

# Ultrafast magnetization dynamics in half-metallic Co<sub>2</sub>FeAl Heusler alloy

R. S. Malik, E. K. Delczeg-Czirjak, R. Knut, I. Vaskivskyi, S.  
Jana, R. Stefanuik, Y. O. Kvashnin, J. Söderström, and O. Karis

*Department of Physics and Astronomy,  
Uppsala University, Box 516, SE-75120, Uppsala, Sweden*

D. Thonig

*School of Science and Technology, Örebro University, SE-70182 Örebro, Sweden*

R. Gupta, S. Husain, A. Kumar, and P. Svedlindh

*Department of Materials Science and Engineering,  
Uppsala University, Box 534, SE-75121, Uppsala, Sweden*

O. Eriksson

*Department of Physics and Astronomy, Uppsala University,  
Box 516, SE-75120, Uppsala, Sweden and*

*School of Science and Technology, Örebro University, SE-70182 Örebro, Sweden*

## Abstract

We report on optically induced, ultrafast magnetization dynamics in the Heusler alloy Co<sub>2</sub>FeAl, probed by time-resolved magneto-optical Kerr effect. Experimental results are compared to results from electronic structure theory and atomistic spin-dynamics simulations. Experimentally, we find that the demagnetization time ( $\tau_M$ ) in films of Co<sub>2</sub>FeAl is almost independent of varying structural order, and that it is similar to that in elemental 3d ferromagnets. In contrast, the slower process of magnetization recovery, specified by  $\tau_R$ , is found to occur on picosecond time scales, and is demonstrated to correlate strongly with the Gilbert damping parameter ( $\alpha$ ). Our results show that Co<sub>2</sub>FeAl is unique, in that it is the first material that clearly demonstrates the importance of the damping parameter in the remagnetization process. Based on these results we argue that for Co<sub>2</sub>FeAl the remagnetization process is dominated by magnon dynamics, something which might have general applicability.

Studies of ultrafast demagnetization was pioneered by Beaurepaire *et al.*<sup>1</sup>, who demonstrated that the optical excitation of a ferromagnetic material - using a short pulsed laser could quench the magnetic moment on sub-picosecond timescales. The exact underlying microscopic mechanisms responsible for the transfer of angular momentum have been strongly debated for more than 20 years<sup>2-4</sup>. Ultrafast laser-induced demagnetization has now become an intense field of research not only from fundamental point of view but also from a technological aspect, due to an appealing possibility to further push the limits of operation of information storage and data processing devices<sup>5</sup>. Both experiment<sup>4-15</sup> and theory<sup>16-23</sup> report that all of the 3d ferromagnets (Fe, Ni and Co) and their alloys, show characteristic demagnetization times in the sub-picosecond range, while 4f metals exhibit a complicated two-step demagnetization up to several picoseconds after the excitation pulse<sup>4,24</sup>.

In this work, we have made element specific investigations of the ultrafast magnetization dynamics of a half-metallic Heusler alloy. The half-metallic Heusler alloys have a unique electronic structure, where the majority spin-channel has a metallic character while the minority spin-channel has a band gap and ideally exhibit 100% spin-polarization at the Fermi level. Heusler alloys are also appealing for spintronic applications due to the low Gilbert damping which is related with the half-metallicity<sup>25,26</sup>. The origin of the band gap and the mechanism of half-metallicity in these materials have been studied by using first principle electronic structure calculations<sup>27-30</sup>. The half-metallic property is furthermore known to be very sensitive to structural disorder<sup>28-32</sup>. From a fundamental point of view, it is intriguing to ask, how the band gap in the minority spin channel effects the ultrafast magnetization dynamics of Heusler alloys<sup>33,34</sup>. It has already been reported that some of the half-metals like CrO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub> exhibit very slow dynamics, involving time-scales of hundreds of picoseconds,<sup>33,34</sup> while several Co-based Heusler alloys show a much faster demagnetization, similar to the time-scales of the elemental 3d-ferromagnets<sup>35-37</sup>. The faster dynamics of these Heuslers has been discussed in Ref.<sup>33</sup> to be due to the fact that the band gap in the minority spin channel is typically around 0.3 – 0.5 eV, which is smaller than the photon energy (1.5 eV) of the exciting laser. It is also smaller than the band gap of CrO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub>. Importantly, the Heusler alloys offer the possibility to study magnetization dynamics, as a function of structural order, since they normally can be prepared to have a fully ordered *L*2<sub>1</sub> phase, a partially ordered *B*2 phase, and a completely disordered *A*2 phase. The structural relationships of these phases are described in the Supplemental Material (SM)<sup>38</sup>.

We have here studied the optically induced, ultrafast magnetization dynamics of Co<sub>2</sub>FeAl (CFA) films, using time-resolved magneto-optical Kerr effect (TR-MOKE) as described in Ref.<sup>39</sup>. By control of the growth temperature, CFA alloy forms with varying degree of structural order, in a continuous way between the *A*2 and *B*2 phases, as well as between the *B*2 and *L*2<sub>1</sub> phases<sup>40,41</sup>. We present data from four CFA samples, grown at 300 K, 573 K, 673 K, and 773 K respectively. We henceforth denote each sample by its growth temperature as a subscript, e.g. CFA<sub>300K</sub>. As evidenced by X-ray diffraction, the sample grown at 300 K is found to exhibit the *A*2 phase, while the samples grown at 573 K and 673 K predominantly exhibit the *B*2 phase. The sample grown at 773 K is found to exhibit a pure *B*2 phase<sup>41</sup>. The value of the Gilbert damping  $\alpha$  is found to monotonously decrease with annealing temperature and is thus lowest for the sample grown at 773 K<sup>42</sup>.

Calculations based on density functional theory (DFT) of the magnetic moment, Heisenberg exchange interaction and the Gilbert damping parameter are described in detail in (SM)<sup>38</sup>. These parameters were used in a multiscale approach to perform atomistic magnetization dynamics simulations, described in Sec.S1 of (SM)<sup>38</sup>. Here we employed the two

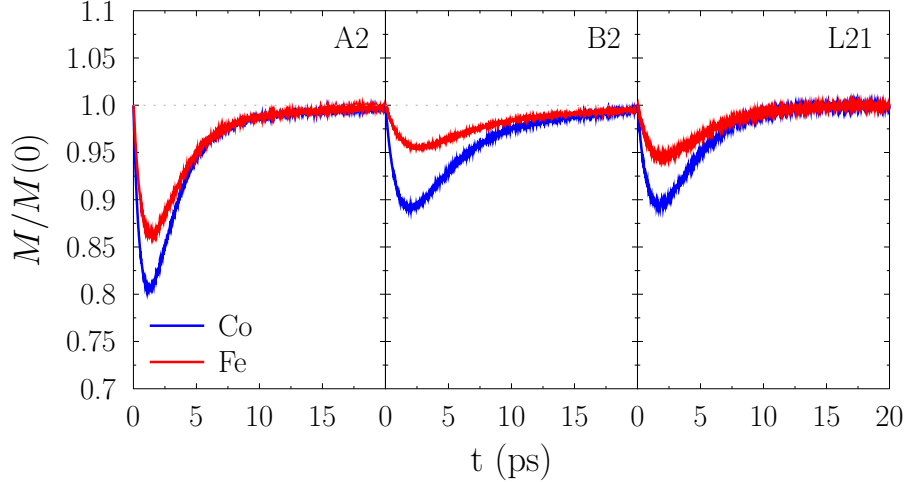


FIG. 1. (Color online) Simulations of ultrafast dynamics of  $\text{Co}_2\text{FeAl}$  in the different structural phases  $A2$  (left panel),  $B2$  (central panel), and  $L2_1$  (right panel). The demagnetization is shown element resolved (blue line - Co, red line - Fe). The peak temperature is 1200 K. The dotted line indicates the equilibrium magnetization at  $T = 300$  K.

temperature model (2TM) for the temperature profile of the spin-system. In the 2TM, the spin temperature increases due to the coupling to the hot-electron bath, that is excited by the external laser pulse. In the simulations we used a peak temperature in the 2TM of 1200 K. A full description of the 2TM and the details of all spin-dynamics simulations are described in Sec.S2 of SM<sup>38</sup>.

The results of the simulations are shown in Fig. 1, for the  $A2$ ,  $B2$  and  $L2_1$  phases. It can be seen that the different phases react differently to the external stimulus. In general, this model provides a dynamics that is controlled by *i*) the temperature of the spin-subsystem, *ii*) the strength of the magnetic exchange interaction and *iii*) the dissipation of angular momentum and energy during the relaxation of the atomic magnetic moments (Gilbert damping)<sup>43</sup>. Before continuing the discussion, we note that the average magnetization,  $M$ , of element  $X$  is calculated as  $M^X = \sum_i c_i^X M_i^X / 4 \sum_i c_i^X$ , where  $c_i^X$  is the concentration of the particular element  $X$  in the particular phase and  $i$  runs over the four nonequivalent sites of the unit cell. After the material demagnetizes, the spin temperature eventually drops and the average magnetization returns to its initial value after 10 – 20 ps (cf. Fig. 1).

To estimate the time constants of the demagnetization  $\tau_M$  and remagnetization ( $\tau_R$ ) processes, in an element-specific way, we fit both the theoretical and experimental transient magnetizations by a double exponential function<sup>44</sup>. We show results of  $\tau_M$  and  $\tau_R$  in Fig. 2 for the  $A2$  and  $B2$  phase, as well as for alloys with intermediate degree of disorder (described in Sec.S2 of SM<sup>38</sup>).

The theoretical demagnetization time is seen from Fig. 2 to typically be around 1 ps, whereas the remagnetization time is 2 – 5 ps. In the theoretical data of Fig. 2, it can be seen that as the  $B2$  ordering increases (moving to the right along the x-axis), both the demagnetization and remagnetization times increases. However, the simulations show a stronger increase of the remagnetization time as function of  $B2$  ordering. We also note that the relevant time scale is somewhat larger for Fe than for Co, and the ratio between them,  $\tau_{Fe}/\tau_{Co}$ , grows when going from  $A2$  to  $B2$  phase.

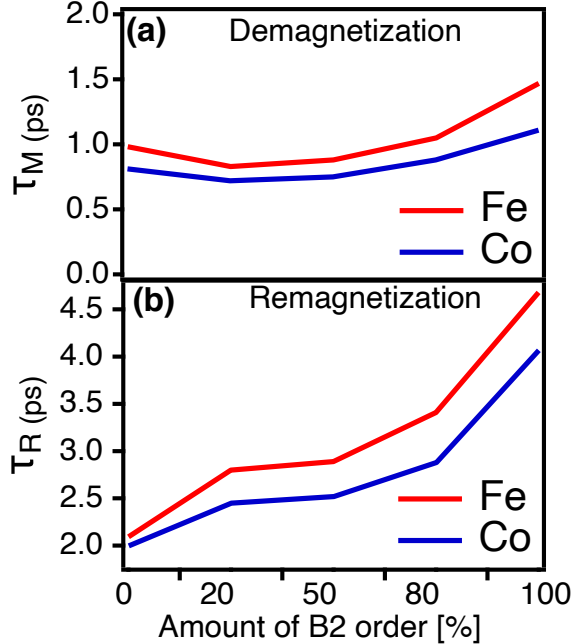


FIG. 2. (Color online) Element resolved relaxation times of  $\text{Co}_2\text{FeAl}$ , from simulations of alloys with varying amounts of  $A2 \rightarrow B2$  phase. 0 corresponds to pure  $A2$  phase while 100 corresponds to pure  $B2$  phase. Panel (a) shows the demagnetization time and panel (b) shows the remagnetization time. Both time constants are obtained from fitting the time trajectory of  $M^X(t)$  by a double-exponential function (see text).

Figures 3 (a-d) shows the element specific magnetization dynamics of CFA films that have different structure ordering, depending on their growth temperatures. All of the measurements are conducted in time-resolved transverse magneto-optical Kerr effect (TR-MOKE) spectrometer. (For details, see SM, Sec. S3 for thin films synthesis, and Sec. S4 for the experimental setup and measurements<sup>38</sup>). The inset of Fig. 3 shows the observed magnetization dynamics up to  $\sim 1$  ps. All of the films are demagnetized ( $\sim 25\%$ ) with a similar pump fluence. For all samples, the data for Fe (red) and Co (blue) show similar demagnetization dynamics in the first few hundred femtoseconds, whereupon differences in the remagnetization dynamics become visible, especially on the picosecond timescale. A significant change in the remagnetization dynamics are more obvious for the film with  $A2$  phase in Fig.3(a) and film with 100%  $B2$  phase in Fig.3(d) respectively.

Figure 4 (a-b) shows the measured values of the demagnetization and remagnetization time constants, for the four different growth temperatures, representing different degree of disorder in  $\text{Co}_2\text{FeAl}$ , along the alloy path  $A2 \rightarrow B2$ . It may be seen that the  $\tau_M$  for Fe and Co is the same within the error bars for all four samples, regardless of the degree of structural ordering (Fig. 4a). It may also be noted that the measured  $\tau_M$  for CFA is similar to that of 3d transition metals<sup>35,36</sup> and very much shorter than that of  $\text{CrO}_2$  or  $\text{Fe}_3\text{O}_4$ .

Demagnetization times that are independent on degree of structural ordering is interesting, since it can be expected that the presence of structural disorder in Heusler alloys ought to result in a lower degree of spin polarization of the electronic states (i.e. an increased density of states (DOS) at the Fermi level in the minority band). This is expected to enhance

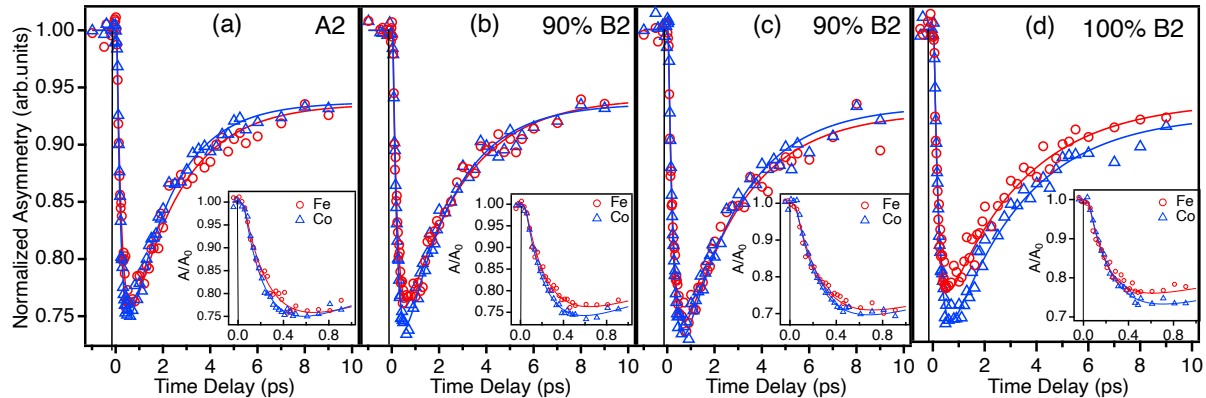


FIG. 3. Measured element-specific Fe (red) and Co (blue) magnetization dynamics of  $\text{Co}_2\text{FeAl}$ . Samples are denoted by the growth temperature in each case. The red and blue lines correspond to fitted data (see text). (a) 300 K (100%  $A_2$  phase), (b) 573 K (90%  $B_2$  phase), (c) 673 K (90%  $B_2$  phase), and (d) 773 K (100%  $B_2$  phase). The insets show the demagnetization dynamics up to  $\sim 1$  ps. All of the measurements were performed with similar pump-fluence (for details, see Sec.S4 of SM).

spin-flip scattering, with an accompanying speed-up of the demagnetization dynamics<sup>33,34</sup>. The electronic structure calculation of CFA also shows that the DOS at the Fermi level varies with different structural phases (analyzed in the Sec.S1 of SM<sup>38</sup>). The  $A_2$  phase has a large number of states at the Fermi level, while the  $L2_1$  phase, and to some extent the  $B_2$  phase, has a low amount<sup>41</sup>. Despite these differences in the electronic structure, the measured demagnetization dynamics shown in Fig. 4(a) is essentially independent on degree of structural ordering.

On longer time-scales, there is a significant effect of structural ordering on the observed magnetization dynamics, which becomes particularly relevant for the remagnetization process. As seen in Fig. 4b, there is a monotonous increase of remagnetization time,  $\tau_R$ , with increasing growth temperature and hence the degree of ordering along the  $A_2 \rightarrow B_2$  path. The sample grown at 300 K with  $A_2$  phase, exhibits the fastest remagnetization dynamics ( $\tau_R$ ). With increasing growth temperature and corresponding increase in the structural ordering along the  $A_2 \rightarrow B_2$  path, a distinct trend of increasingly slower remagnetization dynamics is observed.

The most conspicuous behaviour of the measured magnetization dynamics, and its dependence on the degree of ordering, concerns the remagnetization time (Fig. 4b). The time-scale of the remagnetization process is sufficiently long to allow for an interpretation based on atomistic spin-dynamics. Two materials specific parameters should be the most relevant to control this dynamics; the exchange interaction, as revealed by the local Weiss field, and the damping parameter. In the Sec.S2 of SM<sup>38</sup>, we report on the calculated Weiss fields and damping parameters. It is clear from these results that the trend in the experimental data shown in Fig. 4b, can not be understood from the Weiss field alone, whereas an explanation based on the damping is more likely. In order to illustrate this, we show in Fig. 5 the inverse of the measured remagnetization time compared to the theoretically calculated damping and experimental measured damping (intrinsic) through ferromagnetic resonance (FMR) (described in Sec.S6 of SM)<sup>38</sup>. The figure shows that the damping is large in the completely disordered  $A_2$  phase and for a large range of structural orderings, which comes out from

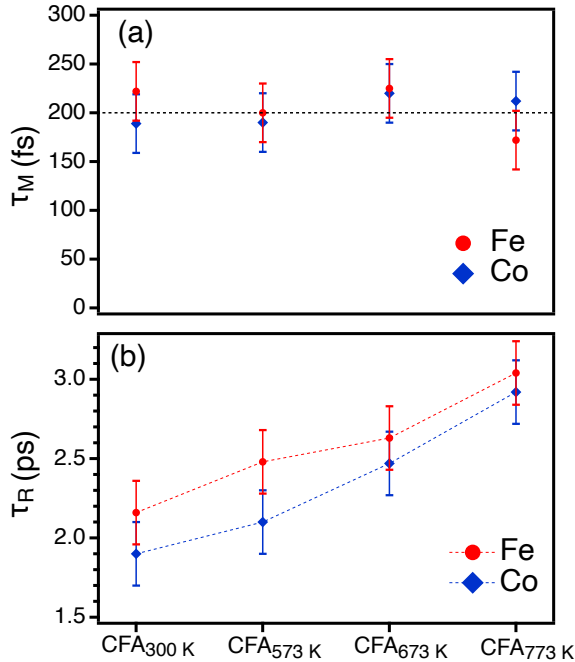


FIG. 4. Measured magnetization times for the investigated  $\text{Co}_2\text{FeAl}$  alloys. In (a) the demagnetization time,  $\tau_M$ , is shown and in (b) the remagnetization time,  $\tau_R$ , is plotted.

both theory and experiment. The figure also demonstrates that the inverse of the measured remagnetization time scales very well with both the calculated damping and experimentally measured damping. According to the figure, a large damping parameter corresponds to faster remagnetization dynamics in the measurements.

$\text{Co}_2\text{FeAl}$  is, to the best of our knowledge, the first system where experimental observations and theory point to the importance of damping in the process of ultrafast magnetization dynamics. We note that this primarily is relevant for the remagnetization process; the initial part of the magnetization dynamics (first few hundred fs) is distinctly different. In the demagnetization we observe a similar behaviour for Fe and Co in all samples, and an insensitivity of the demagnetization times in relation to structural ordering. Also, the measured and theoretical demagnetization times evaluated from atomistic spin-dynamics simulations, do not agree. Other mechanisms, of electronic origin, most likely play role in this temporal regime.

The remagnetization process of  $\text{Co}_2\text{FeAl}$  alloys with varying degree of structural order, highlights clearly the importance of the Gilbert damping and that magnon dynamics dominates the magnetization at ps time-scales. The relevance of the Gilbert damping parameter for ps dynamics is natural, since this controls angular momentum (and energy) transfer to the surrounding. What is surprising with  $\text{Co}_2\text{FeAl}$  is the fact that other interactions (e.g. the Weiss field) show such a weak dependence on the amount of structural disorder. This is fortuitous, since it allows to identify the importance of the Gilbert damping. A picture emerges from the results presented here, that the magnetization dynamics in general have two regimes; one which is primarily governed by electronic processes, and is mainly active in the first few hundred fs ( $\tau_M$ ), and a second regime where it is primarily magnons that govern the remagnetization dynamics ( $\tau_R$ ).

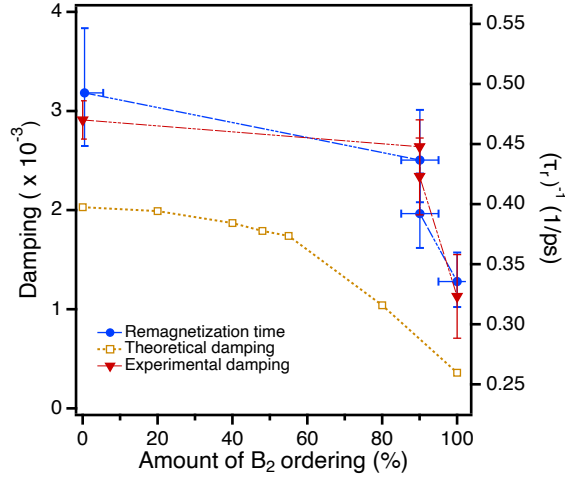


FIG. 5. The relationship of inverse of the measured remagnetization time (right y-axis) and theoretically calculated and experimentally measured Gilbert damping (left y-axis) in  $\text{Co}_2\text{FeAl}$  for varying amount of B2 order along the  $A2 \rightarrow B2$  path, i.e. 0 corresponds to pure A2 phase while 100 corresponds to pure B2 phase.

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