Brillouin light scattering study of Co$_{2}$Cr$_{0.6}$Fe$_{0.4}$Al and Co$_{2}$FeAl Heusler compounds

O Gaier$^{1,3}$, J Hamrle$^{1}$, S Trudel$^{1}$, A Conca Parra$^{1}$, B Hillebrands$^{1}$, E Arbelo$^{2}$, C Herbort$^{2}$ and M Jourdan$^{2}$

$^{1}$ Fachbereich Physik and Forschungszentrum OPTIMAS, Technische Universität Kaiserslautern, Erwin-Schrödinger-Straße 56, D-67663 Kaiserslautern, Germany

$^{2}$ Institut für Physik, Johannes-Gutenberg-Universität Mainz, Staudinger Weg 7, D-55099 Mainz, Germany

E-mail: gaier@physik.uni-kl.de

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Abstract

The thermal magnonic spectra of Co$_{2}$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) and Co$_{2}$FeAl were investigated using Brillouin light scattering (BLS) spectroscopy. For CCFA, the exchange constant $A$ (exchange stiffness $D$) is found to be $0.48 \pm 0.04 \, \mu \text{erg cm}^{-1} (203 \pm 16 \, \text{meV} \, \text{Å}^2)$, while for Co$_{2}$FeAl the corresponding values of $1.55 \pm 0.05 \, \mu \text{erg cm}^{-1} (370 \pm 10 \, \text{meV} \, \text{Å}^2)$ were found. The observed asymmetry in the BLS spectra between the Stokes and anti-Stokes frequencies was assigned to an interplay between the asymmetrical profiles of hybridized Damon–Esbach and perpendicular standing spin-wave modes, combined with the optical sensitivity of the BLS signal to the upper side of the CCFA or Co$_{2}$FeAl film.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Studies of ferromagnetic (FM) half-metals are driven mainly by the possible applications of such materials in spintronic devices as a potential source of a 100% polarized spin current. Some Heusler alloys are promising candidates due to their high Curie temperatures [1]. For example, it was recently proved that spin injection from Co$_{2}$FeSi into the semiconductor (Al,Ga)As can be achieved with a 50% efficiency [2].

Amongst the different Heusler systems studied in recent years, the compound Co$_{2}$Cr$_{0.6}$Fe$_{0.4}$Al (CCFA) has attracted significant experimental [3–8] and theoretical [9–11] attention. CCFA is an interesting candidate for spintronics applications due to its high Curie temperature of 760 K [3] and its high value of volume magnetization of $\approx 3 \, \mu \text{B}$ per formula unit [5, 12] at 5 K (the theoretical value is $3.8 \, \mu \text{B}$ per formula unit [13]). Using spin resolved photoemission, the spin polarization of CCFA at the Fermi level was found to be 45% at room temperature (RT) [14]. CCFA-based spin-dependent transport devices showing large magnetoresistance effects were recently reported. For example, in the simple CCFA/Cu/Co$_{90}$Fe$_{10}$ trilayers a large giant magnetoresistance of 6.8% at RT [15] was found. Tunnelling magnetoresistance ratios (TMRs) of 52% at RT and 83% at 5 K were reported for the CCFA/AlO$_{x}$/Co$_{95}$Fe$_{25}$ magnetic tunnel junction (MTJ) [12]. In the epitaxial CCFA/MgO/Co$_{50}$Fe$_{50}$ MTJ structure, TMR ratios of 109% at RT and 317% at 4.2 K [16] were presented, whereas for the epitaxial CCFA/MgO/CCFA structure TMR ratios of 60% at RT and 238% at 4.2 K are reported [17].

Thin Co$_{2}$FeAl films and MTJ structures containing the Co$_{2}$FeAl electrode have been a subject of extensive studies as well [12, 18–21], even though initial band structure calculations predicted a much lower degree of spin polarization compared with CCFA [9]. However, recent band structure calculations by Felser et al have predicted 100% spin polarization at the Fermi level for bulk Co$_{2}$FeAl [11, 22]. The TMR ratios reported in [12, 18–21] are about 50% at RT and are comparable to those reported for CCFA.

In this paper we report on our study of the thermal spectrum of spin waves in CCFA and Co$_{2}$FeAl films using...
Brillouin light scattering (BLS) spectroscopy. From our BLS spectra we have determined the values of the exchange constants for CCFA and Co2FeAl.

2. Sample preparation and characterization

The CCFA structure under investigation is an Al(2.5 nm)/CCFA(80 nm)/Cr(8 nm)/MgO(1 0 0) epitaxial structure [5, 23]. The buffer layers were deposited by electron beam evaporation onto a single-crystalline MgO(0 0 1) substrate, while the epitaxial CCFA films were subsequently deposited by dc magnetron sputtering. A more detailed description of the sample preparation can be found elsewhere [5]. The films grow with the B2 structure, as there is full disorder between the Cr and Al positions, but order on the Co positions [5]. A volume magnetization of $\mu_{CCFA} \approx 2.5\mu_B$ per f.u. (formula unit) (i.e. $M_S = 490 \text{ emu cm}^{-3}$) was measured at 300 K by SQUID magnetometery. 4-circle x-ray diffraction (XRD) scans yielded lattice parameters $a = 0.570 \pm 0.005 \text{ nm}$ and $b = c = 0.583 \pm 0.012 \text{ nm}$, where the $a$-axis is perpendicular to the sample surface and $b$ and $c$ are in-plane axes [23]. The magnetic reversal of the structure is also described in [23].

The Co2FeAl sample under investigation consists of an 80 nm thick Co2FeAl layer which was epitaxially grown on a single-crystalline MgO(0 0 1) substrate covered with a 10 nm thick MgO buffer layer. Magnetron sputtering was employed for the deposition of both the MgO buffer and the Co2FeAl layer. A post-growth anneal at 550 °C provided a Co2FeAl film with the B2 structure, as confirmed by XRD measurements. A 3 nm thick AlO capping layer was deposited on top of the structure to prevent sample oxidation. The sample exhibits a saturation magnetization of 4.66 $\mu_B$/f.u. measured by SQUID magnetometry at RT. This is in good agreement with the previously reported value of 4.96 $\mu_B$/f.u. determined at 4.2 K [24].

All BLS measurements presented in this paper were performed using a diode pumped, frequency doubled Nd: YVO4 laser with a wavelength of $\lambda = 532 \text{ nm}$ as a light source. Unless specified otherwise, the light impinges on the sample at an angle of incidence of $\varphi = 45^\circ$, corresponding to a transferred wave vector of detected magnons $q_i = 4\pi/\lambda \sin \varphi = 1.67 \times 10^5 \text{ cm}^{-1}$. The external magnetic field $H$ was applied in the so-called magnetostatic surface mode geometry, wherein $H$ is applied in the plane of the sample and is perpendicular to the plane of light incidence (i.e. $H \perp \vec{q}_i$). A more detailed description of the BLS setup used in this work can be found in [25, 26].

3. Experimental results

3.1. CCFA

Typical BLS spectra measured on the investigated CCFA film are presented in figure 1 for several values of the external magnetic field. As is clearly visible, the positions of the peaks in both the Stokes (creation of magnons, negative frequencies in BLS spectra) and anti-Stokes (annihilation of magnons, positive frequencies in BLS spectra) parts of the spectrum move to higher values with an increasing magnetic field. This field dependence is evidence of the magnonic nature of the observed peaks, whereas the position of a phononic peak is not expected to be field dependent. The observed peaks correspond to the Damon–Eshbach (DE) mode and to the perpendicular standing spin-wave (PSSW) modes. As will be discussed below, one of the observed peaks in the BLS spectra results from the hybridization of the DE and the second PSSW mode. Therefore, this mode will be referred to as PSSW2 + DE in the following discussion. The other observed peaks are pure PSSW modes.

Figures 2(a) and (b) provide the dependence of the observed spin-wave frequencies on the value of the external magnetic field which was applied along the $[110]$ (figure 2(a)) and the $[100]$ (figure 2(b)) directions of the CCFA film. The field dependences are nearly identical for both orientations of the external magnetic field. However, the observed spin-wave frequencies are about 1 GHz smaller when $H$ is aligned parallel to the $[100]_{CCFA}$ direction, compared with when the field is applied along the $[110]$ direction. This is also illustrated in figure 2(c), where the dependence of the observed spin-wave frequencies on the in-plane sample orientation (i.e. the angle between the $[100]$ direction and the applied magnetic field) is presented. The lower frequency observed when the field is along the $[100]_{CCFA}$ direction indicates that this direction is an in-plane magnetically hard axis [27], in agreement with previous MOKE investigations [23].

The solid lines in figures 2(a)–(c) are a fit to the experimental data using a theoretical model described in [28]. The found parameters are exchange constant $A = 0.48 \pm 0.04 \text{ meV cm}^{-1}$, cubic volume anisotropy $K_1 = -20 \pm 10 \text{ kerg cm}^{-3}$, Landé g-factor $g = 1.9 \pm 0.1$ and saturation magnetization $M_S = 520 \pm 20 \text{ emu cm}^{-3}$. Note that each of the fitted parameters is determined rather independently by a particular experimental dependence or feature, making the

![Figure 1. (Color online) Selected BLS spectra measured on the CCFA(80 nm) film in an external magnetic field of $H = 400$, 1000 and 1500 Oe. Negative frequencies are related to Stokes processes (creation of magnons), whereas positive frequencies are related to anti-Stokes processes (annihilation of magnons). The dashed lines are guides to the eye.](image-url)
fit reliable and providing results with a relatively small error. In particular, \( M_s \) is obtained from the frequency of the DE mode (here PSSW2 + DE mode) and agrees well with the SQUID value \( M_S = 490 \text{emu cm}^{-3} \) [5]. Landé \( g \)-factor is provided by the slope of BLS frequency on the external field. \( K_1 \) is determined from the variation of BLS frequencies on the sample orientation (figure 2(c)). Finally, the exchange constant \( A \) is determined by frequencies of PSSW modes. Note that the corresponding exchange stiffness of CCFA is \( D = 2 A g \gamma \hbar / M_S = 203 \pm 16 \text{meV Å}^2 \), where \( h \) and \( \gamma = g \gamma_0 \) are the reduced Planck constant and the gyromagnetic ratio, respectively.

For the PSSW2+DE mode we observe a large (between 0.5 and 1 GHz) splitting between the Stokes (▲) and the anti-Stokes (▼) frequencies (figures 2(a) and (b)). This splitting is particularly pronounced for small values of the external field in the range 100–400 Oe. A careful investigation of figures 2(a) and (b) further reveals that the lower frequency component of the split PSSW2 + DE mode is observed only in the Stokes part of the BLS spectrum, whereas the higher frequency component appears only in the anti-Stokes part. Moreover, the slope \( df/dH \) is slightly different for frequencies determined from the Stokes and anti-Stokes parts of the BLS spectra. In the case of a DE mode, such an asymmetry usually indicates different pinning conditions of the dynamic magnetization on each interface. However, in our particular case, the observed asymmetry is related to the hybridization (also called mode repulsion) of the DE and the PSSW2 modes, which is elaborated in detail in the Discussion section.

3.2. Co2FeAl

The BLS spectra collected from the Co2FeAl sample are shown in figure 3. In (a) spectra recorded at a transferred wave vector \( q_f = 1.67 \times 10^3 \text{cm}^{-1} \) and different values of the external magnetic field are displayed, while in (b) BLS spectra measured at \( H = 1 \text{kOe} \) and at different \( q_f \) are presented. As is clearly visible from figure 3(a), the positions of the peaks move to higher values as the magnetic field increases, confirming the magnonic origin of the observed peaks.

Performing BLS measurements at different angles of incidence \( \psi \) (i.e. at different values of \( q_f \)) allows for an unambiguous separation of the dipole dominated magnetostatic surface wave (the DE mode), from the exchange dominated PSSWs. In contrast to PSSW modes, the frequency of the DE mode exhibits a much stronger dependence on the in-plane direction of the wave vector. Therefore, it provides substantial shifts in the collected spectra upon the variation of \( \psi \) (and thus \( q_f \)), while the spectral positions of the PSSW modes are not significantly affected by this variation. Therefore, the peak originating from the DE mode excitation can be easily identified (figure 3(b)).

The extracted peak positions as a function of external magnetic field and incidence angle are presented by symbols in figures 4(a) and (b), respectively. The solid lines present the simulation from which the following parameters were determined: exchange constant \( A = 1.55 \pm 0.05 \mu \text{erg cm}^{-1} \), saturation magnetization \( M_S = 1027 \pm 10 \text{emu cm}^{-3} \) and Landé \( g \)-factor \( g = 2.1 \pm 0.1 \). The corresponding exchange stiffness is \( D = 370 \pm 10 \text{meV Å}^2 \). The value of the cubic volume anisotropy \( K_1 \) was not determined and hence its value is assumed to be zero in the simulations. Note that including \( K_1 \) into the model only slightly changes the frequency of the spin waves (by about 1 GHz). Therefore, setting \( K_1 \) to zero in the model used does not detrimentally affect the determination of \( M_S \) and \( A \).

Note that in the Stokes part of the spectrum (i.e. negative frequencies) shown in figure 3(a), the PSSW2 mode is difficult to recognize. This is due to a relatively small spacing between the PSSW2 and DE modes, as well as the much stronger intensity of the adjacent DE mode. In the anti-Stokes region of the spectrum, the DE mode has intensities that are comparable...
Figure 3. (Color online) (a) BLS spectra of Co$_2$FeAl recorded at a transferred wave vector $q_{\parallel} = 1.67 \times 10^5$ cm$^{-1}$ and different magnetic fields. The change in peak positions upon the variation of the magnetic field reveals the magnonic origin of the peaks. (b) BLS spectra of Co$_2$FeAl recorded in an external applied field $H$ of 1 kOe and different incidence angles $\phi$. In (a) and (b), the dashed lines are guides to the eye. The DE mode exhibits a strong dependence on $\phi$, whereas the frequency of the PSSW modes does not significantly change with $\phi$. A clear distinction between the DE and PSSW modes thus becomes possible. For the assignment of the standing spin-wave peaks to different modes see the text.

Figure 4. (Color online) (symbols) Experimental BLS frequencies of Co$_2$FeAl, corresponding to the peak positions in the BLS spectra presented in figures 3(a) and (b). (solid lines) Simulations for the exchange constant $A = 1.55 \mu$erg cm$^{-1}$, saturation magnetization $M_s = 1027$ emu cm$^{-3}$ and Landé $g$-factor $g = 2.1$. (a) shows the dependence of BLS frequencies on the external magnetic field, whereas (b) demonstrates the dependence of BLS frequencies on the angle of incidence $\phi$.

to, or lesser than the PSSW2 mode. The origin of this asymmetry of the DE mode intensities will be discussed in what follows.

4. Discussion

The values of the exchange constant (exchange stiffness) of CCFA were found to be $A = 0.48 \pm 0.04 \mu$erg cm$^{-1}$ ($D = 203 \pm 16$ meV Å$^2$), whereas the corresponding value for Co$_2$FeAl was found to be $A = 1.55 \pm 0.05 \mu$erg cm$^{-1}$ ($D = 370 \pm 10$ meV Å$^2$). This exchange constant found for Co$_2$FeAl is smaller than both the exchange constants of bcc Fe ($A_{Fe} = 2.0 \mu$erg cm$^{-1}$, $D_{Fe} = 280$ meV Å$^2$ [28–30]) and of bcc Co ($A_{Co} = 2.12 \mu$erg cm$^{-1}$, $D_{Co} = 430$ meV Å$^2$ [31]). However, the exchange stiffness of Co$_2$FeAl is between the exchange stiffness of bcc Fe and bcc Co. On the other hand, the value of the exchange constant (exchange stiffness) of CCFA is very small, being only one-third (one-half) of the exchange constant (exchange stiffness) of Co$_2$FeAl, and about one-half of the exchange constant or exchange stiffness of fcc Ni ($A_{Ni} = 0.85 \mu$erg cm$^{-1}$, $D_{Ni} = 420$ meV Å$^2$). The large discrepancy between the values of exchange determined for Co$_2$FeAl and CCFA demonstrates how these can vary substantially in Co$_2$-based Heusler compounds, even for compounds with the same ordering (B2) and similar compositions.

The value of the cubic volume anisotropy $K_1$ in CCFA is found to be $-20 \pm 10$ kerg cm$^{-3}$, being about 24 times smaller than $K_1$ of bcc Fe ($K_{1,Fe} = 480$ kerg cm$^{-3}$ [32]). However, it is roughly of the order of bcc Co ($K_{1,Co} = 0 \pm 10$ kerg cm$^{-3}$ [31]). The small value of $K_1$ in CCFA found here is consistent with previous investigations showing that the magnetic anisotropy is weak in Heusler compounds [33, 34], reflecting the small anisotropy of the spin–orbit coupling in these materials [35].

To explain the aforementioned asymmetries between the Stokes and anti-Stokes peak positions in the BLS spectra, we
calculated the depth profiles of the dynamic magnetization for CCFA and Co$_2$FeAl films, using the model presented in [28]. The results for CCFA are presented in figure 5 for CCFA thicknesses $t = 60$ and 80 nm, along with the calculated dependence of the spin-wave frequencies on the CCFA thickness. This figure shows that in the vicinity of $t = 80$ nm, i.e. the thickness of the investigated CCFA film, a crossing between the DE and the PSSW modes occurs, resulting in a hybridization of these two modes. Due to the hybridization, a gap of 0.3 GHz is created, which corresponds to the splitting between the Stokes and anti-Stokes frequencies.

The hybridization is also visible in the calculated depth profiles of the dynamic magnetization in figure 5. The left part of each profile image shows the trajectories of the magnetization vector along the film depth (the trajectory at a given depth is an ellipse). In the corresponding right part shows the corresponding profile of the amplitude of the dynamic magnetization in the out-of-plane (dashed line) and the in-plane (solid line) directions, respectively. The sketch in the bottom-right corner shows the used geometry.

In the Co$_2$FeAl film (figure 4), (i) The DE and PSSW modes observed in the Co$_2$FeAl are rather close to each other (about 2 GHz) (ii) the DE and PSSW mode frequencies have different slopes $d f / d H$. (iii) In the Stokes part of the BLS spectra, only the DE mode is visible, since the BLS intensity of the DE mode is much larger than the BLS intensity of the PSSW mode.
of CCFA (figure 5). In particular, the DE and PSSW2 modes for the DE and PSSW2 modes (shown in figure 6). Profiles of dynamic magnetization for the DE and PSSW2 modes, calculated for $t = 80\, \text{nm}$.

(iv) The dependence of the BLS frequency on the CoFeAl thickness (presented in figure 6(a)) is similar to that of CCFA (figure 5). In particular, the DE and PSSW2 modes cross each other at a thickness of about 80 nm in both cases. (v) The calculated profiles of the dynamic magnetization in CoFeAl for the DE and PSSW2 modes (shown in figure 6(b)) demonstrate that they are hybridized. Furthermore, the profiles of those modes are very similar to the profiles of hybridized modes at $t = 80\, \text{nm}$ found in the CCFA film (figure 5).

Therefore, the BLS intensities of each mode are expected to be similar too, which is indeed observed experimentally. While the DE and PSSW2 waves are indistinguishable in CCFA due to a small difference in frequency between these two modes, the frequency difference between these same two modes is larger in the anti-Stokes part of the BLS spectra for CoFeAl, which allowed us to differentiate them. As such, in the case of CoFeAl, we can see that the intensity of the DE mode is smaller than the intensity of the PSSW2 mode on the anti-Stokes side. It confirms our discussion regarding CCFA, showing that the observed mode splitting in CCFA is an interplay between asymmetrical profiles of hybridized modes and the optical depth selectivity of the BLS signal.

5. Conclusion

We have investigated thermal spin waves in CCFA and CoFeAl using BLS spectroscopy. The values of the exchange constant (exchange stiffness) of CCFA was found to be $A = 0.48 \pm 0.04 \, \text{meV} \, \text{Å}^2$ whereas the exchange of CoFeAl was found to be $A = 1.55 \pm 0.05 \, \text{meV} \, \text{Å}^2$. The found cubic volume magnetic anisotropy of CCFA $K_1 = -20 \pm 10 \, \text{meV} \, \text{Å}^2$ is in agreement with small values of $K_1$ found for other Heusler compounds.

The observed asymmetry in BLS spectra between Stokes and anti-Stokes frequencies was assigned to an interplay between asymmetrical profiles of hybridized DE and PSSW2 modes, combined with the optical sensitivity of the BLS signal to the upper side of the CCFA or CoFeAl film, due to a limited probing depth.

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