SOLID-STATE PHYSICS

LONG-PERIOD STATES OF ORDERED METAL ALLOYS.
1. ANALYSIS OF STRUCTURAL FEATURES

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An analysis of empirical data establishes the presence at least two different types of long-period structures. The first type of structure is seen in alloys of the system $Cu-Au$, Cu_3Pd, Cu_3Pt, etc., while the second type is found in alloys Ag_3Mg and Al_3Ti and their analogs. It was concluded that the nature of the long-period states is different in the two types of structures.

Ordered alloys with a long period are one of the most interesting and promising classes of metal alloys. They differ from ordinary ordered systems with a simple superstructure in the periodic or quasi-periodic disruption of the ordered arrangement of atoms by antiphase boundaries (APB). Antiphase boundaries are usually energetically unfavorable in ordered alloys, but they are equilibrium structural elements in systems with a long-period structure (LPS). Distinct regions of stability exist on the temperature − composition phase diagrams of ordered alloys with a long period.

Studies of the mechanical properties of alloys of this type [1-10] have shown that it is possible, by means of aging, to obtain highly disperse stable alloys based on long-period ordered phases. Strengthening of the alloy by the decomposition of a supersaturated solid solution can be effectively combined with strengthening from atomic ordering, which in turn makes it possible to create unusual dispersed decomposition structures that are stable. Alloys formed on this basis have high mechanical properties and are stable throughout the temperature range in which the matrix remains ordered [11-14].

It has been reliably established [15] that a long period is seen in alloys with fcc and cph lattices. A wealth of experimental data [15, 18] has by now been accumulated on the structural features of LPSs and the laws governing the formation and behavior of long-period alloys. However, no consistent physical representations have yet been presented on the nature of LPSs, and it has been quite a long time since the available information on LPSs was generalized. For these reasons, the present article is devoted to analyzing existing information on the long-period state of metallic systems and understanding the nature of the formation and behavior of such states.

Due to the large volume of material with which we are dealing, the article is divided into two parts. The first part surveys the experimental data obtained to date, while the second part analyzes existing physical representations on the nature of LPSs.

Since most of the information has been obtained on fcc alloys, most of the data we will present will concern LPSs in an fcc structure.

FEATURES OF THE STRUCTURE AND BEHAVIOR OF
ALLOYS WITH A LONG PERIOD

In the current literature, the term "long-period state" as applied to an ordered alloy means a state in which the translation cell includes more than one cell of the base superstructure. A region of an ordered crystal bounded by two nearest-
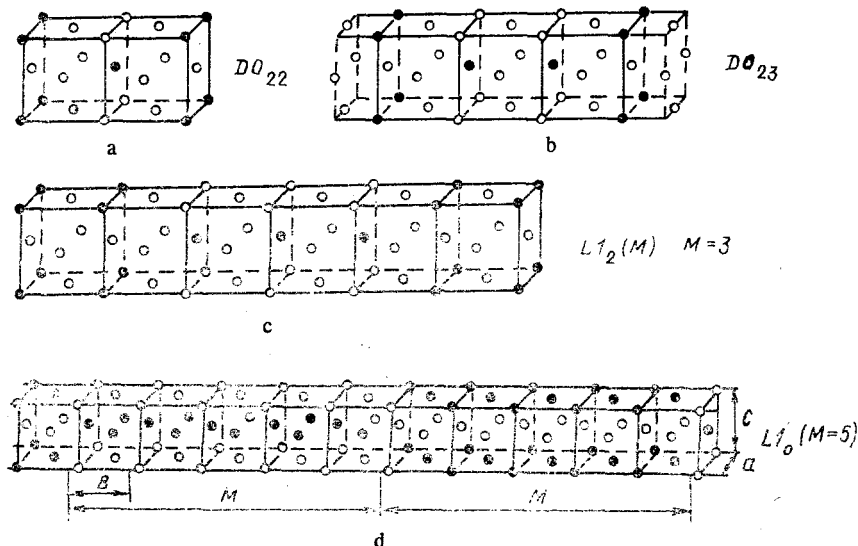
Fig. 1. Examples of long-period ordered superstructures DO_{22} (a), DO_{23} (b), $L1_2(M)$ with $M = 3$ (c), and $L1_0(M = 5)$, i.e., of the type CuAu II (d).

neighbor APBs is called an anti-phase domain. The size of such a domain M (see Fig. 1d, for example) is measured in the parameters of the lattice of the base superstructure, while $P = 2 \cdot M$ is the long period of the LPS.

Geometrically different superstructures within a given crystalline structure may undergo mutual transformation through the introduction of periodic displacements characterized by a suitable vector. In an fcc structure, for example, the mutual transformation of superstructures can be represented by means of periodic APBs [17].

A detailed analysis of possible APBs [19-21] in A_3B alloys with an fcc lattice shows that there are two cases. In the first case, the displacement vector $<uvw>$ lies within the plane of the APB $\{hkl\}$. The condition $hu + kv + lw = 0$ is satisfied for such APBs. Anti-phase boundaries of this type do not lead to the appearance of additional "irregular" bonds with nearest neighbors. Disruption of the regular ordering of atoms is seen only beginning with the second neighbors. In the second case, the displacement vector lies outside the plane of the APB and $hu + kv + lw \neq 0$. Such boundaries are called second-order or nonconservative APBs. When an APB of the second kind is formed, not only must a displacement take place, but the removal or insertion of one of the atomic planes parallel to the APB must also occur. The formation of a second-order APB leads to the formation of additional "irregular" bonds within the first coordination sphere, causing the energy of these APBs to be approximately an order of magnitude greater than the energy of first-order APBs [16, 19].

Computer modeling of the orientation dependence of the energy of single APBs in alloys with an L12 superstructure has shown [22, 23] that the APB $1/2 < 110 > \{100\}$ has the lowest energy in the alloy Cu_3Au.

The first LPSs were observed in a Cu−Au alloy of equiatomic composition [24, 25]. This alloy was designated CuAu II. Its unit cell is shown in Fig. 1d. Since LPSs were first discovered in this alloy, its LPSs have been the most thoroughly investigated. Similar LPSs have since been seen in many binary alloys with fcc and cph lattices [15, 17, 18, 26]. The term "unidimensional" is used to describe LPSs in which antiphase boundaries alternate in one direction. Long-period structures with APBs alternating in two directions are called two-dimensional, biperiodic, or columnar. Alloys in which the periodicity of the APBs is three-dimensional have yet to be observed experimentally. The conclusion reached in [16] was that such a state would be unstable and, in the general case, cannot exist.

It was found experimentally in [15, 16] that only first-order APBs are realized in unidimensional LPSs and that they are located within one of the faces of the cube in alloys with an fcc structure.

Studies of alloys with a columnar structure have shown that they are not always a superposition of a unidimensional LPS in two directions [27, 28]. Not only do they occur as first-order APBs forming in both directions, but also as an alternation of first-order APBs in one direction and second-order APBs in the other direction [17, 19].

It has been firmly established [16, 20, 26, 29] that a LPS represents a stable equilibrium state of the alloy and that the period does not change during isothermal annealing. Moreoever, it was shown in [30-34] that the structure of CuAu II is formed both from the low-temperature ordered state of CuAu I ($L1_0$ superstructure) and from the disordered A1 phase. This
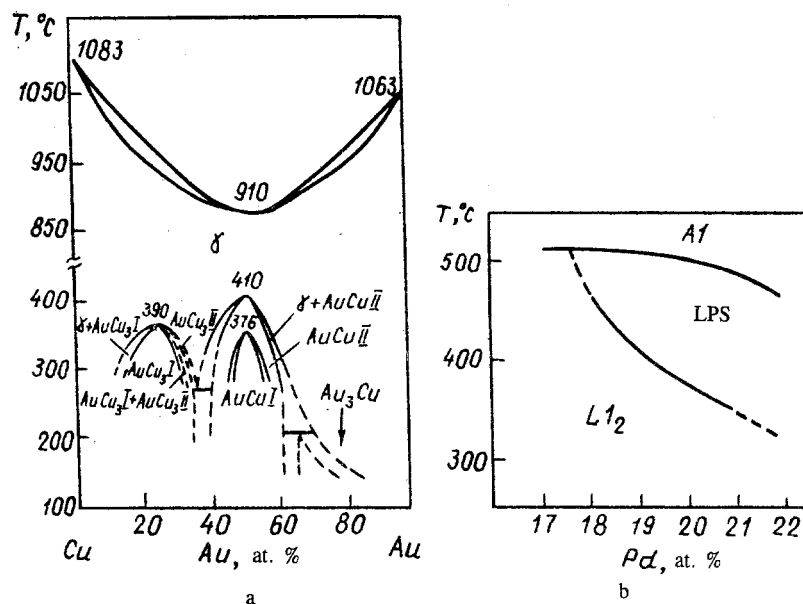
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Fig. 2. Phase diagram of system: a) Cu—Au [35, 36], b) Cu—Pd [37, 38].

fact is further proof of the equilibrium nature of LPSs. Thus, LPSs occupy definite locations on the constitution diagram. As an illustration, Fig. 2 shows the constitution diagrams of Cu—Au (Fig. 2a) [35, 36] and Cu—Pd (Fig. 2b) [37, 38].

Experimental studies [15-17, 26, 39] often reveal fractional values for the long period. This means that some averaging is necessary over a set of integral values and shows that there is a state that is disproportionate to the base superstructure [40].

The indicated diagrams show that the low-temperature phase is realized in the form of base superstructures $L1_2$ and $L1_0$. A disproportionate long-period structure is formed in the alloy with an increase in temperature [15, 16, 26, 39, 41]. We will henceforth refer to such structures as a "LPSs of type one."

On the other hand, LPSs with a different temperature dependence have been noted in the literature. For example, the superstructure DO_{22} is realized in Pt_3V [42] below 900°C. Thus, the low-temperature phase is proprotionate to the base superstructure of the LPS [17]. An increase in temperature is accompanied by the formation of a disproportionate state. We will henceforth refer to structures of this type as "LPSs of type two."

The expediency of separating LPSs into two types was noted in [37, 43]. In [43], it was pointed out that the behavior of alloys with a LPS of the second type is very similar to the behavior of polytypes. The usefulness of such a subdivision based on structural features and the behavior of LPSs of different types under changing external conditions will become evident in the subsequent discussion.

Let us examine alloys in which LPSs of the first type are realized. Cu—Au alloys are classical examples of such alloys [35, 36]. For the alloy CuAu, of equiatomic composition, the order-disorder transition temperature T_c is 410°C. Below 376°C, there is an ordered phase with a layered arrangement of gold and copper atoms in the (100) planes. The superstructure of this phase has been designated CuAu I. Due to the layering of atoms, the well-ordered alloy is characterized by tetragonality c/a = 0.92 [22, 44]. The structure CuAu II forms within the interval 376-410°C [25, 45-49]. This structure is orthorhombic. Its unit cell is shown in Fig. 1d. If the transition to the ordered state is accompanied by anisotropic distortion of the alloy, then there will be several regions with different c-axis directions within one crystallite [16]. These regions are referred to as c-domains and are different from the periodic domains. It has been found experimentally [50-53] that a certain relationship exists between the size of a c-domain and the probability of formation of a LPS. In CuAu, for example, the periodic antiphase structure is stable up to a certain minimum size of c-domain. When the latter is smaller than 80 Å, no periodic APBs are formed [50-53].

When a long period forms in a crystal, the entire lattice is placed in tension or compression in the direction of this period. The lattice parameter along it b differs from the corresponding values along the other axes (see Fig. 1d). When a LPS forms in an alloy, the ratio b/a may be greater than or less than unity [16]. This ratio changes from alloy to alloy. No clear relationship between b/a and the period of the antiphase structure has been established.

Experimental data [15, 16, 26, 39] indicates that phases with a long period are formed at temperatures within the neighborhood of T_c. For equiatomic alloy CuAu, for example, all transformations CuAu I $\rightleftarrows$ Cu Au II $\rightleftarrows$ A1 are first-order transformations [16].

High-resolution electron microscopy was used in [43] to study typical disproportionate statistical structures in the systems Cu$-$Au and Cu$-$Pd. Such structures $-$ LPSs $-$ were seen in nearly all of the compositions for Cu$-$Au when the gold content was greater than 25 at. %. The minimum mean value of the long period corresponded to the composition CuAu. The value of the long period increased as gold content increased or decreased from this value.

The dependences of the size of the antiphase domain M and the tetragonality parameter c/a on composition for Cu alloys with 36-65 at. % Au were reported in [54]. Both curves have minima in the region of the equiatomic composition. The value of $\bar{M}$ decreases monotonically from ~ 6.5 at 36 at. % Au to ~ 5.07 at the equiatomic composition, increasing with a further increase in gold content (~ 7.3 at 65 at. % Au). A relation of similar form is seen for the tetragonality parameter, with $c/a \approx 0.97$ at 35 at. % Au, $c/a \approx 0.92$ at 50 at. % Au, and $c/a \approx 0.99$ at 65 at. % Au. Evidence of the presence of a LPS is provided by the splitting of superstructure reflections of the 110 type in the direction $<100>$ [47].

When the composition deviates from stoichiometric CuAu, excess atoms are concentrated mainly near the antiphase boundaries [55-57].

The formation of coherent ordered lamellae with a structure different from the structure of the disordered matrix creates stresses [58]. It was concluded on this basis that a period of the martensitic type is realized in the form of fine and regular displacements below T_c. The latter structure is also regarded as CuAu II.

The authors of [59-63] demonstrated the existence of the stable periodic antiphase structure Cu$_3$Au II within the composition interval from 31.6 to 29.2 at. % Au at temperatures below T_c. Domain size is nine unit cells of the base superstructure. The results obtained in [63] were corroborated by electron-diffraction methods in [64], although domain size was estimated to be 10 unit cells. The methods of field ion microscopy showed in [65] that the width of the boundary region of the antiphase surface in stoichiometric Cu$_3$Au $-$ with a high degree of long-range order in the domains $-$ is equal to the atomic spacing, regardless of the density of the antiphase boundaries.

The existence of LPSs in stoichiometric CuAu$_3$ and alloys with a higher gold content was shown in [66-68]. It was emphasized that there is a close similarity between CuAu$_3$ and Cu$_3$Au in terms of structure. The ordering of the structure is similar in each case: the energy of the APBs has a deep minimum for the {100} planes in the L1$_2$ superstructure. In addition, the measured domain sizes are within the same range of values as for Cu$_3$Au. The method of x-ray diffractometry was used in [69] to analyze the dimensions of antiphase domains in the alloy Cu $-$ 72.5 at. % Au. A later investigation [70] emphasized that a two-dimensional long-period structure is formed by long annealing within the range 103-205°C.

Let us examine the effect of a third component on the long period of an alloy of the system Cu$-$Au. The effect of the alloying element on the long period of CuAu II in the region of existence of the long-period phase was detailed in [16, 19-21]. Researchers alloyed vacuum-deposited films with 19 different elements $-$ including Ag, Al, Ga, In, Sn, Zn, and Pd $-$ in different concentrations. It was found that additions of elements of the third group (Al, Ga, In, Mn, Zn) decreases the mean size of the antiphase domains when the concentration of the alloying element is increased. The introduction of Ni, Pd, and Ag increases the long period in a manner analogous to the above-noted deviations from equiatomic CuAu. The changes in domain size with a change in the concentration of dissolved Ga and Al are nearly identical. It was noted that for CuAu$-$Al alloys, the region of stability of CuAu II decreases with an increase in the concentration of Al. The CuAu I $\rightarrow$ CuAu II transformation temperature also decreases (for a concentration of 5 at. % Al or more, it is located in the region of 370°C). The effect of additions of gallium was also studied later [71]. It was confirmed that small additions of gallium have a stabilizing effect on the unidimensional long-period structure of CuAu II in Au$_{50}$Cu$_{50-x}$Ga$_x$ alloys, and it was noted that the long period decreases with an increase in the concentration of gallium.

It was found in [19-21] that no CuAu II structure is formed in foils of CuAu containing 3.6-17.3 at. % Ge with the transition from CuAu I to a disordered phase. The addition of small quantities of Sb, Bi, Be, Mg, and Cr had no significant effect on domain size in CuAu II. Most of the elements that did not significantly affect the long period simply did not dissolve in the alloy in an appreciable amount (this applies to Sb, Bi, Be, Cr, and possibly Mg). The long-period phase disappeared with the addition of 3 at. % Pd, but equilibrium APBs reappeared with additional alloying by 5 at. % Al. The addition of aluminum to an alloy that contained 27.5 at. % Au [20] but had no detectable LPS led to the formation of a phase with equilibrium APBs.

The effect of alloying alloys near the composition Cu$_3$Au was studied in [72-78]. In Cu$_3$Au$-$In alloys additionally alloyed with Pd and Ag, the formation of nuclei of an ordered phase in the initial matrix solid solution is accompanied by its layering and the subsequent formation of particles of a precipitated phase in the rich interlayers between domains [72]. The

sequence of phase transformations reverses with an increase in treatment temperature or the concentration of alloying components: particles of the precipitated phase, with high concentrations of Pd or Ag, are formed first; heterogeneous nucleation of domains of the ordered phase then takes place on these particles. A comparative study of the kinetics of phase transformations (breakdown of a solid solution and ordering) and the resulting structures was made in [73] using model alloy Cu_3Au − 8.24 at. % Ag and another alloy having an excess of gold relative to stoichiometric Cu_3Au. A long-period superstructure was observed in the latter when any treatment was administered below T_c. Ordering preceded decomposition in the model alloy, and the resulting structure was nonuniform. A long-period superstructure was seen in the ordered matrix until silver began to rapidly precipitate from the matrix. No lattice modulation associated with decomposition of the solid solution was observed in the aged alloy [74]. In Cu_3Au alloys alloyed with Ag, the nucleation of a phase precipitated during decomposition can take place either homogeneously throughout the volume of the thermal domains or heterogeneously at their boundaries. The nucleation mechanism is determined by the atomic order and the degree of supersaturation of the solution in bulk and near the boundaries of the domains. Heterogeneous nucleation of a phase based on Ag precedes concentrative, thermal, and vacancy "blurring" of the antiphase boundaries, accompanied by alignment of the boundaries in the {100} planes.

Structural changes that occur during the aging of alloy Au − 55.2 at. % Cu − 17.4 at. % Ag were studied in [78], where two stages were observed. The first stage involves spinodal decomposition of a supersaturated solid solution into two fcc phases: a copper-rich phase and a silver-rich phase. This stage is characterized by the formation of a modulated structure along < 100 >. Discontinuous decomposition from the grain boundaries is seen in the second stage. The long-period structure of Cu_3Au II is formed only within the regions in which decomposition is discontinuous.

Thus, if an alloying element is dissolved in the alloy, its addition may either increase or decrease the long period of the LPS. The presence of the alloying element is also capable of changing the region in which the phase with equilibrium APBs is stable.

Diffraction patterns obtained in structural studies of LPSs show splitting of reflections of the 110 type in the direction < 100 > [15, 54]. The intensity of the split components of the superstructure reflections are asymmetric. In certain cases, satellite maxima are present near the principal reflections [54]. The asymmetry of intensity is connected with displacements of atoms from their lattice positions, while the satellite reflections are related either to periodic displacements of the atomic planes (phase modulation) or to a periodic change in density (amplitude modulation) [54]. Study of the antiphase structure of CuAu II alloys by microscopy and electron diffraction within the concentration interval 56-65 at. % revealed an anomaly in the distribution of the superstructure reflections. The most significant finding was that the intensity of the high-order branches was very low. Also, instead of of being parallel to [100], the splitting of the reflections deviated somewhat (up to 6°) from it this direction [54]. It was noted that the anomalies occurred only in the nonstoichiometric composition. Here, domain size deviated from the value 5 and increased regularly with an increase in the content of Cu or Au.

We should point out the important local features of the structural characteristics of Cu−Au alloys with an LPS. Disordering along the antiphase boundaries of CuAu II alloys has been seen at 390°C, within the region in which the CuAu II phase is stable [79-81].

The formation of satellites near the principal reflections and the asymmetric intensity of the split peaks of the superstructure maxima [48, 55, 68, 77, 82] could be explained by the displacement of atoms from the points of the ideal lattice. The occurrence of such displacements in the direction of the long period was demonstrated by Fourier synthesis of electron density [55, 83-88]. Nevertheless, the nature of these shifts or modulations remains somewhat unclear [41]. We should note that modulation of the lattice occurs in the direction of the long period: atoms of one species are displaced toward APBs, while atoms of another species are displaced away from APBs [55, 83-88]. The displacements of atoms from the points of the ideal lattice were first evaluated on the basis of experimental data [86] for Au_3Cd with an annealing temperature of 300°C. It was found that the planes of gold atoms closest to the APBs had been shifted closer to the latter by 0.015 ± 0.005 Å, while the cadmium atoms had been shifted away from the APBs by 0.028 ± 0.005 Å. It was emphasized that periodic atomic displacements are integral characteristics of phases with equilibrium APBs. In the alloys that were studied (two compositions were investigated: Cu − 50 ± 0.4 at. % Au and Cu − 48 ± 0.5 at. % Au), the copper atoms closest to the APBs were shifted 0.079 Å toward the latter, while the gold atoms were displaced 0.048 Å farther from the anti-phase boundaries [55, 85, 87]. It was emphasized that atomic disordering occurs primarily in the immediate vicinity of the APBs and that disordering is negligible within the domain. It was reported in [79] that disordering along APBs is a characteristic of the structure of CuAu II even for the stoichiometric composition of the alloy. On the atomic planes closest to the APBs, the probability of substitution at a point which is regular for a copper atom is 0.7 for Cu and 0.3 for Au, while the analogous probabilities for the planes

second from the respective APBs are 0.94 and 0.06. It was pointed out that the disordering occurs over the entire atomic plane on each side of the LPS boundary.

There is yet another feature of alloys of the given system that might play an important role. The possibilities of calculating phase equilibria in metallic systems of the Cu−Au type were analyzed in [89]. The conclusion reached emphasizes the key role of relaxation processes in the lattice when thermodynamic properties and phase equilibria are being calculated [89-91].

The authors of [92] studied the structure of ordering twins formed in the pseudoalloy AuCu − 3 at. % Ag during annealing at temperatures below T_c. Tetragonal ordering of the phase CuAu II was observed. It was noted that the structure that is formed ensures maximal relaxation of the stresses that develop in the course of the fcc → cph transformation.

Alloys of the Cu−Pd system with a LPS have recently come into wide use. This system is interesting because it permits alteration of the structure of the phase with equilibrium APBs by changing the composition of the alloy.

Up to 10 at. % Pd, ordering occurs without the formation of periodic APBs. The unidimensional LPS is stabilized with an increase in the palladium content of the alloy, the period of the LPS decreasing with an increase in Pd concentration [20, 28, 37, 38, 54, 93-102]. At a palladium content greater than 27 at. %, the unidimensional structure is replaced by a two-dimensional structure [99, 100]. The antiphase boundaries of the second system of APBs that is formed are nonconservative. The situation that is realized is similar to that seen in the alloy Au_3Zn [26]. In both cases, general deformation of the lattice occurs only in one direction ($c/a < 1$ in Cu−Pd, while $c/a > 1$ in Au−Zn [20], c being the lattice parameter in the direction of alternation of the first-order APB). Thus, the appearance of a second-order APB does not lead to a change in the lattice parameter along the direction of alternation of such APBs [20]. The authors of [103] noted the presence of four diffuse maxima near the superstructure point 110 on the dark-field image of alloy Cu − 29.8 at. % Pd quenched from the region of the disordered phase. The appearance of the maxima is connected with the realization of a periodic arrangement of the antiphase boundaries parallel to (100) and (010). The period of alternation of APBs in the Cu − 21 at. % Pd alloy is greater than in Cu − 29.8 at. % Pd. In [102], a study was made of the structure of conservative APBs in Cu − 22 at. % Pd and Cu 25.2 at. % Pd having a ordered unidimensional long-period structure based on $L1_2$. In contrast to the available literature data on the diffuse character of the boundaries in Cu−Pd alloys with a palladium content lower than 26 at. %, researchers in [102] observed sharp boundaries with random jogs. The number of jogs depended on the composition of the alloy. Later studies [104-106] gave considerable attention to this matter. The method of dark-field electron-microscope images (with the use of the 100, 010, and 110 reflections) was used in [104] to study broadening of the boundary of an APB near the order−disorder transition temperature. In the one-phase region of the ordered phase of Cu − 17 at. % Pd, the degree of broadening of the APB changed little with temperature and remained less than 7 nm. The authors of [107] studied the mechanism of formation of a two-dimensional ordered structure in the alloy Cu_3Pd. A columnar LPS developed on the basis of the unidimensional LPS as a result of the formation of a series of parallel nonconservative APBs in the direction perpendicular to the axis of alternation of the boundaries that existed in the unidimensional LPS. An analysis of one- and two-dimensional LPSs in Cu_3Pd alloys containing 17.3-30 at. % Pd [37] revealed that the half-period M of the superstructure changes not only with a change in composition, but also with temperature (decreasing with an increase in temperature). In Cu_3Pd, a change in the concentration of palladium from 18 to 27 at. % led to a change in the mean size of the antiphase domain from 12 to 3 [40]. The values of M were disproportionate to the base superstructure when palladium content was low. An increase in palladium led to a decrease in M, while at high concentrations M had values of 4.5, 3.5 and 3, corresponding to proportionate structures. The authors emphasized the existence of four ordered fcc-based phases near the composition Cu_3Pd:

a) a cubic $L1_2$ superstructure (Cu_3Au type) realized at low palladium concentrations (to ~ 18 at. %); this superstructure is also present as a low-temperature phase throughout the concentration interval;

b) a unidimensional tetragonal LPS with conservative APBs, seen within the composition interval 18-28 at. % Pd;

c) a two-dimensional orthorhombic LPS that exists within a narrow temperature-concentration interval in the palladium-rich region of the unidimensional LPS;

d) a unidimensional LPS with nonconservative APBs, located between the two-dimensional LPS and the solid solution.

In studying the patterns detected by high-resolution microscopy, researchers [37, 99, 100] have made special mention of the fact that no information on the probabilities of substitution at lattice points along APBs can be obtained if no allowance is made for relaxation effects. The same investigations showed that the two atomic planes closest to the respective APBs have some disordering or small fluctuations in positions. This situation is very close to that seen in Cu−Au alloys. It was emphasized that the given phenomenon is an integral characteristic of unidimensional LPSs. As suggested in [101], the transition "superstructure $L1_2$ − LPS" is a first-order transition.

The disordering of the alloy $Cu - 40$ at % Pd, with the reverse transformation of a bcc lattice back into an fcc lattice, can occur by the martensitic mechanism with the formation of an ordered metastable intermediate phase of the type CuAu II. This phase forms in the transitional two-phase region [108]. Evidence of this comes from the four split 110 reflections that are seen. The degree of tetragonality of the alloy in this case is 0.97. The orthorhombic lattice includes four fcc unit cells. The suggestion was made that a structure of the type CuAu II might be stabilized as a result of heat treatment or alloying. An anomalous increase in the rms displacements of atoms and a nonmonotonic temperature-related change in the lattice parameter were observed in an alloy of this composition [109] prior to $A1-B2$ phase transformations.

The authors of [10, 11] examined the effect of nonstoichiometry of the matrix on ordering and decomposition in the alloy $Cu - 30$ at. % $Pd - 10$ at. % Ag. As in the corresponding binary $Cu-Pd$ alloy, an increase in the content of palladium in the ternary alloy reduces the period of the antiphase domain structure to seven. Stratification of the solid solution, initiating the precipitation of another phase in the disordered layers between the domains, occurs to a considerably lesser extent in this alloy than in stoichiometric alloy $Cu-Au-Ag$.

An analysis of empirical data [84] led to the conclusion that in alloy Cu_3Pd (α'') $(25.8 \pm 1$ at. % Pd), the palladium (heavier) atoms are displaced toward the APB, while the copper (lighter) atoms are displaced away from it. The behavior of the atoms in this alloy differs from the behavior of the atoms in Au_3Cd and CuAu II, in which the heavier atoms are moved farther from the APB and the lighter atoms closer. Thus, the directions and magnitudes of the atomic displacements are not determined by the masses of the atoms that comprise the alloy.
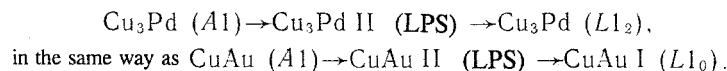
Let us briefly discuss disordering near APBs. Some disordering near APBs on three atomic planes filled with atoms of a different species was seen in [84]. We will henceforth refer to such planes as mixed planes. During the disordering of the alloy, palladium atoms preferentially change places with copper atoms in the mixed planes. The probability of substitution of a copper atoms in unmixed planes changes negligibly.

In studying the structure of alloys near the composition Pd_3Mn [83, 112], the authors of [83] found that with a manganese content lower than 24 at. %, the alloy has an fcc lattice without periodic APBs. Within the concentration interval 24-80 at. % manganese, the alloy is seen to have a unidimensional LPS with a period equal to four lattice parameters of the base superstructure. In this case, the periodic displacements are similar to the displacements in alloy Cu_3Pd (α''). The magnitude of the displacements is ~ 0.01 Å for atoms of both species. Both x-ray and neutron diffraction studies were made of the alloy. The neutron diffraction studies were conducted to answer the question as to whether or not the nuclei of the atoms are displaced together with the associated electron clouds. However, no definitive answer was obtained in this regard.

We should also note that while the lattice is elongated in Au_3Cd and CuAu II alloys during the formation of the LPS, it undergoes contraction in Cu_3Pd (α'') and Pd_3Mn. The maximum displacements of the atoms and the greatest disordering (if such occurs) are seen near the APB and decrease rapidly with increasing distance from it [15].

Several studies (see [88], for example) have confirmd that the presence of periodic displacements of atoms is one feature of ordered long-period structures. Meanwhile, we note the experimental fact that the magnitude of the relative displacements of atoms in LPSs in the direction of the long period is proportional to the ratio of the atomic radii of the components of the alloy.

It was found in [41] that the following series of transformations occurs with a decrease in temperature:

$$Cu_3Pd \ (A1) \rightarrow Cu_3Pd \ II \ (LPS) \rightarrow Cu_3Pd \ (L1_2),$$

in the same way as $CuAu \ (A1) \rightarrow CuAu \ II \ (LPS) \rightarrow CuAu \ I \ (L1_0)$.

We emphasize that this sequence is more general than the sequence considered earlier.

While there is a wealth of literature data on the systems $Cu-Au$ and $Cu-Pd$, the information available on other systems is only fragmentary.
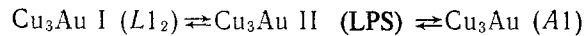
A long-period structure is realized in alloys of the system $Cu-Pt$. A LPS is formed in the region of the composition Cu_3Pt [113-120] as a stable phase within a certain temperature and concentration interval (from 24.5 to 26 at. % Pt at 827 K). The APB lies in $\{100\}$. The low-temperature phase has the superstructure $L1_2$. It was noted in [117] that Cu_3Pt with the structure $L1_2$ has very low phase stability relative to the superstructure DO_{22}. It was concluded that the low phase stability relative to the long-period phases is responsible for the positive temperature dependence of the strengthening of ordered alloys with the superstructure $L1_2$.

The sequence of transformations $L1_2 \rightleftarrows LPS \rightleftarrows A1$ is seen with variation of temperature, the phase transformation $L1_2-LPS$ having been determined to be of second order [118, 119]. The behavior of alloy Cu_3Pt with an increase in
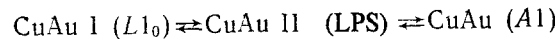
temperature is close to the behavior of Cu_3Au [17, 113]. The transformation LPS−A1 is close to second order. The size of the antiphase domain changes within the interval from 8.5 to 4.5 [17, 120].

It is apparent that a long-period structure of the first type is also realized in Pd_3Mn [121], Au_3In [122], and others.

Let us briefly focus on the effect of temperature and external pressure on LPSs of the first type. The low-temperature phase is realized in the form of a base superstructure − such as $L1_2$ or $L1_0$ − that can be designated as a LPS with the domain $M = \infty$ [17, 41, 118]. A sequence of transformations of the type

$$Cu_3Au \ I \ (L1_2) \rightleftharpoons Cu_3Au \ II \ (LPS) \rightleftharpoons Cu_3Au \ (A1)$$

or

$$CuAu \ I \ (L1_0) \rightleftharpoons CuAu \ II \ (LPS) \rightleftharpoons CuAu \ (A1)$$

is seen with variation of temperature. In the course of the temperature increase, the disproportionate LPS continuously decreases the average size of the antiphase domain. The value of $\bar{M}$ decreases continuously from ~ 8.8 to ~ 8.4 in Cu − 32.3 at. % Au within the one-phase region of the LPS Cu_3Au II [17, 123].

The analogous interval in Cu_3Pt includes values of $\bar{M}$ from ~ 8.5 to ~ 4.5 [17]. In alloys of the system Cu−Pd, the region of values differs appreciably according to composition [37, 100]. For example, for the alloy Cu − 20 at. % Pd, with a unidimensional LPS, the variation takes place within the interval 8-9 [17]. It was noted in [37] that $\bar{M}$ is substantially dependent on temperature. For example, for Cu − 19.3 at. % Pd, it varies as follows: $M = \infty$ at 350°C, $\bar{M} = 14.3$ at 400°C, and $\bar{M} = 10.1$ at 500°C.

As regards alloys of the system Cu−Pd, it was pointed out in [37] that the APBs are of a "diffuse" nature within the region of the unidimensional LPS (18-21.3 at. % Pd) and do not lie precisely within the planes {100}, while $\bar{M}$ changes continuously with temperature and composition, i.e. these LPSs are disproportionate. At higher palladium contents (21.3-30 at. %), the APBs are well-defined and lie exactly within {100}, while the half-period has rational values, i.e., M = 3, 3.5, 4.5, ... The value of M is independent of temperature within this concentration interval, and the changes in composition are discrete in character. Thus, proportionate LPSs are seen within the given interval.

A disproportionate LPS is realized at low palladium concentrations (18.5-21 at. %) and temperatures below the order−disorder transition, while the APBs are of a wave-like character and finite thickness [38, 101]. The appreciable smoothing of the LPS modulation profile with an increase in temperature was emphasized in [101].

The effect of pressure on the state of alloys with LPSs has been studied only in the alloy CuAu [124, 125]. It was found that pressure increases the order-disorder transition temperature. The long period of the structure of CuAu II increases with pressure: $\bar{M} = 5.1$ at 1 bar and 5.5 at 50 kbar. The structure of CuAu II becomes unstable at pressures above 50 kbar and only CuAu I exists. No other structures have been discovered except for those stable at atmosphere pressure. The significant effect of pressure is manifest in the fact that the phase CuAu II appears at higher temperatures with an increase in pressure, while the temperature interval within which it exists narrows.

In conclusion, let us briefly mention the features of alloys with LPSs of the first type:

1) a low-temperature phase is realized in the form of a base superstructure (such as $L1_2$ or $L1_0$, i.e., $M = \infty$);

2) the LPS is an equilibrium disproportionate structure within the temperature region below T_c. The half-period is generally $\bar{M} \geq 5$;

3) the dependence on external conditions (composition, pressure, alloying, temperature) is substantial. The mean size of the antiphase domain changes continuously with changes in composition and temperature. It decreases with an increase in temperature to T_c and a change in the composition of the alloy to equiatomic;

4) relaxation effects − local changes in composition, variation of order, lattice modulation − are seen in the neighborhood of periodic APBs;

Let us examine the features of alloys with the LPSs of the second type. These LPSs are sometimes referred to as "rational structures" and are found in such well-studied alloys as Ag_3Mg and Cu_3Al (α'') [40, 126-138]. Variation of the composition of these alloys is accompanied by a discrete change in the long period.

Alloys Ag_3Mg, with Mg contents of 22, 23.5, 24, and 25 at. %, were studied in [131]. In the interval from 20 to 22 at. % Mg, the half-period of the antiphase remains a constant M = 2. The latter figure corresponds to the superstructure DO_{23}. Some researchers [132] place this superstructure within the interval 20-28 at. % Mg, while others [133] believe that $\bar{M}$

continuoussly decreases from 2 to 1.67 with an increase in Mg concentration from 22 to 28% at. %. The continuous change is determined by averaging over a set of domains of different sizes (M = 1 and 2). The mixture of domains should be uniform, since only such uniformity could produce distinct antiphase reflections of the type observed experimentally [131]. However, the distribution is not regular, since there is a certain degree of disorder in the sequencing of the set of domains M = 1 and 2 along the antiphase direction. Nevertheless, the antiphase boundaries are clearly established planes.

The most important result reported in [131] was that the ordered phase with the $L1_2$ superstructure was never seen.

The main difference between the x-ray diffraction patterns of Ag_3Mg and the standard distribution of LPS patterns of the first type (such as Cu_3Au) is that the satellites around the principal maxima are much weaker. The superstructure reflections are split into a certain set which is comparable to the set of antiphase domains [131]. The following was obtained in [131] with a change in composition: 22 at. % Mg − M = 2; 23.5 at. % Mg − $\bar{M}$ = 13/7 = 1.86; 24 at. % Mg − M = 1.8; 25 at. % Mg − $\bar{M}$ = 1.75. High-resolution electron microscopy was used in [134, 135] to obtain a similar result in alloys of the Ag−Mg system with an Mg content of 22-27 at. %. Researchers found a mixture of structures with M = 1 and 2, so that M ceased to be an integral value. The latter fact is the basis for regarding the actual structure as disproportionate. With heating to 325°C, we observed a restructuring that led to the formation of a alternating pattern of boundaries corresponding to a proportionate structure with M = 2.

It was emphasized in [136] that $\bar{M}$ always has rational values. For example, the values 10/6, 7/4, 32/18, 18/10, 26/14, and 4/2 were found experimentally with a decrease in concentration from 26.5 to 22 at. % Mg. It was determined that Au_4Zn, Au_3Cd, and Au_3Mn are similar types of alloys [136]. In a study of the ordered structure of Ag_3Mg − which is stable within the concentration region 22-27 at. % Mg at temperatures below 370°C − it was found [137] that there is a series of long-period superstructures which are stable within a very narrow concentration interval. Each phase corresponds to its own value of M, so that $\bar{M}$ changes continuously with a change in concentration.

The method of high-resolution transmission electron microscopy was subsequently [138] used to confirm the above findings. The structure $<2^5 1>$ was obtained on an alloy with 24 at. % Mg at 384°C (this notation denotes the sequence of domains 222221, where 2 and 1 represent the domains M = 2 and 1). In terms of disproportionate structures, this corresponds to $\bar{M}$ = 1.83. A structure similar to 2H was observed at 219°C. The mean value of $\bar{M}$ was ∼1.82, while mathematical averaging gave ∼1.89. There is clearly no unambiguous temperature dependence, but it was found in [131] that the transformation Ag_3Mg (A1) → Ag_3Mg takes place within the composition interval 20-28 at. % Mg (monoperiodic LPS). It was emphasized in [15] that the transition DO_{23} → A1 occurs with an increase in temperature. In addition, $\bar{M}$ increases from ∼1.92 to ∼2.0 with a decrease in long-range order [17].

A similar series of proportionate structures and similar results were obtained in [122, 139-143] for Au_3Zn.

The clearest results have been obtained for alloys Al_3Ti [143-146]. Transmission electron microscopy was used to study Al_3Ti alloys with an Al content of 71-73 at. % [143]. High- and low-temperature phases with long-period structures were detected. Several relatively simple proportionate structures were observed at low temperatures, while several relatively complex configurations were identified at high temperatures. For example, the configuration $<211>$ was seen at T = 700°C. In terms of disproportionate structures, this corresponds to $\bar{M}$ ≈ 1.33. At T = 900°C, the configuration $<21>$ was seen at T = 900°C and, accordingly, $\bar{M}$ ≈ 1.5; the corresponding results for T = 1150°C and 1200°C were $<221>$, $\bar{M}$ ≈ 1.68 and $<22221>$, $\bar{M}$ ≈ 1.76. It is emphasized that $\bar{M}$ clearly increases with temperature, while the low-temperature phase is realized in the form of a mixture of antiphase LPSs M = 2 and M = 1. Elements of the superstructure DO_{22} (M = 1) are clearly dominant.

The simple long-period structure DO_{22} was seen in the alloy of stoichiometric composition. Structures with 4/3 ≤ $\bar{M}$ ≤ 2 were seen at concentrations within the range 69-73 at. % Al, with $\bar{M}$ depending on composition and temperature. At T < 900°C, the value of $\bar{M}$ was characterized by the rational ratio 4.3. At T > 1000°C, the value of $\bar{M}$ could be either rational (5/3; 7/4) or more complex (37/23, for example). The dependence of $\bar{M}$ on composition and temperature was also noted in [145, 146].

An increase in the antiphase period in Au_4Zn alloys in which a two-dimensional LPS is formed was observed with a decrease in the degree of order [17] − which in the final analysis corresponds to an increase in temperature.

In analyzing high-resolution images, the authors of [146] pointed out that the APBs present in Pt_3V and Al_3Cu have the form of rectangular jogs. They encompass the entire atomic plane in Al_3Ti and extend over several atomic planes in Cu_3Pd. The LPSs in alloys of the system Al−Ti are a set of proportionate structures with an APB "width" that is ∼0.2 lattice parameters for the temperature interval 1150-1200°C.

In [147], a study was made of Pt−V alloys with a LPS at compositions close to Pt_3V. The ordered structure DO_{22} (Al_3Ti type) was transformed into an ordered structure of the $L1_2$ type during annealing at 930°C, while it became a disordered

state at 1036°C. It was noted in [148] that the high-temperature (higher than 1000°C) phase has the structure $L1_2$, while the low-temperature (lower than 900°C) phase has the structure DO_{22}. An increase in M was seen with an increase in temperature.

A structure in which the mean distance between antiphase boundaries $\tilde{M}$ changed from 4 to 5 periods DO_{22} of the lattice was seen in [149] in Cu−Al alloys with 20-24 at. % Al. The value of $\tilde{M}$ increased with an increase in aluminum content. In an alloy with $\tilde{M} \approx 4.3$, the relative number of domains with M = 5 differed in different regions of the specimen. Several modulated long-period antiphase structures based on the superstructure $L1_2$ were observed in later studies [150, 151]. The mean distance $\bar{M}$ between the APBs, expressed in the lattice parameters of the superstructure $L1_2$, was equal to 4/3. Long-wave structures with the periods 9/7 and 5/4 are also formed, these being proportionate modulated structures.

Ending our examination of alloys with LPSs of the second type, we briefly summarize their features:

1) a low-temperature phase is realized in the form of a long-period structure that is proportionate to the base superstructure (for example, DO_{22} or DO_{23} in relation to $L1_2$);

2) the high-temperature state is formed as a regular set (if "disproportionate" LPSs are formed) of proportionate structures that are well-resolved in diffraction patterns obtained in structural studies;

3) the "mean" size of the antiphase domain takes on rational values with a change in the composition of the alloy and temperature. The value of $\bar{M}$ tends to increase with temperature. As a rule, $\bar{M} \leq 2.5$ in the disproportionate representation.

4) no significant relaxation effects have been observed in the neighborhood of periodic APBs.

Thus, two types of alloys can clearly be distinguished among alloys with a long-period structure. These types differ in both structure and behavior with variation of composition and temperature.

CONCLUSION

Our analysis of experimentally observed features of ordered alloys with a long-period structure established the presence of at least two different types of LPSs. The first type is seen in alloys of the system Cu−Au and in Cu_3Pd and Cu_3Pt. Their most important characteristics are as follows:

− the low-temperature phase is realized in the form of a base superstructure (such as $L1_0$ or $L1_2$). A disproportionate state characterized by a decrease in the mean size of the antiphase domain is seen in the high-temperature region near T_c;

− lattice modulation, redistribution of the components, and segregation of alloying elements are seen in the long-period state. The characteristics of these features undergo the largest deviations from the means near periodic APBs;

− long-period structures exist within a wide range of concentration intervals; here, the long period changes continuously.

The second type of LPS is seen in alloys Al_3Ti, Ag_3Mg, and Cu_3Al. This type also has several characteristic features:

− the low-temperature phase is realized in the form of a proportionate superstructure (DO_{22}, DO_{23}, etc.). The high-temperature state is a mixture of proportionate elements and is sometimes accompanied by an increase in the mean half-period with an increase in temperature;

− the antiphase boundaries are distinct planes, with no significant structural changes occurring in the nearby regions;

− several mutually transforming proportionate structures are seen with a change in the composition of the alloy; here, half the long period of the mixed state is an irrational number.

It can be concluded on the basis of the above analysis that long-period states of the first and second types are different in nature.

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