FAR INFRARED SPECTRA OF TETRAHEDRAL QUATERNARY ALLOYS

B.V.Robouch[1], R.Hus[2], J.Polit[2], E.M.Sheregii[2], E.Ściesińska[3], J.Ściesiński[3], A.Kisiel[4]

[1]Ex-Associazione Euraton-ENEA sulla Fuzione,Via Lucidi 3 00044 Frascati (rm), Italy
[2]Institute of Physics, Pedagogical University Rzeszow, Rejtana 16A, Poland
[3]Institute of Nuclear Physics n. Henryk Niewodniczański, Cracow, Poland.
[4.]Institute of Physics, Jagiellonian University, Cracow, Poland

Far infrared reflection spectra (FIR) of five distinct compositions of quaternary $Zn_xCd_yHg_{1-x-y}Te$ (ZMCT) are observed. These spectral curves treated by the Kramers–Kronig (KK) method, give the imaginary part of dielectric functions resolved into the Lorentzian oscillator set. Generally, 18 lines were distinguished in the FIR spectra of five compositions of ZMCT. The present analysis shows that the quaternary ZMCT has more frequencies in the observed spectra and expands the span of frequency region – from 108 to 190 cm^{-1} – withp respect to ternary solid solutions (118 to 180 cm^{-1}). The oscillator strength of a particular mode enables to determine the fraction of cells with each configuration.

1. Introduction

Random homogenous substitution of matrix cations by another metal atoms in solid solution lattices with common anion (as is the case of GaAs-AlAs or HgTe-CdTe) is known to cause a continuous reconstruction of electronic structure and phonon spectra with composition. It may be assumed that the introduction of the third cation will enable us to obtain a supplementary degree of freedom in the controlling of material parameters. It has an additional advantage in the case of HgTe-CdTe (MCT), because the introducing of Zn atoms stabilizes the weak Hg-Te bonds in the crystal lattice of this solid solution, whereas the presence of Cd atoms destabilizes them [1].

Quaternary $Zn_xCd_yHg_{1-x-y}Te$ (ZMCT) solid solutions have been first noted by A. Rogalski and J.Piotrowski [2] .

In the obtained epilayers magnetophonon resonance which is a powerful tool for the investigation of electron and hole spectra [3], was observed.

Introduction of the fourth component Zn causes not only simple extension of the physical properties of MCT, but new phenomena are also observable in connection with the multimodeness of crystal lattice. Multimodeness is an issue of primary significance in the study of four component solid solutions.

Verleur and Barker [4, 5]were the first to consider (for $GaAs_yP_{1-y}$) the contributions of five basic elemental tetrahedra to explain the featuers of vibrational spectra of solid solution.

Kozyrev at el [6] applied this approach to interpret the observed CdHgTe far infrared (FIR) reflection spectra.

The present paper discusses the results of the investigation of phonon spectra of $Zn_xCd_yHg_{1-x-y}Te$ (ZMCT).

2. Experimental Method

Reflection spectra were obtained by infrared Fourier spectrometer FTS-14 Digilab in the region 30–250 cm^{-1} ($\lambda = 40$–330 μm) at 300 K.

The CdTe substrates of the studied samples have been removed by mechanical grinding with subsequent chemical etching. Thus separated by this way $Zn_xCd_yHg_{1-x-y}Te$ (ZMCT) layers of $d = 4$–5 μm thickness.

The measurements of the reflectivity were performed at the angle of incidence $0°$ and $45°$.
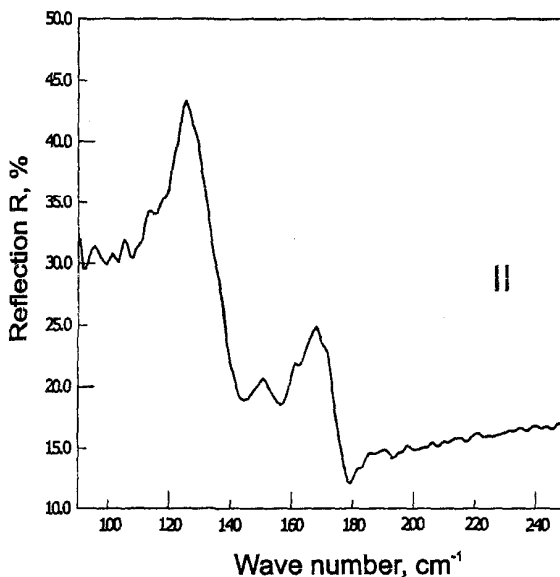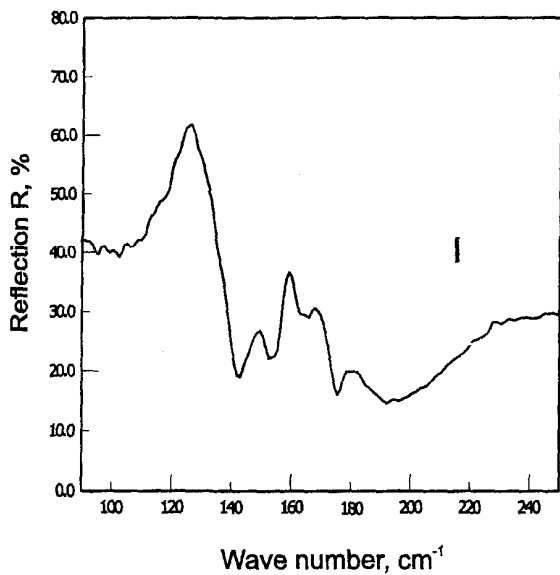
Fig. 1. Reflection spectra of $Zn_{0.07}Cd_{0.21}Hg_{0.81}Te$ (I) and $Zn_{0.18}Cd_{0.12}Hg_{0.7}Te$ (II) solid solutions.

The obtained curves of reflection spectra for five compositions of ZMCT are presented in Fig. 1. The presented curves are similar to typical reflection spectra [1, 7], on the other hand, much richer structure of the spectra can be observed. There are six lines for composition II and eight lines for composition V. However, lines 1 to 6 remain the main lines owing to their intensity. Fig. 1 also shows that the intensity of lines 3 to 5 grows with the increase of common content of Cd and Zn. Specifically, for composition IV (contents of Zn and Cd are approximately equal) the intensity of these lines is comparable. For composition V (content of Zn is

greater than that of Cd) line 5 becomes predominant for that group of lines. From this follows that line 5 should be attributed to ZnTe-like sublattice.

It is seen that in the spectra three main lines at 130 cm^{-1}, 160 cm^{-1}, 180 cm^{-1} can be pointed out. However, each of these lines has additional subtle structure, what indicates the superposition of greater number of lines.

3. Reconstruction of the Dielectric Function

Especially important for us is the imaginary part of dielectric function Im $\varepsilon(\omega)$ because it can be approximated in straight manner by Lorentzian oscillators. To determine the complex dielectric function $\varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)$ the Kramers–Kronig analysis was applied.

The real part of the dielectric function for $Zn_xCd_yHg_{1-x-y}Te$ can be calculated with the help of the following equation :

$$\text{Re}[\varepsilon(\omega)] = \varepsilon_\infty + \sum_{i=1}^{n} \frac{S_i(\omega_{TOi}^2 - \omega^2)}{(\omega_{TOi}^2 - \omega^2)^2 + \omega^2\gamma_i^2} \quad (1)$$

where ω is frequency [cm^{-1}], ω_{TO} – phonon mode frequency [cm^{-1}], S_i – oscillator strengths [cm^{-2}], γ – damping factor [cm^{-1}].

The statistical weight of the tetrahedron generated phonon modes should be involved into the oscillator strengths S_i as

$$S_{AZ} = N_0 f_{AZ} P_k(x) \quad , \quad (2)$$

where f_{AZ} is the oscillator strength of a single ionic pair. N_0 – the total number of the ionic pairs per unit volume, and $P_k(x)$ is the population of A cations in the k-th tetrahedtral configuration.

The imaginary part of the dielectric function for $Zn_xCd_yHg_{(1-x-y)}Te$ solid solutions can also be presented as the superposition of Lorentzians:

$$\text{Im}[\varepsilon(\omega)] = \sum_{i=1}^{n} \frac{S_i\gamma_i\omega}{(\omega_{TOi}^2 - \omega^2)^2 + \omega^2\gamma_i^2} \quad . \quad (3)$$

4. Approximation of the Reflection Spectra by Lorentzians

The curve of $Im\varepsilon(\omega)$ obtained after Kramers-Kronig analysis of the experimental reflection spectrum is shown in Fig. 2 by continuous upper curve

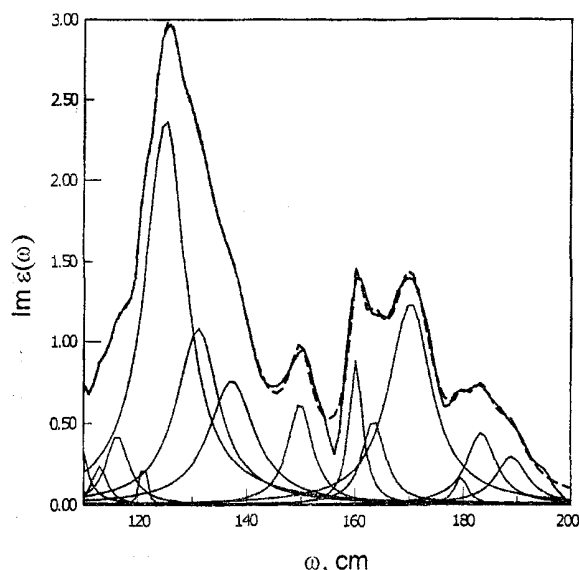Fig.2. $Im\varepsilon(\omega)$ (continuous upper curve) reconstructed by the Kramers-Kronig analysis of the $R(\omega)$ spectrum ,and the Lorentzian profiles (continuous curves) of the harmonic oscillators. and fitted curve (dashed curves).

Table 1. The Lorentzian parameters used for the approximation of $Zn_{0.07}Cd_{0.21}Hg_{0.81}Te$ phonon spectrum.

	ω $[cm^{-1}]$	S $[cm^{-2}]$	γ $[cm^{-1}]$
1.	108.3	220	2.9
2.	112.9	75	3.3
3.	116.2	237	5.1
4.	121.0	48	1.0
5.	125.0	2730	9.1
6.	131.1	1370	9.5
7.	137.3	1120	10.2
8.	150.0	495	5.5
9.	160.2	433	3.0
10.	163.5	433	5.3
11.	170.2	1940	9.5
12.	179.6	104	2.9
13.	183.4	484	6.6
14.	188.9	402	7.1

To reconstruct this curve of $Im\varepsilon(\omega)$ we use the superposition of 14 Lorentzian , according to the formula (3).

The obtained best result is shown in Fig. 2 by dashed curve. The Lorentzians, used for the approximation, are presented by continuous curves of $Im\varepsilon(\omega)$. The parameters of these Lorentzians are listed in Table 1.

It can be seen from Fig. 2 that from three above mentioned main lines can be approximated by several Lorentzians. Namely, the line at 130 cm^{-1} can be approximated by four oscillators, the line at 160 cm^{-1} – by four oscillators, and the line at 180 cm^{-1} – by three oscillators. Besides, there is a singularity in range of frequencies below 120 cm^{-1}, which requires three oscillators more.

5. Analysis of Tetrahedral Quaternary $Zn_xCd_yHg_{1-x-y}Te$ Solid Solutions

In ZMCT quaternary tetrahedral structures several observable configurational tetrahedra coexist simultaneously: 3 strict-binary (HgTe, CdTe, ZnTe), 9 strict-ternary (three for each of HgCdTe, HgZnTe, CdZnTe), and 3 strict-quaternary ($Hg^2Cd^1Zn^1Te$, $Hg^1Cd^2Zn^1Te$, $Hg^1Cd^1Zn^2Te$), resulting together in 30 modes:

$$3\times1 + 9\times2 + 3\times3 = 3 + 18 + 9 = 30$$

The contributions of these modes can increase the number of the observable lines in the reflection spectra. Indeed, the present analysis shows that the analysed quaternary ZMCT has more resonance frequencies in the observed spectra compared to the ternary solid solutions (118 to 180 cm^{-1}) and extends the frequency region from 108 to 190 cm^{-1}.

The oscillator strength of a particular mode enables the fraction of basic tetrahedral cells to be determined.

Therefore, it is necessary to consider the probability of finding cations in each tetrahedron. On the base stochastic homogeneous distribution of cations in the crystal lattice of $A_x B_y C_{1-x-y}Z$ solid solution this probability can be recorded in the form of Bernoulli polynomials:

$$P_{n,m}(x,y) = \binom{4-n}{m}\binom{4}{n}(1-x-y)^{4-n-m}x^{n}y^{m} \quad (5)$$

containing n A atoms, m B atoms and $4-n-m$ C atoms.

We can substitute the probability (5) into formula (2) and calculate the oscillator strength for each of the 30 above mentioned phonon modes. Then it is possible to attribute these modes to the 14 observable oscillators. This analysis has shown that the magnitudes of the experimental oscillator strengths, listed in Table 1, are insufficient to explain the experimental phonon spectra by 30 tetrahedral modes.

References

1. A.Sher, A.B.Chen, W.E.Spicer, C.K.Shih, *J. Vac. Sci. Technol. A* **3**, 105(1985).
2. A.Rogalski, J.Piotrowski, *Prog. Quantum Electron.* **87**, 12 (1988).
3. J.Cebulski, W.Gębicki,V.I.Ivanov-Omskii, J.Polit, E.M.Sheregii, *J. Phys. Condens. Matter* **10**, 8587 (1998).
4. H.W.Verleur, A.S.Barker, *Phys.Rev.* **149**, 715 (1966).
5. H.W.Verleur, A.S.Barker, *Phys.Rev.* **155**, 750 (1967).
6. S.P.Kozyrev, L.K.Vodopyanov, R.Tribulet, *Phys. Rev. B*, **58**, 1374 (1998).

СПЕКТРИ ЧЕТВІРНИХ ТЕТРАЕДРИЧНИХ ТВЕРДИХ РОЗЧИНІВ У ДАЛЕКІЙ ІНФРАЧЕРВОНІЙ ОБЛАСТІ

Б.В.Робуш[1], Р.Гус[2], Я.Політ[2], Є.М.Шерегій[2], Е.Сьцєсіньська[3], Й.Сьцєсіньський[3], А.Кісєль[4]

[1] Асоціація Euraton-ENEA, Фраскаті, Італія
[2] Інститут фізики, Педагогічний університет, Жешув, Польща
[3] Інститут ядерної фізики ім. Г.Нєводнічанського, Краків, Польща
[4.] Інститут фізики, Ягеллонський університет, Краків, Польща

Отримано спектри п'яти складів четвірних твердих розчинів $Zn_xCd_yHg_{1-x-y}Te$ (ЦКРТ) в далекій інфрачервоній області. Методом Крамерса-Кроніга з цих спектральних кривих одержано уявну частину діелектричної проникності, яку розділено на набір лоренцівських осциляторів. Всього виявлено 18 ліній у далеких інфрачервоних спектрах п'яти складів ЦКРТ. Проведений аналіз показує, що в спектрах ЦКРТ спостерігається більше частот і розширюється частотна область – від 108 до 190 $см^{-1}$ – у порівнянні з потрійними твердими розчинами (від 118 до 180 $см^{-1}$). Сила осцилятора для конкретної моди дозволяє визначити частку комірок з кожною з конфігурацій.