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**REGULAR ARTICLE**

# Calculation of Magnetization of Multicomponent Metal Alloys: Paramagnetic Approach

I.Yu. Protsenko* ✉, A.K. Rylova, N.I. Shumakova

*Sumy State University, Sumy 40007, Ukraine*

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The method of paramagnetic magnetization ($M$) calculations of bulk equiatomic HEA based on atoms Cr, Cu, Fe, Ni and Co is proposed. The methodology is based on the Langevin theory for paramagnetic materials. Calculations $M$ were carried out during induction $B$ = 1 і 5 T, that correspond to the fields of ordinary ($M$) or saturation magnetization ($M_s$ ). In addition, for such fields, the parameter of the Langevin function ($L(a)$), $a = \frac{\vec{\mu}\vec{B}}{k_B T}$ satisfies the condition $a \ll 1$, which simplifies function to form $L(a) \approx \frac{1}{3}a$. $M$ and $M_s$ have order $10^{-3}$ emu/cm$^3$, but $M_s$ approximately 5 times greater than $M$. The calculated data agree satisfactorily with the literature data obtained for NEAs based on Al, Cr, Cu, Fe, Ni, and Co in the bulk (calculated data) or film (experimental data) state. The proposed technique can be used for preliminary estimation of values $M$ and $M_s$ in the case of other multicomponent alloys.

**Keywords**: HEA, Magnetization, Paramagnetic approach, Average magnetic moment, Langevin function, Saturation magnetization.

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## 1. INTRODUCTION

The increased interest of scientists in studying the properties of multicomponent metal alloys (MCA), including high-entropy alloys (HEA), was connected with the expectation of their unique physical properties. In principle, these expectations have been fulfilled when it comes to mechanical and tribological properties (see, for example, [1, 2]). Along with this research of electrophysical, magnetoresistive and magnetic properties MCA and HEA are still insufficiently represented in the literature. Most likely, we can be talking about episodic studies of these properties, as opposed to studies of mechanical properties. In this regard, the publications [3-5], which present experimental and calculated results regarding the electrical and magnetic properties of HEAs, can be considered pioneering works in this regard: $Al_xCoCrFeNi$ [3] or FeCoNiCr (Cu or Al, or Ti) [4]. In work [5], for the first time, studies of the crystal structure, electrophysical (resistivity, strain coefficient, TCR) and magnetoresistive properties of film HEAs based on Co, Fe, Ni, Cr and Al. Further, these studies were continued by the authors of works [6-8]. It was established [5] that during simultaneous or layer-by-layer condensation of components with subsequent homogenization of the phase composition, a single-phase FCC alloy with a lattice parameter is formed $a$ = 0,3604 nm, although weak lines of the BCC phase based on Fe and Cr are recorded in the diffraction patterns. In addition, the authors [5] observed resistivity values typical for metal and HEAs films $\rho \sim 10^{-6}$ Ohm.m and TCR ~ $10^{-3}$ K$^{-1}$ and increased values strain coefficients compared to single-component films: to the value of SC = 12 [5] or to the SC = 5 units [8].

Measurement of magnetoresistance (MR) [5-7] indicate its anisotropic character with an amplitude of 0,02 % [5] to 0,50 % [6]. At the same time, giant magnetoresistance (GMR) with an amplitude of up to 0,20%. Since GMR is not observed in films obtained by simultaneous condensation of individual components [5,6], then it can be concluded that in the considered case [7] the GMR effect is realized as a result of spin-dependent scattering of electrons at internal interfaces. The authors of this work have prepared a cycle of works [9-12], in which calculated concentration dependences of electronic (resistivity, mean free path of electrons) and dynamic characteristics (lattice parameter HEAs and Debye temperature). It should be noted that the calculations took into account the principle of additivity of physical quantities, which can be correctly applied only in the case of individual, not collective characteristics (size of atoms, their mass, relative change in resistance, magnetic moment of an atom, etc.) (see in more detail [9]). Note that in the case of [7,8] the use of the additivity principle was correct.

Taking into account [10-12] the dependence of the lattice parameter of the solid solution (s.s.) and the Debye temperature ($\theta_D$) from the atomic fraction of the doping element, as which all the elements of the multicomponent alloy are alternately used. It was established that the lattice parameter s.s. changes by an $\Delta a$ = (0,01 – 0,03) nm, and

* Correspondence e-mail: i.protsenko@aph.sumdu.edu.ua





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$\Delta\theta_D = (40 - 80)$ K, if the atomic fraction of the doping element changes in the interval $\Delta x = 0 - 0{,}3$ (5-7 component HEAs based on Fe, Ni, Co, Cu, Al and Ti). In works [11,12] it is proposed to consider the integral and differential concentration coefficients for the lattice parameter and the Debye temperature as a quantitative measure, the value and sign of which allows characterizing the influence of each dopping element on the specified dynamic characteristics.

## 2. METHODOLOGICAL BASES OF PARAMAGNETIC MAGNETIZATION CALCULATIONS

The method of magnetization calculations in the paramagnetic approach was applied to HEAs based on Co, Ni, Fe, Cu and Cr at the equiatomic composition of the main (phase-forming 4 elements) and atomic $x = (0 - 0{,}3)$ doping element, which all 5 components performed in turn HEAs.

Let us indicate that the equiatomicity of the composition HEAs does not allow analyzing the influence of the degree of entropy on magnetization, although the results [13] clearly indicate that the resistivity and TCR are correlated with the entropy value.

The magnetization of HEAs per cm$^3$ (sometimes denoted cc) was calculated according to a ratio similar to Langevin's theory:

$$M = N_A\left[\frac{\rho_1}{A_1}\, c_1\mu_1(\overline{\cos\theta})_1 + \frac{\rho_2}{A_2}c_2\mu_2(\overline{\cos\theta})_2 + \cdots + \frac{\rho_{n-1}}{A_{n-1}}c_{n-1}\mu_{n-1}(\overline{\cos\theta})_{n-1}\right] + N_A\frac{\rho_n}{A_n}x\mu_n\,\overline{\cos\theta_n}, \qquad (1)$$

Where $N_A$ is Avogadro's number; $\rho$ – density; $A$ – atomic mass of elements; $c$ and $x$ – atomic fraction (concentration of the main and doping element); $\mu$ – magnetic moment of atoms; $(\overline{\cos\theta})$ – average value $(\cos\theta)_i$ ($\theta$ is angle between the direct external magnetic field and the magnetic moments of other components).

If introduced for consideration $(\overline{\cos\theta})$, which is equal to $\frac{\sum_{i=1}^{m}\overline{(\cos\theta)i}}{m}$ ($m$ – the number of magnetic atoms), is a formula (1) will be simplified to the form:

$$M = N_A\,\overline{\cos\theta}\left[\left(\sum_{i=1}^{n-1}\frac{\rho_i}{A_i}c_i\mu_i\right) + \frac{\rho_n}{A_n}x\mu_n\right], \qquad (1')$$

Where $\overline{\cos\theta} = \frac{\int_{-1}^{+1}\cos\theta e^{a\cos\theta}d\cos\theta}{\int_{-1}^{+1}e^{a\cos\theta}d\cos\theta} = ctg\ a - \frac{1}{a} = L(a)$ is the Langevin function;

$a = \frac{B\bar{\mu}\,\mu_B}{k_B\,T}$ is function parameter $L(a)$;

$\bar{\mu}$ and $\mu_B$ – the average magnetic moment of HEAs atoms in units of the Bohr magneton (i.e., the number of unpaired electrons in atoms) and the Bohr magneton;

$B$ – induction of external magnetic (in our calculations $B = 1$ and 5 T);

$k_B$ – Boltzmann's constant;

$T$ – temperature.

Value $\bar{\mu}$ for HEAs in the equiatomic approach was calculated by the formula:

$$\bar{\mu} = \frac{1-x}{n-1}\sum_{i=1}^{n-1}\mu_i + x\mu_n, \qquad (2)$$

Where multiplier $\frac{1-x}{n-1}$ indicates the equiatomic composition of the main $(n-1)$ elements.

Under the condition $a << 1$ the Langevin function becomes:

$$L(a) \approx \frac{1}{3}a, \qquad (3)$$

which greatly simplifies magnetization calculations.

## 3. CONCENTRATION DEPENDENCE OF MAGNETIZATION

Using the literature data [14, 15] regarding the magnetic moments of atoms, calculations were made of the average magnetic moment of HEAs atoms based on the ratio (2) (Fig. 1). These results allow us to calculate the value of the Langevin function (Table 1). The results of magnetization calculations are presented in Table 2 (for $B = 1$ T) and in Fig. 1 (for $B = 5$ T).

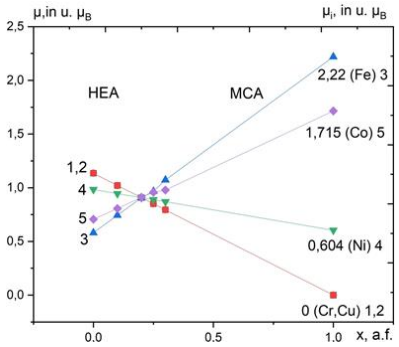


**Fig. 1** – Concentration dependence of the average magnetic moment of HEAs atoms on the atomic fraction of the doping element. MCA – multicomponent alloys

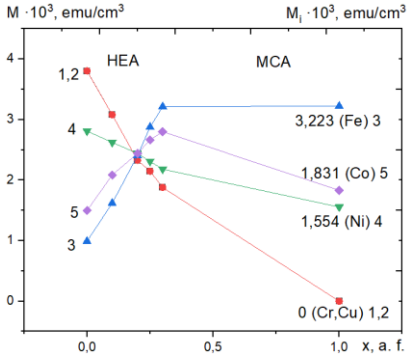


**Fig. 2** – Concentration dependence of M on the atomic fraction of the doping element

Let us point out that the calculated values of the paramagnetic $M_i$ for individual components (Fig. 2) at $x = 1$ were obtained based on the following ratio:

$$M_i = N_A L(a_i^*)\frac{\rho_i}{A_i}\mu_i\mu_B,$$

where parameter $a_i^* = \frac{\mu_i \mu_B B}{k_B \theta_c}$ corresponds to the paramagnetic temperature interval $T > \theta_c$ ($\theta_c$ = 1393 K (Co); 1042 (Fe) and 631 K (Ni) [14, 15]).

**Table 1** – Values of the Langevin function $L(a)\cdot 10^3$ at the $a << 1$

| Doping element | Magnetic field $B$, T | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | 1,0 | | | | | 5,0 | | | | |
| | Atomic fraction, $x$ | | | | | | | | | |
| | 0 | 0,10 | 0,20 | 0,25 | 0,30 | 0 | 0,10 | 0,20 | 0,25 | 0,30 |
| Cr | 0,85 | 0,76 | 0,68 | 0,64 | 0,60 | 4,25 | 3,82 | 3,40 | 3,20 | 3,00 |
| Cu | 0,85 | 0,76 | 0,68 | 0,64 | 0,60 | 4,25 | 3,82 | 3,40 | 3,20 | 3,00 |
| Fe | 0,43 | 0,55 | 0,67 | 0,73 | 0,80 | 2,15 | 2,75 | 3,35 | 3,67 | 4,00 |
| Ni | 0,73 | 0,70 | 0,68 | 0,65 | 0,63 | 3,65 | 3,52 | 3,40 | 3,27 | 3,15 |
| Co | 0,53 | 0,60 | 0,68 | 0,70 | 0,73 | 2,65 | 3,02 | 3,40 | 3,52 | 3,65 |

**Table 2** – Magnetization M $10^3$, emu/(cm$^3$) 5-component HEA based on Cr, Cu, Fe, Co and Ni at $B$ = 1 T

| $x$, u.a. | Cr | Cu | Fe | Ni | Co |
|---|---|---|---|---|---|
| 0 | 0,7605 | 0,7605 | 0,1979 | 0,5619 | 0,2994 |
| 0,10 | 0,6160 | 0,6160 | 0,3240 | 0,5244 | 0,4169 |
| 0,20 | 0,4653 | 0,4653 | 0,4810 | 0,4881 | 0,4880 |
| 0,25 | 0,4295 | 0,4295 | 0,5751 | 0,4618 | 0,5330 |
| 0,30 | 0,3758 | 0,3758 | 0,6775 | 0,4360 | 0,5800 |

The obtained $M_i$ values are plotted in Fig. 2 (right axis). It follows from Fig. 2 that the largest value $M = 3{,}8\cdot 10^{-3}$ emu/cm$^3$ is realized in the case when non-magnetic metals Cu or Cr act as doping elements. Despite the fact that in the presence of three magnetic components, a ferromagnetic state can be realized (see, for example, [3]), it can be asserted that the quantities $M \sim 10^{-3}$ emu/cm$^3$ paramagnetic state and very far from magnetization for ferromagnets. It should be thought that the non-magnetic atoms of Cu or Cr, which are quantitatively in an equiatomic ratio with the magnetic components, act as a kind of blocker of the formation of the domain structure. We also note that the extrapolation of $M$ by the value $x = 1$ allows comparing the values of $M$ for HEAs and individual components, which is important for the analysis of the problem, which can be formulated as low-temperature paramagnetism base on ferromagnetic metals.

## 4. COMPARISON WITH EXPERIMENTAL DATA

Since we start from the assumption that HEAs are in a paramagnetic state, it is very important to compare our calculations with the experimental data of other authors. From this point of view, the most relevant results can be considered those presented in works [3, 6], which present data for HEAs on Al, Cr, Fe, Co, Ni, and Cu. Although Al is not included in the elemental composition of our HEAs, the comparison we made seems quite correct, since Al atoms cannot significantly affect the magnetization. According to the data [3], homogenized HEAs with atomic fraction $x$ = 0 and 0.25 for Al are in a paramagnetic state and have the value $M \cong 0$ emu/cm$^3$ (authors indicated that «not detestable»). More specific data are given in work [6] on the example of film ones HEAs. First, from the above dependencies $M$ від $B$ при 300 K it follows that $M_s = 1{,}32\cdot 10^{-3}$ emu/cm$^3$ at the $B = (0{,}6 – 5)$ T, which allows us to talk about the satisfactory correspondence of our calculated and experimental data [6].

Secondly, the dependencies are given $M$ from $B$ allow to make an assessment $M_s$ in our case by comparing: $M(x, 5\text{ T})/M(x,1\text{ T}) = 5$, that is $M_s$ (300 K;5 T) ≅ 5 M (300 K; 0,6 T).

## 5. CONCLUSIONS

1. Within the framework of Langevin's classical theory of paramagnetism, the calculation of the paramagnetic magnetization of HEAs under the induction of a magnetic field was carried out $B$ = 1 and 5 T.

2. It was established that $M$ and $M_s$ have an order of magnitude $10^{-3}$ emu/cm$^3$, which is consistent with the experimental and calculated data of other authors, who obtained results on the example of a 6-component film system or a bulk HEA.

3. The largest value $M = 3{,}8\cdot 10^{-3}$ emu/cm$^3$ observed in the case of dopping elements Cr or Cu $B$ = 5 T, while at $B$ = 1 T the largest value $M = 0{,}760\cdot 10^{-3}$ emu/cm$^3$.

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## Розрахунок намагніченності багатокомпонентних металевих сплавів: парамагнітне наближення


І.Ю. Проценко, А.К. Рилова, Н.І. Шумакова

*Сумський державний університет, 40007 Суми, Україна*



Запропонована методика розрахунків парамагнітної намагніченості ($M$) масивних еквіатомних HEA на основі атомів Cr, Cu, Fe, Ni Co. В основі методики покладена теорія Ланжевена для парамагнітних матеріалів. Розрахунки $M$ здійснювались при індукції $B$ = 1 і 5 T, що відповідають полям звичайної $M$ або намагніченості насичення ($M_s$). Крім того, при таких полях параметр функції Ланжевена ($L(a)$), $a = \frac{\vec{\mu} B}{k_B T}$ задовольняє умові $a \ll 1$, що спрощує функцію до вигляду $L(a) \approx \frac{1}{3} a$. Величини $M$ та $M_s$ мають порядок $10^{-3}$ emu/cm$^3$, але $M_s$ приблизно у 5 разів більша за $M$. Розрахункові дані задовільно узгоджуються із літературними, отриманими для HEAs на основі Al, Cr, Cu, Fe, Ni та Co у масивному (розрахункові дані) або плівковому (експериментальні дані) станах. Запропонована методика може бути використана для попередньої оцінки величин $M$ і $M_s$ у випадку інших багатокомпонентних сплавів.



**Ключові слова**: BEC, Намагніченість, Парамагнітне наближення, Середній магнітний момент, Функція Ланжевена, Намагніченість насиченняю.