

Multilayer Composite Epitaxial CuCo Single Crystals

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Abstract—The ion-plasma spraying method was used to synthesize new phases of metastable atomic-ordered layered CuCo single crystals and single crystals of $\text{Cu}_c\text{Co}_{1-c}$ solid solutions via epitaxial layer-by-layer crystallization, and some of their physical properties were studied. © 2000 MAIK “Nauka/Interperiodica”.

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The study of layered compositionally modulated crystal structures exhibiting promising solid-state properties is one of the “hot” areas of physical metallurgy. The nature of magnetic ordering and the giant magnetoresistance of the artificial periodic superlattices comprised of alternating magnetic and nonmagnetic layers generate considerable interest among researchers. At present, it is generally believed that the transition-metal layers in multilayer systems are magnetically ordered, while the interlayer exchange interactions, e.g., of the RKKY type, can only result in oscillations of the ferromagnetic or antiferromagnetic ordering, depending on the thickness of the nonmagnetic layers [1, 2]; i.e., the multilayer effect on the intralayer exchange interactions is *a priori* ignored. The experimental results obtained in our work [3, 4] allow one to suggest a somewhat different point of view on the nature of magnetic ordering, at least in multilayer monatomic polycrystalline $(\text{Cu}/\text{Co})_x$ structures.

Unexpected results were obtained when we synthesized artificial epitaxial composite multilayer $(\text{Cu}/\text{Co})_x$ crystal superlattices to continue our experiment. The crystal structures revealed the cooperative atomic ordering phenomenon.

Single-crystal multilayer film structures $(n_1\text{Cu}/n_2\text{Co})_x$, where n_1 , n_2 , and x are the number of monatomic Cu layers, the same for Co, and the number of bilayers, respectively, were grown via the epitaxial technique through layer-by-layer crystallization on the MgO and LiF (001) crystal surfaces obtained by cleaving bulky single crystals along the atomic cleavage planes. The ion-plasma spraying technique was applied to deposit metals on supports heated to the epitaxial temperatures.

The results of the X-ray structural and magnetic studies carried out in this work can be generalized by

the following epitaxial relation between the MgO, LiF, Cu, and Co crystal lattices:

$$\text{FCCMgO}(001)[001] \parallel \{ \text{FCC}n_1\text{Cu}(001)[001] \parallel \times \text{FCC}n_2\text{Co}(001)[001] \}_x, \quad (1)$$

$$\text{FCCLiF}(001)[001] \parallel \{ \text{FCC}n_1\text{Cu}(001)[001] \parallel \times \text{FCC}n_2\text{Co}(001)[001] \}_x. \quad (2)$$

X-ray structural studies were performed by X-ray diffractometry on a DRON-4 spectrometer at room temperature with $\text{CuK}\alpha$ radiation.

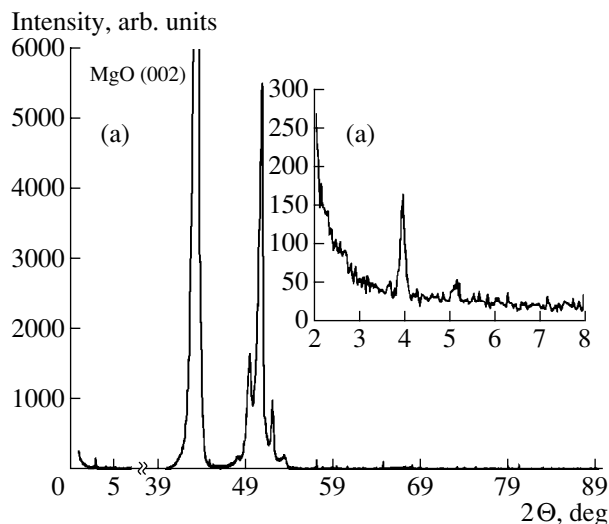


Fig. 1. X-ray pattern of a multilayer $[\text{Cu}(36 \text{ \AA})/\text{Co}(38 \text{ \AA})]_{30}$ single crystal ($\text{CuK}\alpha$, room temperature): (a) is the small-angle scattering region, and MgO(002) indicates the reflection from a single-crystal support.

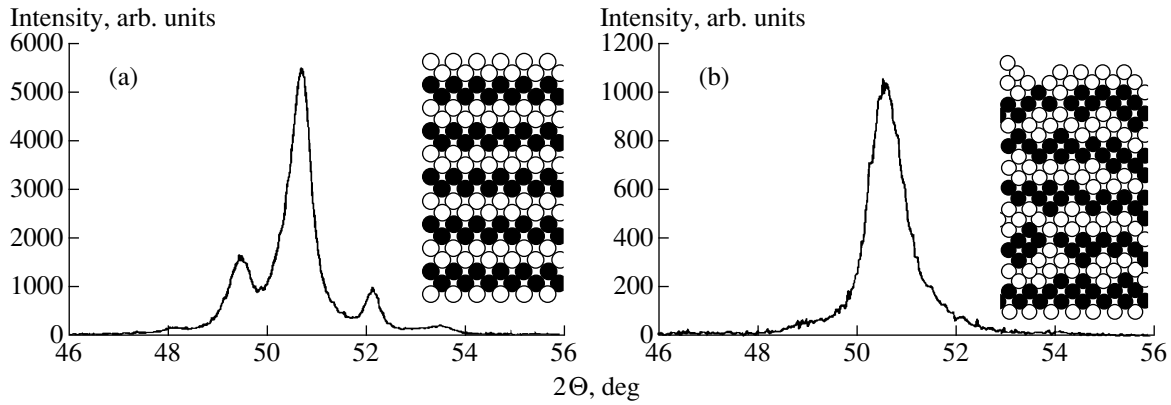


Fig. 2. Portions of the X-ray patterns of two multilayer single-crystal samples ($\text{CuK}\alpha$, room temperature, large-angle scattering region): (a) $[\text{Cu}(36 \text{ \AA})/\text{Co}(38 \text{ \AA})]_{30}$ and (b) $[\text{Cu}(28 \text{ \AA})/\text{Co}(25 \text{ \AA})]_{30}$. The interfaces between the layers (fragments of computer simulation) are schematically represented in the insets illustrating the distinctions in X-ray diffraction.

The superstructure composition modulation periods were determined from the superstructure diffraction reflections for the small (small-angle scattering) and large (large-angle scattering) X-ray scattering vectors, as well as by the X-ray fluorescence method. The positions of diffraction peaks in the X-ray patterns fit the following relationships.

Small-angle scattering:

$$\sin \Theta = \frac{\lambda_0}{2\Lambda} N \quad (\text{principal maxima}), \quad (3)$$

$$\sin \Theta = \frac{\lambda_0}{2\Lambda} \left(N + \frac{2n+1}{x} \right) \quad (\text{subsidiary maxima}), \quad (4)$$

where λ_0 is the radiation wavelength, Θ is the Bragg angle, Λ is the superlattice spacing, and N and n are the reflection periods.

Large-angle scattering:

$$\sin \Theta = \frac{\lambda_0}{2d} N \quad (\text{central maximum}), \quad (5)$$

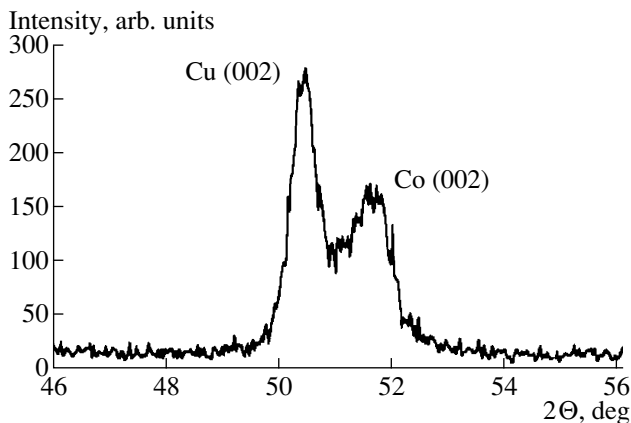


Fig. 3. X-ray diffraction spectrum of a two-layer composite single crystal $\text{Cu}(680 \text{ \AA})/\text{Co}(580 \text{ \AA})$.

$$\sin \Theta = \frac{\lambda_0}{2} \left(\frac{N}{d} \pm \frac{k}{\Lambda} \right) \quad (\text{satellites of the central maximum}), \quad (6)$$

$$\sin \Theta = \frac{\lambda_0}{2} \left(\frac{N}{d} \pm \frac{2n+1}{\Lambda x} \right) \quad (\text{subsidiary maxima}), \quad (7)$$

where d is the mean interplanar spacing and k is the order of reflection.

It is customarily believed (theory and experiment) [5–7] that the position of the central peak in the X-ray patterns of the multilayer superstructures corresponds to the mean d value (weighted mean of the number of individual lattice planes). This rule is violated for the structures considered in this work. The position of the central peak corresponds to the interplanar spacing d_{002} in a pure copper single crystal (Figs. 2, 3). This and other results of our X-ray measurements, as well as the results of the magnetic, galvanomagnetic, electrical, and thermal studies allow the following conclusion to be drawn. The layered atomic-ordered single-crystal CuCo structures, metastable at room temperature (on vacuum heating to 800°C , the metastable phase converts into a stable two-phase structure representing a mixture of pure copper and cobalt phases, in accordance with the phase diagram for the cobalt–copper alloy), were grown by the epitaxial technique. We associate the metastability of the close-packed FCC crystal structure of CuCo with a peculiar (excited) electronic state of the Co atoms, which is experimentally manifested as an increase in the metallic radius of 2%.

We also synthesized and studied the structures with atomic disorder (single crystals of solid solutions). Such structures can be prepared by the epitaxial layer-by-layer crystallization of alternating Cu and Co layers with effective thicknesses of less than one monatomic layer. The concentrations of components in the alloy are experimentally predetermined by the ratio of effective thicknesses. According to the phase diagram of a cobalt–copper alloy prepared by cooling (including

superfast cooling [8]) liquid solutions, the solid solutions synthesized in our work cannot exist. Similar to the layered single crystals described above, the metallic radius of Co in the single crystals of CuCo solid solution is 2% larger than in a pure cobalt phase. The solid solution is thermodynamically unstable (metastable). On vacuum heating, the solid solution is layered into pure Co FCC and Cu FCC phases.

Single crystals of intermediate structures were also prepared and studied. They can be regarded as partially disordered layered single crystals or partially ordered solid solutions. The physical properties of such structures are widely diversified and can even be quite unusual.

The general regularity (sequence) of the structural transformations during the heating \longleftrightarrow cooling thermal cycle (with the retention of the close-packed single-crystal FCC structure) is as follows: the metastable structure with atomic disorder ($\text{Cu}_c\text{Co}_{1-c}$ solid solution) \longrightarrow metastable atomic-ordered structure [layered $(\text{Cu/Co})_x$ single crystal] \longrightarrow two-phase stable structure (composite single-crystal Co + Cu).

The layered single crystals are ferromagnetic with cubic magnetocrystalline anisotropy in the (001) plane. For the cobalt layers of thickness 15 Å and over, the saturation magnetization I_s and the first magnetocrystalline anisotropy constant K_1 are close to the analogous values for a pure cobalt FCC single crystal (per the same cobalt amount in the CuCo structure). For cobalt layers of thickness less than 15 Å, the ferromagnetic ordering depends on the thickness of the copper layer [3].

The room-temperature transverse magnetoresistance anisotropy $d\rho/pdH$ is positive for the layered CuCo FCC single crystals, whereas the anisotropy of the Co FCC single crystal has negative sign.

Single crystals of the $\text{Cu}_c\text{Co}_{1-c}$ solid solutions are ferromagnetically ordered and magnetically anisotropic over a wide range of copper concentrations c . The saturation magnetization I_s is a linear function of copper concentration. The atomic magnetic moment μ changes as

$$d\mu/\mu_b dc = 2; \quad (8)$$

i.e., the $\text{Cu}_{0.86}\text{Co}_{0.14}$ alloy is nonmagnetic at any temperature [expression (8) was obtained by extrapolating

the experimentally measured dependence of the room-temperature saturation magnetization I_s on the copper concentration c to the temperature $T = 0$]. The experimental result (8) agrees well with the theoretical model of two "hard" bands, one of which is only partially filled [9].

The magnetocrystalline cubic anisotropy in the (001) plane depends on the copper concentration, in accordance with the relation $K_1 \approx I_s^2$. The results obtained in this work for the $\text{Cu}_c\text{Co}_{1-c}$ alloys are similar to those previously obtained for the $\text{Cu}_c\text{Ni}_{1-c}$ alloys exhibiting infinite mutual solubility [10].

We plan to systematically study the synthesized new materials in future works.

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