of the corresponding component – is fulfilled if the observed additional modes will be involved in sums of the oscillator strengths.

Application of the Bernoulli equation to the ternary and quaternary alloys enable us to decipher the tangled phonon spectra in these case of semiconductor solid solutions.

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