Unveiling the Mechanism for the Split Hysteresis Loop in Epitaxial Co$_{2}$Fe$_{1-x}$Mn$_{x}$Al Full-Heusler Alloy Films

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Utilizing epitaxial Co$_{2}$Fe$_{1-x}$Mn$_{x}$Al full-Heusler alloy films on GaAs (001), we address the controversy over the analysis for the split hysteresis loop which is commonly found in systems consisting of both uniaxial and fourfold anisotropies. Quantitative comparisons are carried out on the values of the twofold and fourfold anisotropy fields obtained with ferromagnetic resonance and vibrating sample magnetometer measurements. The most suitable model for describing the split hysteresis loop is identified. In combination with the component resolved magnetization measurements, these results provide compelling evidences that the switching is caused by the domain wall nucleation and movements with the switching fields centered at the point where the energy landscape shows equal minima for magnetization orienting near the easy axis and the field supported hard axis.

Magnetic anisotropy is one of the fundamental properties of magnetic materials which governs their applications. According to the symmetry of ferromagnet, magnetic anisotropies can be classified into twofold anisotropy (also called uniaxial anisotropy) $K_2$, fourfold anisotropy $K_4$, and higher order anisotropies. Typically, they are mixed in one material. The coexistence of $K_2$ and $K_4$ is the most common case for magnetic systems mainly due to the competition between the crystalline anisotropy and the uniaxial anisotropy induced by lattice mismatch, miscut and inclined deposition etc. Several interesting phenomena such as the split hysteresis loop (Fig. 1), and the field-induced spin reorientation transition (SRT) have been observed$^{1-14}$. In order to obtain the magnetic anisotropy constants quantitatively from the hysteresis loop, several models have been proposed$^{1,4,6}$. Weber et al.$^{1}$ pointed out the uniaxial and fourfold anisotropies can be directly given by the split field $H_s$ and the linear slope $k$ of the split hysteresis loop at zero field [For the definition of $H_s$ and $k$, see Fig. 1]. Through minimizing the in-plane free energy with respect to the angle between magnetization and magnetic field and assuming $K_2 \gg K_4$, the authors derived that $K_2 = M_s H_s$ and $K_4 = M_s^2/2k$, where $M_s$ is the saturation magnetization. Dumm et al.$^{5}$, recognized that $K_2$ and $K_4$ depend on both $k$ and $H_s$. Meanwhile, Oepen et al.$^{6}$, analyzed the split loops in the framework of the field-driven SRT. Their results showed that the switching fields in the split loops are directly related to the SRT and thus $K_2$ and $K_4$ are obtained by the switching fields and $k$. The different models have been adopted by many groups$^{11,13-14}$. However, they have not been cross-checked with additional magnetic anisotropy sensitive techniques such as ferromagnetic resonance (FMR) and magnetic torque measurements.

In this paper, we identified the model which best describes the system by comparing the uniaxial anisotropy field $H_1 = 2K_2/M_s$ and fourfold anisotropy field $H_4 = 2K_4/M_s$ obtained with FMR and vibrating sample magnetometer (VSM) measurements on the Co$_{2}$Fe$_{1-x}$Mn$_{x}$Al films with different Mn concentrations. Co$_{2}$Fe$_{1-x}$Mn$_{x}$Al is a kind of Co-based full-Heusler alloys, which possess high spin polarization and have great potential in spintronics application$^{25-34}$. Interestingly, electronic structure calculations have revealed that Co$_{2}$MnAl can retain half-metallic...
property for different levels of Fe doping. The magnetic anisotropy however shows strong variations on concentration which mainly due to a competition between uniaxial and four-fold anisotropies. Thus, it provides an interesting system to crosscheck the different models used in the split hysteresis analysis besides the fundamental interest in the exploration of the concentration dependent properties. We first measured the hysteresis loops with VSM and calculated the anisotropy fields from the split loops using different models. The results are quantitatively compared with the measured values utilizing FMR which allows us to identify the most suitable model used in the split loop analysis. The underlying physics is discussed. And the concentration dependent important material parameters such as effective magnetization, anisotropy fields, and damping constants are also given.

### Results

**Controversy over the analysis for the split hysteresis loop.** Figure 1 shows a typical split hysteresis loop of a 45 nm Co$_2$FeAl film measured by VSM with the magnetic field being applied within the sample plane and along the [110] direction. A discontinuity appears around $H_s$. This discontinuity is assumed to be the consequence of the superposition of the uniaxial and the fourfold anisotropy in the case that the hard axis of the uniaxial anisotropy coinciding with an easy axis of the fourfold component. With the measured split loops, we calculated $H_2$ and $H_4$ utilizing the three models mentioned above. The results are listed in Table 1 for Co$_2$FeAl and Co$_2$Fe$_{0.7}$Mn$_{0.3}$Al films. Both films have the same thickness of 45 nm. We can find that the results obtained with different models are significantly different. This raises an interesting question which model describes the magnetic anisotropies of the system best.

**FMR measurements and analysis.** To obtain independent results for crosschecking the different models, we performed angular dependent measurements utilizing FMR. The samples were positioned on a coplanar waveguide (CPW) fixture with the [110] direction of the film parallel to the x-direction as sketched in Fig. 2. The CPW is connected with a Vector Network Analyzer (VNA), which generates microwave with tunable frequencies. The VNA also detects the rf signal via the change of the forward transmission coefficient in scattering parameters, $S_{21}$. The external magnetic field $H$ is applied within the film plane. The orientation of magnetic field is controlled by a servo motor with high accuracy of positioning (error margin < 0.15°). We note that all the graphs are plotted with the raw data without any mathematical smoothing. The magnitude of the magnetic field is first set to be 1548 Oe, which is larger than the saturation field. The microwave frequency is swept to find the resonance condition where the maximum absorption occurs. Rotating the magnet yields an angular-dependent resonance frequency for Co$_2$FeAl which is plotted in Fig. 3(a). From the plot of the angular dependence one can easily identify twofold and fourfold symmetries with the maximum frequency located at 0° and 180°. Since the resonance frequency is proportional to the effective magnetic field, the maximum (minimum) value is found when the field is parallel to the easy (hard) axis. The angular dependence of the resonance frequency immediately shows that the easy axis

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**Figure 1.** Typical split-hysteresis loop obtained using VSM on a 45 nm epitaxial Co$_2$FeAl film along the [110] direction at room temperature. $H_s$ and $k$ are the split field and the slope at the zero field, respectively.

| Material     | Weber et al. $H_2$ (Oe) | Dumm et al. $H_2$ (Oe) | Oepen et al. $H_2$ (Oe) |
|--------------|------------------------|------------------------|------------------------|
| Co$_2$FeAl   | 154                    | 140.4                  | 259.7                  |
| Co$_2$Fe$_{0.7}$Mn$_{0.3}$Al | 92.2                  | 87.3                  | 239.5                  |

| Material     | Weber et al. $H_4$ (Oe) | Dumm et al. $H_4$ (Oe) | Oepen et al. $H_4$ (Oe) |
|--------------|------------------------|------------------------|------------------------|
| Co$_2$FeAl   | 444.4                  | 304.1                  | 184.7                  |
| Co$_2$Fe$_{0.7}$Mn$_{0.3}$Al | 434.8               | 347.5                  | 195.4                  |
Figure 2. Sketch of the FMR measurement configurations in our experiment. The sample was stuck on a CPW fixture to be excited by a rf magnetic field. VNA is used to generate microwave magnetic field and detect the $\delta_{31}$ parameters. The external magnetic field can be rotated in-plane by a servo motor.

Figure 3. Angular-dependent ferromagnetic resonance frequency with a constant magnetic field applied parallel to the film for the sample. The magnetic field is 1548 Oe for (a), while 1166 Oe for (b). The solid line is fitting results with the parameters in Table 2.

is along the [110] direction, consistent with the hysteresis loop. The uniaxial easy axis coincides with one of the fourfold axes and is along the [110] direction. Therefore, [110] bears the easy character of the fourfold cubic anisotropy and the hard character of the uniaxial anisotropy, which causes the split hysteresis loop (Fig. 1).

By summing up the Zeeman, the demagnetization and the anisotropy energy densities, the total free energy density of the system can be written as:

$$E = -M_H \sin \theta_M \sin \theta_H \cos (\varphi_M - \varphi_H) + \cos \theta_M \cos \theta_H - (2\pi M_s^2 - K_1) \sin^2 \theta_M$$

$$+ K_2 \sin^2 \varphi_M \sin^2 \theta_M + \frac{K_4}{4} \sin^2(2\varphi_M) \sin^2 \theta_M$$

(1)

where $K_1$, $K_2$ and $K_4$ are the out-of-plane uniaxial, in-plane uniaxial and fourfold anisotropy constants, respectively. And $\theta_M$, $\varphi_M$, $\theta_H$ and $\varphi_H$ are the angles between $M(H)$ and the axis perpendicular to the film plane (y-axis), while $\varphi_M$ are the in-plane angles between $M(H)$ and the easy axis (x-axis), see Fig. 2. Here the exchange energy and the higher-order anisotropy energies are neglected since the magnetic field used in the measurements is larger than the saturation field and the measured data do not show higher-order anisotropy. According to the FMR theory, the resonance
The numbers in the parentheses are the experimental error margins.

Table 2. The best fitting results of the magnetic parameters for the FMR measurements and anisotropy fields obtained from hard-axis loops. The numbers in the parentheses are the experimental error margins.

| $x$ | 0      | 0.3    | 0.7    | Method   |
|-----|--------|--------|--------|----------|
| $H_s$(Oe) | 1548   | 1166   | 1166   | 1166     |
| $g$ | 2.0 (0.01) | 2.0 (0.01) | 2.0 (0.01) | 2.0 (0.01) |
| $M_s$(Oe) | 15300 (40) | 15320 (30) | 14800 (40) | 14273 (40) |
| $H_c$(Oe) | 145.6 (1.8) | 146 (1.5) | 86.7 (2) | 12 (0.5) |
| $H_K$(Oe) | 307.2 (4.1) | 306.1 (5) | 340.7 (3.5) | 257.1 (2.8) |
| $H_V$(Oe) | 140.5 (2) | 85.3 (4) | N.A.    | VSM      |
| $H_I$(Oe) | 304.1 (3) | 349.5 (6) |

Comparison of FMR and VSM measurements along different directions. To further check the accuracy of the fitted results, we also performed field dependent FMR measurements along different directions. In the left column of Fig. 4, we show the two dimensional (2D) gray scale mapping of the $S_{11}$ signal of the microwave absorption measured via VNA as a function of the microwave frequency $f$ and magnetic field $H$ with the field applied along the [110], [100] and [110], respectively. The right column displays the corresponding hysteresis loops obtained via VSM. In the FMR measurements we swept the magnetic field from $-1400$ Oe to zero. The brightness of the grayscale shows the amplitude of the absorption and the brightest spots represent the position of FMR. Since the anisotropy fields cause additional contributions to the torque on the magnetization, FMR spectrum exhibits different behavior in these three directions. The easy axis of magnetization is along the [110] direction, as can be recognized from the rectangular shape of the hysteresis loop in Fig. 4(d). The data for the field along the [110] direction [Fig. 4(a)] show a typical FMR spectrum. It can be easily fitted with Kittel formula (red curve). In this case, the magnetization precesses around the direction of external magnetic field. The curve along the [100] direction, Fig. 4(b), is however more complicated. With the increased of the magnetic field, the resonance frequency first decreases then increases after an inflection field at $\sim 450$ Oe. Consequently, two resonance modes at different fields can be obtained for a given frequency. For instance, a 7 GHz microwave field causes resonance at either 200 Oe or 700 Oe. From Fig. 4(e), it is obvious that [100] is the hard axis of the sample and it saturates at the field higher than 500 Oe. The remanence is about 0.7 of the saturation value which can be easily understood as the field is applied 45° with respect to the easy axis, i.e., the [110] direction and the magnetization is expected to align close to the easy axis at low external field. With the increasing of the magnetic field, the magnetization will align along the field direction eventually. In such case, $\varphi_M$ is no longer constant, but is determined by the total energy minimum of equation (1). In fact, it changes from 45° to 0°. From equation (2), we see that the resonance frequency is proportional to $(H_a \cdot H_b)^{1/2}$ with $H_a$ consisting of four terms, $H_s \cos(\varphi_M - \varphi_H)$, $4\pi M_{eff}$, $H_s(3 + \cos 4\varphi_M)$ and $H_s(1 + \cos 2\varphi_M)$. Among them, $4\pi M_{eff}$ is a constant of about $\sim 15300$ Oe and is much larger than the applied field and the anisotropy fields. Therefore, $H_s$ shows only a weak dependence on field. $H_s$ consists of three terms, $H_s \cos(\varphi_M - \varphi_H)$, $H_s \cos 4\varphi_M$ and $H_s \cos 2\varphi_M$. When the field is not applied along the easy axis, the term $H_s \cos 4\varphi_M$ and $H_s \cos 2\varphi_M$ decrease with increase of the magnetic field as the magnetization deviates from the easy axis, while $H_s \cos(\varphi_M - \varphi_H)$ behaves oppositely. Hence the resonance frequency is determined by the competition between anisotropy fields and magnetic field. As listed in Table 2, both $H_a$ and $H_b$ are comparable with $H$ at low magnetic field which can lead to a decrease of resonance frequency $f$ with increasing field in a certain field range, as shown in Fig. 4(b). Taking the same parameters in the angular dependent FMR measurements as in Fig. 3 and using equations (1) and (2), we have computed the field dependent $f$ (red curve) and found that it agrees well with the experimental data. Similarly, we can understand the field dependence of $f$ for the field applied along the [110] direction [Fig. 4(c)] which is also not an easy axis. The resonance frequency decreases with increasing the field up to 80 Oe. After a sharp drop, the spectrum increases with field as expected. The
Experimental data can be well produced with the calculation (red curve) except the jump at ~80 Oe. We note that the hysteresis loop along the [110] direction shows similar behavior. At large field, the magnetization $M$ is forced by the field $H$ to align along the [110] direction. At zero field $M$ is oriented along the easy axis and perpendicular to $H$. Depending on either increase or decrease of the field, the magnetization curve shows two discontinuities at two different switching fields around $H_s$. We further performed the field dependent FMR measurements with ascending field and found that the jumping field in the FMR occurs at exactly the same field as the switching field in the field ascending branch of hysteresis loop. The one-to-one correspondence identifies that the origin of the jump in the field dependent FMR measurements shown in Fig. 4(c) is the magnetization switching. We emphasize that all the three resonance curves shown in Fig. 3(a–c) can be well reproduced with the parameters in Figure 4.

Figure 4. Left column (a–c) are 2D gray scale mapping of the $S_{21}$ absorption signal as function of the frequency and $H$ with the magnetic field along the [110], [100], [110] directions, respectively. The white indicate the position of FMR. The red lines are fitting results with the parameters in Table 2. Right column (d–f) are the corresponding longitudinal hysteresis loops of Co$_2$FeAl film measured by VSM at RT.
Table 1 according to equations (1) and (2) [red lines in Fig. 3(a–c)] once again, proving the validity of the method used for anisotropy field analysis in the FMR measurements.

Underlying mechanisms for the split hysteresis loop analysis. Comparing the results obtained via FMR with the values derived from the VSM measurements utilizing different models (Table 1), we recognize that the values obtained with the model by Dumm et al. gives the closest agreement. It is not too surprising to find that the model by Weber et al., could not yield similar results since their model is applicable only for $K_2 \gg K_4$.

In the model proposed by Oepen et al., the field driven reorientation for a strong uniaxial behavior is discussed. The assumption of the model is that without field the only minimum of the energy landscape appears for magnetization parallel to one axis only while the field applied along the hard axis drives the system through a state of coexisting phases (or metastability). We note here that, in the original paper, the free energy is expressed as $E = a \sin^2 \varphi + b \sin^4 \varphi$ and a translation of $K_2 = a + b$ and $K_4 = b$ is therefore required for comparison. The main assumption of the model is that in descending field the magnetization switches back into the easy axis when the local minimum created by the field is erased. With values of the anisotropy obtained via FMR it is evident that $-6 < -1 - \frac{K_4}{K_2} < -2$. We note that there is no indication in the original paper that the field assumed to cause an energy degeneracy of the two states. Apparently the latter is the appropriate assumption to describe the situation in the system investigated here.

In ref. 6 a reversible rotation near the zero field in the potential that is determined by the sum of two- and fourfold anisotropy contribution is assumed. In case of strong shape anisotropy the magnetization reversal appears fully within the sample plane, the total energy of the system [equation (1)] can be simplified by taking $\theta_M = 90^\circ$. As the magnetic field is applied along the [1 1 0] direction, i.e., $\varphi_H = 90^\circ$, an analytic expression for $H(m)$ can be derived by minimizing the total energy $E$, i.e.,

$$H(m) = \frac{2K_2}{M_s} m - \frac{2K_4}{M_s} (2m^3 - m),$$

where $m = \sin \varphi_M$ is the normalized magnetization component along the applied field direction. By differentiating equation (3), one can obtain the inverse slope of $m(H)$ for $m = 0$ as

$$\frac{1}{k} = \frac{\partial H}{\partial m} \bigg|_{m=0} = H_2 + H_4$$

Figure 5. The normalized value of the magnetization calculated from the two in-plane components. The magnetic field is applied along the [1 1 0] direction. Due to the strong shape anisotropy, the perpendicular component is negligible.
The linearization is a valid approximation which is proven experimentally by the split hysteresis loop. Dumm et al. further assumed that the energies are the same for two magnetization configurations at split field $H_s$ in the split loop, $m_H(H_s)$ and $m_H(H_s)$. Plotting the angle dependent free energy with a magnetic field along the $[110]$ direction for 80 Oe, i.e. the field value of the jump in the FMR measurement, and $H_2 = 145.6$ Oe and $H_4 = 307.2$ Oe (FMR results) we obtain three local energy minima around 0°, 90° and 180° values (see Fig. 6). The minima have the same value at 80 Oe while for fields that are either smaller (76 Oe) or larger (84 Oe), the local minimum at 90° is higher or lower than the other two minima. Hence the FMR data verify the assumption of equal energy states at the switching field. We note that $H_s = 77$ Oe, the small variance with 80 Oe may origin from the error bar of the measurements. Dumm et al. made reasonable settings for the magnetization component along the hard axis of $m_H(H_s) = kH_s$. The latter in an extrapolation utilizing the zero-field slope which is a good assumption as long as $m_H(H_s)$ is small. For the sake of a more general treatment, we propose to take the measured value of $m_H(H_s)$ and from the hysteresis curve. Combining it with equation (4), the twofold and fourfold anisotropy fields can be derived as:

$$ H_2 = \frac{m_H^2(H_s) + m_H^2(H_s) + 2kH_s}{k[m_H^2(H_s) + m_H^2(H_s) + m_H(H_s) + 1]} $$

$$ H_4 = \frac{1 - 2kH_s + m_H(H_s)}{k[m_H^2(H_s) + m_H^2(H_s) + m_H(H_s) + 1]} $$

With equation (5), we can obtain the twofold and fourfold anisotropy fields from $H_s, m_H(H_s)$ and slope $k$. The obtained results, $H_2 = 140.5$ Oe and $H_4 = 304.1$ Oe, are consistent with the fitted data from the FMR measurements.

**Confirmation with different Mn concentration.** Above, we have investigated the magnetic properties of Co$_2$FeAl film via FMR and VSM measurements. In the following, we continue to discuss the property variation with different Mn doping, i.e., Co$_{0.7}$Mn$_{0.3}$Al and Co$_{0.3}$Mn$_{0.7}$Al. Figure 7(a,b) present the angular dependent FMR measurements. They can be fitted very well with equation (2) and the fitted twofold and fourfold anisotropy fields are listed in Table 2. For Co$_{0.7}$Mn$_{0.3}$Al, the fitted results agree very well with VSM measurements using the analysis mentioned above. The good agreement once again proves the validity of the model used in the split loop analysis. From the angular dependent FMR measurements in Fig. 7(b), we can find that Co$_{0.7}$Mn$_{0.3}$Al shows almost pure fourfold symmetry. Meanwhile, hysteresis loops along the [110] and [110] directions become similar. Due to the weak twofold anisotropy, the VSM measurements for Co$_{0.3}$Mn$_{0.7}$Al did not show any split hysteresis loop. Therefore, the quantitative comparison between FMR and VSM measurements is not applicable. Besides, we found that the effective magnetization decreases from 15300 Oe to 14273 Oe and twofold anisotropy field decreases from 145 Oe to 12 Oe with the Mn composition increasing from 0 to 0.7. Recent X-ray magnetic circular dichroism measurements show that Co, Fe, Mn all exhibit net ferromagnetic states and contribute ferromagnetism to the films. Since the magnetic moment of Mn atom is generally larger than that of Fe atom, it would be expected that the magnetization of the system would increase with increasing Mn concentration if Mn, Fe and Co atoms are completely ferromagnetic coupled. Our FMR measurements, however, show the opposite behavior. This strongly suggests that there must be antiferromagnetic interaction among the system. The evolution of $H_2$ and $H_4$ with the changing Mn concentration could be associated with the competition between ferromagnetic Ruderman–Kittel–Kasuya–Yoshida exchange and antiferromagnetic superexchange, as reported by Şaşıoğlu et al. Through the FMR linewidth measurements, we also obtain the damping factor of samples with...
different Mn concentration. For Co$_2$FeAl, the damping factor is $7.7 \times 10^{-3}$, while for Co$_{0.7}$Fe$_{0.3}$Mn$_{0.3}$Al and Co$_{0.5}$Fe$_{0.5}$Mn$_{0.2}$Al, this value decreases to $6.5 \times 10^{-3}$ and $5.9 \times 10^{-3}$, respectively.

Summary
Combining VSM and FMR measurements on the full-Heusler alloy Co$_2$Fe$_{1-x}$Mn$_x$Al epitaxially grown on GaAs(001), three different models for the interpretation of split hysteresis loop are checked. The most suitable model is identified as the one that assumes the switching fields centered at the point where the energy landscape shows equal minima near the easy axis and the field supported hard axis. Our studies reveal that $H_e$ decreases rapidly with increasing Mn concentration and almost vanishes at $x = 0.7$, while $H_s$ shows much less concentration dependence. The decreasing effective magnetization with adding Mn component strongly suggests the existence of antiferromagnetic coupling among the system.

Methods
The Co$_{2-x}$Mn$_x$Al samples were prepared by molecular-beam epitaxy on GaAs (001) at 553 K. All the films have the same thickness of 45 nm and the Mn composition $x$ varies from 0 to 0.7. Before being taken out of the ultra-high vacuum chamber, the films were protected by 2 nm of aluminum capping layer. The crystal structure and the quality of order were analyzed by double-crystal X-ray diffraction as described previously. The hysteresis loops along different directions were obtained via VSM. The FMR measurements were performed with a Vector Network Analyzer, which generates microwave with tunable frequencies (20 MHz to 20 GHz). The VNA also detects the signal via the change of the forward transmission coefficient in scattering parameters, $S_{11}$. The external magnetic field $H$ was applied within the film plane. The orientation of magnetic field was controlled by a servo motor with high accuracy of positioning (error margin < 0.15°).

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Figure 7. Angular-dependent ferromagnetic resonance frequency with a constant magnetic field applied in the plane of films from (a) $x = 0.3$ and (b) $x = 0.7$. $H = 1166$ Oe.
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