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Abstract

We have carried out an experimental investigation of the Heusler Alloy CoFeVAl and its two variants $Co_{1.5}Fe_{0.5}VAl$ and $CoFeVAl_{0.5}Si_{0.5}$ for their potential application in the field of spintronics. Heusler alloys are investigated for their many remarkable properties, including half-metallicity and spin-gapless semi-conductivity. Spintronic technology utilizes the intrinsic spin of an electron for information storage and manipulation in solid state devices. We synthesized these alloys using arc-melting and annealing. All three alloys were found to have cubic crystal structures with varying disorders. The parent alloy CoFeVAl shows a magnetic transition at 65 K. However, after modifying the elemental composition by either replacing 50% of Al with Si or the same amount of Fe with Co the magnetic properties substantially changed. The ferromagnetic to paramagnetic transition temperature increased to 215 K. The doped alloys $Co_{1.5}Fe_{0.5}VAl$ and $CoFeVAl_{0.5}Si_{0.5}$ show saturation magnetizations of 37 emu/g and 35 emu/g, respectively at 100 K.

I. Introduction

Heusler alloys have recently attracted much attention in the material science community due to their many interesting properties. Hesuler alloys are investigated for half-metallic and spin-gapless semiconducting properties. In addition, these materials can be investigated for their magnetocaloric effect, topological magnetic order, shape memory effect, and high perpendicular magnetic anisotropy. [1-6] Due to the nature of the electronic band structure, half-metals and spin-gapless semiconductors can produce fully (100%) spin polarized electron currents and can be applied to the field of spintronics. [7] This 100% spin polarization means the alloys are completely conducting for one spin channel, and completely insulating for the other.[8] Such alloys exhibiting ferro- or ferrimagnetic order with the Curie temperature above room temperature can retain a high degree of spin polarization at room temperature. This, in turn, makes them ideal for room temperature spintronic applications.[9] One of the interesting features of Heusler alloys is that their magnetic and electronic band properties can be altered by adjusting elemental compositions.[10] That is the main objective of this work.

Heusler alloys are mainly composed of three elements such as XYZ and X_2YZ and crystallize in the cubic structure.[11] Here, X and Y are transition metals, and Z is the main group element. A structural variant of the Heusler alloy family is the quaternary Heusler alloy ($XX'YZ$) in which one of the X atoms in X_2YZ is replaced with another transition metal (X').[12] An ideal, perfectly ordered quaternary Heusler alloy crystallizes in the Y cubic structure.[13] Figure 1 shows such a structure for CoFeVAl where Co, Fe, V and Al respectively occupy the (3/4, 3/4, 3/4), (1/4, 1/4, 1/4), (1/2, 1/2, 1/2), and (0, 0, 0) lattice sites. However, it is very difficult to synthesize Heusler alloys in completely ordered structures and many of them crystallize in a disordered $L2_1$, B2, or A2 structure.[14] These disorders arise from the mixing of the X and X' atoms ($L2_1$), Y and Z atoms

(B2), or the mixing of all atoms (A2). Heusler alloys in the disordered structures do not exhibit the theoretically predicted properties.[15] Therefore is it important to refine materials synthesis to reduce structural disorder in this class of materials. Doping external elements can help to improve crystalline order with improved magnetic properties.

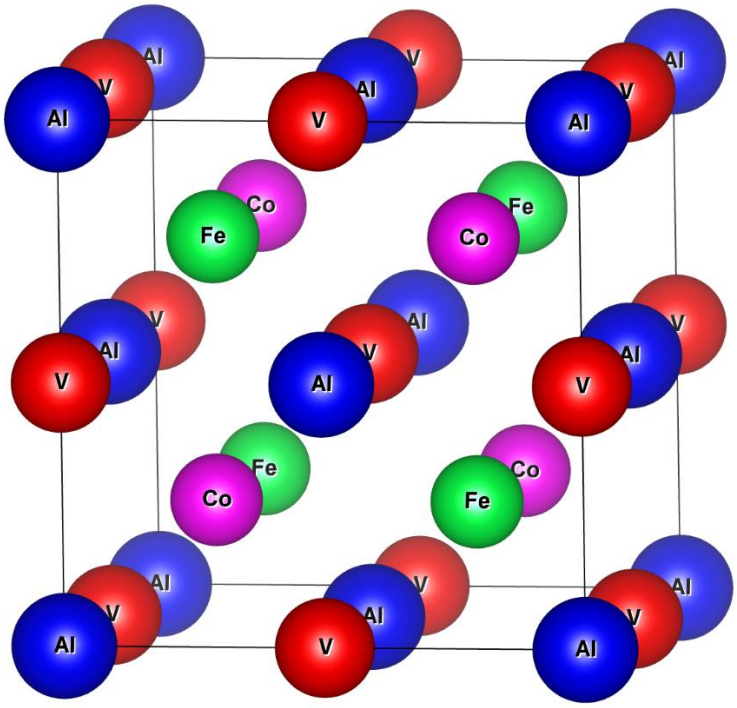

Figure 1 displays the crystal cubic structure of the parent alloy CoFeVAl with no disorder present. Lattice site occupancy: Co(3/4, 3/4, 3/4), Fe(1/4, 1/4, 1/4), V(1/2, 1/2, 1/2), and Al(0, 0, 0).

In this work, we have used the same strategy where CoFeVAl is doped with Fe and Si. The parent alloy CoFeVAl is predicted to exhibit nearly half-metallic properties. Unfortunately, it is paramagnetic above 60 K making it undesirable for room temperature applications. [20] We found that replacing 50% of Al with Si or the same amount of Fe with Co in CoFeVAl produced significant change in magnetic properties where the values of both magnetization and Curie

temperature increased. In addition, partially replacing Fe with Co also improved the crystalline order of the alloy.

Experimental Methods

Bulk alloys of CoFeVAl, $Co_{1.5}Fe_{0.5}VAl$, and $CoFeVAl_{0.5}Si_{0.5}$ were synthesized using arc-melting and high vacuum annealing techniques. The metal pieces used for synthesis were cut from highly pure (99.9%) commercially available pellets. They were then placed on a water-cooled copper crucible in the base plate of the arc furnace. The materials were subject to an argon environment during the melting process. The formed bulk ingots were then placed in a high vacuum tube furnace and annealed at 850ºC for 72 hours. Crystal structures of the powder samples prepared from the annealed ingots were studied using a Rigaku MiniFlex600 X-Ray Diffractometer. The Rigaku uses Cu-Kα radiation (λ = 1.54Å). Magnetic measurements were done with a Quantum Design VersaLab Vibrating Sample Magnetometer. All data analysis was done using OriginLab Software.

II. Results and Analysis

Structural Properties

Figure 2 shows the combined X-Ray Diffraction (XRD) patterns of the samples recorded at room temperature. The indexed peaks in the patterns represent corresponding lattice planes in the cubic crystal structures of the alloys. These XRD patterns indicate that the parent alloy CoFeVAl and the Si doped alloy $CoFeVAl_{0.5}Si_{0.5}$ crystallize in A2 type disordered cubic structure. This can be seen from the lack of both superlattice peaks (111) and (200) in the patterns. However, the sample with an excess of Co, $Co_{1.5}Fe_{0.5}VAl$, crystallized in a cubic structure with B2 type disorder. This is apparent from the absence of only one superlattice peak (111). This indicates that partially replacing Fe with Co helped to improve the crystalline order of CoFeVAl.

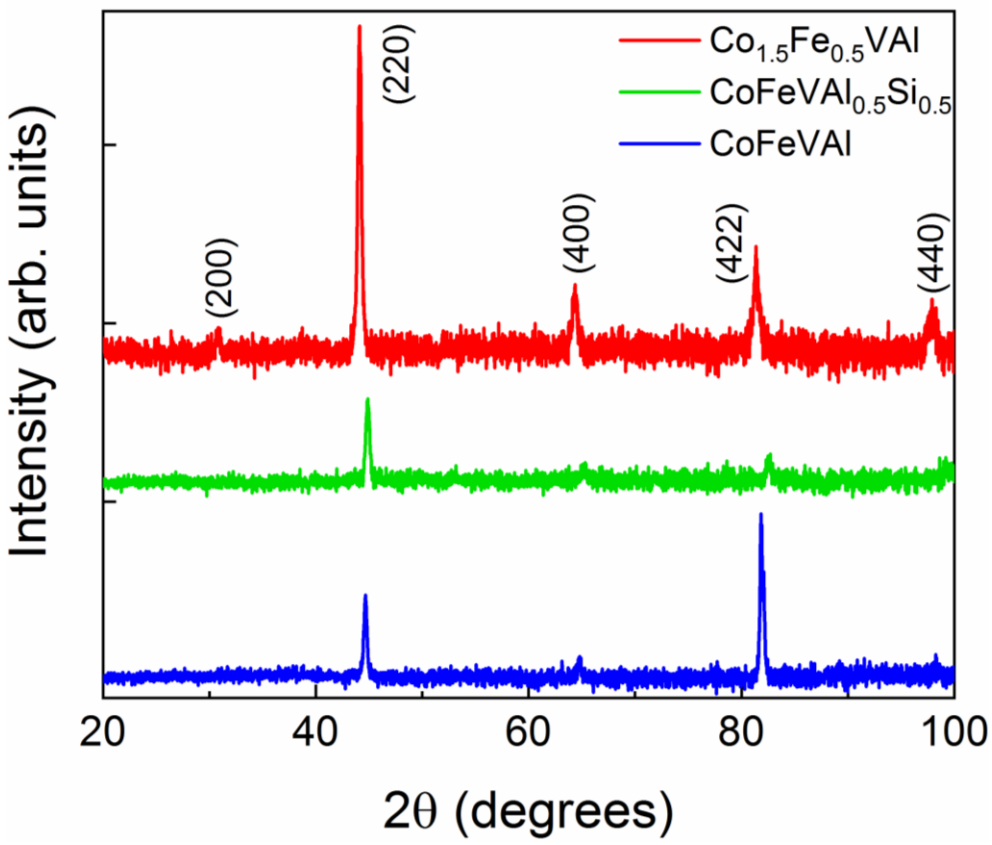

Figure 2 shows plotted room temperature XRD data for CoFeVAl and the two daughter alloys of $CoFeVAL_{0.5}Si_{0.5}$ and $Co_{1.5}Fe_{0.5}VAl$.

Magnetic Properties

Figure 3 shows the thermomagnetic M(T) curves for the alloys studied. The M(T) measurements were done at a constant magnetic field of 1 kOe between 50 K and 350 K. As shown in the inset of Fig. 3, the M(T) curve of the parent alloy shows paramagnetic-like behavior. Since we could not measure M(T) below 50 K due to instrumental limitations, it is not clear if the material is paramagnetic or if it has different magnetic ordering below 50 K. However, after partially replacing Al with Si or Fe with Co in CoFeVAl, we see a clear magnetic transition from a ferro-or ferrimagnetic to paramagnetic order at approximately 215 K. The Curie temperatures determined from the first derivative of magnetization with respect to temperature ($dM/dT \ versus \ T$) are 210

K and 220 K for $CoFeVAl_{0.5}Si_{0.5}$ and $Co_{1.5}Fe_{0.5}VAl$, respectively. Theoretically, CoFeVAl is predicted to show nearly half metallic behavior with a magnetic moment of approximately $1\mu_B$/f.u. We believe that its magnetic order below 50 K is either ferro or ferrimagnetic. However, further measurements at lower temperatures are required to confirm this claim.

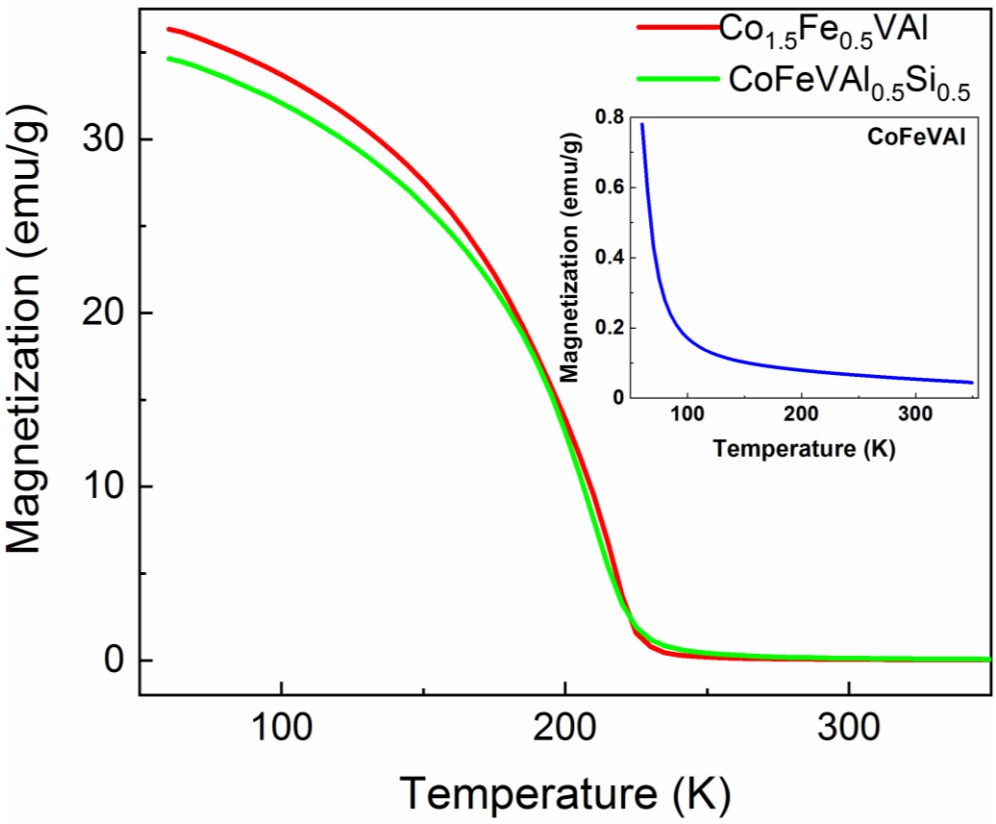

Figure 3 shows the M(T) plots for $CoFeVAl_{0.5}Si_{0.5}$ and $Co_{1.5}Fe_{0.5}VAl$ and the inset shows the plot for the parent alloy. These measurements were all done at a constant magnetic field of 1kOe.

Figure 4 displays the plotted isothermal magnetization curves M(H) for the studied alloys. The measurements were done at 100 K with a magnetic field varying from -30 kOe to 30 kOe. The inset of Figure 4 shows the isothermal magnetization plot of the parent alloy. The M(T) curve shown in the inset of Figure 3 indicates that the parent alloy is paramagnetic at 100 K. Therefore, an almost linear M(H) curve is expected at 100 K. The other two alloys $Co_{1.5}Fe_{0.5}VAl$ and

$CoFeVAl_{0.5}Si_{0.5}$ show hysteresis curves similar to those of ferro- or ferrimagnetic materials with saturation magnetization values of 37 emu/g (1.30μ_B/f.u.) and 35 emu/g (1.20μ_B/f.u.), respectively. Due to the extremely small coercivities present in the hysteresis curves, these materials can be classified as soft magnetic materials. As the magnetic field changes, their magnetization changes accordingly with minimal resistance to demagnetization. According to the Slater Pauling rule ($M_T = Z_T - 24$), the expected saturation magnetization value for each of these alloys is 1.50 μ_B/f.u [13]. For CoFeVAl with 25 valence electrons the expected magnetic moment is 1 μ_B/f.u. Here we used 1 $\mu_B = 9.274\times 10^{-21}$emu for these conversions. The experimental saturation magnetizations for these alloys are close to the predicted values. The small discrepancy between the theoretical and experimental magnetization values can be attributed to the structural disorder present in the alloys.

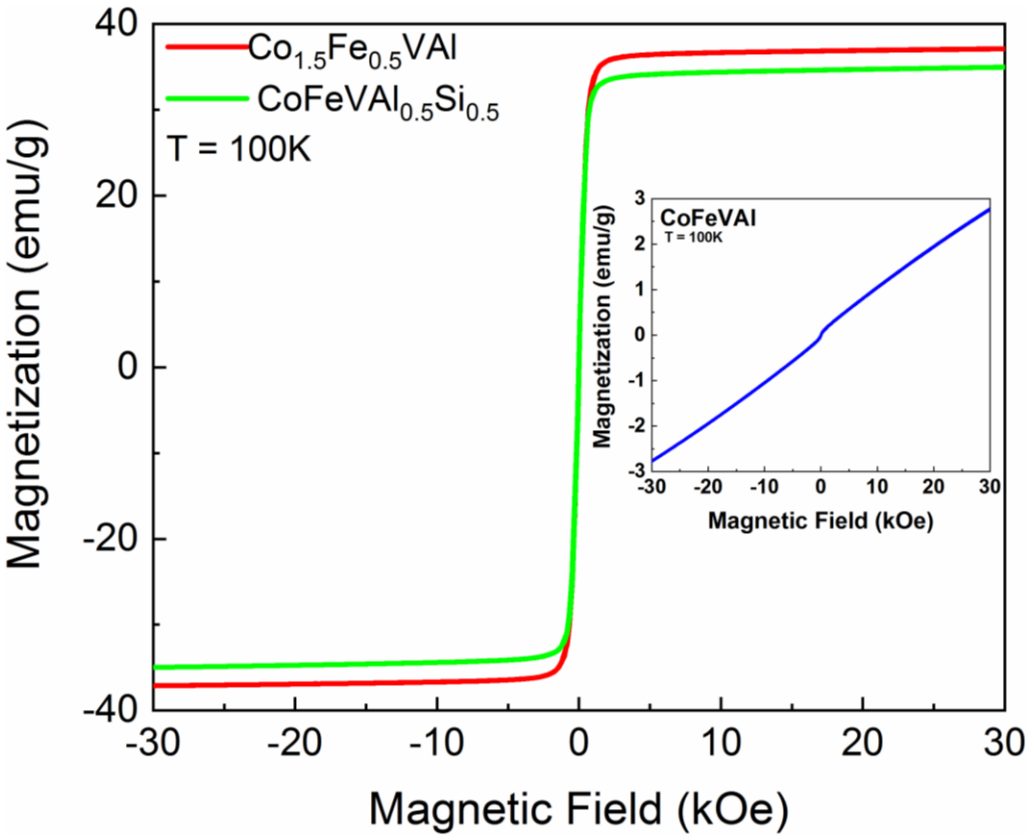

Figure 4 gives a visual representation of the magnetization of the alloys with respect a changing magnetic field at constant temperature of 100 K. Inset is the isothermal magnetization plot for the parent alloy.

III. Conclusion

The structural and magnetic properties of the Heusler alloys CoFeVAl, $Co_{1.5}Fe_{0.5}VAl$, and $CoFeVAl_{0.5}Si_{0.5}$, that were prepared using arc melting and annealing, were investigated. The XRD patterns revealed that the studied alloys crystallized in cubic structure with A2 type disorder (CoFeVAl and $CoFeVAl_{0.5}Si_{0.5}$) and B2 type disorder ($Co_{1.5}Fe_{0.5}VAl$). Magnetic measurements show that all three studied alloys exhibit Curie temperatures below room temperature. Partially replacing Al with Si or Fe with Co in CoFeVAl substantially improved its magnetic properties. The parent compound is observed to be paramagnetic at the temperatures studied, and the other two doped alloys show ferro- or ferrimagnetic magnetic properties below their respective Curie

temperatures. Despite not seeing the ideal case of a T_C value above room temperature, it is worth noting that by engineering the original alloy, CoFeVAl, we were able to drastically increase the magnetization and Curie temperatures of the doped compounds. These alloys may have an application for below room temperature devices that require moderate saturation magnetization. Further investigation is needed to determine their spin polarization, density of states, and their applicability to the field of spintronics.

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Author Biography

Gavin and Jax are in their senior year of studies in physics. They have been working in the physics materials and nanoscience lab since their sophomore years of study. Gavin is working to earn his BS in physics with minors in mathematics and aerospace engineering. He intends to obtain his PhD in aerospace engineering with a focus on propulsion and plasmas. Jax is majoring in physics and mathematics. Jax aims to earn his PhD in physics with focuses on astrophysics and cosmology. Matthew is a sophomore majoring in physics and mathematics. Matthew aims to earn his PhD in biophysics.