THERMAL CONDUCTIVITY 0F $In_{1-x}Ga_xAs$ SOLID SOLUTION

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The thermal conductivity of $In_{1-x}Ga_xAs$ (with $0 \le x \le 0.08$) solid solutions has been investigated in the 80÷300K range. The thermal conductivity as a function of temperature and the alloy composition has been analyzed on the basis of the theory taking into account boundary phonon scattering, three – phonon normal and three – phonon Umklapp process, phonon resonance scattering and also phonon scattering on point defects with regard to the local change of density and elastic properties of crystals. The revealed dip in the temperature dependence of the thermal conductivity is explained by the phonon resonance scattering on various complexes.

1. Introduction

$In_{1-x}Ga_xAs$ solid solutions are perspective materials for solid-state devices and studies of their thermal properties may be used at computations necessary on designing. Besides experimentally investigating the phonon thermal conductivity of the solid solutions versus the content in the wide range of temperatures enables one to study the influence of distortion of the crystal lattice periodicity (lattice imperfections) and the phonon scattering and to estimate applicability of available theories of the phonon scattering on various defects. The thermal conductivity of this system was investigated in a number of works [1-7]. Abrahams, Braunstein, and Rossi [1] investigated the thermal conductivity of the $In_{1-x}Ga_xAs$ system except regions near initial components. Ohmer et al. [2] measured the thermal conductivity of GaAs-InAs alloys for the alloy concentration less than 1%. In a paper of Szmulowicz et all. [3] on a basis of theories of Klemens and Callaway gave a theoretical estimation for the Ohmer's and et al. experimental result. Their calculation considers both normal interactions and Umklapp anharmonic ones, as well as mass differences and size distortions for the scattering by point defects. The results indicate that it is the size-distortion scattering, which leads to a very abrupt decrease in the thermal conductivity for dilute alloys. In a work of Adachi [4] the lattice thermal resistivity of $Ga_{1-x}In_xAs$ alloys was analyzed with a theoretical prediction based on a simplified model of the alloy-disorder scattering. It was shown a quite good agreement with experimental data on various III-V ternary compounds.

It should be noted that in all above-mentioned works the thermal conductivity dependence on the indium and gallium contents is analyzed at room temperature.

Arasly et al.[5] investigated the thermal conductivity of the $In_{1-x}Ga_xAs$ solid solutions at high temperatures by the light flash method.

In the present work as a continuation of our earlier conducted investigations, the thermal conductivity of the $In_{1-x}Ga_xAs$ alloys near the InAs initial components in the range of 80 to 300K has been investigated.

2. Experimental

The investigated samples, which grown by the Czochralski method, had the identical charge carriers concentration of $2.4 \cdot 10^{17} cm^{-3}$ and the dislocation density 10^3 to $10^4 cm^{-2}$. The thermal conductivity was measured by the absolute stationary method and flash light method between 80 and 300K.

The thermal conductivity of the initial InAs and $In_{1-x}Ga_xAs$ solid solution (with $0 \le x \le 0.08$) as function of temperature is plotted in fig. I. As it is seen, with increasing the GaAs content the value of the thermal conductivity decreases and its temperature dependence is weaken that is typical for alloys. In the temperature dependence of the thermal conductivity is observed the obvious dip within the narrow interval of 90 to120K.

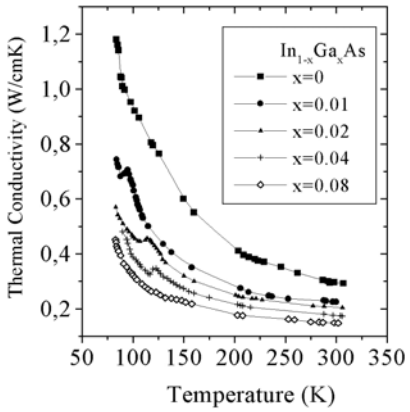

Fig.1. Thermal conductivity of $In_{1-x}Ga_xAs$ alloys versus temperature.

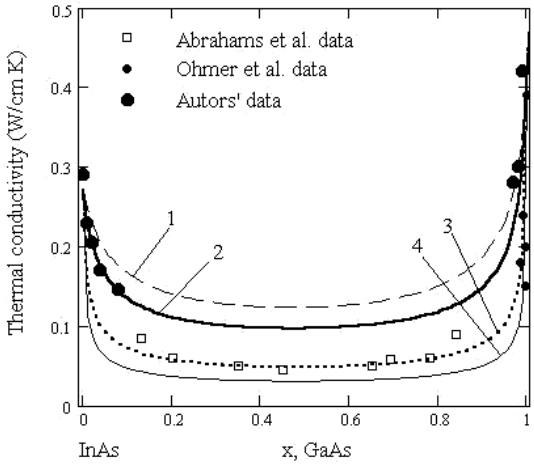

Fig.2. Thermal conductivity of $In_{1-x}Ga_xAs$ alloys as a function of the alloy concentration (x). The theoretical curves 1-4 have been calculated with formulas (1)-(4); (5)-(4); (5)-(6) with the value of G=1; and (5)-(6) with G=2, respectively.

The thermal conductivity for $In_{1-x}Ga_xAs$ at room temperature versus the alloy composition and data of Abrahams et al. [1] and Ohmer et al. [2] are presented in fig.2.

3. Discussion

The electron share of the thermal conductivity (K_{el}), determined in accordance with the Widemann-Franz formula, is insignificant and the observed change in the thermal conductivity is related with phonon processes.

The lattice thermal conductivity are analyzed in the framework of the well-known Callaway-Klemens model.

Despite that in the Callaway's model [8] it does not take into account the nonlinearity of the phonon spectrum dispersion and also makes no differences for longitudinal and transverse phonons in phonon-phonon interactions, in the literature widely use this model for the thermal conductivity analysis.

For a quantitative estimation of the phonon scattering on point defects in $In_{1-x}Ga_xAs$ at 300K first was computed the thermal conductivity taking into account U-processes and the phonon scattering on point defects by the given formula [9]:

$$K_p = K_\theta \left(\frac{\omega_\theta}{\omega_D}\right) arctan\left(\frac{\omega_D}{\omega_\theta}\right) \qquad (1)$$

where

$$\left(\frac{\omega_\theta}{\omega_D}\right)^2 = \frac{\hbar}{2\pi^2 v K_\theta \theta A} \qquad (2)$$

$$A = \frac{\Gamma . V}{4\pi v^3} \qquad (3)$$

$$\Gamma = x(1-x)\left[\left(\frac{\Delta M}{\bar{M}}\right)^2 + \varepsilon\left(\frac{\Delta\delta}{\delta}\right)^2\right] \qquad (4)$$

Here K_0 is the lattice thermal conductivity in the absence of defects, v is the speed of sound, V is the primitive cell volume, θ is Debye temperature, Γ is the disorder parameter including local changes of the density and elastic properties of the alloy when one atom is replaced with another, ω_D is the ponon frequency maximum in the Debye model, and ω_0 is the frequency at which the value of relaxation times, due to the phonon scattering on point defects $\tau_D^{-1} = A\omega^4$ and Umklapp prosesses $\tau_U^{-1} = B_U \omega^2 T$, are equal.

In the case of the A^3B^5 semiconductor compound, parameter ε is determined from the relation between elastic constants and the atomic volume. As for the $In_{1-x}Ga_xAs$ solid solution the Vegard`s law remains in force, the change of the lattice constant, in accordance with Ref.6, has been considered as a measure of the local change of material elastic properties. The parameters, which are necessary for the computation, have been taken from the work [3] and they have been linearly extrapolated for the solid solutions. It should be noted that in the $In_{1-x}Ga_xAs$ solid solution, the term connected with the local change of the elastic properties of medium makes its significant contribution to the disorder parameter, Γ.

As it is seen from fig.2, experimental data laid beneath the computed curve 1. It may be supposed that the significant quantitative divergence between the computation and the experimental data is connected with that the three-phonon normal process is not taking into consideration.

It is known that three-normal phonon processes themselves directly do not result in the thermal resistivity. At the same time the *N*- processes can determine the structure of the stationary nonequilibrium phonon distribution. As a result, their role turns out to be rather essential and they render the important influence on the thermal conductivity magnitude.

At high temperatures in solid solutions the high frequency phonons strongly scatter on point defects, and interaction between longitudinal phonons with conservation of the quasi-impulse, may influence on the lattice thermal conductivity.

The influence of *N*-processes on the thermal conductivity at $T>\theta$ in alloys is considered in Ref. 6, 10. Abeles [6] has proposed a phenomenological approach to a lattice thermal conductivity of disordered semiconductor alloys at high temperatures. His theory is based on the model presented by Klemens and Callaway and successfully used for alloys. Assuming that the temperature and frequency dependences of the relaxation time for three-phonon *N*- and *U*- processes, are the same, namely, $\tau_N^{-1} = B_N \omega^2 T$ and $\tau_U^{-1} = B_U \omega^2 T$, and for the scattering on point defects $\tau_i^{-1} = V\omega^4 \Gamma / 4\pi v^3$, in [6] it was obtained the following formula for the lattice thermal conductivity:

$$K = K_\theta \left(\frac{1}{1 + \frac{5C}{9}}\right)\left[\frac{arctan\, y}{y} + \frac{\left(1 - \frac{arctan\, y}{y}\right)}{\left(\frac{1+C}{C}\right)\frac{y^4}{5} - \frac{y^2}{3} + 1 - \frac{arctan\, y}{y}}\right] \qquad (5)$$

where $y^2 = \frac{(\omega_D / \omega_0)^2}{1 + 5C/9}$, $C = B_N/B_U$

C is the adjustable parameter indicating in how many times the *N*-processes are stronger than the *U*-processes.

At computations the parameter of disorder, Γ, is calculated with the formula (4), where the local change both the density and elastic properties of the alloy is taken into account. The curve 2 in fig.2, calculated by the formula (5) with the adjustable parameter value of C=2, well fits our

experimental data. It shows the essential role of the *N* processes in the phonon scattering in the $In_{1-x}Ga_xAs$ solid solution.

In Ref.3 the role of the *N* processes in the thermal conductivity in the GaAs-InAs alloy for the dilute alloy (with the InAs concentrations less than 1%) is appreciated and the disordered parameter Γ is expressed as:

$$\Gamma = \sum_i f_i \left[\frac{M_i - M}{M} + 2G \frac{V_1 - V}{V} \frac{1 + (4\mu / 3E)}{1 + (4\mu / 3E_i)} \right]^2 \quad (6)$$

where the sum is over the two alloy components; f_i is the alloy fraction for each constituent, M_i its molecular mass, V_i its molecular volume, E_i its bulk modulus; G is the Gruneissen constant, *M*,*V*, and E are averages of mass, volume, and bulk modulus; the μ is averages shear modulus.

The computed curves by the formula (5) and (6) for C=2 and G=2 (curve 3), G=1 (curve 4) are presented in Fig. 2 As it is seen, the curve 3 well fits Ohmer et al. (for dilutely alloyed) experimental data [2], but does not fit Abrahams and et al. [1] and our experimental data.

Thus our experimental data on the In-rich and Ga-rich $In_{1-x}Ga_xAs$ solid solutions well follow the Abeles expression (5).

The dips observed for various GaAs concentrations in the temperature dependence of the thermal conductivity in the of 90 to 120K (Fig.1) are of great interest. The observed dips evidently indicate the resonance phonon scattering realization. The *K*(T) behavior like this was also observed for pure A^3B^5 compounds at temperature lower 50K [11-14] and it is related, in the authors' opinion, with the resonance phonon scattering. However, the nature of the resonance phonon scattering center is not clear as yet. The resonance phonons scattering is observed at comparatively "high" temperatures in the range of 90 to 120 K in A^3B^5 alloys, in contrast with binary compounds.

The analysis of the temperature dependence of the thermal conductivity is carried out by the following formula:

$$K = \frac{k}{2\pi v} \left(\frac{2\pi k}{\hbar} \right)^3 T^3 \int_0^{\theta/T} \frac{\tau_c z^4 e^z}{(e^z - 1)^2} dz \quad (7)$$

where, $z = \frac{\hbar\omega}{kT}$.

The combined relaxation time (τ_c) includes all probable phonon scattering processes, the crystal boundary scattering, three –phonon normal process, three –phonon *Umklapp* process, Rayleigh scattering on point defects, phonon resonance scattering

$$\tau_c^{-1} = v / L + A\omega^4 + (B_N + B_U)\omega^2 T^3 + R \frac{\omega^2 T}{(\omega^2 - \omega_R^2)^2}, \quad (8)$$

where *L* is the length of a sample, ω_R is the resonance frequency. For comparison of calculated data with experiments we have used the method of least squares.

In a fig.3 the experimental data and computed by formula (7) curves for two of $In_{1-x}Ga_xAs$ alloys (with x=0,01 and x=0,04) are plotted. Values of the approximation coefficients are given in table 1. As it is seen from fig.3, including in the combined relaxation time also the relaxation time of the resonance phonon scattering, it is possible to quantitively circumscribe the dips observed in the temperature dependence of the thermal conductivity.

It should be noted that the adjustable parameter, A, is considerably differs from the computed parameter of the alloy-disorder scattering, determined by formula (3). Probably, it is connected with the presence of the other defects. It is possible that in solid solutions compositions of various atoms turn out to be in an environment of the "impurity" center owing to the chaotic distribution of components of the atoms in the crystal lattice. The nonequivalence of the nearest environment of the same atom can result in formation of various complexes, the presence of which brings about features of the phonon scattering. Such complexes can be also impurity – vacancy complexes

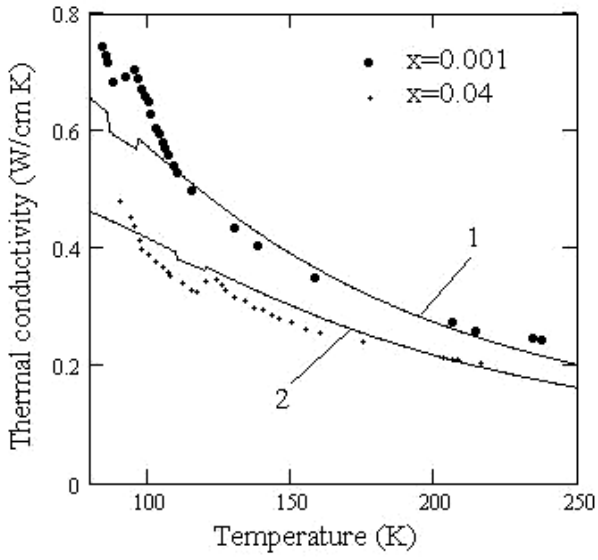

Fig.3. Thermal conductivity of $In_{1-x}Ga_xAs$ for x=0.01, x=0.04 alloys as a function of temperature The curves 1, 2 have been calculated with Formulas (7), (8).

Table 1.

Parameters used at computation of the phonon thermal conductivity of $In_{1-x}Ga_xAs$ with x=0,01; 0,04.

	InAs	GaAs	$In_{0.99}Ga_{0.01}As$	$In_{0.96}Ga_{0.04}As$
θ (K)	280	370	281	283
v (cm/s)	3.09×10^5	3.8×10^5	3.1×10^5	3.12×10^5
V, cm^3	5.56×10^{-23}	4.52×10^{-23}	5.55×10^{-23}	5.52×10^{-23}
μ (dyn/cm^2)	1.9×10^{11}	3.26×10^{11}	1.91×10^{11}	1.95×10^{11}
E (dyn/cm^2)	5.79×10^{11}	7.55×10^{11}	5.81×10^{11}	5.86×10^{11}
A_{exp} (s^3)			3.988×10^{-42}	7.507×10^{-42}
A_{cal} (s^3)			0.78×10^{-43}	3.01×10^{-43}
B_U ($s\ K^{-3}$)			0.718×10^{-25}	0.637×10^{-25}
B_N/B_U			2	2
ω_R (s^{-1})			1.327×10^{13}	1.54×10^{13}
v/L (s^{-1})			6.2×10^5	6.5×10^5
R ($s^{-3}K^{-2}$)			1.73×10^{28}	1.796×10^{26}

4. Summary

The investigation of the thermal conductivity of the $In_{1-x}Ga_xAs$ alloys at temperature between 80 and 300K has been shown that with increasing the Ga content the value of the thermal conductivity decreases and its temperature dependence is weaken. It has been established that for the solid solution in the phonon scattering on point defects together with *Umklapp* processes the normal processes also play essential role. The best good agreement with the theory is obtained with the quadratic frequency dependence for the relaxation time of the *N*-processes. In the temperature dependence of the thermal conductivity is revealed the obvious dips within the narrow interval of 90 to120K. The analysis of the thermal conductivity temperature dependence has displayed that in the combined relaxation time of the phonon scattering, including also the relaxation time of the resonance phonon scattering in the Klemens-Callaway formula, it is possible to quantitively describe the dip observed in the temperature dependence of the thermal conductivity. It is assumed that in the solid solutions resonance phonon scattering centers can be complexes related with the nonequivalence of the nearest environment of the same atom in the crystal lattice.

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$In_{1-x}Ga_xAs$ BƏRK MƏHLULLARININ İSTİLİK XASSƏLƏRİ

$In_{1-x}Ga_xAs$ bərk məhlullarının istilik keçiriciliyi 80-300K temperatur bölümündə tədqiq olunmuşdur. Kristalların tərkibindən və temperaturdan asılılığı olaraq istilik keçiriciliyinin dəyişməsi fononların müxtəlif mexanizmlərdən - üç-fononlu *N* və *U*-prosesləri, fononların rezonans səpilməsi, kristalın sərhədindən, sıxlığın və elastiki xassələrin dəyişməsini nəzərə alan nöqtəvi defektlərdən səpilməsini nəzərə alan nəzəriyyə əsasında araşdırılmışdır. İstilik keçiriciliyin temperatur asılılığında müşahidə olunan çuxurlar fononların müxtəlif mərkəzlərdən-komplekslərdən rezonansla səpilməsi ilə izah edilir.

Р.Н. Рагимов

ТЕПЛОПРОВОДНОСТЬ ТВЕРДЫХ РАСТВОРОВ $In_{1-x}Ga_xAs$

Исследование теплопроводности твердых растворов $In_{1-x}Ga_xAs$ проведено в области 80-300К. Теплопроводность сплавов как функция от температуры и состава анализирована на основе теорий, учитывающих рассеяние фононов на границах, трехфононные N и U - процессы, резонансное рассеяние фононов, рассеяние фононов на точечных дефектах с учетом локального изменения плотности и упругих свойств кристалла. Обнаруженное углубление в температурной зависимости теплопроводности объясняется резонансным рассеянием фононов на различных центрах- комплексах.

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