Hindawi
Journal of Nanomaterials
Volume 2022, Article ID 2862439, 5 pages
https://doi.org/10.1155/2022/2862439

Research Article

Strain Properties of Multicomponent Nanosize Film Materials

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Received 28 June 2022; Revised 16 August 2022; Accepted 9 September 2022; Published 12 November 2022

Academic Editor: Leander Tapfer

The results of the correlation between the longitudinal strain coefficient (SC) and the values of the Poisson coefficient and Grüneisen parameter for nanosize film multicomponent alloys ($d \cong 30$–50 nm, $c_i \cong 11$–20 at%) are presented. It is established that in the region of elastic or quasi-elastic deformation (less than or equal to 0.4%), the value of SC is insensitive to changes in the Grüneisen parameter in the range of 1.5–2.5 units during the transition to plastic formation (more than 0.4%). The value of SC decreases from 4–5 units to 3–1 units. Similarly, SC is insensitive to changes in the Poisson coefficient less than or equal 0.4 but decreases sharply in the region of plastic deformation. It is concluded that a sensitive element based on a multicomponent nanoscale film of a solid solution, including a high-entropy one, has advantages over others due to phase stability in a wide temperature range. These materials may be used to develop the architecture of sensitive elements of sensors for various functional purposes.

1. Introduction

Considerable attention of researchers to the study of the strain effect in film materials is due to the possibilities of their practical application for the sensor-sensitive elements formation with improved operating characteristics (see, e.g., [1–5]). Sensitive elements are formed based on nanoparticles [1], granular solid solutions [4], metal films [6], nanocomposits [7], and multicomponent film materials [8]. The strain properties of single- and multilayer metal films have been studied in considerable detail (see, e.g., our generalization ref. [8]), and only some aspects of the problem need to be clarified (e.g., the temperature dependence of the longitudinal (γ_l) or transverse (γ_t) strain coefficient (SC); comparative characteristics of the strain effect in the area of elastic or plastic deformation, etc.). It should be noted that in our case, we are talking about film materials that can be classified as low- or medium-entropy materials (see more details in [9]). They are formed as single-phase films with a lateral phase, the diffraction lines from which are of very low intensity. The polycrystalline structure of the films is characterized by the average crystallite size L, which is approximately equal to the sample thickness d, i.e., $L \cong d$. Figure 1 shows examples of the crystal structure and diffraction patterns for film samples.

Note that, based on the definition of γ_l as a relative change in the film resistance at a single deformation, the basic relationship can be written in the following form:

$$\gamma_l = \frac{d\ln \rho}{d\varepsilon_l} + 1 + 2\mu_f, \quad (1)$$

where $\rho = \sum_{(i)} c_i \rho_i + \rho_r$ is the resistivity of the film material; $\rho_r \ll \rho$ is residual resistivity; $d\varepsilon_l = d\ln l$ is the longitudinal deformation; l is the sample length; μ_f is the Poisson coefficient of the film.

From relation (1), provided that $d\ln \rho / d\varepsilon_l = 0$, the limiting value of SC γ_l^b is equal to 1.5–1.7 at $\mu_f = 0.25$–0.35 under conditions of elastic deformation. Since the experimental values of γ_l are always greater than γ_l^b, the redundant value of γ_l is associated with the first term in relation (1). Its value is determined by the internal (scattering of conduction electrons at grain boundaries and interfaces) and external (electron scattering at external surfaces of the sample) size effects (SE) [8], as well as the deformation dependence of not only the mean free path (MFP) of electrons in the film volume, but also the electrons transmission coefficients of interfaces and the surface scattering coefficient [8].

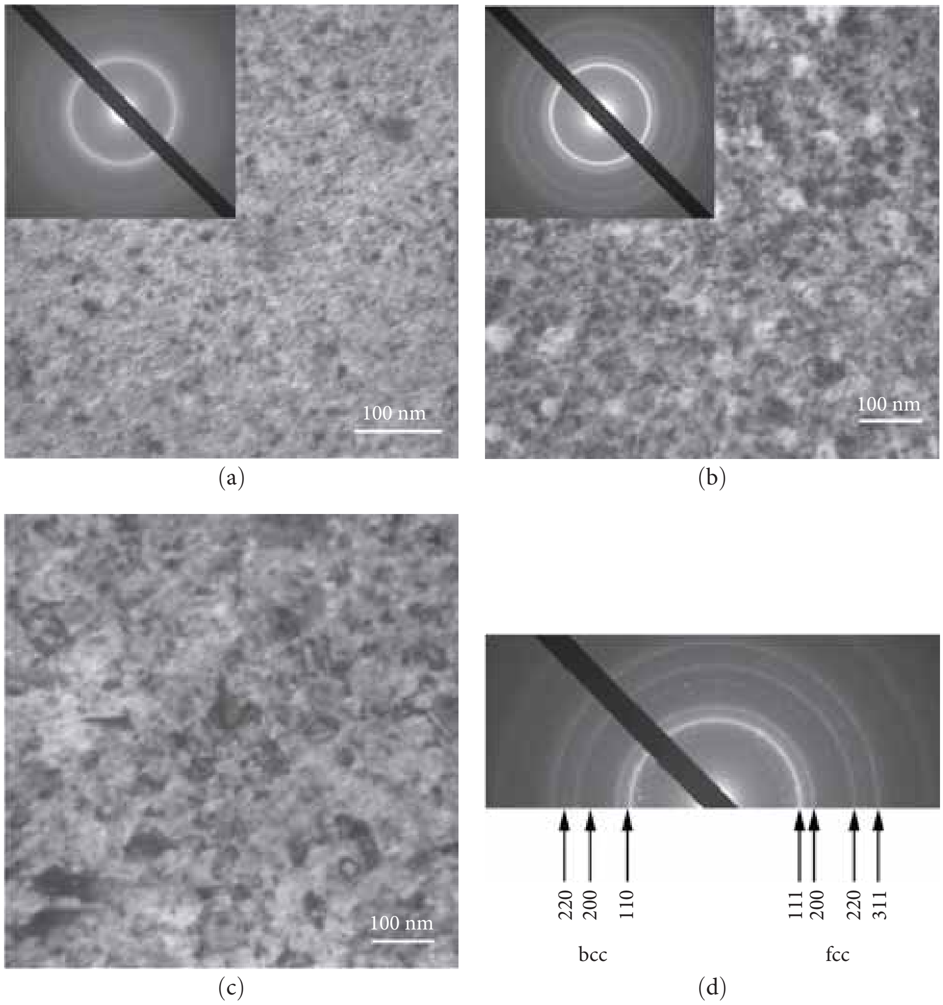

Figure 1: Microstructure and diffraction patterns from sample based on Cr(19), Al(8), Co(21), Cu(12), Ni(19), and Fe(20) (a, b) and based on Cr(18), Al(9), Co(20), Cu(19), Ni(19), and Fe(18) (c, d). The concentration of elements in at% is indicated in parentheses. The sample thickness $d \cong 38$ nm (a, b) and 40 nm (c, d).

At the same time, when analyzing the strain properties of multicomponent nanosize film alloys belonging to medium- or high-entropy solid solutions (s.s.) in the crystalline state, the role of internal and external SE can be minimized and only the deformation dependence of λ_0 can be taken into account. In other words, such a film material can be considered as a single-layer film of a solid solution. This approach is fully justified if film s.s. are obtained by the method of simultaneous condensation of individual components (for more details, see [10]) or by the method of layer-by-layer condensation followed by annealing.

The aim of this work is to develop a method for predicting the strain properties of high-entropy film alloys not based on the individual characteristics of the components, but using the integral characteristics of the alloy, such as the Poisson coefficient (μ_f) or Grüneisen parameter (γ). This opens the possibility to propose the architecture of a strain-sensitive element, temperature, or magnetic field sensors without studying the properties of individual components in detail, but only by calculating the value of the Poisson coefficient $\mu_f = \sum_{(i)} c_i \mu_{fi}$ or the Grüneisen parameter $\gamma = \sum_{(i)} c_i \gamma_i$.

2. Basic Relationships

The addendum $d\ln \rho / d\varepsilon_l$ in relation (1) can be represented in the form:

$$\frac{d\ln \rho}{d\varepsilon_l} = -\left(\frac{d\ln \lambda_0}{d\varepsilon_l} + \frac{d\ln n}{d\varepsilon_l}\right) = 2\frac{d\ln \theta_D}{d\varepsilon_l} + 1, \quad (2)$$

where λ_0 MFP in film volume; n concentration of conduction electrons; $\theta_D = \sum_{(i)} c_i \theta_{Di}$ is the Debye temperature of the film material.

Taking into account Equation (2), ratio (1) can be rewritten as follows:

$$\gamma_l = 2\frac{d\ln \theta_D}{d\varepsilon_l} + 2(1 + \mu_f). \quad (3)$$

Nepijko et al. [4] rewrote the ratio (3) using the Grüneisen parameter (γ) in the following form:

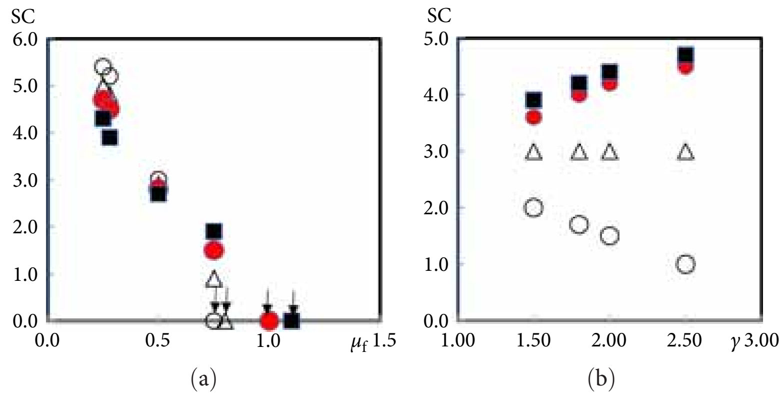

Figure 2: Dependence of SC versus μ_f (a) or γ (b) at: $\gamma = 1.8$ (black squares); 2 (red circles); 2.5 (white triangles) and 2.8 (white circles) and $\mu_f = 0.25$ (black squares); 0.28 (red circles); 0.50 (white triangles) and 0.75 (white circles). Arrows show values μ_{f0}.

Table 1: Literature data on γ and μ_f.

Metal	γ [12]	μ_f [13]	Calculated quantities γ and μ_f based on data [10]
Cu	1.96	0.35	
Al	2.34	0.34	$\gamma = \sum_{(i)} c_i \gamma_i = 1.80$
Cr	1.80	0.20	
Fe	1.66	0.29	$\mu_f = \sum_{(i)} c_i \mu_i = 0.28$
Co	1.87	0.32	
Ni	1.88	0.28	$c_i = (11\text{–}20)$ at%, $d = (30\text{–}50)$ nm

$$\gamma_l = (2\gamma - 4\mu_f \gamma) + 2(1 + \mu_f). \tag{4}$$

Note that, we declare the additivity of γ, μ_f, and θ_D based on the formula of the additivity of the resistivity $\rho = \sum_{(i)} c_i \rho_i$.

This assumption worked successfully when calculating the thermal resistance coefficient

$$\beta_T = \frac{\partial \ln \rho}{\partial T}, \tag{5}$$

and in the calculation of the magnetic coefficient of isotropic magnetoresistance (GMR) β_B^{GMR} for a high-entropy film alloy based on Fe, Co, Ni, Al, and Cu. In both cases, the calculated data agree well with the experimental data [11]. This is the basis for using this approach in the analysis of the strain properties of multicomponent film alloys with the same elemental composition as in the case calculations of β_T and β_B^{GMR}.

3. Calculation of the Dependences of γ_l versus γ or μ_f

The dependences were calculated (Figure 2) using ratio (4). Since there are no data in the literature on the values γ_i and μ_{fi} for single-layer films, we used, assuming a certain error in the calculations, the corresponding values for solid metals (Table 1). Calculated data are presented in Figure 2. Note that, in the case of $\gamma = 1.80$ and $\mu_f = 0.28$, the calculated value γ_l is compared with the experimental value from [10]:

$$\gamma_{lexp} = \frac{\Delta\left(\dfrac{\Delta R}{R(0)}\right)}{\Delta \varepsilon_l} = 4.80 \text{ and } \gamma_{lcalc} = 4.30, \quad \text{that is, } \frac{\Delta \gamma_l}{\gamma_{lexp}} = \frac{\left|\gamma_{lexp} - \gamma_{lcalc}\right|}{\gamma_{lexp}} = 10\%. \tag{6}$$

From the data in Figure 2(a), we can conclude that within the elastic and quasi-elastic deformation $\varepsilon_l \cong 0.4\%$, the Grüneisen parameter has a very insignificant effect on the γ_l. The same conclusion is largely confirmed by calculations of the dependence of γ_l on γ (Figure 2(b)). Since, it follows from Formula (1) that γ_l can be represented by terms, the first of which is responsible for the electronic properties (the so-called internal factor), and the second is associated with a change in the geometric dimensions of the sample (geometric factor), it is theoretically possible that they, acting in opposite directions, can cause zero value γ_l.

Figure 2(a) shows these limits, which denote as μ_{f_0}. The physical nature of zero value γ_l follows Equation (1):

$$\eta_{\lambda_0} + 2(1 + \mu_f) = 0, \mu_{f0} = \frac{\gamma - 1}{2\gamma + 1}. \tag{7}$$

A negative value η_{λ_0} (i.e., the conduction electrons are accelerated during longitudinal deformation) can practically provide $\gamma_l = 0$.

4. Results and Discussion

Based on Equation (1), a point of view was formed long ago that the value of SC is determined by two factors: geometric and internal. The first is associated with a change in geometric dimensions (length, width, and thickness of the sample) and, as already noted, can contribute to the value of SC by no more than 1.7 units under conditions of elastic deformation. It should be borne in mind that during the deformation of the film on the substrate, the Poisson coefficient μ'_{f} is slightly smaller than the corresponding value for the free film μ_{f}. However, from the relation that connects these quantities follows that

$$\mu'_{\mathrm{f}} = \mu_{\mathrm{f}} \frac{1 - \mu_{\mathrm{S}}}{1 - \mu_{\mathrm{f}}}, \tag{8}$$

where μ_{S} is the Poisson coefficient of the substrate material, it follows that $\mu'_{\mathrm{f}} \cong \mu_{\mathrm{f}}$.

The second factor is related to the electronic properties of the film, and the main contribution to, it is given by the deformation coefficient of the MFP as the main tensometric characteristic of the film material. Modern theoretical models of the strain effect (for more details, see [8]) in the theoretical analysis take into account that not only the MFP, but also the specularity coefficients of the outer surfaces of the film and the passage of grain boundaries depend on the deformation. In the case of a multilayer film, it is also necessary to take into account the deformation dependence of the electron transfer coefficient of the interface. It is obvious that this approach does not allow prediction of the value of SC due to the complexity of the analytical relations, which contain a large number of electrical transfer parameters.

In this sense, it can be considered great merit of Kuczynski [14], who proposed a different approach to calculating the value of the internal factor. He related the change in the MFP and the Debye temperature (or the phonon spectrum) during the film deformation. In other words, Nepijko et al. [4] reduced the consideration of complex electronic processes to the consideration of the deformation of the phonon spectrum using the Grüneisen parameter (relation (4)). Despite the fact that Formula (4) does not take into account more subtle effects (changes in the specularity coefficients and the passage of electrons across crystallite boundaries), it is absolutely suitable for a fairly accurate prediction of the value of SC of a high-entropy film alloy. Since the ranges of μ_{f} and γ values are relatively narrow, this simplifies the choice of film materials in the form of high-entropy s.s.

It should be emphasized that sensors based on high-entropy film alloys will be characterized by high-temperature stability. This feature is due to the fact that in a solid solution, atoms whose radii differ by no more than 5% will replace each other isomorphically without changing the phase composition in the process of thermal diffusion.

5. Conclusions

For the first time, the dependence of the SC of multicomponent film materials on the Poisson coefficient μ_{f} and Grüneisen parameter γ in the region of elastic or quasi-elastic ($\mu_{\mathrm{f}} \leq 0{,}4$) and plastic ($\mu_{\mathrm{f}} > 0{,}4$) deformation was performed. In the first case, the value of SC is not very sensitive to changes in the value of γ within the range of 1.5–2.5, while during plastic deformation in the same range of the Grüneisen parameter, the value of SC decreases from 3 to 1 unit. Within the framework of the phenomenological model for SC, the values of μ_{f} were calculated, at which $\gamma_{\mathrm{l}} \cong 0$, that is associated with the mutual competition of the so-called internal and geometric factors. Based on calculations [10], we concluded that the maximum value of SC in multicomponent nanosize film alloys obtained by simultaneous condensation of components can reach a value of 5 units. At the same time, in the case of layer-by-layer condensation of individual components, the value of SC in the region of elastic deformation can reach several tens of units due to the interface scattering of conduction electrons. From the point of view of tensometry, the sensitive element of a sensor based on a multicomponent film alloy, including high entropy, has a clear advantage over other nanoscale film materials, which is associated with their phase stability [10] in a wide temperature range.

Data Availability

The authors declare that all data supporting the findings of this study are available within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Funding

The work was supported by basic funding (No. BF/25-2021) and a grant from the Ministry of Education and Science of Ukraine for 2022–2024 (No. 0122U000785).

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