

Potential energies of characteristic atoms on basis of experimental heats of formation of AuCu and AuCu₃ compounds (II)

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Received 30 March 2009; accepted 2 June 2009

Abstract: The potential energy sequences of characteristic atoms were separated out by nine potential energy E -functions on the basis of larger experimental heats of formation of the $L1_0$ -AuCu and $L1_2$ -AuCu₃ compounds only. According to these potential energy sequences of characteristic atoms, the potential energies and heats of formation of disordered $Au_{1-x}Cu_x$ alloys were calculated by corresponding E -functions at 0 K; and the potential energies, heats of formation and critical T_c -temperatures of the order-disorder transitions of $L1_0$ -AuCu, $L1_2$ -Au₃Cu and $L1_2$ -AuCu₃ compounds, Au₃Cu-, AuCu- and AuCu₃-type ordered alloys with maximal ordering degrees were also calculated at 0 K. The results obtained by both the first and present parts of this investigation were compared. Comparing the results obtained by nine E -functions, the 5th $E(x, 0, \sigma)$ function may be chosen for describing thermodynamic properties of the compounds, ordered and disordered phases and for establishing the phase diagram of the Au-Cu system in the future.

Key words: systematic science of alloys; Au-Cu system; potential energy; heat of formation; phase transition temperature

1 Introduction

In the energetic aspect, the systematic science of alloys(SSA) is a framework of the total energy able to be separated out. It can not only accurately describe phase regions in the phase diagram, but also precisely describe the variations in thermodynamic properties, such as general vibration heat capacity $C_p^v(x, T, \sigma)$, general vibration energy $U^v(x, T, \sigma)$, general vibration entropy $S^v(x, T, \sigma)$, enthalpy $H(x, T, \sigma)$, characteristic Gibbs energy $G^*(x, T, \sigma)$ without containing configuration entropy and Gibbs energy $G(x, T, \sigma)$ containing configuration entropy $S^c(x, \sigma)$ as functions of composition, temperature and ordering degree, within the phase regions. Therefore, it is very important to separate out potential energy sequences of characteristic atoms on the basis of accurate heats of formation of several compounds in the alloy system.

In the first part of this investigation, the potential energy sequences of the A_i^{Au} and A_i^{Cu} characteristic atoms in Au-Cu system were separated out respectively by nine E -functions on the basis of smaller experimental heats of formation of the AuCu and AuCu₃ compounds: $\Delta H_{exp}(AuCu, 298, 1) = -8746$ J/mol and $\Delta H_{exp}(AuCu_3, 298, 1) = -7164$ J/mol[1–2]. According to these potential energy sequences of the A_i^{Au} and A_i^{Cu} characteristic atoms, the potential energies $E(x, 0, 0)$ and heats of formation $\Delta H(x, 0, 0)$ of disordered $Au_{1-x}Cu_x$ alloys, the potential energies, heats of formation and order-disorder transition T_c -temperatures of Au₃Cu, AuCu and AuCu₃ compound as well as the Au₃Cu-, AuCu- and AuCu₃-type ordered alloys with maximal ordering degree were calculated. And finally, the 5th E -function was determined to be suitable for describing Au-Cu system.

In the present part of this investigation, the potential energy sequences of the A_i^{Au} and A_i^{Cu} characteristic atoms in Au-Cu system were separated out respectively by

nine E -functions on the basis of larger experimental heats of formation of the AuCu and AuCu₃ compounds: $\Delta H_{\text{exp}}(\text{AuCu}, 298, 1) = -9\,337 \text{ J/mol}$ and $\Delta H_{\text{exp}}(\text{AuCu}_3, 298, 1) = -7\,268 \text{ J/mol}$ [3–4]. According to these potential energy sequences of the A_i^{Au} and A_i^{Cu} characteristic atoms, the potential energies $E(x, 0, 0)$, heats of formation $\Delta H(x, 0, 0)$ of disordered Au_{1-x}Cu_x alloys, the potential energies, heats of formation and order-disorder transition T_c -temperatures of Au₃Cu, AuCu and AuCu₃ compound as well as the Au₃Cu-, AuCu- and AuCu₃-type ordered alloys with maximal ordering degree were calculated. And finally, the 5th E -function was still determined to be suitable for describing Au-Cu system.

2 Results

2.1 Potential energy sequences of characteristic atoms

On the basis of larger experimental heats of formation of the $L1_0$ -AuCu and $L1_2$ -AuCu₃ compounds, the potential energy $E_i^{\text{Au}}(0)$ and $E_i^{\text{Cu}}(0)$ sequences of the A_i^{Au} and A_i^{Cu} characteristic atoms in the Au-Cu system are separated out by nine E -functions, listed in Table 1 and shown in Fig.1, from which it can be known that the potential energy sequences obtained from nine E -functions are different each other, and different to ones obtained from the first part of this investigation.

2.2 Average potential energies $E(x, 0, 0)$, $E_{\text{Au}}(x, 0, 0)$ and $E_{\text{Cu}}(x, 0, 0)$ of disordered Au_{1-x}Cu_x alloys and their components at 0 K

According to the potential energy sequences of the characteristic atoms, the $E(x, 0, 0)$, $E_{\text{Au}}(x, 0, 0)$ and $E_{\text{Cu}}(x, 0, 0)$ potential energies of disordered Au_{1-x}Cu_x alloys and their components as well as their $\Delta H(x, 0, 0)$ heats of formation calculated by nine E -functions are listed in Table 2 and shown in Fig.2, from which the following knowledge can be obtained.

1) The average $E(x, 0, 0)$ potential energies of the disordered Au_{1-x}Cu_x alloys obtained from the nine functions are different each other. The average $E_{\text{Au}}(x, 0, 0)$ and $E_{\text{Cu}}(x, 0, 0)$ potential energies of the Au and Cu components obtained by nine E -functions are different each other too. But there is no E -function, which can well describe the experimental heats of formation in the whole compositional range. It has been discovered that the experimental heats of formation of so-called disordered alloys are referred to partly-ordered alloys with short-range ordering(SRO) degrees[6–7]. The effect of ordering degrees on the heats of formation of the alloy phases are discussed in Section 3.

2) The heats of formation of the disordered

Au_{1-x}Cu_x alloys obtained by the 5th E -function are higher than corresponding experimental values in the compositional range $25\% \leq x(\text{Cu}) \leq 75\%$. But it may be used to describe the Au-Cu system, if the experimental heats of formation of disordered alloys are referred to ones of partly-ordered alloys with SRO degrees.

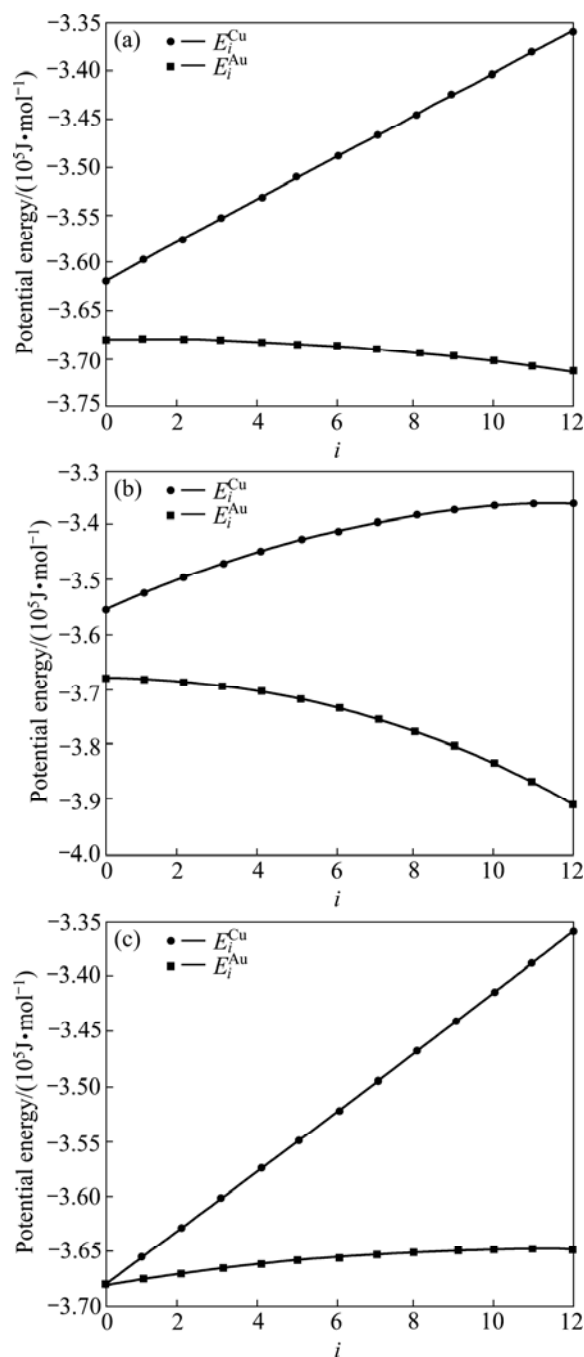


Fig.1 Potential energy $E_i^{\text{Au}}(0)$ and $E_i^{\text{Cu}}(0)$ sequences of A_i^{Au} and A_i^{Cu} characteristic atoms in Au-Cu system ($E_i^{\text{Au}}(0)$ and $E_i^{\text{Cu}}(0)$ sequences obtained by the 4th (a), 5th (b) and 7th (c) E -functions are different, even average potential energies of compounds, ordered and disordered alloys calculated by them are, respectively, equivalent (see results of 2.2, 2.3 and 2.4 in the present section))

Table 1 Potential energy sequences of A_i^{Au} and A_i^{Cu} characteristic atoms in Au-Cu system calculated by nine E -functions at 0 K (J/mol)

No.	i	0	1	2	3	4	5	6
1**	E_i^{Au}	-368 000*	-369 183	-370 366	-371 549	-372 732	-373 915	-375 098
	E_i^{Cu}	-350 196	-349 013	-347 830	-346 647	-345 464	-344 281	-343 098
2	E_i^{Au}	-368 000	-370 511	-373 022	-375 533	-378 044	-380 555	-383 067
	E_i^{Cu}	-332 817	-333 325	-333 790	-334 210	-334 585	-334 917	-335 204
3	E_i^{Au}	-368 000	-369 981	-371 961	-373 942	-375 922	-377 903	-379 884
	E_i^{Cu}	-339 183	-339 161	-339 095	-338 984	-338 829	-338 630	-338 387
4	E_i^{Au}	-368 000	-368 022	-368 088	-368 199	-368 354	-368 553	-368 796
	E_i^{Cu}	-361 889	-359 732	-357 574	-355 417	-353 259	-351 102	-348 945
5	E_i^{Au}	-368 000	-368 157	-368 628	-369 412	-370 511	-371 924	-373 650
	E_i^{Cu}	-355 417	-352 315	-349 484	-346 922	-344 630	-342 607	-340 854
6	E_i^{Au}	-368 000	-367 752	-367 010	-365 772	-364 039	-361 811	-359 087
	E_i^{Cu}	-374 834	-374 564	-373 755	-372 406	-370 519	-368 092	-365 125
7	E_i^{Au}	-368 000	-367 492	-367 027	-366 607	-366 232	-365 900	-365 613
	E_i^{Cu}	-368 255	-365 567	-362 879	-360 191	-357 503	-354 815	-352 128
8	E_i^{Au}	-368 000	-375 219	-381 811	-387 775	-393 111	-397 819	-401 900
	E_i^{Cu}	-287 618	-295 345	-302 401	-308 785	-314 497	-319 537	-323 904
9	E_i^{Au}	-368 000	-369 424	-370 723	-371 899	-372 951	-373 880	-374 684
	E_i^{Cu}	-348 096	-348 012	-347 760	-347 340	-346 752	-345 996	-345 072

No.	i	7	8	9	10	11	12
1**	E_i^{Au}	-376 281	-377 464	-378 647	-379 830	-381 013	-382 196
	E_i^{Cu}	-341 915	-340 732	-339 549	-338 366	-337 183	-336 000*
2	E_i^{Au}	-385 578	-388 089	-390 600	-393 111	-395 622	-398 133
	E_i^{Cu}	-335 447	-335 646	-335 801	-335 912	-335 978	-336 000
3	E_i^{Au}	-381 864	-383 845	-385 825	-387 806	-389 786	-391 767
	E_i^{Cu}	-338 100	-337 768	-337 393	-336 973	-336 508	-336 000
4	E_i^{Au}	-369 083	-369 415	-369 790	-370 210	-370 675	-371 183
	E_i^{Cu}	-346 787	-344 630	-342 472	-340 315	-338 157	-336 000
5	E_i^{Au}	-375 690	-378 044	-380 712	-383 694	-386 990	-390 600
	E_i^{Cu}	-339 371	-338 157	-337 214	-336 539	-336 135	-336 000
6	E_i^{Au}	-355 869	-352 155	-347 947	-343 243	-338 044	-332 350
	E_i^{Cu}	-361 619	-357 574	-352 990	-347 866	-342 203	-336 000
7	E_i^{Au}	-365 370	-365 171	-365 016	-364 905	-364 839	-364 817
	E_i^{Cu}	-349 440	-346 752	-344 064	-341 376	-338 688	-336 000
8	E_i^{Au}	-405 352	-408 177	-410 375	-411 944	-412 886	-413 200
	E_i^{Cu}	-327 600	-330 624	-332 976	-334 656	-335 664	-336 000
9	E_i^{Au}	-375 365	-375 922	-376 356	-376 665	-376 851	-376 913
	E_i^{Cu}	-343 980	-342 720	-341 292	-339 696	-337 932	-336 000

* The values are taken from Ref.[5]; ** Up till now, we cannot give a reasonable method to separate the potential energies of characteristic atoms by the first function. Therefore, it cannot be used at present.

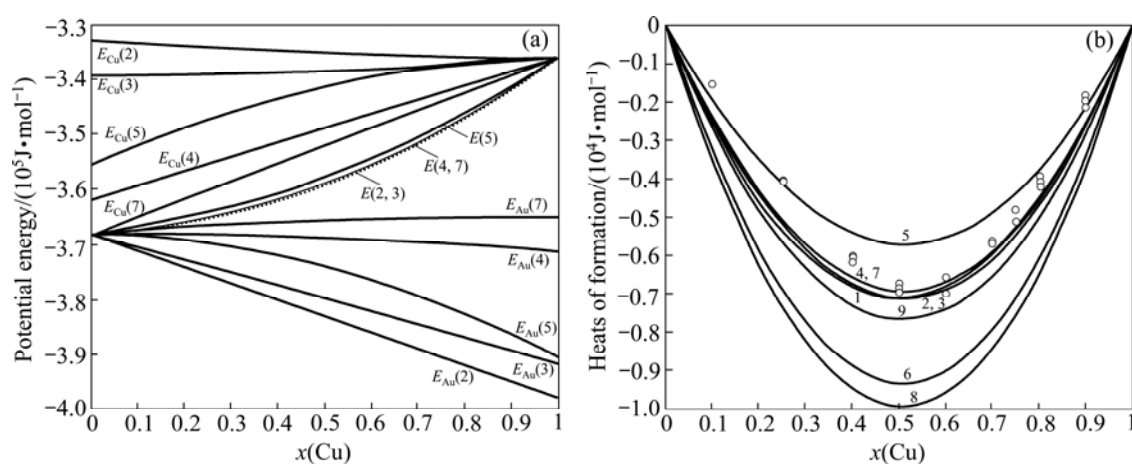


Fig.2 Average potential energies $E(x, 0, 0)$, $E_{Au}(x, 0, 0)$ and $E_{Cu}(x, 0, 0)$ of disordered $Au_{1-x}Cu_x$ alloys and their components calculated from the 2nd, 3rd, 4th, 5th and 7th E -functions (a); and heats of formation $\Delta H(x, 0, 0)$ for disordered $Au_{1-x}Cu_x$ alloys calculated from nine E -functions, together with experimental heats of formation at 320 K [4] (denoted by circles) (b)

Table 2 Potential energies $E(x, 0, 0)$, $E_{Au}(x, 0, 0)$, $E_{Cu}(x, 0, 0)$ and heats of function $\Delta H(x, 0, 0)$ of disordered $Au_{1-x}Cu_x$ alloys calculated by nine E -functions at 0 K (J/mol)

No.		$x(Cu)$						
		0	0.1	0.2	0.25	0.3	0.4	0.5
1	$E_{Au}(x)$	-368 000	-369 420	-370 839	-371 549	-372 259	-373 679	-375 098
	$E_{Cu}(x)$	-350 196	-348 777	-347 357	-346 647	-345 938	-344 518	-343 098
	$E(x)$	-368 000	-367 355	-366 143	-365 324	-364 363	-362 014	-359 098
	$\Delta H(x)$	0	-2 555	-4 543	-5 324	-5 963	-6 814	-7 098
2	$E_{Au}(x)$	-368 000	-371 013	-374 027	-375 533	-377 040	-380 053	-383 067
	$E_{Cu}(x)$	-332 817	-333 398	-333 920	-334 160	-334 385	-334 790	-335 138
	$E(x)$	-368 000	-367 252	-366 005	-365 190	-364 243	-361 948	-359 102
	$\Delta H(x)$	0	-2 452	-4 405	-5 190	-5 843	-6 748	-7 102
3	$E_{Au}(x)$	-368 000	-370 377	-372 753	-373 942	-375 130	-377 507	-379 884
	$E_{Cu}(x)$	-339 183	-339 127	-339 013	-338 934	-338 841	-338 610	-338 321
	$E(x)$	-368 000	-367 252	-366 005	-365 190	-364 243	-361 948	-359 102
	$\Delta H(x)$	0	-2 452	-4 405	-5 190	-5 843	-6 748	-7 102
4	$E_{Au}(x)$	-368 000	-368 056	-368 170	-368 249	-368 342	-368 573	-368 862
	$E_{Cu}(x)$	-361 889	-359 300	-356 711	-355 417	-354 122	-351 533	-348 945
	$E(x)$	-368 000	-367 180	-365 878	-365 041	-364 076	-361 757	-358 903
	$\Delta H(x)$	0	-2 380	-4 278	-5 041	-5 676	-6 557	-6 903
5	$E_{Au}(x)$	-368 000	-368 395	-369 205	-369 766	-370 429	-372 068	-374 121
	$E_{Cu}(x)$	-355 417	-351 873	-348 686	-347 225	-345 854	-343 378	-341 259
	$E(x)$	-368 000	-366 743	-365 101	-364 131	-363 057	-360 592	-357 690
	$\Delta H(x)$	0	-1 943	-3 501	-4 131	-4 657	-5 392	-5 690
6	$E_{Au}(x)$	-368 000	-367 376	-366 099	-365 215	-364 168	-361 583	-358 345
	$E_{Cu}(x)$	-374 834	-374 154	-372 762	-371 800	-370 659	-367 843	-364 316
	$E(x)$	-368 000	-368 054	-367 431	-366 861	-366 115	-364 087	-361 330
	$\Delta H(x)$	0	-3 254	-5 831	-6 861	-7 715	-8 887	-9 330
7	$E_{Au}(x)$	-368 000	-367 419	-366 897	-366 657	-366 432	-366 027	-365 679
	$E_{Cu}(x)$	-368 255	-365 029	-361 804	-360 191	-358 579	-355 353	-352 128
	$E(x)$	-368 000	-367 180	-365 878	-365 041	-364 076	-361 757	-358 903
	$\Delta H(x)$	0	-2 380	-4 278	-5 041	-5 676	-6 557	-6 903

Continue

No.		$x(\text{Cu})$						
		0	0.1	0.2	0.25	0.3	0.4	0.5
8	$E_{\text{Au}}(x)$	−368 000	−376 249	−383 669	−387 069	−390 261	−396 024	−400 958
	$E_{\text{Cu}}(x)$	−287 618	−296 447	−304 390	−308 029	−311 446	−317 615	−322 896
	$E(x)$	−368 000	−368 269	−367 813	−367 309	−366 616	−364 660	−361 927
	$\Delta H(x)$	0	−3 469	−6 213	−7 309	−8 216	−9 460	−9 927
9	$E_{\text{Au}}(x)$	−368 000	−369 627	−371 090	−371 760	−372 389	−373 526	−374 499
	$E_{\text{Cu}}(x)$	−348 096	−347 884	−347 451	−347 151	−346 795	−345 918	−344 820
	$E(x)$	−368 000	−367 452	−366 362	−365 608	−364 711	−362 483	−359 659
	$\Delta H(x)$	0	−2 652	−4 762	−5 608	−6 311	−7 283	−7 659
No.		$x(\text{Cu})$						
		0.6	0.7	0.75	0.8	0.9	1	
1	$E_{\text{Au}}(x)$	−376 518	−377 938	−378 647	−379 357	−380 777	−382 196	
	$E_{\text{Cu}}(x)$	−341 679	−340 259	−339 549	−338 839	−337 420	−336 000	
	$E(x)$	−355 614	−351 563	−349 324	−346 943	−341 755	−336 000	
	$\Delta H(x)$	−6 814	−5 963	−5 324	−4 543	−2 555	0	
2	$E_{\text{Au}}(x)$	−386 080	−389 093	−390 600	−392 106	−395 120	−398 133	
	$E_{\text{Cu}}(x)$	−335 427	−335 658	−335 751	−335 830	−335 944	−336 000	
	$E(x)$	−355 688	−351 688	−349 463	−347 085	−341 862	−336 000	
	$\Delta H(x)$	−6 888	−6 088	−5 463	−4 685	−2 662	0	
3	$E_{\text{Au}}(x)$	−382 260	−384 637	−385 825	−387 014	−389 390	−391 767	
	$E_{\text{Cu}}(x)$	−337 973	−337 568	−337 343	−337 103	−336 581	−336 000	
	$E(x)$	−355 688	−351 688	−349 463	−347 085	−341 862	−336 000	
	$\Delta H(x)$	−6 888	−6 088	−5 463	−4 685	−2 662	0	
4	$E_{\text{Au}}(x)$	−369 210	−369 615	−369 840	−370 080	−370 602	−371 183	
	$E_{\text{Cu}}(x)$	−346 356	−343 767	−342 472	−341 178	−338 589	−336 000	
	$E(x)$	−355 497	−351 521	−349 314	−346 958	−341 790	−336 000	
	$\Delta H(x)$	−6 697	−5 921	−5 314	−4 558	−2 590	0	
5	$E_{\text{Au}}(x)$	−376 588	−379 469	−381 065	−382 765	−386 475	−390 600	
	$E_{\text{Cu}}(x)$	−339 495	−338 087	−337 517	−337 036	−336 340	−336 000	
	$E(x)$	−354 332	−350 502	−348 404	−346 181	−341 353	−336 000	
	$\Delta H(x)$	−5 532	−4 902	−4 404	−3 781	−2 153	0	
6	$E_{\text{Au}}(x)$	−354 453	−349 907	−347 390	−344 708	−338 856	−332 350	
	$E_{\text{Cu}}(x)$	−360 077	−355 125	−352 383	−349 462	−343 087	−336 000	
	$E(x)$	−357 827	−353 560	−351 135	−348 511	−342 664	−336 000	
	$\Delta H(x)$	−9 027	−7 960	−7 135	−6 111	−3 464	0	
7	$E_{\text{Au}}(x)$	−365 390	−365 159	−365 066	−364 987	−364 873	−364 817	
	$E_{\text{Cu}}(x)$	−348 902	−345 676	−344 064	−342 451	−339 226	−336 000	
	$E(x)$	−355 497	−351 521	−349 314	−346 958	−341 790	−336 000	
	$\Delta H(x)$	−6 697	−5 921	−5 314	−4 558	−2 590	0	
8	$E_{\text{Au}}(x)$	−405 064	−408 341	−409 668	−410 789	−412 409	−413 200	
	$E_{\text{Cu}}(x)$	−327 291	−330 799	−332 220	−333 420	−335 153	−336 000	
	$E(x)$	−358 400	−354 061	−351 582	−348 893	−342 879	−336 000	
	$\Delta H(x)$	−9 600	−8 461	−7 582	−6 493	−3 679	0	
9	$E_{\text{Au}}(x)$	−375 308	−375 955	−376 216	−376 437	−376 757	−376 913	
	$E_{\text{Cu}}(x)$	−343 499	−341 957	−341 103	−340 193	−338 207	−336 000	
	$E(x)$	−356 223	−352 156	−349 881	−347 442	−342 062	−336 000	
	$\Delta H(x)$	−7 423	−6 556	−5 881	−5 042	−2 862	0	

2.3 Potential energies and heats of formation of $L1_2$ - Au_3Cu , $L1_0$ - $AuCu$ and $L1_2$ - $AuCu_3$ compounds

According to the potential energy sequences of the characteristic atoms in Table 1, the potential energies ε_i^{Au} and ε_i^{Cu} of characteristic atoms occupied at the i th site in the cell and the average potential energies, heats of formation and order-disorder transition temperatures of the $L1_2$ - Au_3Cu , $L1_0$ - $AuCu$ and $L1_2$ - $AuCu_3$ compounds can be calculated by the additive law of properties of characteristic crystals and are listed in Table 3, from which the following knowledge can be obtained.

1) The average potential energies of these compounds obtained from the 2nd to 9th E -functions are equal to each other, but their potential energies of characteristic atoms are different each other.

2) The average potential energies and heats of formation of the $L1_0$ - $AuCu$ and $L1_2$ - $AuCu_3$ compound obtained by the 5th E -function are equal to corresponding experimental values, and their T_c -temperatures are closer by experiment values than ones obtained by other E -functions.

3) The average potential energies and heats of formation of the $L1_2$ - Au_3Cu compound calculated from the 2nd to 9th E -functions are equal to each other and closer by experiment values, but only the T_c -temperature obtained by the 5th E -function is closer by the experiment value than ones obtained by other E -functions.

2.4 Energetic properties of ordered $Au_{1-x}Cu_x$ alloys

According to the potential energy sequences and concentrations $x_i^{Au}(x, \sigma)$ and $x_i^{Cu}(x, \sigma)$ in the ordered $Au_{1-x}Cu_x$ alloys with ordering degree σ , their energetic properties can be calculated at 0 K. The average potential energies $E(x, 0, \sigma_{max})$ and heats of formation $\Delta H(x, 0, \sigma_{max})$ of the Au_3Cu -, $AuCu$ - and $AuCu_3$ -type ordered $Au_{1-x}Cu_x$ alloys with maximal ordering degree are listed in Tables 4–6. Their heats of formation $\Delta H(x, 0, \sigma_{max})$ calculated by nine E -functions are shown in Fig.3.

2.5 $T_c(x, \sigma_{max})$ temperatures of order-disorder phase transformations

The potential energy differences, $E_{Au_3Cu}(x, 0, \sigma_{max}) - E_{dis}(x, 0, 0)$, $E_{AuCu}(x, 0, \sigma_{max}) - E_{dis}(x, 0, 0)$ and $E_{AuCu_3}(x, 0, \sigma_{max}) - E_{dis}(x, 0, 0)$ between the ordered and disordered alloys, and the critical temperatures $T_c(x, \sigma_{max})$ of Au_3Cu -, $AuCu$ -, and $AuCu_3$ -type $Au_{1-x}Cu_x$ alloys translated into the disordered alloys described by nine

Table 3 Potential energies of characteristic atoms (ε , in 10^{-19} J/atom), total potential energies (E , in J/mol), heat of formation (ΔH , in J/mol) and order-disorder transformation temperatures (T_c , in K) of $L1_2$ - Au_3Cu , $L1_0$ - $AuCu$ and $L1_2$ - $AuCu_3$ compounds at 0 K

No.	$L1_2$ - Au_3Cu				
	ε_4^{Au}	ε_0^{Cu}	E	ΔH	T_c
1	-6.187 66	-5.813 55	-367 098	-7 098	379.55
2	-6.275 84	-5.525 03	-366 738	-6 738	331.00
3	-6.240 62	-5.630 71	-366 738	-6 738	331.00
4	-6.114 97	-6.007 65	-366 738	-6 738	362.92
5	-6.150 79	-5.900 21	-366 738	-6 738	557.58
6	-6.043 34	-6.222 54	-366 738	-6 738	-26.42
7	-6.079 74	-6.113 33	-366 738	-6 738	362.92
8	-6.525 96	-4.774 69	-366 738	-6 738	-122.15
9	-6.191 30	-5.778 67	-366 738	-6 738	241.65
Exp.			-365 736 ^a	-5 736 ^b	473 ^c

No.	$L1_0$ - $AuCu$				
	ε_8^{Au}	ε_4^{Cu}	E	ΔH	T_c
1	-6.26 622	-5.73 499	-361 464	-9 464	410.56
2	-6.44 259	-5.55 439	-361 337	-9 337	387.77
3	-6.37 213	-5.62 484	-361 337	-9 337	387.77
4	-6.13 259	-5.86 439	-361 337	-9 337	422.29
5	-6.27 584	-5.72 113	-361 337	-9 337	632.86
6	-5.84 607	-6.15 091	-361 337	-9 337	1.15
7	-6.06 213	-5.93 485	-361 337	-9 337	422.29
8	-6.77 608	-5.22 090	-361 337	-9 337	-102.41
9	-6.24 062	-5.75 636	-361 337	-9 337	291.12
Exp.			-361 337 ^a	-9 337 ^b	683 ^c

No.	$L1_2$ - $AuCu_3$				
	ε_{12}^{Au}	ε_8^{Cu}	E	ΔH	T_c
1	-6.344 77	-5.656 43	-351 098	-7 098	379.55
2	-6.609 33	-5.572 00	-351 268	-7 268	385.96
3	-6.503 65	-5.607 23	-351 268	-7 268	385.96
4	-6.161 94	-5.721 13	-351 268	-7 268	417.88
5	-6.484 27	-5.613 69	-351 268	-7 268	612.54
6	-5.517 27	-5.936 02	-351 268	-7 268	28.54
7	-6.056 26	-5.756 36	-351 268	-7 268	417.88
8	-6.859 45	-5.488 63	-351 268	-7 268	-67.19
9	-6.257 06	-5.689 43	-351 268	-7 268	296.61
Exp.			-351 268 ^a	-7 268 ^b	663 ^c

“a” means data calculated from experimental cohesive energy of elemental solids[5] and ΔH [3–4]; “b” means data taken from Refs.[3–4]; “c” means data taken from Ref.[8].

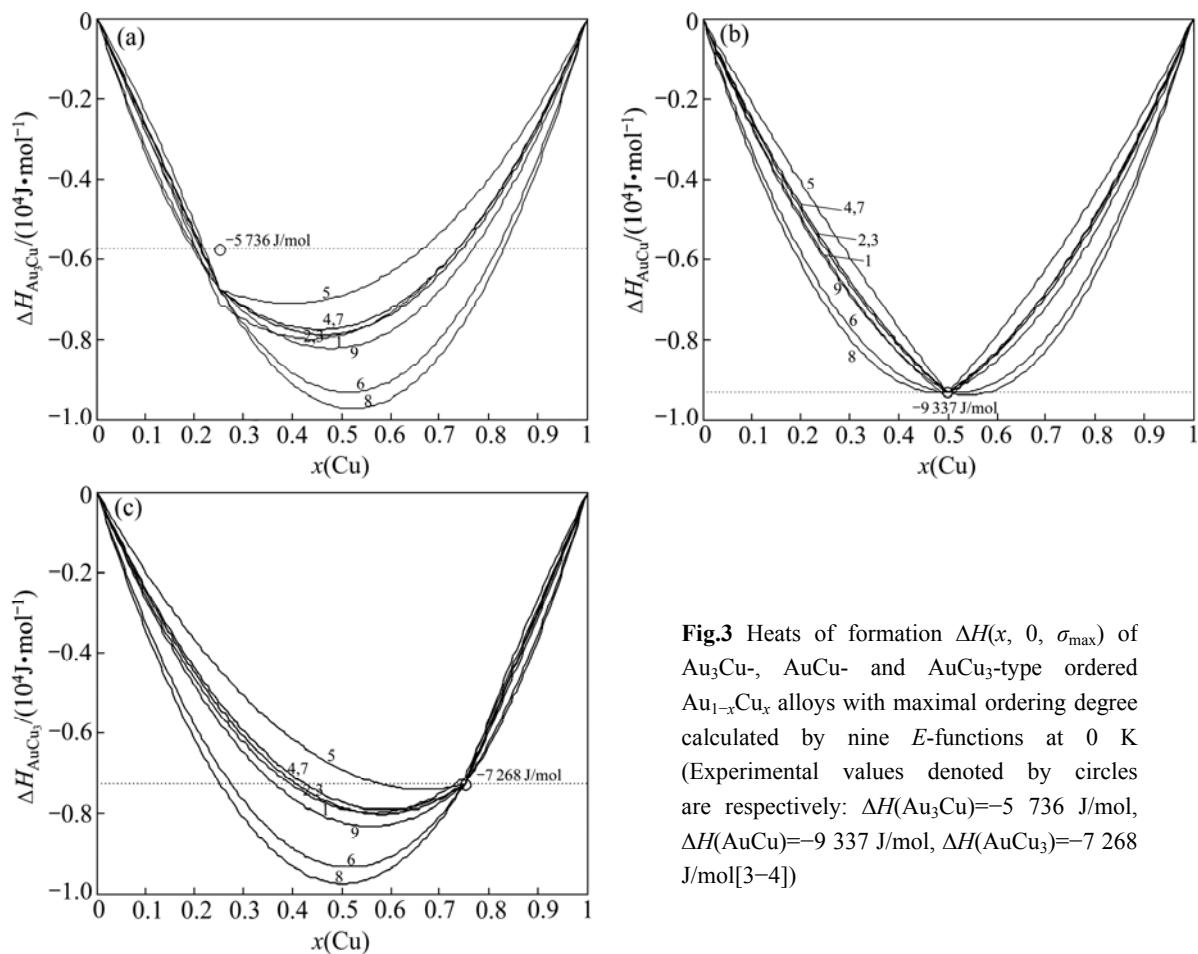


Fig.3 Heats of formation $\Delta H(x, 0, \sigma_{\max})$ of Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with maximal ordering degree calculated by nine E -functions at 0 K (Experimental values denoted by circles are respectively: $\Delta H(\text{Au}_3\text{Cu}) = -5\,736$ J/mol, $\Delta H(\text{AuCu}) = -9\,337$ J/mol, $\Delta H(\text{AuCu}_3) = -7\,268$ J/mol[3–4])

E -functions have been calculated. The results are shown in Fig.4. The T_c -temperatures of these compounds described by the 5th E -function are closer by the experimental values than ones by other E -functions.

2.6 $\Delta H_{\text{ord}}(0) - \Delta H_{\text{dis}}(0) - T_c$ interrelated patterns

Here, we have made a comparison for the $\Delta H_{\text{ord}}(0) - \Delta H_{\text{dis}}(0) - T_c$ interrelated patterns of the AuCu , AuCu_3 and Au_3Cu compounds relative to corresponding disordered alloys obtained respectively in the first part and in the present part of this investigation. From Fig.5 the following knowledge can be obtained.

1) For the AuCu compound, although $\Delta H_{\text{ord}}(\text{AuCu}, 0, 1) = -9\,337$ J/mol of the AuCu compound obtained from the 2nd–9th E -functions in the present part are equivalent each other, $\Delta H_{\text{dis}}(\text{AuCu}, 0, 0) = -5\,690$ J/mol obtained from the 5th E -function is higher (smallest absolute value) than that obtained from other E -functions. Therefore, the order-disorder transition temperature $T_c(\text{AuCu}) = 633$ K obtained from the 5th E -function is the highest (see in Fig.5(a)). These results are, respectively, larger than $\Delta H_{\text{ord}}(\text{AuCu}, 0, 1) = -8\,746$ J/mol, $\Delta H_{\text{dis}}(\text{AuCu}, 0, 0) = -5\,330$ J/mol and $T_c(\text{AuCu}) =$

593 K obtained in the first part of this investigation (see in Fig.5(a')).

2) For the AuCu_3 compound, there are analogous results to ones of the AuCu compounds. Although the $\Delta H_{\text{ord}}(\text{AuCu}_3, 0, 1) = -7\,268$ J/mol obtained from the 2nd–9th E -functions in the present part are equivalent each other, $\Delta H_{\text{dis}}(\text{AuCu}_3, 0, 0) = -4\,404$ J/mol and $T_c(\text{AuCu}_3) = 613$ K obtained from the 5th E -function are the highest (see in Fig.5(b)). These results are, respectively, larger than $\Delta H_{\text{ord}}(\text{AuCu}_3, 0, 1) = -7\,164$ J/mol, $\Delta H_{\text{dis}}(\text{AuCu}_3, 0, 0) = -4\,309$ J/mol and $T_c(\text{AuCu}_3) = 611$ K obtained from the 5th E -function in the first part of this investigation (see in Fig.5(b')).

3) For the Au_3Cu compound, there are analogous results to ones for the AuCu and AuCu_3 compounds. Although $\Delta H_{\text{ord}}(\text{Au}_3\text{Cu}, 0, 1) = -6\,738$ J/mol obtained from the 2nd–9th E -functions in the present part are equivalent each other, the $\Delta H_{\text{dis}}(\text{Au}_3\text{Cu}, 0, 0) = -6\,738$ J/mol and $T_c(\text{Au}_3\text{Cu}) = 558$ K obtained from the 5th E -function are the highest (see in Fig.5(c)). These results are, respectively, larger than $\Delta H_{\text{ord}}(\text{Au}_3\text{Cu}, 0, 1) = -5\,956$ J/mol, $\Delta H_{\text{dis}}(\text{Au}_3\text{Cu}, 0, 0) = -3\,686$ J/mol and $T_c(\text{Au}_3\text{Cu}) = 485$ K obtained from the 5th E -function in the first part of this investigation (see in Fig.5(c')).

Table 4 Potential energies $E(x, 0, \sigma_{\max})$ and heats of formation $\Delta H(x, 0, \sigma_{\max})$ of Au_3Cu -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with maximal ordering degrees calculated by nine E -functions at 0 K (J/mol)

No.		x(Cu)						
		0	0.1	0.2	0.25	0.3	0.4	0.5
1	E(x)	-368 000	-367 639	-367 279	-367 098	-365 908	-363 150	-359 887
	ΔH(x)	0	-2 839	-5 679	-7 098	-7 508	-7 950	-7 887
2	E(x)	-368 000	-367 495	-366 990	-366 738	-365 609	-362 977	-359 835
	ΔH(x)	0	-2 695	-5 390	-6 738	-7 209	-7 777	-7 835
3	E(x)	-368 000	-36 7495	-366 990	-366 738	-365 609	-362 977	-359 835
	ΔH(x)	0	-2 695	-5 390	-6 738	-7 209	-7 777	-7 835
4	E(x)	-368 000	-367 447	-366 958	-366 738	-365 572	-362 882	-359 702
	ΔH(x)	0	-2 647	-5 358	-6 738	-7 172	-7 682	-7 702
5	E(x)	-368 000	-367 156	-366 764	-366 738	-365 345	-362 299	-358 893
	ΔH(x)	0	-2 356	-5 164	-6 738	-6 945	-7 099	-6 893
6	E(x)	-368 000	-368 030	-367 347	-366 738	-366 025	-364 047	-361 320
	ΔH(x)	0	-3 230	-5 747	-6 738	-7 625	-8 847	-9 320
7	E(x)	-368 000	-367 447	-366 958	-366 738	-365 572	-362 882	-359 702
	ΔH(x)	0	-2 647	-5 358	-6 738	-7 172	-7 682	-7 702
8	E(x)	-368 000	-368 173	-367 442	-366 738	-366 136	-364 333	-361 718
	ΔH(x)	0	-3 373	-5 842	-6 738	-7 736	-9 133	-9 718
9	E(x)	-368 000	-367 629	-367 079	-366 738	-365 713	-363 245	-360 206
	ΔH(x)	0	-2 829	-5 479	-6 738	-7 313	-8 045	-8 206

No.		x(Cu)					
		0.6	0.7	0.75	0.8	0.9	1
1	E(x)	-356 119	-351 846	-349 521	-347 069	-341 787	-336 000
	ΔH(x)	-7 319	-6 246	-5 521	-4 669	-2 587	0
2	E(x)	-356 169	-351 965	-349 658	-347 211	-341 894	-336 000
	ΔH(x)	-7 369	-6 365	-5 658	-4 811	-2 694	0
3	E(x)	-356 169	-351 965	-349 658	-347 211	-341 894	-336 000
	ΔH(x)	-7 369	-6 365	-5 658	-4 811	-2 694	0
4	E(x)	-356 020	-351 822	-349 525	-347 095	-341 825	-336 000
	ΔH(x)	-7 220	-6 222	-5 525	-4 695	-2 625	0
5	E(x)	-355 114	-350 948	-348 716	-346 383	-341 404	-336 000
	ΔH(x)	-6 314	-5 348	-4 716	-3 983	-2 204	0
6	E(x)	-357 832	-353 569	-351 143	-348 518	-342 666	-336 000
	ΔH(x)	-9 032	-7 969	-7 143	-6 118	-3 466	0
7	E(x)	-356 020	-351 822	-349 525	-347 095	-341 825	-336 000
	ΔH(x)	-7 220	-6 222	-5 525	-4 695	-2 625	0
8	E(x)	-358 278	-353 999	-351 541	-348 869	-342 873	-336 000
	ΔH(x)	-9 478	-8 399	-7 541	-6 469	-3 673	0
9	E(x)	-356 584	-352 366	-350 029	-347 538	-342 087	-336 000
	ΔH(x)	-7 784	-6 766	-6 029	-5 138	-2 887	0

Table 5 Potential energies $E(x, 0, \sigma_{\max})$ and heats of formation $\Delta H(x, 0, \sigma_{\max})$ of AuCu-type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with maximal ordering degrees calculated by nine E -functions at 0 K (J/mol)

No.		x(Cu)						
		0	0.1	0.2	0.25	0.3	0.4	0.5
1	E(x)	−368 000	−367 450	−366 521	−365 915	−365 214	−363 529	−361 464
	ΔH(x)	0	−2 650	−4 921	−5 915	−6 814	−8 329	−9 464
2	E(x)	−368 000	−367 334	−366 341	−365 720	−365 014	−363 349	−361 337
	ΔH(x)	0	−2 534	−4 741	−5 720	−6 614	−8 149	−9 337
3	E(x)	−368 000	−367 334	−366 341	−365 720	−365 014	−363 349	−361 337
	ΔH(x)	0	−2 534	−4 741	−5 720	−6 614	−8 149	−9 337
4	E(x)	−368 000	−367 270	−366 245	−365 620	−364 919	−363 285	−361 337
	ΔH(x)	0	−2 470	−4 645	−5 620	−6 519	−8 085	−9 337
5	E(x)	−368 000	−366 882	−365 663	−365 013	−364 336	−362 897	−361 337
	ΔH(x)	0	−2 082	−4 063	−5 013	−5 936	−7 697	−9 337
6	E(x)	−368 000	−368 047	−367 410	−366 834	−366 084	−364 062	−361 337
	ΔH(x)	0	−3 247	−5 810	−6 834	−7 684	−8 862	−9 337
7	E(x)	−368 000	−367 270	−366 245	−365 620	−364 919	−363 285	−361 337
	ΔH(x)	0	−2 470	−4 645	−5 620	−6 519	−8 085	−9 337
8	E(x)	−368 000	−368 238	−367 697	−367 132	−366 370	−364 253	−361 337
	ΔH(x)	0	−3 438	−6 097	−7 132	−7 970	−9 053	−9 337
9	E(x)	−368 000	−367 512	−366 608	−365 998	−365 282	−363 527	−361 337
	ΔH(x)	0	−2 712	−5 008	−5 998	−6 882	−8 327	−9 337
No.		x(Cu)						
		0.6	0.7	0.75	0.8	0.9	1	
1	E(x)	−357 129	−352 414	−349 915	−347 321	−341 850	−336 000	
	ΔH(x)	−8 329	−6 814	−5 915	−4 921	−2 650	0	
2	E(x)	−357 148	−352 526	−350 051	−347 465	−341 959	−336 000	
	ΔH(x)	−8 348	−6 926	−6 051	−5 065	−2 759	0	
3	E(x)	−357 148	−352 526	−350 051	−347 465	−341 959	−336 000	
	ΔH(x)	−8 348	−6 926	−6 051	−5 065	−2 759	0	
4	E(x)	−357 084	−352 431	−349 952	−347 370	−341 895	−336 000	
	ΔH(x)	−8 284	−6 831	−5 952	−4 970	−2 695	0	
5	E(x)	−356 696	−351 848	−349 345	−346 787	−341 507	−336 000	
	ΔH(x)	−7 896	−6 248	−5 345	−4 387	−2 307	0	
6	E(x)	−357 861	−353 596	−351 165	−348 535	−342 672	−336 000	
	ΔH(x)	−9 061	−7 996	−7 165	−6 135	−3 472	0	
7	E(x)	−357 084	−352 431	−349 952	−347 370	−341 895	−336 000	
	ΔH(x)	−8 284	−6 831	−5 952	−4 970	−2 695	0	
8	E(x)	−358 052	−353 882	−351 464	−348 821	−342 863	−336 000	
	ΔH(x)	−9 252	−8 282	−7 464	−6 421	−3 663	0	
9	E(x)	−357 326	−352 794	−350 330	−347 733	−342 137	−336 000	
	ΔH(x)	−8 526	−7 194	−6 330	−5 333	−2 937	0	

Table 6 Potential energies $E(x, 0, \sigma_{\max})$ and heats of formation $\Delta H(x, 0, \sigma_{\max})$ of AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with maximal ordering degrees calculated by nine E -functions at 0 K (J/mol)

No.		$x(\text{Cu})$						
		0	0.1	0.2	0.25	0.3	0.4	0.5
1	$E(x)$	−368 000	−367 387	−366 269	−365 521	−364 646	−362 519	−359 887
	$\Delta H(x)$	0	−2 587	−4 669	−5 521	−6 246	−7 319	−7 887
2	$E(x)$	−368 000	−367 279	−366 118	−365 368	−364 503	−362 421	−359 859
	$\Delta H(x)$	0	−2 479	−4 518	−5 368	−6 103	−7 221	−7 859
3	$E(x)$	−368 000	−367 279	−366 118	−365 368	−364 503	−362 421	−359 859
	$\Delta H(x)$	0	−2 479	−4 518	−5 368	−6 103	−7 221	−7 859
4	$E(x)$	−368 000	−367 210	−366 001	−365 235	−364 360	−362 273	−359 727
	$\Delta H(x)$	0	−2 410	−4 401	−5 235	−5 960	−7 073	−7 727
5	$E(x)$	−368 000	−366 790	−365 289	−364 426	−363 486	−361 367	−358 918
	$\Delta H(x)$	0	−1 990	−3 689	−4 426	−5 086	−6 167	−6 918
6	$E(x)$	−368 000	−368 052	−367 425	−366 853	−366 107	−364 085	−361 345
	$\Delta H(x)$	0	−3 252	−5 825	−6 853	−7 707	−8 885	−9 345
7	$E(x)$	−368 000	−367 210	−366 001	−365 235	−364 360	−362 273	−359 727
	$\Delta H(x)$	0	−2 410	−4 401	−5 235	−5 960	−7 073	−7 727
8	$E(x)$	−368 000	−368 259	−367 775	−367 251	−366 537	−364 531	−361 743
	$\Delta H(x)$	0	−3 459	−6 175	−7 251	−8 137	−9 331	−9 743
9	$E(x)$	−368 000	−367 472	−366 445	−365 739	−364 904	−362 837	−360 231
	$\Delta H(x)$	0	−2 672	−4 845	−5 739	−6 504	−7 637	−8 231

No.		$x(\text{Cu})$					
		0.6	0.7	0.75	0.8	0.9	1
1	$E(x)$	−356 750	−353 108	−351 098	−348 079	−342 039	−336 000
	$\Delta H(x)$	−7 950	−7 508	−7 098	−5 679	−2 839	0
2	$E(x)$	−356 804	−353 243	−351 268	−348 246	−342 155	−336 000
	$\Delta H(x)$	−8 004	−7 643	−7 268	−5 846	−2 955	0
3	$E(x)$	−356 804	−353 243	−351 268	−348 246	−342 155	−336 000
	$\Delta H(x)$	−8 004	−7 643	−7 268	−5 846	−2 955	0
4	$E(x)$	−356 709	−353 206	−351 268	−348 214	−342 107	−336 000
	$\Delta H(x)$	−7 909	−7 606	−7 268	−5 814	−2 907	0
5	$E(x)$	−356 126	−352 979	−351 268	−348 020	−341 816	−336 000
	$\Delta H(x)$	−7 326	−7 379	−7 268	−5 620	−2 616	0
6	$E(x)$	−357 874	−353 659	−351 268	−348 603	−342 690	−336 000
	$\Delta H(x)$	−9 074	−8 059	−7 268	−6 203	−3 490	0
7	$E(x)$	−356 709	−353 206	−351 268	−348 214	−342 107	−336 000
	$\Delta H(x)$	−7 909	−7 606	−7 268	−5 814	−2 907	0
8	$E(x)$	−358 160	−353 770	−351 268	−348 698	−342 833	−336 000
	$\Delta H(x)$	−9 360	−8 170	−7 268	−6 298	−3 633	0
9	$E(x)$	−357 072	−353 347	−351 268	−348 335	−342 289	−336 000
	$\Delta H(x)$	−8 272	−7 747	−7 268	−5 935	−3 089	0

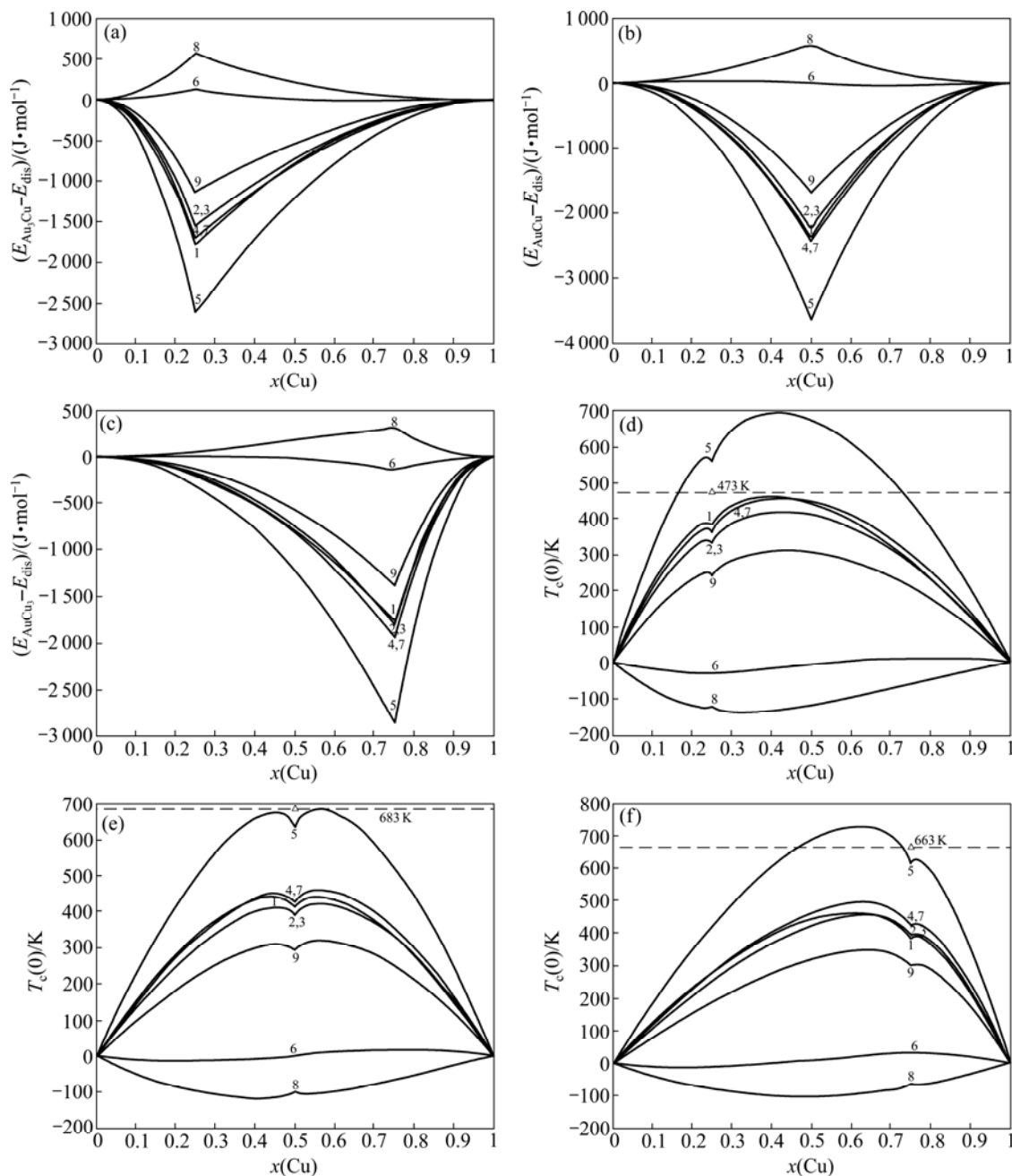


Fig.4 Potential energy differences of Au_3Cu -, AuCu - and AuCu_3 -type ordered alloys with the maximum ordering degrees relative to disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys ((a), (b), (c)); critical temperatures of Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys translated into disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys described by nine E -functions ((d), (e), (f)) together with experimental data[8] (denoted by triangles)

3 Discussion on effect of ordering degree on heat of formation

In Au-Cu system containing ordered phases the effective interaction bond energy between the different kinds of atoms is bigger than one between the same kind of atoms, it leads to a tendency of atoms to be surrounded by atoms of the other kind and to a deviation of the distribution of concentration x_i^{Au} and x_i^{Cu} of characteristic atoms A_i^{Au} and A_i^{Cu} in a real alloy

from the distribution in the completely disordered state. If this influence is long-range(LR) order, the original lattice splits into sublattices and each is occupied by only one kind of atom and the additional superstructure lines appear in Bragg scattering. If this influence is short-range(SR) order, it only affects the probability of finding an atom of a specific kind in the neighboring coordination shells around a certain atom, i.e., the concentration x_i^{Au} and x_i^{Cu} of characteristic atoms A_i^{Au} and A_i^{Cu} in a certain alloy. Therefore, the LR order degree σ^{LR} describes not only the deviation of the

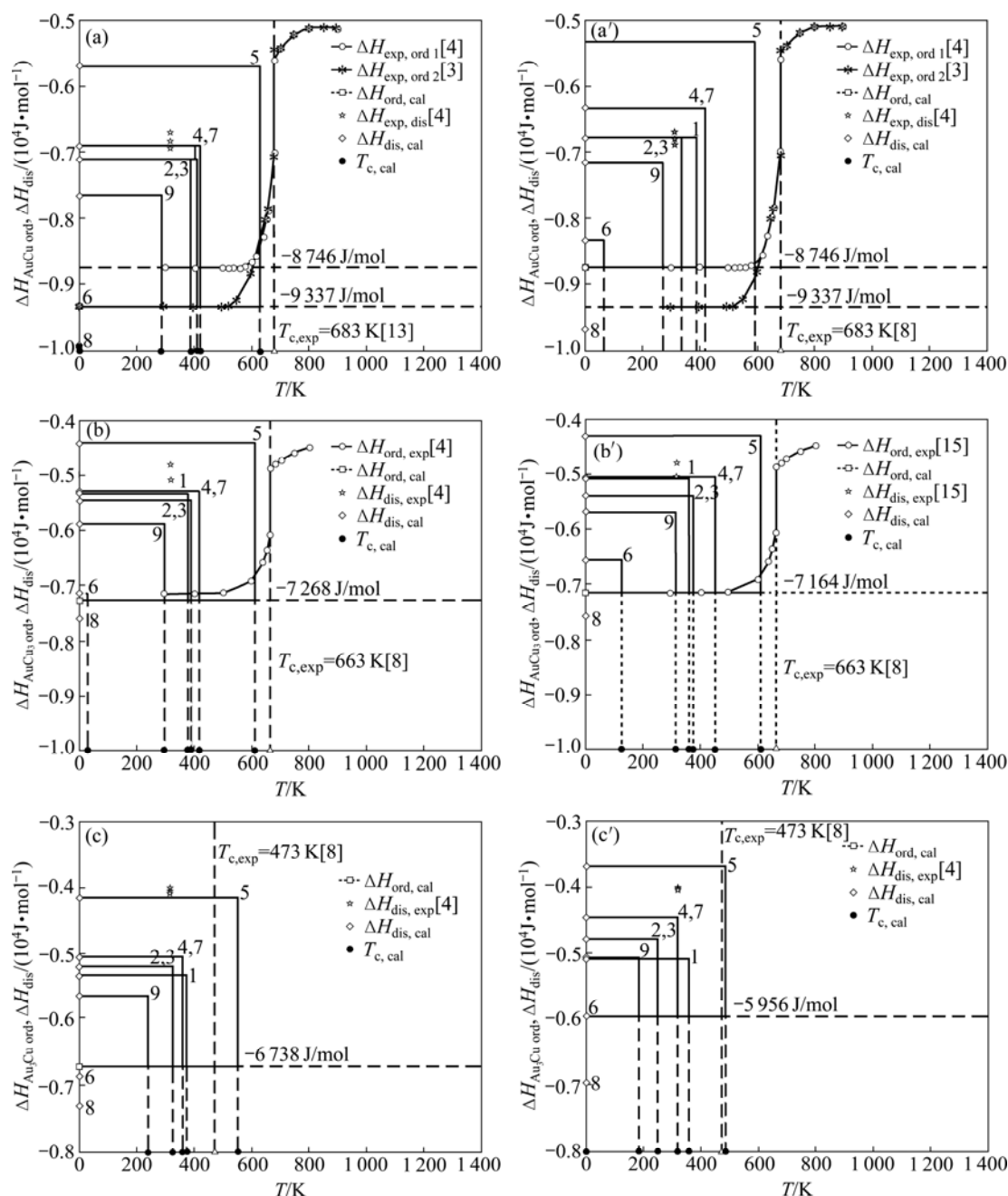


Fig.5 Comparison for $\Delta H_{\text{ord}}(0) - \Delta H_{\text{dis}}(0) - T_c$ interrelated patterns of AuCu, AuCu₃ and Au₃Cu compounds relative to corresponding disordered alloys obtained respectively from nine E -functions in present part ((a), (b), (c)) and the first part ((a'), (b'), (c')) of this investigation

concentration x_i^{Au} and x_i^{Cu} of characteristic atoms A_i^{Au} and A_i^{Cu} in the LR ordered alloy from one in the disordered alloy, but also the relationship between atoms occupying right and wrong sublattices. But the SR order degree σ^{SR} describes only the deviation of the concentration of characteristic atoms in the SR ordered alloy from one in the disordered alloy.

Above critical temperatures, the stoichiometric AuCu and AuCu₃ are statistically face-centered cubic and a SR order exists. The experimental measurements of the SR order and the evaluation of X-ray data were

carried out by ROBERTS[9], KEATING and WARREN [10]. The results are concluded as follows: 1) For the AuCu samples held at 698 K and 778 K above the critical temperature 683 K, there is high SR order degree, as evidenced by the first neighbor parameters which are respectively 37% and 36% of the way from complete disorder to perfect order. 2) The SR order degree for the quenched AuCu sample from 773 K is considerably higher than that for the sample held at this temperature, as evidenced by the first neighbor parameter which is 47.4% of the way from complete disorder to perfect

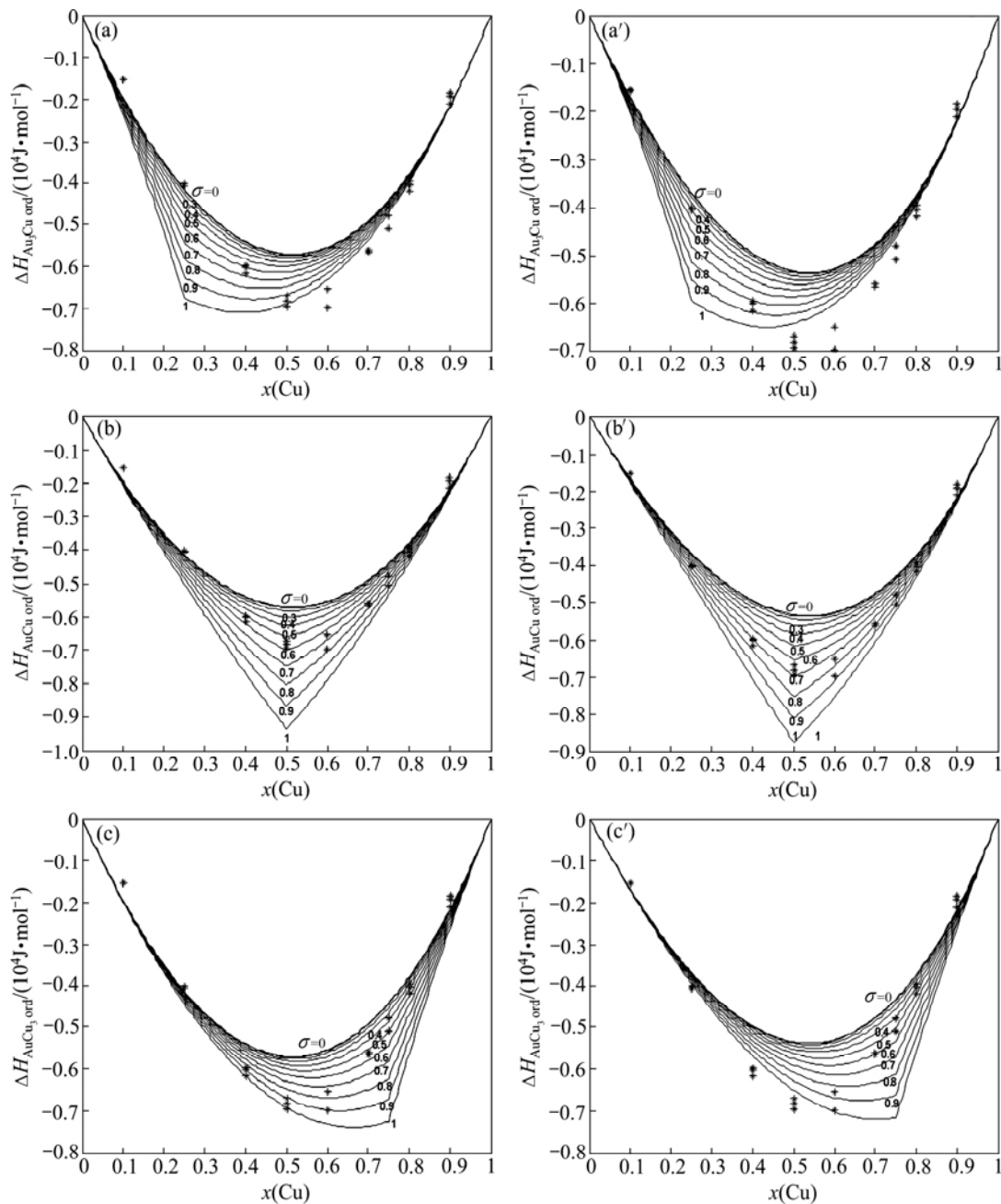


Fig.6 Effect of ordering degree on heats of formation of Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys described by the 5th E -function in present part ((a), (b), (c)) and the first part ((a'), (b'), (c')) of this investigation together with experimental data at 320 K [4]

order, indicating that the quenching of a large single crystal is inadequate to suppress increase in SR order degree. These results suggest that it is more difficult to get completely disordered alloy than to get fully ordered alloy in an alloy system containing ordered phases, that also has been demonstrated by the recent investigations [9–11].

Whether the heats of formation of disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys calculated from the 5th E -function on the basis of the smaller or larger heats of formation of the $L1_0$ - AuCu and $L1_2$ - AuCu_3 compounds are higher than

the experimental ones. This means that the so-called disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys contain SRO degrees.

In the SSA framework, the ordering of disordered alloys is the degeneracy of the energetic states of the characteristic atoms, and the disordering of ordered alloys is the splitting of the energetic states of the characteristic atoms. In this significance, both the LRO degree and the SRO degree can be described by the probabilities of the characteristic atoms occupied at the sublattices. This means that there are the same probabilities of characteristic atoms in the LRO and SRO

alloys, if the $\sigma^{\text{LRO}} = \sigma^{\text{SRO}}$.

The effects of ordering degrees on the heats of formation $\Delta H(x, 320, \sigma)$ of the Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys described by the 5th E -function in the first part and the present part of this investigation are shown in Fig.6, from which it can be known that the experimental heats of formation of the so-called disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with the SRO degree should be described by three $\Delta H(x, T, \sigma)$ functions of the Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys in the three compositional ranges.

In the present part of this investigation, the SRO degrees of the experimental $\text{Au}_{1-x}\text{Cu}_x$ disordered alloys described by the 5th $\Delta H(x, T, \sigma)$ function are shown in Figs.6(a), (b) and (c). For examples,

$$\begin{cases} \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.5}\text{Cu}_{0.5}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.5}\text{Cu}_{0.5}, 0) = 0.5 - 0.6 \\ \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.25}\text{Cu}_{0.75}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.25}\text{Cu}_{0.75}, 0) = 0.3 - 0.4 \\ \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.75}\text{Cu}_{0.25}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.75}\text{Cu}_{0.25}, 0) = 0 \end{cases}$$

In the first part of this investigation, the SRO degrees of the experimental $\text{Au}_{1-x}\text{Cu}_x$ disordered alloys described by the 5th $\Delta H(x, T, \sigma)$ function are shown in Figs.6(a'), (b') and (c'). For examples,

$$\begin{cases} \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.5}\text{Cu}_{0.5}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.5}\text{Cu}_{0.5}, 0) = 0.6 - 0.7 \\ \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.25}\text{Cu}_{0.75}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.25}\text{Cu}_{0.75}, 0) = 0.4 - 0.5 \\ \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.75}\text{Cu}_{0.25}, 320) = \sigma_{\text{dis}}^{\text{SRO}}(\text{Au}_{0.75}\text{Cu}_{0.25}, 0) = 0.2 - 0.3 \end{cases}$$

From this discussion, it can be concluded that it is more difficult to get completely disordered alloys than to get fully ordered alloys in an alloy system containing ordered alloy phases.

4 Conclusions

1) Based on the larger experimental heats of formation of the $L1_0$ - AuCu and $L1_2$ - AuCu_3 compounds, the potential energy sequences for nine E -functions have been determined. Although the potential energies $E(x, 0, 0)$ of the $L1_2$ - Au_3Cu , $L1_0$ - AuCu and $L1_2$ - AuCu_3 compounds calculated from these E -functions are, respectively, equivalent each other, the potential energies of their characteristic atoms are different each other.

2) The T_c -temperatures of the Au_3Cu , AuCu and AuCu_3 compounds obtained from the 5th E -function are closer by the experimental values than from other E -functions. The T_c -temperatures of the three compounds

obtained from the 4th and 7th E -functions are greatly lower than experimental ones.

3) In an alloy system containing ordered alloy phases, it is more difficult to get completely disordered alloys than to get fully ordered alloys. The experimental heats of formation of the so-called disordered $\text{Au}_{1-x}\text{Cu}_x$ alloys with SRO degree should be described by three $\Delta H(x, T, \sigma)$ -functions of the Au_3Cu -, AuCu - and AuCu_3 -type ordered $\text{Au}_{1-x}\text{Cu}_x$ alloys in the three compositional ranges.

4) The $\Delta H_{\text{ord}}(0) - \Delta H_{\text{dis}}(0) - T_c$ interrelated patterns of the AuCu , AuCu_3 and Au_3Cu compounds relative to corresponding disordered alloys obtained respectively from nine E -functions in the present part and the first part of this investigation are compared. The potential energy sequences determined from the 5th E -function in both the first and the present parts may be chosen and developed into the energetic information chain. It will be necessary to make further investigation for understanding effects of potential energy sequences obtained on the basis of both smaller and larger experimental heats of formations on full thermodynamic properties within the phase regions and of phase transformations.

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(Edited by YANG Hua)