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Co_2FeSi crystallizes in the ordered $L2_1$ structure as proven by x-ray diffraction and Möβbauer spectroscopy. The magnetic moment of Co_2FeSi was measured to be about $6\mu_B$ at 5 K. Magnetic circular dichroism spectra excited by soft x-rays were taken to determine the element-specific magnetic moments of Co and Fe. The Curie temperature was measured with different methods to be (1100 ± 20)K. Co_2FeSi was found to be the Heusler compound as well as the half-metallic ferromagnet with the highest magnetic moment and Curie temperature. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2167330]

I. INTRODUCTION

Heusler compounds are ternary intermetallics with the composition X_2YZ.[1] They order in the $L2_1$ structure, space group $F\,m\bar{3}m$. The Co_2-based Heusler compounds are of particular interest due to their high Curie temperatures and the high magnetic moments per unit cell (for a review see Refs. 2 and 3).

Half-metallic ferromagnets (HFM) are predicted to be 100% spin polarized.[4] In those materials, the majority electrons are metallic, whereas the minority electrons are semiconducting or insulating. A high degree of spin polarization is interesting for various applications in magnetoelectronics (for examples see Ref. 5). Several materials in the class of Heusler compounds were predicted to be half-metallic ferromagnets.[4,6,7]

This work reports about the Heusler compound Co_2FeSi that is found to be, at present, the Heusler compound exhibiting the highest magnetic moment as well as the highest Curie temperature.

II. RESULTS AND DISCUSSION

Co_2FeSi samples were prepared by arc-melting stoichiometric quantities of pure metals in an argon atmosphere, followed by annealing in sealed quartz tubes at 1300 K for 21 days. The crystal structure of the polycrystalline ingots was investigated by x-ray diffraction and was proven to exhibit the correct $L2_1$ structure. The lattice constant was found to be 5.64 Å by Rietveld refinement. The Rietveld refinement allowed for a $B2$-type disorder of only $<10\%$ (mixing between Fe and Si). ^{57}Fe Möβbauer spectroscopy was performed to gain additional structural information. The derived spectrum (not shown here) exhibited a single sextet consisting of sharp lines with a width of 0.15 mm/s being in the order of the α-Fe linewidth (0.136 mm/s). This observation confirms the occupation of a single site for Fe and thus a well-ordered system. DO_3-like disorder (mixing of Co and Fe at X and Y sites) can be excluded from a comparison of the measured $(26.3\times10^6$ A/m) and calculated values (21×10^6 A/m for Fe in Y and 11×10^6 A/m for Fe in X positions) of the hyperfine field.

Low-temperature magnetometry was performed using a superconducting quantum interference device (SQUID). The results are displayed in Fig. 1(a).

The field dependence of the magnetization is typical for a soft magnetic material. The measured magnetic moment in saturation is $(5.97\pm0.05)\mu_B$ at 5 K, resulting in $6\mu_B$ at 0 K by extrapolation. This value is in agreement with the spin moment expected from the Slater-Pauling rule.[8] The measured magnetic moment is an integer within the experimental uncertainty, as expected for a half-metallic ferromagnet.

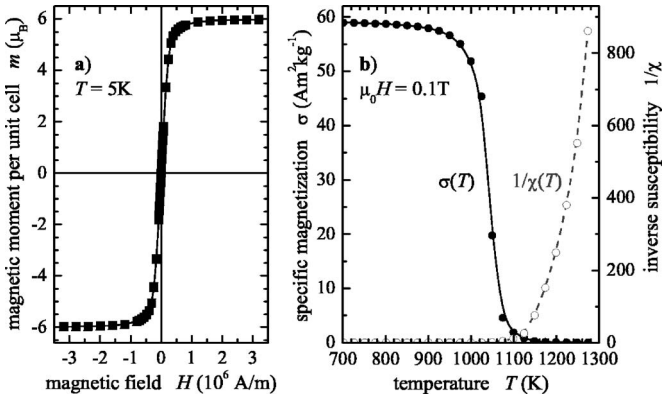

FIG. 1. Magnetization of Co_2FeSi. (a) shows the low-temperature and (b) the high-temperature magnetic properties as measured by SQUID and VSM, respectively.

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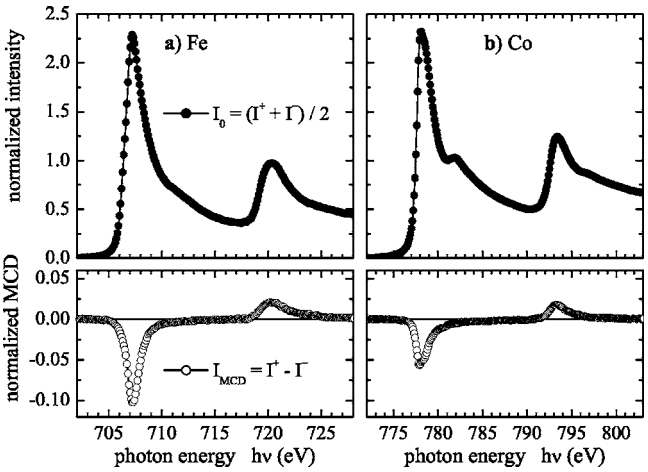

FIG. 2. Site-resolved magnetic properties of Co_2FeSi. Shown are the XAS (I_0) and XMCD (I_{MCD}) spectra taken at the $L_{2,3}$ absorption edges of Fe (a) and Co (b) after subtracting a constant background.

X-ray magnetic circular dichroism (XMCD) in photoabsorption [x-ray absorption spectroscopy (XAS)] was measured at the First Dragon beamline of NSRRC (Hsinchu, Taiwan). The XAS and XMCD spectra were taken at the $L_{2,3}$ absorption edges of Fe and Co. The results are shown in Fig. 2. The two white lines corresponding to the L edges are clearly seen for Fe (a) and Co (b). An additional spectral feature is visible 3.5 eV below the L_3 absorption edge of Co and a weaker one at the Fe L_3 edge. This feature is related to the $L2_1$ structure and demonstrates the high structural order of the sample (it vanishes for $B2$-like disorder).[9]

The magnetic moments per atom derived from a sum-rule analysis[10,11] are $(2.6\pm0.1)\mu_B$ for Fe and $(1.2\pm0.1)\mu_B$ for Co at $T=300$ K and $\mu_0 H=0.4$ T. The error arises mainly from the unknown number of holes in the $3d$ shell and the disregard for the magnetic dipole term in the sum-rule analysis. The orbital-to-spin magnetic moment ratios are about 0.05 for Fe and 0.1 for Co. The measured ratio between the Fe and Co spin moments of 2.16 is fully reproduced by the calculated ratio of 2.18.

A seeming linear dependence of the Curie temperature as a function of the magnetic moment is observed in Co_2-based Heusler compounds as shown in Ref. 8. One expects that T_C is highest for those HMFs exhibiting a large magnetic moment. T_C should be above 1000 K in Co_2FeSi with a magnetic moment of $6\mu_B$. Following this suggestion, the ferromagnetic Curie temperature of Co_2FeSi was measured with a vibrating-sample magnetometer (VSM) equipped with a high-temperature stage. The result obtained in a constant induction field of $\mu_0 H=0.1$ T is shown in Fig. 1(b). A value of (1100 ± 20)K is obtained from the measurement.

The paramagnetic Curie-Weiss temperature Θ was estimated from a plot of the inverse susceptibility $(1/\chi)$ as a function of temperature [see Fig. 1(b)]. The Curie-Weiss temperature is found by interpolating $1/\chi(T)$ to be (1150 ± 50)K. A true linear behavior for $1/\chi$ as a function of temperature is not observed here because the experiment was performed in a temperature range close to the Curie temperature. A linear dependence can be expected from molecular-field theory only for temperatures far above T_C.

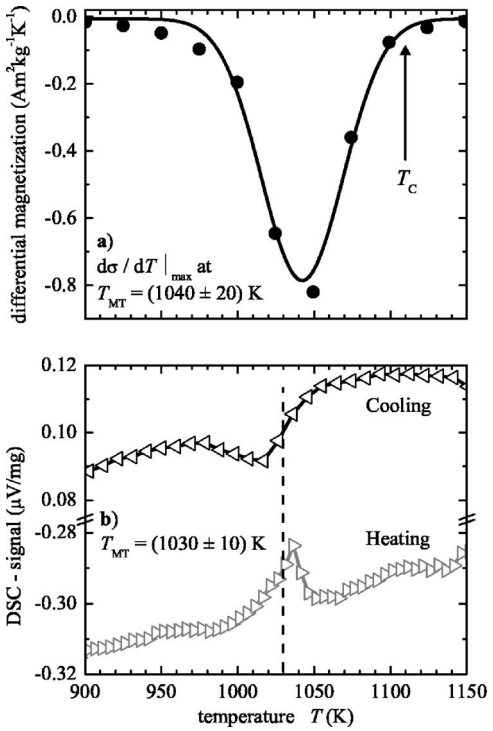

FIG. 3. Phase transition in Co_2FeSi. The differential magnetization (a) close to T_C is compared to differential scanning calorimetry (b).

The properties of Co_2FeSi observed here are in agreement with those reported previously by Niculescu *et al.*[12,13] for a higher degree of disorder (10% $B2$ plus 16% DO_3). (Please note that Niculescu *et al.* reported the following in Ref. 12: $a=5.66$Å, $T_C>980$K, and $m=5.9\mu_B$ at 10 K; the lattice parameter (5.64 Å) found here agrees with the one reported in Ref. 12 for Fe_2CoSi. The values for a and m reviewed later[3,14] are considerably different, for unknown reasons.)

SQUID magnetometry does not oftenly allow to determine high-temperature magnetic-phase transitions. The low-temperature requirement of the instrumental setup may not be met, in particular, if T_C is very high. Differential scanning calorimetry (DSC) is well established to investigate various kinds of phase transitions in solid materials (for example, see (Ref. 15). Here it was used to examine the magnetic-phase transition in order to support the Curie temperature received by the VSM experiments. Figure 3 compares the DSC signal (b) with the derivative of the specific magnetization (a) with respect to the temperature [compare Fig. 1(b)]. The minimum at (1040 ± 20)K in Fig. 3(a) corresponds to the maximum change of the magnetization with temperature. With DSC, a pronounced shift of the signal is observed during cooling or heating at about 1017 and 1037 K, respectively. This shift is due to the hysteresis of the DSC method and depends on the temperature gradient $\dot{T}(t)$. The signals are attributed to changes of the magnetic properties, as no structural transitions were observed in this temperature range. Therefore, one expects a mean value of (1030 ± 5)K for the magnetic-phase transition of Co_2FeSi. The melting point was also observed by DSC and found to be $T_m=(1517\pm5)$K (not shown in Fig. 3).

The observation of the magnetic transition by DSC is verified by the comparison of the structures in DSC with the differential magnetization. The latter is clearly observed at the point of maximum change of the magnetization with temperature. It is seen that the value obtained by DSC is slightly lower than T_C determined from the VSM measurement. For Co_2FeSi one finds it to be only 4.5% below the Curie temperature, and thus the DSC value may be a simple estimate for T_C if other methods are not available.

III. SUMMARY

The structural and magnetic properties of the Heusler compound Co_2FeSi were reported. Co_2FeSi has a lattice parameter of 5.64 Å and crystallizes in the $L2_1$ structure with very low disorder. Its melting point appears to be (1517 ± 5)K. The material is soft magnetic and its specific saturation magnetization of 0.166 A m^2 kg^{-1} at 0 K exceeds the one of pure Co by about 9%. The Curie temperature of 1100 K is 5% higher than that of pure Fe. The magnetic moment of $6\mu_B$ per unit cell points clearly on a half-metallic ferromagnet.

As a practical application, it was shown how to estimate the lower temperature limit of the magnetic-phase transition for materials with very high Curie temperatures by means of differential scanning calorimetry.

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