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Elastic moduli of Al–Si and Al–Ge solid solutions

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The elastic moduli of Al–Si and Al–Ge alloys obtained by solid-solution under pressure are investigated theoretically using a previous treatment based on the microscopic electronic theory. The obtained results for elastic coefficients such as bulk modulus, shear modulus, Young's modulus and Poisson's ratio for matrix Al are in good agreement with temperature-dependent experimental data. These moduli of Al–Si and Al–Ge alloys are calculated, and the concentration dependence of the elastic data is presented quantitatively.

1. Introduction

By rapid quenching from the liquid state under pressure, a substantial increase in the solubility of Si and Ge in Al has been observed experimentally [1–3]. Recently, the present authors have reported the solubility limit of Si and Ge in Al under pressure, using the microscopic electronic theory based on pseudopotentials and the virtual crystal approximation [4]. The bulk properties and solid solubility of Al–Si and Al–Ge systems obtained from first principles were consistent with experimental data. In the present work, we introduce the dynamic contribution to free energy, in addition to the static term, and formulate the temperature-dependent elastic moduli by Fuch's method [5]. Next, we calculate the temperature variation of elastic moduli for matrix Al, and compare the obtained results with observed data. Finally, we apply our formulation to obtain the concentration dependence of elastic coefficients for Al–Si and Al–Ge solid solutions.

2. Formulations

In the framework of the usual second-order perturbation based on pseudopotentials, the Helmholtz free energy $F(\Omega, x, T)$ per atom in the metallic alloys $Al_{1-x}Si_x$ and $Al_{1-x}Ge_x$ is given by

$$F(\Omega, x, T) = E(\Omega, x) + (1 - x)F_{ph}^{Al}(\Omega, x, T) + xF_{ph}^{Si \text{ or } Ge}(\Omega, x, T) \quad (1)$$

where Ω is the atomic volume at the absolute temperature T. The static crystal energy $E(\Omega, x)$ per atom is given in the virtual crystal approximation by

$$E(\Omega, x) = E_i(\Omega, x) + E^{(0)}(\Omega, x) + E^{(1)}(\Omega, x) + E^{(2)}(\Omega, x) \quad (2)$$

where the model for the pseudopotential, the dielectric screening function and the notation are the same as those in our previous work [4]. In the harmonic approximation, the lattice vibrational free energy $F_{ph}^{j}(\Omega, x, T)$ for the band (Al) and local (Si or Ge) mode is given by

$$F_{ph}^{j}(\Omega, x, T) = kT\sum_{i,\mathbf{q}} \ln\left\{2\sinh\left[\frac{h\nu_i^j(\mathbf{q}, x)}{kT}\right]\right\} \quad (3)$$

where j = Al, Si or Ge, the suffix i refers to the polarization and $\mathbf{q}$ the wave vector in the Brillouin zone. The normal vibrational modes with the wave vector $\mathbf{q}$ and the frequency $\nu_i^j(\mathbf{q}, x)$ are determined by solving the secular equation

$$|D_j^{\alpha\beta}(\mathbf{q}, x) - M_j\nu_i^j(\mathbf{q}, x)^2\delta_{\alpha,\beta}| = 0 \quad (4)$$

where the details of the model and the notation are the same as in our previous work [6, 7].

The second-order Brugger elastic constants are given as follows:

$$C_{ijkl} = \frac{1}{\Omega}\left(\frac{\partial^2 F}{\partial\eta_{ij}\partial\eta_{kl}}\right)_{\eta_{ij}=0} \quad (5)$$

The Voigt abbreviation is used to denote the Brugger elastic constants, namely $C_{ijkl} = C_{JP}$, where ij's and J's are related by 11-1, 22-2, 33-3, 23-4, 31-5 and 12-6. Fuchs [5] used three kinds of deformation parameter: v, γ_1 and ε_1. The isothermal second-order elastic constants B, C and C', which correspond to uniform volume expansion, shear deformation in one plane, and expansion and contraction with the corresponding constant cross-sectional area, respectively, are related to the derivatives of the Helmholtz free energy as follows:

$$B = \frac{1}{\Omega}\frac{d^2F}{dv^2} \quad (6)$$

$$C = \frac{1}{\Omega}\frac{d^2F}{d\gamma_1^2} \quad (7)$$

$$C' = \frac{1}{4\Omega}\frac{d^2F}{d\varepsilon_1^2} \quad (8)$$

The elastic constants have two contributions, one arising from the static crystal energy part $E(\Omega, x)$ and

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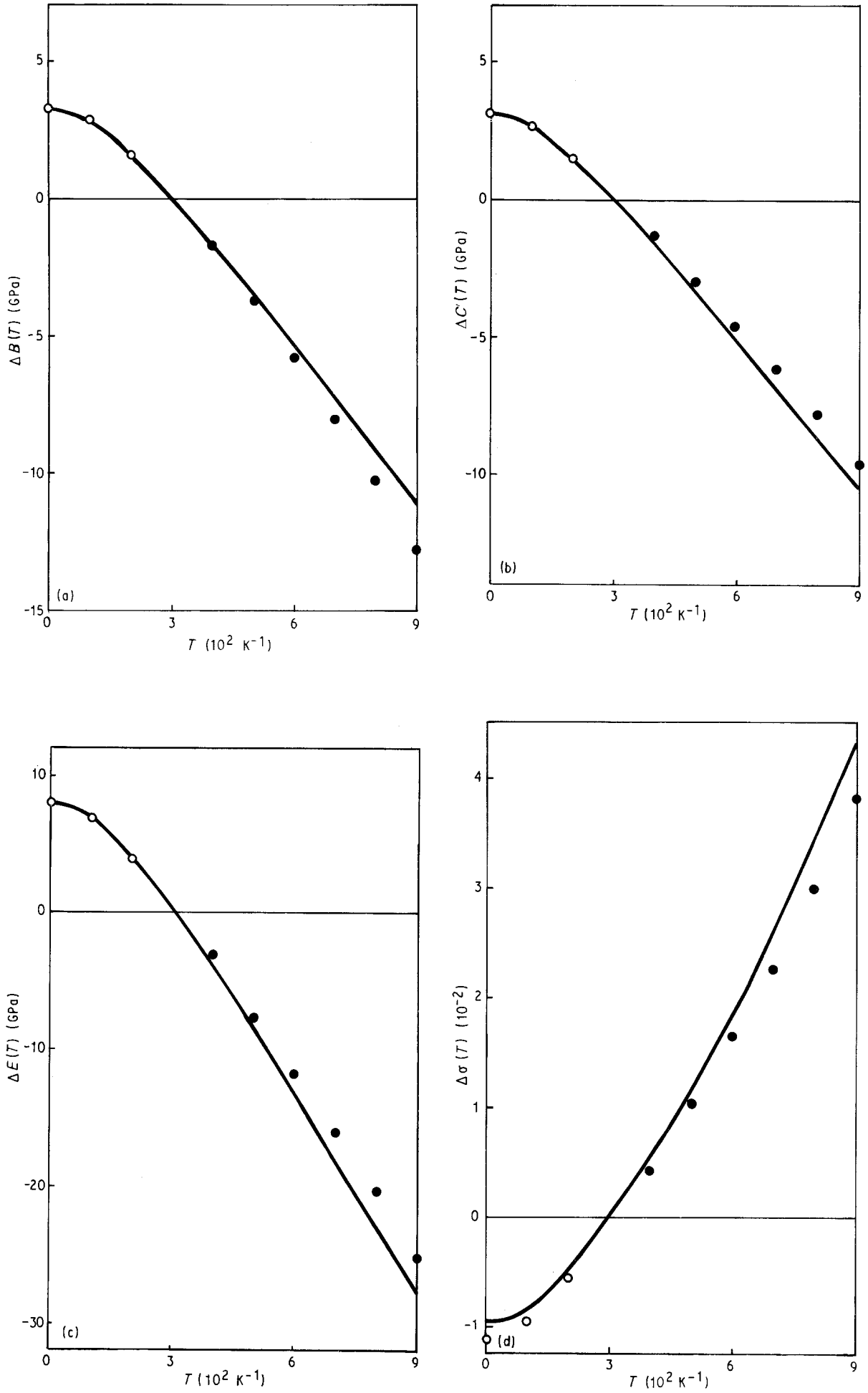

Figure 1 Temperature shift of elastic moduli $\Delta C_i(T)$ against temperature T for Al. (a) Bulk modulus; (b) shear modulus; (c) Young's modulus; (d) Poisson's ratio. ○ [10] and ● [11], observed data.

the other from the lattice vibrational part $F^{j}_{ph}(\Omega, x, T)$. The contribution from the crystal energy part to the elastic constants is calculated by the homogeneous deformation method [8]. First, considering the lattice vector component $R'_x = v^{1/3}R_x$, $R'_y = v^{1/3}R_y$, $R'_z = v^{1/3}R_z$, the static bulk modulus $B_s(\Omega, x)$ is given by

$$B_s(\Omega, x) = \Omega \frac{d^2 E(\Omega, x)}{d\Omega^2} \tag{9}$$

where the static energy E depends on the volume in three ways such as direct crystal volume Ω, reciprocal lattice vectors $\mathbf{G}$ and electron density $n(= Z/\Omega)$. Next, considering $R'_x = R_x + \gamma_1 R_y$, $R'_y = R_y$, $R'_z = R_z$, namely, $G'_x = G_x$, $G'_y = G_y - \gamma_1 G_x$, $G'_z = G_z$ in wavenumber space, where the crystal volume is maintained constant, the static shear coefficient $C_s(\Omega, x)$ is given only by contributions through reciprocal lattice vectors $\mathbf{G}$ by

$$C_s(\Omega, x) = \frac{1}{2\Omega} \sum_{\mathbf{G} \neq 0} G_x^2 \frac{\partial^2 \Phi(G, x)}{\partial G_y^2} \tag{10}$$

and

$$\Phi(G, x) = \frac{4\pi Z^2 e^2}{\Omega G^2} - \Omega \frac{V_b^{VCA}(G)^2}{\varepsilon(G)} \frac{\chi_0(G)}{1 - 4\pi e^2 f(G)\chi_0(G)/G^2} \tag{11}$$

where the notation and details are as those in our previous work [4]. Thirdly, considering $R'_x = (1 + \varepsilon_1)R_x$, $R'_y = R_y/(1 + \varepsilon_1)$, $R'_z = R_z$, namely, $G'_x = G_x/(1 + \varepsilon_1)$, $G'_y = (1 + \varepsilon_1)G_y$, $G'_z = G_z$ in wavenumber space where the crystal volume is also maintained constant, the static shear modulus $C'_S(\Omega, x)$ is given by

$$C'_S(\Omega, x) = \frac{1}{4\Omega} \sum_{\mathbf{G} \neq 0} \left[G_x^2 \frac{\partial^2 \Phi(G, x)}{\partial G_x^2} + G_y^2 \frac{\partial^2 \Phi(G, x)}{\partial G_y^2} - 2G_x G_y \frac{\partial^2 \Phi(G, x)}{\partial G_x \partial G_y} \right] \tag{12}$$

The contribution from the lattice vibrational part involves the derivatives of the normal-mode frequencies $\nu_i^j(\mathbf{q}, x)$ with respect to the deformation parameters v, γ_1, ε_1, and their calculation is complicated from first principles. In the present work, we adopt the approximation by Leibfried and Ludwig [9], where every normal-mode frequency is replaced by the square root of the second moment μ_2 of the frequency distribution function. This second moment, μ_2, is obtained by taking the trace of the dynamical matrix $D_j^{\alpha\beta}(\mathbf{q}, x)$ and by averaging over the wave vectors. Then the contribution $B_D(\Omega, x)$, $C_D(\Omega, x)$ and $C'_D(\Omega, x)$ from the lattice vibrational part is obtained from the derivatives of μ_2 with respect to Fuch's strain parameters. Using the total bulk modulus $B(\Omega, x)$ and shear modulus $C'(\Omega, x)$, we obtain Young's modulus $E(\Omega, x)$ and Poisson's ratio $\sigma(\Omega, x)$ of $Al_{1-x}Si_x$ and $Al_{1-x}Ge_x$ alloys by

$$E = \frac{9BC'}{3B + C'} \tag{13}$$

and

$$\sigma = \frac{E}{2C'} - 1 \tag{14}$$

3. Numerical results and discussion

Firstly, we show the obtained results of elastic moduli for matrix Al, and give the temperature variation $\Delta C_i(T) \equiv C_i(T) - C_i(T = 300\,\mathrm{K})$ of three elastic moduli such as bulk modulus, shear modulus, Young's modulus and Poisson's ratio in Fig. 1. The results with the Vashishta–Singwi screening function [4] are shown below. Results with other screening functions [4] are almost the same, and the numerical results for the elastic modulus C_i and its temperature-derivative $d[\ln C_i]/dT$ at $T = 300$ K for Al are summarized, together with observed data [10, 11], in Table I, where the calculated error is due to the different screening function. From Fig. 1 and Table I, we see that our obtained data of elastic moduli for matrix Al are in good agreement with the observed data [10, 11] from low to high temperatures.

Next, we calculate the elastic moduli of $Al_{1-x}Si_x$ and $Al_{1-x}Ge_x$ alloys, and show the concentration variation $\Delta C_i(x) \equiv C_i(x) - C_i(x = 0)$ of three elastic moduli and Poisson's ratio at $T = 300$ K in Fig. 2. The obtained data with various screening functions are qualitatively the same as in Fig. 2, and the numerical results about the concentration derivative $d[\ln C_i]/dx$ at $T = 300$ K and $x = 0$ for Al–Si and Al–Ge alloys are presented in Table II. From Fig. 2 and Table II, we see that all of bulk modulus, shear modulus, and Young's modulus decrease with the increase of solute Si or Ge atom and Poisson's ratio increases inversely. For $Al_{1-x}Ge_x$ alloy with the same solid solubility x, the shift of elastic moduli from matrix Al is large in comparison with that for $Al_{1-x}Si_x$ alloy. Then, the

TABLE I Elastic moduli (in GPa, except for Poisson's ratio) and its temperature derivative ($10^{-4}\,\mathrm{K}^{-1}$) at $T = 300$ K for matrix Al

		Present	Observed
C_i	B	7.95 ± 0.24	7.608[10], 7.571[11]
	C'	2.29 ± 0.10	2.302[10], 2.304[11]
	E	6.26 ± 0.21	6.273[10], 6.275[11]
	σ	0.363 ± 0.008	0.3626[10], 0.3618[11]
$\frac{d[\ln C_i]}{dT}$	B	− 2.13 ± 0.13	− 2.16[10], − 2.51[11]
	C'	− 6.96 ± 0.14	− 6.95[10], − 6.24[11]
	E	− 6.63 ± 0.32	− 6.50[10], − 5.70[11]
	σ	1.62 ± 0.14	1.65[10], 0.73[11]

TABLE II Calculated concentration derivative of elastic moduli $d[\ln C_i]/dx$ at $T = 300$ K and $x = 0$ for $Al_{1-x}Si_x$ and $Al_{1-x}Ge_x$ solid solutions

	Al–Si	Al–Ge
B	− 1.91 ± 0.12	− 2.65 ± 0.11
C'	− 1.31 ± 0.06	− 2.34 ± 0.08
E	− 1.13 ± 0.06	− 2.01 ± 0.09
σ	0.48 ± 0.06	1.02 ± 0.08

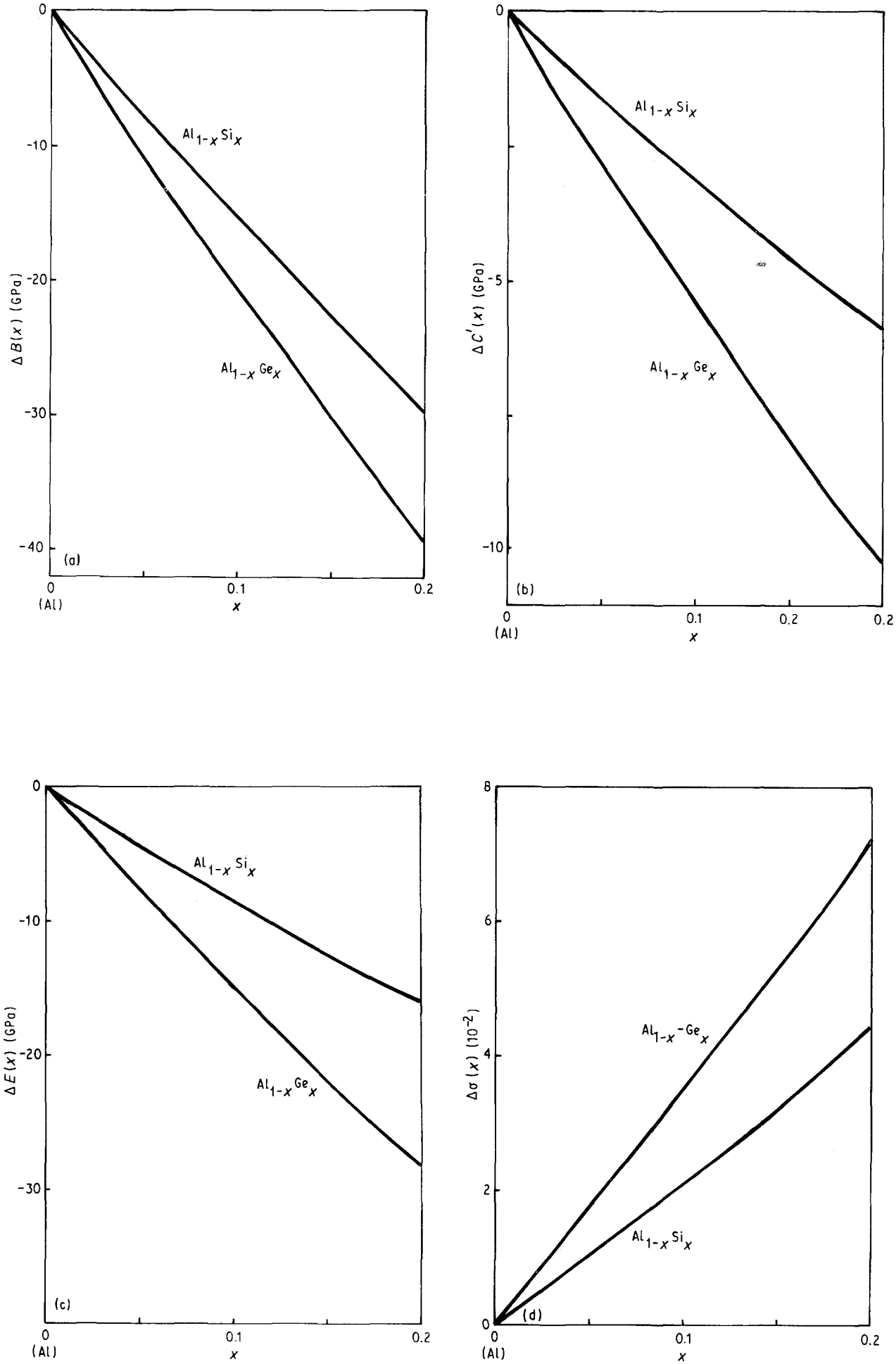

Figure 2 Concentration dependence of elastic moduli $\Delta C_i(x)$ against atomic fraction x at $T = 300$ K for $Al_{1-x}Si_x$ and $Al_{1-x}Ge_x$ solid solutions. (a) Bulk modulus, (b) shear modulus; (c) Young's modulus; (d) Poisson's ratio.

concentration derivatives $d[\ln C_i]/dx$ at $x = 0$ in Table II have a weak temperature dependence, and are decreased at $T = 900$ K by approximately 4% for Al–Si and 2% for Al–Ge system.

4. Conclusions

The elastic moduli of Al–Si and Al–Ge alloys were studied by solid-solutioning under pressure, and the concentration dependence of elastic moduli were predicted for these solid solutions from first principles, based on the microscopic electronic theory. The data obtained are useful in studying the mechanical and thermal properties of Al–Si and Al–Ge systems, and further experimental research in this field could be useful.

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