Determination of exchange constants of Heusler compounds by Brillouin light scattering spectroscopy: application to Co$_2$MnSi

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Abstract

Brillouin light scattering spectroscopy from so-called standing spin waves in thin magnetic films is often used to determine the magnetic exchange constant. The data analysis of the experimentally determined spin-wave modes requires an unambiguous assignment to the correct spin-wave mode orders. Often additional investigations are needed to guarantee correct assignment. This is particularly important in the case of Heusler compounds where values of the exchange constant vary substantially between different compounds. As a showcase, we report on the determination of the exchange constant (exchange stiffness constant) in Co$_2$MnSi, which is found to be $A = 2.35 \pm 0.1 \mu$erg cm$^{-1}$ ($D = 575 \pm 20$ meV Å$^2$), a value comparable to the value of the exchange constant of Co.

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

1. Introduction

The investigation of electron–electron interactions in half-metallic ferromagnetic Heusler compounds is an important issue in order to understand the strong temperature dependence of spin polarization of these materials. One of the key parameters in this context is the magnetic exchange constant (in the following simply referred to as exchange constant) which describes the strength of the exchange interaction between two spins inside a ferromagnetic system. Brillouin light scattering (BLS) spectroscopy from standing spin waves in thin magnetic films is a well-established technique for the study of exchange interaction in various material systems [1–3]. However, the application of this experimental technique for the determination of exchange constants in thin films of Heusler compounds presents some difficulties, which, as discussed in this paper, are mostly related to an ambiguity in the assignment of the measured mode frequencies to the correct standing spin-wave mode orders. The goal of this paper is to show in detail how the values of exchange constants are determined from the BLS spectra measured on thin films of Heusler compounds as well as to discuss the difficulties in the extraction of the exchange constants from the experimental data. For this purpose, we present BLS studies of Co$_2$MnSi films in this paper.

In the following, we briefly describe the investigated Co$_2$MnSi films and the determination of the exchange constant by means of BLS. Thereafter, the experimental results of BLS studies performed on Co$_2$MnSi thin films are presented, and a procedure leading to the correct mode assignment is discussed.

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2. Experimental details

The investigated Co$_2$MnSi films with thicknesses $t = 20, 30, 40, 60$ and $80$ nm were epitaxially grown on a MgO(1 0 0) substrate covered by a 40 nm thick Cr(1 0 0) buffer layer. For $t = 20, 40, 60$ and $80$ nm were epitaxially grown on a MgO(1 0 0) and a transferred wave vector $q = 1.67 	imes 10^5$ cm$^{-1}$ and $q = 80$ nm thick Co$_2$MnSi film measured at $H = 1.5$ kOe and different angles of incidence $\varphi$, i.e. different transferred wave vectors $q$. The solid line is a guide to the eye, showing expected peak positions as follows from the model.

The BLS measurements presented in this paper were performed at room temperature in the magnetostatic surface mode geometry where the magnetic field $H$ is applied in the plane of the sample and perpendicular to the plane of light incidence, i.e. perpendicular to the transferred wave vector $q$. The BLS spectra of Co$_2$MnSi films with varying thicknesses were recorded at an external magnetic field of $5$ kOe and different angles of incidence $\varphi$, where the magnetic origin of the peaks presented in figure 1(a) is compared with simulations which were performed using a theoretical model described in detail in [9]. The exchange constant $A$, the Landé $g$-factor, the saturation magnetization $M_S$ and the magnetic anisotropies are the free parameters in these simulations. Because our previous BLS investigations of Co$_2$MnSi films have shown very small anisotropy in L2$_1$ ordered Co$_2$MnSi films [6], the anisotropy values were set to zero.

As follows from the analytical expressions [6–8], for small spin-wave vector used in our investigations, the frequency of the DE mode depends only marginally on the value of $A$, i.e. DE mode frequency is particularly determined by the values of $M_S$ and $g$. On the other hand, the frequencies of the PSSW modes are particularly determined by the $A/M_S$ ratio and $g$. Furthermore, the Landé $g$-factor is easy to determine independently, as it scales with the slope of the BLS frequency on applied magnetic field, $d/\partial H$. Therefore, the fitting procedure is not underdetermined and the parameters $M_S$, $A/M_S$ and $g$ are found rather independently of each other, providing a high reliability (low correlation) of the fitted values.

3. Results and discussion

Examples of BLS spectra collected from 20, 40, 60 and 80 nm thick Co$_2$MnSi films are shown in figure 1(a). The spectra were recorded at an external magnetic field of $H = 1.5$ kOe and a transferred wave vector of $q = 1.67 	imes 10^5$ cm$^{-1}$ (i.e. $\varphi = 45^\circ$). The magnetic origin of the peaks presented in figure 1(a) is confirmed by $H$-dependent measurements, demonstrated in figure 1(b) for the case of the 40 nm thick Co$_2$MnSi film. The peak positions in both the Stokes (negative frequencies) and the anti-Stokes (positive frequencies) part of the spectrum move towards higher frequencies upon increasing
Triangles up (down) represent Stokes (anti-Stokes) frequencies. Calculations were performed using the exchange constant $(\Delta A)$ for two different values of the exchange constant $(\Delta A) = 2.35 \pm 0.1 \, \mu\text{erg cm}^{-1}$ and $(\Delta A) = 0.6 \pm 0.1 \, \mu\text{erg cm}^{-1}$. The remaining parameters used in the calculations are the saturation magnetization $(M_s = 970 \, \text{emu cm}^{-1})$ and the Landé $g$-factor $(g = 2.05)$. The magnetic anisotropies are neglected.

The field, revealing their magnetic origin. Figure 1(c) shows the BLS spectra recorded for the 80 nm thick Co$_2$MnSi film for different spin-wave wavevectors $q_1$. Compared with the PSSW modes, the DE mode exhibits a much stronger dependence on the wavevector. Therefore, the peaks originating from the DE mode excitation can be easily identified, whereas the spectral positions of the PSSW modes remain nearly unchanged (figure 1(c)). As expected, the frequency of the DE mode increases with increasing film thickness whereas the frequencies of the PSSW modes shift to lower values (figure 1(a)).

Results of numerical simulations (solid lines) are shown in figure 2 along with the experimentally determined BLS frequencies (▲, ▼ represent Stokes, anti-Stokes frequencies, respectively) for two different values of the exchange constant $A$. The saturation magnetization $(M_s = 970 \, \text{emu cm}^{-1})$ (corresponding to $\mu = 4.72 \, \mu_B/\text{f.u.}$) and Landé $g$-factor $(g = 2.05)$ are found for both values of $A$ and are in agreement with previous investigations (e.g. [10–12]). An equally good agreement between the simulations and the experimental data points is achieved for $A = 2.35 \pm 0.1 \, \mu\text{erg cm}^{-1}$ ($D = 575 \pm 20 \, \text{meV Å}^2$) (figures 2(a)–(c)) and $A = 0.60 \pm 0.05 \, \mu\text{erg cm}^{-1}$ ($D = 145 \pm 10 \, \text{meV Å}^2$) (figures 2(d)–(f)), respectively. When $A = 2.35 \pm 0.1 \, \mu\text{erg cm}^{-1}$, the calculations describe all observed PSSW modes. This $A$ value, however, is surprisingly large, being nearly as large as the exchange constant reported for Co(fcc) $(A = 2.73 \, \mu\text{erg cm}^{-1}, D = 466 \, \text{meV Å}^2)$ or Co(hcp) $(A = 2.85 \, \mu\text{erg cm}^{-1}, D = 435 \, \text{meV Å}^2)$ [2]. In the second case, where $A = 0.6 \, \mu\text{erg cm}^{-1}$, only even PSSW modes seem to be observed in the experiment.

Obviously from the fit alone, the correct value of the exchange constant cannot be obtained due to the ambiguity in mode order assignment. To find out which value of the exchange constant is the correct one, we have performed analytical calculations of the BLS intensities. In particular, we have examined under which conditions the BLS intensity becomes zero for odd PSSW modes only. The used analytical approach (presented in appendix A) combines an expression for the BLS intensity, as proposed by Buchmeier et al. [3, 13], and the analytical expression of the depth selectivity of the MOKE effect by Hamrle et al. [14]. The calculations indicate that when conditions (A8) and (A9) derived in appendix A are fulfilled, the BLS intensity will be zero for all odd modes. Using the complex refractive index of Co$_2$MnSi $N^{\text{CMS}} = 1.1 + 1.1i$ [15], an incidence angle $\varphi = 45^\circ$ and a thickness $t = 20 \, \text{nm}$, the value of conditions (A8) and (A9) becomes $4\varphi(N_z)t/\lambda = 0.15$ and $4\varphi(N_z)t/\lambda = 0.18$, respectively. Those values are far from the required values of an odd integer (condition (A8)) and zero (condition (A9)), respectively. This proves that all BLS modes in our Co$_2$MnSi films should contribute with significant scattering cross section. Hence the exchange constant of Co$_2$MnSi is $A = 2.35 \pm 0.1 \, \mu\text{erg cm}^{-1}$ (i.e. exchange stiffness $D = 575 \pm 20 \, \text{meV Å}^2$), which was
determined from the calculations presented in figures 2(a)–(c), that predicted all experimentally observed modes.

Here, we would like to comment on the values of the exchange constant reported for Co$_2$MnSi by different groups. In [16], the value of exchange stiffness was determined to be $D = 466$ meV Å$^2$ (i.e. $A = 1.93$ μerg cm$^{-1}$), using the temperature dependence of the saturation magnetization. This value is rather close to the one we have determined in our BLS investigations. In [10], the value of exchange stiffness was estimated to be $D_B = 1.9 \times 10^{-9}$ Oe cm$^2$ (i.e. $D = 225$ meV Å$^2$, $A = 0.97$ μerg cm$^{-1}$) from FMR investigations, which is much lower compared with our results. This discrepancy might originate from the fact that in [10] asymmetrical pinning conditions were assumed (i.e. the dynamic magnetization is unpinned at the second one). In our samples, this condition does not apply due to the following considerations. As can be seen in figure 2, the frequencies of the DE mode exhibit equal values in both the Stokes and the Anti-Stokes part of the spectrum. Since the Stokes and Anti-Stokes DE modes are bound to opposite interfaces of the FM layer, it shows that in our Co$_2$MnSi films both interfaces are magnetically equivalent.

Finally, we would like to note that in our previous paper reporting on BLS studies of Co$_2$MnSi films [6] the assumption that only even PSSW modes are observed has been used. In view of the results presented here, the conclusions of this paper remain valid. The values of the exchange constant in figure 9(b) in [6], however, must be scaled by a factor of 5 to get the correct values.

4. Conclusion

Using the case of Co$_2$MnSi thin films, we demonstrated in detail how the values of exchange constants are determined from the BLS measurements and carefully discussed different values of $A$ proposed by numerical simulations. The value of Co$_2$MnSi is found to be $A = 2.35 \pm 0.1$ μerg cm$^{-1}$ ($D = 575 \pm 20$ meV Å$^2$), in agreement with the value determined from the temperature dependence of the magnetization [16]. The found value of exchange is comparable to the value of exchange constant of Co. The pinning conditions for the dynamic magnetization are found to be equal for both Cr/Co$_2$MnSi and Al/Co$_2$MnSi interfaces.

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Appendix A. Depth sensitivity of BLS

The BLS intensity from a single FM layer of thickness $t$ is given by [13]

$$I^{(BLS)} = I_0 \left| \int_0^t \left[-L(z)m_L(z) + P(z)m_P(z)\right] \, dz \right|^2,$$  

(A1)

where $m_L(z)$ and $m_P(z)$ are the depth profiles of the dynamic magnetization through the FM layer in longitudinal (i.e. in the plane of the sample and in the plane of light incidence) and polar (i.e. out-of-plane) directions, respectively. $L(z)$, $P(z)$ are complex depth sensitivity functions of the off-diagonal reflectivity coefficient $r_{qp}$ for longitudinal $M_L(z)$ and polar $M_P(z)$ static magnetization profiles [13, 14]

$$r_{qp}(\vec{M}) = r_{qp}(M = 0) + \int_0^t \left[ L(z)M_L(z) + P(z)M_P(z)\right] \, dz.$$  

(A2)

The dependence of $L(z)$, $P(z)$ on depth $z$ is the same for both of them. Assuming that the substrate has a refractive index identical to the refractive index of the FM layer, the analytical terms of $L(z)$ and $P(z)$ may be written as [14]

$$L(z) = L(0) \exp[-4\pi i N_z z/\lambda]$$  

(A3)

$$P(z) = \gamma L(z),$$  

(A4)

where the complex coefficient $\gamma$ is the ratio of $P(0)$ and $L(0)$. $N_z$ is the normalized wave vector in the polar direction $N_z = \sqrt{(N^{(\text{fm})})^2 - (N^{(\text{air})})^2} \sin \varphi$, where $N^{(\text{fm})}$ and $N^{(\text{air})}$ are the refractive indices of the FM layer and air, respectively, and $\varphi$ is the angle of incidence of the probing light beam with respect to the sample’s normal. Note that if the refractive index of the substrate is different from the one of the FM layer, the analytical expressions become more complex. However, the basic features (i.e. continuous decay of the amplitude and continuous shift of the phase) remain valid.

Using the open (anti-pinning) boundary conditions at the interfaces of the FM layer, the depth profile of the dynamic magnetization of a PSSW mode of the $m$th order reads as [8]

$$m_L(z, \tau) = m_0 \cos(m\pi z/t) \cos(\omega_{sw} \tau)$$  

(A5)

$$m_P(z, \tau) = m_0 \varepsilon \cos(m\pi z/t) \cos(\omega_{sw} \tau + \pi/2),$$  

(A6)

where $\omega_{sw}$ and $\tau$ are the frequency of the spin-wave mode and time, respectively. The magnetization vector follows an elliptical trajectory, with ellipticity $\varepsilon$. Therefore, the $m_L$ and $m_P$ magnetization components are shifted by $\pi/2$ in their time dependence.

Combining equations (A1), (A3)–(A6) and integrating over the thickness of the FM layer and averaging over time, we obtain

$$I^{(BLS)} = \frac{1}{2} I_0 \left| \int_0^t \left[-L(0)\alpha t m_0 \right] \, dz \right|^2 \times \left| 1 - \exp(\pm i\alpha)(-1)^m \right|^2,$$  

(A7)

with $\alpha$ being a dimensionless parameter defined as $\alpha = 4\pi N_z t/\lambda$.

The total BLS intensity $I^{(BLS)}$ is zero when the last term in equation (A7) is zero. For odd $m$, the intensity is zero when the following conditions hold:

$$4\pi(N_z t/\lambda) = 2k + 1,$$  

(A8)

$$4\pi(N_z t/\lambda) = 0,$$  

(A9)
where \( k \) is integer. Note that for \( m = 2k + 1 \), the denominator in equation (A7) becomes zero as well, which would also provide a zero BLS intensity. For even \( m \), \( I^{\text{BLS}} \) is zero when

\[
2\pi(N_i) / \lambda = k, \quad (A10)
\]

\[
4\pi(N_i) / \lambda = 0, \quad (A11)
\]

where \( k \) is again integer.

## Appendix B. Units of exchange

Throughout the literature, the exchange is expressed in at least four different variables, making the direct comparison of exchange values difficult. This situation is further complicated by the use of both cgs and SI units, as well as the lack of a unique nomenclature. Here, we give a short overview of different definitions of exchange and the conversion between them.

The energy of a single magnon (i.e. a spin wave, where one single spin in one period of spin wave is reversed) having wave vector \( \vec{k} \) is given by [17]

\[
E = \hbar \omega = DK^2, \quad (B1)
\]

where \( D \) is called the exchange stiffness (which is sometimes referred to as the spin-wave stiffness), \( \omega \) is the angular frequency of the spin wave and \( \hbar \) is the reduced Planck constant. The routinely used units of \( D \) are meV Å² for both cgs and SI units.

Alternatively, the exchange is expressed as the strength of the effective field \( DB \) created by the exchange energy of the spin wave. As \( D \), \( DB \) is also often called the exchange stiffness. Neglecting a dipolar contribution to the spin wave (i.e. assuming a large value of \( k \)), the angular frequency of a spin wave is given by

\[
\frac{\omega}{g\gamma_0} = (B + DBk^2), \quad (B2)
\]

where \( \omega \) is the angular frequency of the spin wave, \( g \) is Landé g-factor, \( B \) is an external field and \( g\gamma_0 \) is the gyromagnetic ratio, where \( \gamma_0 = e/(2mc) = 8.793 \times 10^{-10} \text{s}^{-1} \text{T}^{-1} \) in SI and \( \gamma_0 = e/(2mc) = 8.793 \times 10^{-7} \text{G}^{-1} \) in cgs, \( e \) and \( mc \) being the speed of light, and \( e \) and \( mc \) are the charge and the rest mass of an electron, respectively. Units of \( DB \) are Oe cm² in cgs, and T m² or J A⁻¹ in SI.

Another description of the exchange is provided by the exchange constant \( A \) (sometimes also called the exchange stiffness). Within this description, the energy density \( E/V = \mathcal{E} \) of a given continuous magnetization distribution \( \vec{m}(\vec{r}) \) within space is given by

\[
E/V = \mathcal{E} = A|\nabla \vec{m}|^2, \quad (B3)
\]

where \( \vec{m} = \vec{M}/M_S \) is the reduced magnetization and \( M_S \) is the saturation magnetization. Units of \( A \) are erg cm⁻¹ in cgs and J m⁻¹ in SI. Assuming a reduced magnetization \( \vec{m} \) in the form of a spin wave with wavevector \( k \), the effective field of such a spin wave is

\[
B_{\text{eff}} = -\nabla \mathcal{E} = \frac{2A}{M_S}k^2. \quad (B4)
\]

Comparing equations (B1)–(B4), the conversions between \( D \), \( DB \) and \( A \) are (valid both in SI and cgs)

\[
D_B = 2A/M_S, \quad (B5)
\]

\[
DB = \frac{D}{g\gamma_0\hbar} = \frac{D}{g\mu_B}, \quad (B6)
\]

\[
A = \frac{DM_S}{2g\gamma_0\hbar} = \frac{DM_S}{2g\mu_B}, \quad (B7)
\]

where \( \mu_B = \gamma_0 \) is the Bohr magneton.

Note that the conversion between SI and cgs for the exchange constant is given by 1 erg cm⁻¹ = \( 10^{-5} \) J m⁻¹. For the saturation magnetization 1 emu cm⁻³ = 1000 A m⁻¹. If \( M_S = 1 \) emu cm⁻³, then 4\( \pi M_S = 4\pi G \) (i.e. in unit of Gauss).

Finally, here we mention a description of exchange coming from the effective Heisenberg Hamiltonian with classical spins

\[
E = -\sum_{ij}(J_{ij}\vec{s}_i \cdot \vec{s}_j), \quad (B8)
\]

where \( J_{ij} \) is the exchange interaction energy between two spins at positions \( i, j \) and \( s_i, s_j \) are unit vectors pointing in the direction of local magnetic moments at sites \( i, j \), respectively. Assuming that a unit cell contains a single atom, the spin-wave energy \( E(\vec{k}) \) is related to the exchange parameters \( J_{ij} \) by a simple Fourier transformation [18]

\[
E(\vec{k}) = \frac{4\mu_B}{\mu} \sum_{ij}\langle 1 - \exp(i\vec{k} \cdot \vec{R}_{ij}) \rangle, \quad (B9)
\]

where \( \vec{R}_{ij} = \vec{R}_i - \vec{R}_j \) denotes a lattice vector in real space, \( \vec{k} \) is the spin-wave vector, \( \mu \) is the magnetic moment per atom and \( \mu_B \) is the Bohr magneton. The exchange stiffness \( D \) is given by the curvature of the spin-wave dispersion \( E(\vec{k}) \) at \( \vec{k} = 0 \). When the unit cell contains several atoms, the spin-wave energy is expressed, e.g. in [19].

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