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THEORETICAL STUDY OF THE OSCILLATORY ELECTRONIC HEAT CAPACITY IN QUATERNARY ALLOYS UNDER MAGNETIC QUANTIZATION

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An attempt is made to study theoretically the effect of a quantizing magnetic field on the electronic heat capacity in quaternary alloys at low temperatures. It is found, taking $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP as an example, that the electronic heat capacity shows an oscillatory magnetic field dependence as is expected because of the dependence of the same capacity on the Fermi energy which oscillates with changing magnetic field. The amplitude of oscillations is, however, found to be significantly influenced by the alloy composition whereas the period is independent of the compositional parameter in quaternary alloys. In addition, the corresponding results for parabolic energy bands are also obtained from the expressions derived.

1. Introduction

In recent years, there has been considerable interest in studying the physical features of quaternary alloys specially in the presence of a quantizing magnetic field. Though many new effects associated with magnetic quantization in quaternary alloys having non-parabolic energy bands and obeying Kane's dispersion relation[1] have extensively been investigated, there still remain scopes in the investigations made while the interest for studying the various other physical properties of such alloys is becoming increasingly important. One such important parameter is the electronic heat capacity, which has been investigated in the literature under various physical conditions[2-4]. Incidentally, it is well-known that the heat conduction in alloys mainly takes place through the lattice waves since the electronic contribution to such conduction is normally not significant. However, the electronic contribution to the heat capacity would no longer be negligible particularly in the range of very low temperatures, where the phonon contribution is rather insignificant, and may become even more significant than that of the lattice. Besides, the switching speed of the thermal alloy devices is directly proportional to the electronic heat capacity. Incidentally, though the dependence of the electronic heat capacity in relatively large-gap semiconductors in the presence of a quantizing magnetic field has recently been studied elsewhere[3] in an incorrect manner the same dependence has yet to be correctly derived for quaternary alloys having Kane-type energy bands.

It is worth noting that even at low temperatures where the quantum effects become prominent, the electronic heat capacity in quaternary alloys having Kane-type energy bands is much greater than that of the corresponding III—IV semiconductors having the same E-$\vec{k}$ dispersion relation[5]. Incidentally, at low temperatures it is possible to observe[5] the *SdH* oscillations significantly in quaternary alloys. Besides, the changes in the Fermi energy in such alloys can only be obtained under strong magnetic quantization at low temperatures. Therefore, the changes in the electronic heat capacity in quaternary alloys in the presence of a quantizing magnetic field could be experimentally obtained only under such condition because of the same dependence of the capacity on Fermi energy and also since the corresponding lattice part would be negligible, at such low temperatures[6]. Furthermore, the quaternary alloys have received considerable attention recently as a possible material for the fabrication of heterojunction lasers[7-9], light emitting diodes[10], avalanche photodiodes in the near infrared region of the spectrum[11-13], microwave devices such as field effect transistors[14] and have an additional advantage that the band gap in such materials can be made as narrow as desired by varying the alloy composition. We may also note in this context that our analysis is a generalised one, because we can obtain the corresponding results for parabolic energy bands from our analytical expressions by equating the non-parabolicity parameter with zero. It would, therefore, be of much interest to study the effect of a quantizing magnetic field on the electronic heat capacity in quaternary alloys.

In what follows, we shall show theoretically, that the electronic heat capacity in quaternary alloys oscillates significantly with the reciprocal quantizing magnetic field as is expected because of the dependence of the same capacity on Fermi energy which, in turn, is known in the literature to be an oscillatory function of the quantizing magnetic field. Some of the oscillatory features will also be demonstrated explicitly by numerical calculations, taking $In_{1-x}Ga_x$-As_yP_{1-y} lattice matched to InP as an example.

2. Theoretical background

The heat capacity of the electrons in semiconductors can, in general be defined[4] as

$$C_e = \frac{\partial U}{\partial T} \tag{1}$$

where U is the total energy of the electrons and T is the temperature. It appears then that the derivation of the electronic heat capacity in non-parabolic semiconductors under magnetic quantization requires the corresponding expression of U which in turn, is determined by the density-of-states function. Incidentally, the density-of-states function under magnetic quantization in Kane-type semiconductors having non-parabolic energy bands can be written, following Nag[1], as

$$N_B(E) = \pi \hbar \omega_0 \left(\frac{2m^*}{h^2}\right)^{3/2} \sum_{n=0}^{n_{max}} (1 + 2aE)[\{E(1 + aE) - \Phi_+\}^{-1/2} + \{E(1 + aE) - \Phi_-\}^{-1/2}] \tag{2}$$

where $\hbar = h/2\pi$, h is the Planck's constant, T is the temperature, $\omega_0 = eB/m^*$, e is the electron charge, B is the quantizing magnetic field along z-direction, m^* is the effective electron mass at the edge of the conduction band,

$$a = \frac{1}{E_g}\left[1 - \frac{1}{3}\frac{E_g \Delta}{(E_g + \Delta)\left(E_g + \frac{2}{3}\Delta\right)} - \frac{2m^*}{m_0}\right],$$

E_g is the band-gap, Δ is the spin-orbit splitting, m_0 is the free electron mass, E is the energy in the presence of magnetic quantization as measured from the edge of the conduction band in the absence of any quantization, $\Phi_\pm = \left[\left(n + \frac{1}{2}\right)\hbar\omega_0 \pm \frac{1}{2} g_0 \mu B\right]$, $n\,(= 0, 1, 2, ...)$ is the Landau quantum number, g_0 is the magnitude of the spectroscopic splitting factor at the edge of the conduction band, and μ is the Bohr magneton.

Combining Eq. (2) with the Fermi-Dirac occupation probability factor, the total energy of the electrons under magnetic quantization can be expressed as

$$U(B) = \frac{N_c \Theta K_B T}{2} \sum_{n=0}^{n_{max}} \{[f(\eta_+)/\sqrt{a_+}] + [f(\eta_-)/\sqrt{a_-}]\} \tag{3}$$

where $N_c = 2\,[2\pi m^* k_B T/h^2]^{3/2}$, k_B is the Boltzmann constant, $\Theta = \hbar\omega_0/k_B T$, $f(\eta_\pm) = \left[\frac{9}{8} a k_B T F_{3/2}(\eta_{B,\pm}) + \frac{1}{2} A_\pm F_{1/2}(\eta_{B,\pm}) + C_\pm F_{-1/2}(n_{B,\pm})\right]$,

$F_j(\eta)$ is the Fermi-Dirac integral of order j as defined by Blakemore[15],

$$A_{\pm} = [1 + 3\,a\,b_{\pm}], \quad b_{\pm} = \left[\left(n + \frac{1}{2}\right)\hbar\,\omega_0 \pm \frac{1}{2} g_0\,\mu\,B\right] / a_{\pm},$$

$$a_{\pm} = 1 + a\left[\left(n + \frac{1}{2}\right)\hbar\,\omega_0 \pm \frac{1}{2} g_0\,\mu\,B\right], \; C_{\pm} = \frac{b_{\pm} \pm A_{\pm}}{k_B\,T},$$

$$\eta_{B,\pm} = (k_B\,T)^{-1}(E_{FB} - b_{\pm})$$

and E_{FB} is the Fermi energy under magnetic quantization as measured from the edge of the conduction band in the absence of any quantization. Thus using Eqs. (1) and (3) the magneto-heat capacity of the electrons in Kane-type semiconductors can be written as

$$C_e(B) = t_{1B} + t_{2B}\frac{\partial E_{FB}}{\partial T} \tag{4}$$

where

$$t_{1B} = \{U(B)\,N_c^{-1}\,[N_c' - a\,E_g'] - 2\,a_1\,E_g' \sum_{n=0}^{n_{max}} \{g_+ f(\eta_+)\,a_+^{-3/2} + g_- f(\eta_-)\,a_-^{-3/2}\} +$$

$$+ a_1 \sum_{n=0}^{n_{max}} \{\Psi(\eta_+) + \Psi(\eta_-)\}\}, \; N_c' = \frac{3}{2} N_c\,[1 + a\,T\,E_g'],$$

$$a_1 = \frac{N_c\,\Theta\,k_B\,T}{2}, \quad E_g' = \frac{\mathrm{d}E_g}{\mathrm{d}T},$$

$$g_{\pm} = a\,[a\,\hbar\,\omega_0\left(n + \frac{1}{2}\right) + 1 - a_{\pm}], \; \Psi(\eta_{\pm}) = (a_{\pm})^{-1/2}\left[\frac{9}{8} a_n' F_{3/2}(\eta_{B,\pm}) + \right.$$

$$+ \frac{1}{2} A_{\pm}' F_{1/2}(\eta_{B,\pm}) + C_{\pm}' F_{-1/2}(\eta_{B,\pm}) - G_{\pm}\left\{\frac{9}{8} a\,k_B\,T\,F_{1/2}(\eta_{B,\pm}) + \right.$$

$$\left.\left. + \frac{1}{2} A_{\pm} F_{-1/2}(\eta_{B,\pm}) + C_{\pm} F_{-3/2}(\eta_{B,\pm})\right\}\right],$$

$$a_n' = a\,k_B\,T\left[\frac{1}{T} - a\,E_g'\right],$$

$$A_{\pm}' = [1 - a\,E_g'\,b_{\pm}\,(b_{\pm}')^{-1}]\,3\,a\,b_{\pm}', \quad b_{\pm}' = E_g'\,H_{\pm},$$

$$H_{\pm} = (-a_{\pm})^{-1}\left[a\left(n + \frac{1}{2}\right)\hbar\,\omega_0 + b_{\pm}\,g_{\pm}\right],$$

$$c'_{\pm} = \left[\frac{-C_{\pm}}{T} + \frac{1}{k_B T}\left\{b_{\pm} A'_{\pm} + A_{\pm} b'_{\pm}\right\}\right],$$

$$G_{\pm} = \left[-T^{-1}\,\eta_{B,\pm} + \frac{1}{k_B T} b'_{\pm}\right]$$

and

$$l_{2B} = \left[a_1 (k_B T)^{-1} \sum_{n=0}^{n_{max}} \frac{1}{\sqrt{a_+}} \left\{\frac{9}{8} a\, k_B\, T\, F_{1/2}(\eta_{B,+}) + \frac{1}{2} A_+ F_{-1/2}(\eta_{B,+}) + \right.\right.$$

$$\left. + C_+ F_{-3/2}(\eta_{B,+})\right\} + \frac{1}{\sqrt{a_-}} \left\{\frac{9}{8} a\, k_B\, T\, F_{1/2}(\eta_{B,-}) + \frac{1}{2} A_- F_{-1/2}(\eta_{B,-}) + \right.$$

$$\left.\left. + C_- F_{-3/2}(\eta_{B,-})\right\}\right].$$

It appears then that the determination of $C_e(B)$ from Eq. (4), requires an expression of $\partial E_{FB}/\partial T$ which in turn is determined by the electron concentration. Incidentally, the corresponding electron statistics can be written, following Nag[1], as

$$n_0 = \frac{N_c \Theta}{2} \sum_{n=0}^{n_{max}} \left\{\frac{1}{\sqrt{a_+}} [A_+ F_{-1/2}(\eta_{B,+}) + \frac{3}{4} a\, k_B\, T\, F_{1/2}(\eta_{B,+})] + \right.$$

$$\left. + \frac{1}{\sqrt{a_-}} [A_- F_{-1/2}(\eta_{B,-}) + \frac{3}{4} a\, k_B\, T\, F_{1/2}(\eta_{B,-})]\right\}. \tag{5}$$

Since in the range of very low temperatures $\partial n_0/\partial T = 0$, from Eq. (5) we get

$$\frac{\partial E_{FB}}{\partial T} = l_1/l_2 \tag{6}$$

where

$$l_1 \equiv \left[-n_0 N'_c (N_c)^{-1} + n_0\, a\, E'_g (N_c)^{-1} - \right.$$

$$- \frac{N_c \Theta}{2} \sum_{n=0}^{n_{max}} \left\{\frac{1}{2} a_+^{-3/2} g_+ E'_g \overline{\psi}(\eta_+) + \right.$$

$$+ \frac{1}{2} a_-^{-3/2} g_- E'_g \overline{\psi}(\eta_-) + (a_+)^{-1/2} [A'_+ F_{-1/2}(\eta_{B,-}) +$$

$$+ \frac{3}{4} \{a\, k_B - a^2 k_B\, T\, E'_g\} F_{1/2}(\eta_{B,+})] + (a_-)^{-1/2} [A'_- F_{-1/2}(\eta_{B-}) +$$

$$+ \frac{3}{4} \{a\, k_B - a^2 k_B\, T\, E'_g\} F_{1/2}(\eta_{B,-})] - (a_+)^{-1/2} \{\overline{\psi}(\eta_+)\}' G_+ -$$

$$\left.\left. - \{\overline{\psi}(\eta_-)\}' (a_-)^{-1/2} G_-\right\}\right],$$

$$[\overline{\psi}(\eta_{\pm})] = [A_{\pm} F_{-1/2}(\eta_{B,\pm}) + \frac{3}{4} a k_B T F_{1/2}(\eta_{B,\pm})]$$

$$[\overline{\psi}(\eta_{\pm})]' = [A_{\pm} F_{-3/2}(\eta_{B,\pm}) + \frac{3}{4} a k_B T F_{-1/1}(\eta_{B,\pm})]$$

and

$$l_2 = \frac{N_c \Theta}{2 k_B T} \sum_{n=0}^{n_{max}} [(a_+)^{-1/2} \{\overline{\psi}(\eta_+)\}' + (a_-)^{-1/2} \{\overline{\psi}(\eta_-)\}'].$$

In the absence of magnetic field, the electronic heat capacity in non-parabolic semiconductors can be expressed as

$$C_e(0) = t_{10} + t_{20} \frac{\partial E_{F0}}{\partial T} \quad (7)$$

where

$$t_{10} = U(0) N_c' (N_c)^{-1} + U(0) T^{-1} + \frac{3}{2} k_B T N_c [-n_0 T^{-1} F_{1/2}(\eta_0) +$$

$$+ \frac{25 a k_B T}{4} F_{3/2}(\eta_0) - \frac{25}{4} a k_B \{a T E_g' F_{3/2}(\eta_0) + \eta_0 F_{1/2}(\eta_0)\}]$$

in which

$$U(0) = \frac{3}{2} N_c k_B T [F_3(\eta_0) + \frac{25 a k_B T}{4} F_{5/2}(\eta_0)], \quad (8)$$

$\eta_0 = E_{F0}/k_B T$, E_{F0} is the Fermi energy as measured from the edge of the conduction band in the absence of any quantization,

$$t_{20} = (3 N_c/2) [F_{-1/2}(\eta_0) + (25 a k_B T/4) F_{1/2}(\eta_0)]$$

$$\frac{\partial E_{F0}}{\partial T} = -l_3/l_4 \quad (9)$$

in which

$$l_3 = n_0 N_c^{-1} N_c' + N_c [-n_0 T^{-1} F_{-1/2}(\eta_0) + \frac{15 a k_B}{4} F_{3/2}(\eta_0) -$$

$$- \frac{15 a k_B}{4} \{a T E_g' F_{3/2}(\eta_0) + \eta_0 F_{1/2}(\eta_0)\}]$$

$$n_0 = N_c [F_{1/2}(\eta_0) + \frac{15 a k_B}{4} T F_{3/2}(\eta_0)] \quad (10)$$

and

$$l_4 = \frac{N_c}{k_B T} [F_{-1/2}(\eta_0) + \frac{15 a k_B T}{4} F_{1/2}(\eta_0)].$$

Incidentally, for parabolic energy bands Eqs. (4) and (7) assume the following forms:

$$C_e(B) = t_{1B} + t_{2B} \frac{\partial E_{FB}}{\partial T} \tag{11}$$

and

$$C_e(0) = \left[\frac{3}{2} N_c k_B F_{3/2}(\eta_0)\left(1 + \frac{N_c' T}{N_c}\right) - \frac{3}{2} k_B N_c \{F_{1/2}(\eta_0)\}^2 \{F_{-1/2}(\eta_0)\}^{-1}\right] \tag{12}$$

in which

$$t_{1B} \equiv [U(B) N_c' N_c^{-1} - a E_g' U(B) N_c^{-1} + a_1 \sum_{n=0}^{n_{max}} \{\varrho_+' F_{-1/2}(\eta_{B,+}) + \varrho_-' F_{-1/2}(\eta_{B,-}) - \delta_+ F_{-1/2}(\eta_{B,+}) - \delta_- F_{-1/2}(\eta_{B,-}) - \delta_+ \varrho_+ F_{-3/2}(\eta_{B,+}) - \delta_- \varrho_- F_{-3/2}(\eta_{B,-})\}],$$

$$U(B) = a_1 \sum_{n=0}^{n_{max}} [F_{1/2}(\eta_{B,+}) + F_{1/2}(\eta_{B,-}) + \varrho_+ F_{-1/2}(\eta_{B,-}) + \varrho_- F_{-1/2}(\eta_{B,-})] \tag{13}$$

$$\eta_{B,\pm} = (k_B T)^{-1} [E_{FB} - \Phi_\pm],\ \varrho_\pm = (k_B T)^{-1} \Phi_{,\pm},\ \varrho_\pm' = -T^{-1} [\varrho_\pm + a\left(n + \frac{1}{2}\right) \hbar \omega_0 E_g' k_B^{-1}],\ \delta_\pm = (k_B T)^{-1} [k_B \eta_{B,\pm} - a\left(n + \frac{1}{2}\right) \hbar \omega_0 E_g'],$$

$$t_{2B} = \frac{a_1}{k_B T} \sum_{n=0}^{n_{max}} [F_{-1/2}(\eta_{B,+}) + F_{-1/2}(\eta_{B,-}) + \varrho_+ F_{-3/2}(\eta_{B,+}) + \varrho_- F_{-3/2}(\eta_{B,-})]$$

$$\frac{\partial F_{FB}}{\partial T} = -\Omega_1/\Omega_2 \tag{14}$$

$$\Omega_1 = [N_c' n_0 N_c^{-1} + n_0 \Theta' N_c^{-1} - \frac{N_c \Theta}{2} \sum_{n=0}^{n_{max}} \{F_{-3/2}(\eta_{B,+}) \delta_+ + F_{-3/2}(\eta_{B,-}) \delta_-\},$$

$$\Theta' = -\frac{\Theta}{T}[1 + a T E_g'],\ \Omega_2 = \frac{N_c \Theta}{2 k_B T} \sum_{n=0}^{n_{max}} [F_{-3/2}(\eta_{B,+}) + F_{-3/2}(\eta_{B,-})] \tag{15}$$

and $$n_0 = \frac{N_c \Theta}{2} \sum_{n=0}^{n_{max}} [F_{-1/2}(\eta_{B,+}) + F_{-1/2}(\eta_{B,-})].$$

However, it may be stated in this context that the correct forms of Eqs. (2), (3), (5) and the relation for $C_e(0)$ of Ref. 3 are expressed through Eqs. (13), (11), (14) and (12) together with the allied definitions, respectively, in the present work. Besides, it many also be noted under that the conditions of temperature independent band-gap and $E_{F0} \gg k_B T$, Eq. (12) can be simplified to the well-known form[4)]

$$C_e(0) = \left(\frac{n_0 k_B \pi^2}{2}\right)(k_B T/E_{F0}). \tag{16}$$

3. Results and discussion

Using the appropriate equations one can determine the dependence of the normalized heat-capacity on a quantizing magnetic field in quaternary alloys provided the band-gap, the spin-orbit splitting parameter and the effective electron mass at the edge of the conduction band are known. Thus taking $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP as an example together with the parameters[16–18]

$$E_g = [1.35 - 0.738y + 0.138y^2]\ \mathrm{eV} \tag{17}$$

$$\Delta = [3.11 - 0.87y + 0.30y^2 + 0.007y^3]\ \mathrm{eV} \tag{18}$$

$$m^* = [0.080 - 0.039y]\, m_0$$

we have plotted (neglecting spin-effects) the $C_e(B)/C_e(0)$ versus $1/B$ in $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP for $y = 0.2$ at 4.2 K corresponding to an electron concentration of $2.28 \times 10^{22}\ \mathrm{m^{-3}}$, as shown in Fig. 1. The dotted plot shows the same dependence in the case of isotropic parabolic energy bands for the purpose of comparison.

It is observed from the figure that the electronic heat capacity in $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP exhibits an oscillatory function of the quantizing magnetic field and the band-nonparabolicity affects the amplitude of oscillations rather significantly. The oscillatory dependence is due to the crossing over of the Fermi level by the sub-bands in steps, resulting in a successive reduction in the number of occupied Landau levels as the magnetic field is increased. For each coincidence of a Landau level with the Fermi level, there would be a discontinuity in the density-of-states function resulting in a peak of oscillations. Thus it may be noted that the origin of the oscillations in the electronic heat capacity is the same as that of the Shubnikov de Hass oscillations. Incidentally, the *SdH* period being given by $\Delta'(1/B) = (e/h)(8/3\, n_0 \sqrt{\pi})^{2/3}$ is independent of alloy composition and is a function of electron concentration only[1]. The period as determined graphically has been found to be $0.41\ \mathrm{T^{-1}}$ which is in good agreement with that of $0.3945\ \mathrm{T^{-1}}$ determined from the above relation with the same parameters as used in obtaing Fig. 1. This is, therefore, a check up of the correctness of the data shown in the figure. It may be noted that at low temperatures, the broadening of the Landau levels may be neglected and the effects of spin splitting would simply to be increase the number of spikes of oscillations and to reduce their amplitudes. Besides, since the effect of electron-electron interactions, the spin-splitting effects and the broadening of Landau levels have not been considered in obtaining both the plots of Fig. 1, the essential meaning of the influence of band non-parabolicity on the electronic heat capacity would be meaningful. It may be noted that though the experimental verification of the basic content of our paper is not available to the best of our knowledge the magnetoelectronic heat capacity in quaternary alloys would be rather significant particularly in the range of low temperatures where the quantum effects become prominent and the lattice heat capacity contributes very small to the total heat capacity. Finally, it may be stated that though in a more rigorous treatment the modifications as suggested above should be considered along with a self consistent procedure this simplified analysis exhibits the basic qualitative features of the electronic heat capacity in quaternary alloys in the presence of a quantizing magnetic field.

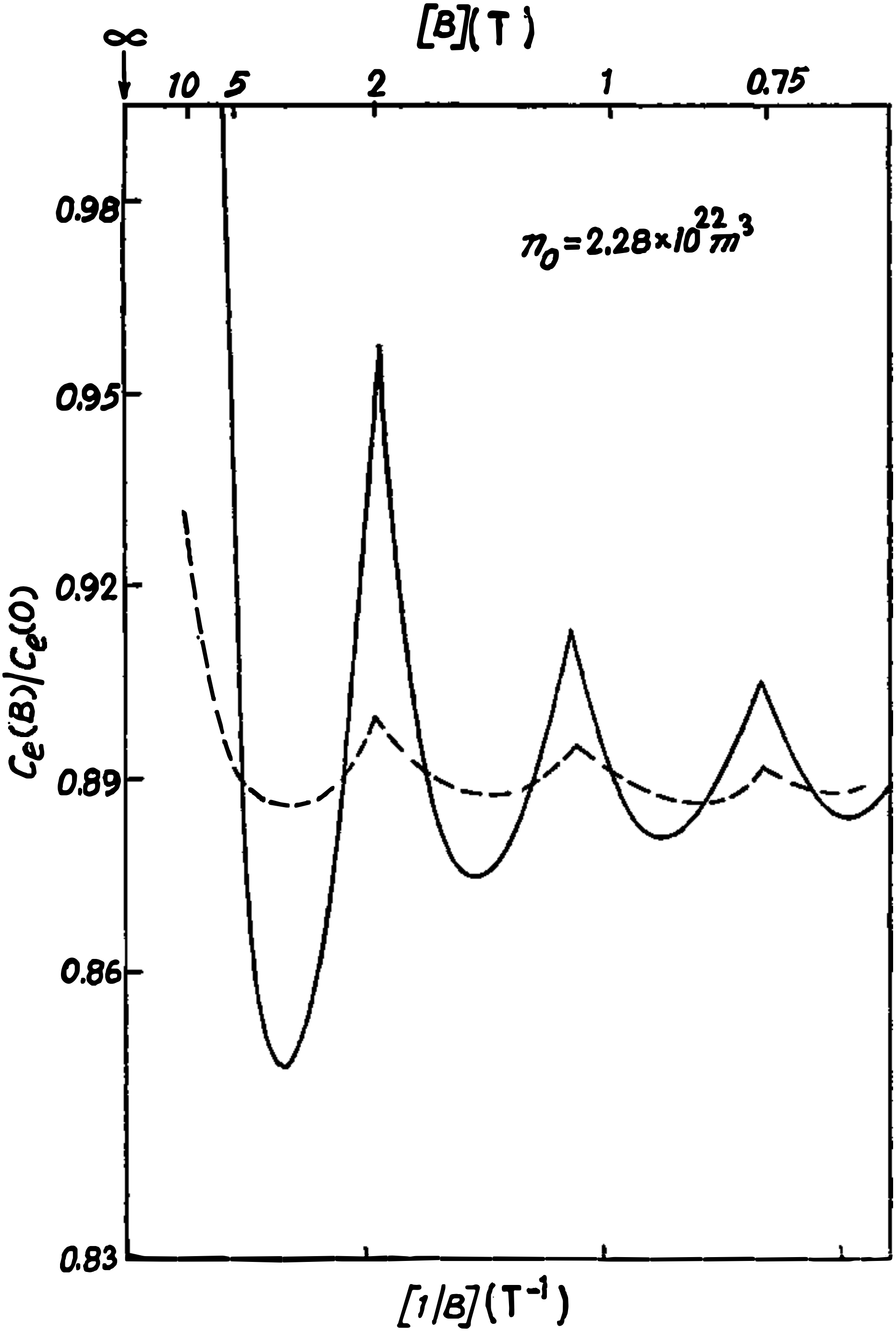

Fig. 1. The plot of the normalized electronic heat capacity as a function of the quantizing magnetic field in $In_{1-x}Ga_xAs_yP_{1-y}$ lattice matched to InP at 4.2 K corresponding to a given value of alloy composition. The dotted plot corresponds to the same dependence for isotropic parabolic energy bands.

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TEORIJSKA STUDIJA ELEKTRONSKOG TOPLINSKOG KAPACITETA U KVATERNARNIM LEGURAMA U UVJETIMA MAGNETSKE KVANTIZACIJE

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Originalni znanstveni rad

U radu je teorijski analiziran utjecaj magnetske kvantizacije na elektronski toplinski kapacitet u kvaternarnim legurama pri niskim temperaturama. Pokazano je, na primjeru $In_{1-x}Ga_xAs_yP_{1-y}$ legure usklađene po konstanti rešetke sa InP, da ovisnost elektronskog toplinskog kapaciteta o magnetskom polju ima oscilatorni karakter, što je očekivan rezultat jer Fermijeva energija (o kojoj ovisi kapacitet) oscilira pri promjeni magnetskog polja. Amplituda izrazito ovisi o sastavu legure, dok je perioda oscilacija nezavisna od kompozicionih parametara kvaternarne legure. Iz izvedenih relacija dobiveni su odgovarajući rezultati za paraboličnu energetsku vrpcu.