Interlayer Exchange Coupling in Full Heusler Alloy-Based Layered Structures

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学位授与番号	工博第4217号
URL	http://hdl.handle.net/10097/62016

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授　与　学　位	博士（工学）
学位授与年月日	平成２１年９月９日
学位授与の根拠法規	学位規則第４条第１項
研究科，専攻の名称	東北大学大学院工学研究科（博士課程）知能デバイス材料学専攻
学位論文題目	Interlayer Exchange Coupling in Full Heusler Alloy-Based Layered Structures （フルホイスラー合金系積層構造における層間交換結合）
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論 文 内 容 要 旨

1. Introduction

The discovery of antiferromagnetic interlayer exchange coupling (IEC) in Fe/Cr/Fe layered structures [1] and accordingly the manifestation of giant magnetoresistance effect [2] gave birth to a new field of research, called spintronics that combines physics, technologies and industrial applications. The precise knowledge of IEC is very important to control the magnetic behavior in a lot of spintronic devices, such as MRAM, reading heads, etc. Cobalt-based full-Heusler alloy such as Co_2MnY(Y = Si, Ge, etc.) is a prospective candidate for the next generation electronics due to their large spin-polarizations and high Curie temperatures. However, IEC in full-Heusler alloy based systems has not been studied sufficiently and the understanding of IEC in such systems remains poor [3,4]. Recently, our group reported the absence of 180° coupling and only the oscillatory 90° coupling with a very long oscillation period (3.3-3.5 nm) in Co_2MnSi(CMS)/Cr/CMS trilayer structures [5,6]. The origin of this anomalous behavior remains an open question, and further systematic study is required.

The main objectives of this study are to investigate the IEC in epitaxially grown full-Heusler alloy-based systems systematically and to find out the mechanism responsible for the unusual behavior of IEC in CMS/Cr/CMS trilayers. First, the IEC has been investigated in different full-Heusler Co_2XY (X = Mn, Fe, and Y = Si, Al) alloy based trilayer structures with a Cr spacer. Then, the IEC has been investigated using different spacer materials (V and Ag) in CMS based epitaxial trilayer structures. Moreover, the effect of the change of chemical composition of Co-Mn-Si has been investigated in Co-Mn-Si/Cr/Co-Mn-Si trilayers. Finally, the IEC has been investigated in binary alloy Co-Fe/Cr/Co-Fe epitaxial trilayer structures for the comparison with that in full-Heusler alloy based structures.

2. Experiments and Numerical simulations

The samples in this study were prepared with a stacking structure of Cr(5 nm)/Au(30 nm)/Cr(15 nm)/Ferromagnet(FM)/metallic spacer/FM/Au-cap on a MgO(001) substarte by a UHV-compatible dc sputtering method (1×10^{-7} Pa). The magnetic properties were measured by vibrating sample magnetometer (VSM), superconducting quantum interference device (SQUID) and polarized neutron reflectivity (PNR) measurements. The structural and surface properties were analyzed by x-ray diffraction (XRD), Atomic force microscope (AFM), high resolution tunnel electron microscopy (HRTEM), whereas, the composition was analyzed by electron probe micro analyzer (EPMA) and inductively coupled plasma (ICP) analysis. The experimental *M-H* loops were fitted by a numerical simulation model based on the three major energy contributions, namely, the anisotropy energy (E_K), the exchange energy (E_J) and the Zeeman energy (E_Z). The total energy can be expressed as [5,6]

$$E_{total} = E_K + E_J + E_Z \quad (1)$$

where,

$E_K = K_a t_a \sin^2\alpha\cos^2\alpha + K_b t_b \sin^2\beta\cos^2\beta$

$E_J = -J_1\cos(\alpha-\beta) - J_2\cos^2(\alpha-\beta)$

$E_Z = -M_a t_a H\cos\alpha - M_b t_b H\cos\beta.$

M, *K* and *t* in eq. (1) are the magnetization, the first-order cubic magnetocrystalline anisotropy constant and the thickness of a FM layer, respectively. The subscripts *a* and *b* represent the top and bottom FM layers, respectively. J_1 and J_2 are the bilinear and biquadratic exchange coupling constants, respectively. *H* is the applied magnetic field and α (or β) represents the angle between the direction of *H* and *M* of the bottom (or top) FM layer in plane.

3. IEC in Co_2XY (X = Mn, Fe, Y = Si, Al)/ Cr based epitaxial trilayers

The trilayers of CMS/Cr/CMS, Co_2MnAl(CMA)/Cr/CMA, Co_2FeSi(CFS)/Cr/CFS, and CMS/Cr/Fe were prepared epitaxially with very small average surface roughness (R_a~ 0.2 to 0.4 nm). The XRD measurements confirmed the presence of long range *B*2 ordering in CMS, CMA and CFS. The annealing temperature dependence for bottom CMS in CMS/Cr/CMS trilayers and the spacer layer thickness dependence of IEC in CMS/Cr/Fe, CMA/Cr/CMA and CFS/Cr/CFS trilayers showed the existence of dominating 90° coupling without any detectable 180° coupling. The t_{Cr} dependence of 90° coupling strength $-J_2$ in different full-Heusler alloy based systems is shown in Fig. 1. Moreover, the presence of only 90° coupling was also confirmed by PNR measurement result in CMS/Cr/CMS trilayer.

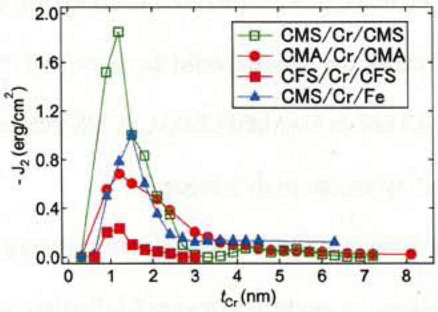

Figure 1: Spacer thickness (t_{Cr}) dependence of 90° coupling strength $-J_2$ is plotted for CMS/Cr/CMS [4], CMA/Cr/CMA, CFS/Cr/CFS, and CMS/Cr/Fe trilayer structures.

4. IEC in Co_2MnSi/X(V, Ag)/Co_2MnSi epitaxial trilayer structures

IEC has been investigated in full-Heusler alloy CMS based structures with different spacer materials. CMS/V/CMS and CMS/Ag/CMS trilayers with different spacer thicknesses were prepared epitaxially with almost flat surface (R_a< 0.4 nm). CMS was grown in long range *B*2 ordered structure. However, neither 90° coupling nor 180° coupling was detected for any of the cases.

5. IEC in Co-Mn-Si/Cr/Co-Mn-Si epitaxial trilayer structures

To elucidate the effect of chemical composition and the degree of $B2$ ordering of full-Heusler alloy on the IEC behavior, the Co-Mn-Si/Cr/Co-Mn-Si trilayers were prepared by increasing the composition of Co from 53.3 to 80 at.%, systematically. The degree of long range $B2$ ordering measured from XRD patterns was gradually decreased with increasing the Co composition and then disappeared for the Co composition over 75.6 at.% (Fig. 2(a)). Likewise, the 90° coupling strength was decreased in a similar fashion and took a minimum at the Co composition 70.3 at.%. Afterwards, the clear contribution of $-J_1$, *i.e.*, 180° coupling with 90° coupling appeared for the Co composition above 73 at.% (Fig. 2(b)). The spacer thickness (t_{Cr}) dependence of IEC was also investigated in highly Co enriched (Co composition 75.6 at.%) Co-Mn-Si based trilayers. The contribution of both the bilinear and 90° coupling was observed in such structures. The coupling angle at remanence was found to vary with t_{Cr}, and a maximum of about 152° was observed at t_{Cr}= 1.5 nm.

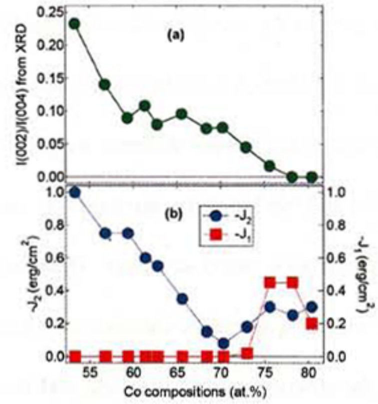

Figure 2: Intensity ratio of the superlattice peak I(002) to the fundamental peak I(004) measured from XRD (a) and the values of J_1 and J_2 evaluated from the comparison between experiments and numerical simulations (b) as a function of the Co composition for a series of samples with different Co compositions in Co-Mn-Si/Cr/Co-MnSi trilayers.

6. IEC in Co_xFe_{1-x}/Cr/Co_xFe_{1-x} epitaxial trilayer structures

To elucidate the effect of the composition of two magnetic elements and the $B2$ ordering on IEC behavior, binary alloy Co_xFe_{1-x}/Cr/Co_xFe_{1-x} trilayer structures were prepared at RT for $x = 0$ to 0.8. The strong 180° coupling with comparatively weak 90° coupling was observed for all the values of x. However, when the bottom Co-Fe in Co_xFe_{1-x}/Cr(1.2 nm)/Co_xFe_{1-x} ($x = 0.4$ and 0.5) trilayers was annealed at 400°C, the strength of $-J_2$ became comparable to that of $-J_1$. Figure 3 shows the change of J_1/J_2 with T_{ann} in $Co_{0.4}Fe_{0.6}$/Cr/ $Co_{0.4}Fe_{0.6}$ trilayers. Therefore, this result suggests that the increase in $-J_2$ is associated with the enhancement of $B2$ ordering by annealing.

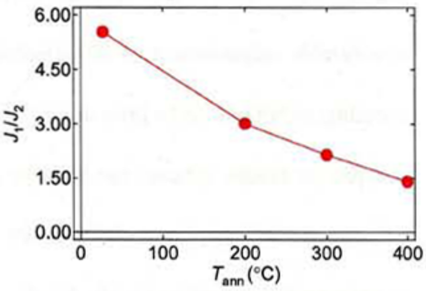

Figure 3: The ratio of the 180° coupling strength ($-J_1$) and the 90° coupling strength ($-J_2$), J_1/J_2 as a function of the annealing temperature T_{ann} for the bottom $Co_{0.4}Fe_{0.6}$(20 nm) layer in $Co_{0.4}Fe_{0.6}$(20 nm)/Cr(1.2 nm)/$Co_{0.4}Fe_{0.6}$ (7 nm) trilayers.

7. Discussion

The experimental investigations of this study suggest that both the degree of $B2$ ordering and the spacer Cr are important for the observation of anomalous behavior of 90° coupling. No existing theoretical models of IEC can explain this observation clearly, since they have some limitations. Very recent theoretical investigation by Inoue in Nagoya University observed that the magnetic frustration at the interface between the Cr spacer and full-Heusler alloy shows completely different behavior [7] from that between Cr and ferromagnetic transition metal. Cr has antiferromagnetic coupling with Fe (with *A*2 structure) at the interface and due to the one monolayer terraces the two dimensional magnetic frustration arises. Whereas, in case of long range and short-range ordered Co and Mn atoms in Co_2MnSi, Cr on the Co and Mn-Si terminated interfaces couples ferromagnetically and antiferromagnetically with Co and Mn, respectively. These different magnetic exchange interactions of Cr with periodically arranged two different magnetic elements (Co and Mn/Fe) at the interface may cause the frustration giving rise to the dominating 90° coupling in ordered binary alloy and full-Heusler alloy based structures. Therefore, theoretical calculations considering full-Heusler alloy structure and the magnetic interaction with Cr spacer at the interface may lead to the understanding of the strong 90° coupling in full-Heusler alloy based systems. However, the absence of 180° coupling and the long period oscillation of only 90° coupling in ordered full-Heusler alloy based structures are still mysterious.

8. Conclusions

In this study, the interlayer exchange coupling in different full-Heusler alloy based structures was investigated, systematically. The dominating 90° coupling and the absence of 180° coupling was observed in all the *B*2 ordered full-Heusler alloy based, CMS/Cr/CMS, CMA/Cr/CMA, CFS/Cr/CFS and CMS/Cr/Fe trilayer structures. However, no non-ferromagnetic coupling was observed for V and Ag spacers in CMS based trilayer structures. In the study of Co composition dependence of IEC in Co-Mn-Si/Cr/Co-Mn-Si trilayers the strength of 90° coupling was found to decrease with Co compositions and contribution of 180° coupling appeared for highly Co antisite defective Co-Mn-Si structure (Co composition over 73 at.%), when the long range *B*2 ordering almost disappeared. The considerable enhancement of 90° coupling strength was also observed in binary alloy based Co-Fe/Cr/Co-Fe trilayers after the annealing of bottom Co-Fe layer at T_{ann} = 400 °C. This might be caused due to the enhancement of *B*2 ordering in Co-Fe by annealing. The present results indicate that both the degree of *B*2 ordering of full-Heusler alloy and the spacer material Cr have significant influence on 90° coupling in full-Heusler alloy based structures. No present theoretical models of 90° coupling could explain the experimental observations clearly. Finally, this work has introduced anomalous behavior of 90° coupling in full-Heusler alloy based systems; the understanding of the underlying mechanism is a challenge for the theoretical physics.

References

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論文審査結果の要旨

2つの強磁性層間に非強磁性層を挟んだ3層構造において発現する層間交換結合は、これまで多くの研究がなされてきた。一般的に層間交換結合は、2つ強磁性層の磁化が平行に揃う強磁性的な結合と反平行に揃う180° 結合が非強磁性層厚に対して振動的に出現する。しかしながら近年、フルホイスラー合金の1つであるCo_2MnSiとCr中間層を用いた3層構造において、180° 結合の寄与が完全に消失し、代わりに非常に大きな90° 結合が生じることが報告され、関心を集めている。フルホイスラー合金は高いスピン分極率を有することからスピントロニクス用の材料として注目されており、フルホイスラー合金の層間交換結合を理解することは、応用上重要である。本論文は、フルホイスラー合金を用いた積層構造における層間交換結合について、フルホイスラー合金の種類や非強磁性層材料の種類、および非強磁性層厚に対する依存性を系統的に評価し、その層間交換結合の起源について考察したものであり、全編8章から成る。

第1章は序論であり、本研究の背景および目的を述べている。

第2章では、本研究における試料の作製法と評価法、並びに磁化曲線のシミュレーション手法とそれによる交換結合の評価手法について述べている。

第3章では、Cr中間層とCo_2MnSiおよびCo_2MnAl, Co_2FeSiを用いた場合における層間交換結合についての実験結果を述べている。まず90° 結合の強さがCo_2MnSiの$L2_1$規則度に依存しないという重要な知見を得ている。次に、Co_2MnAlやCo_2FeSiを用いた場合においても、同様に90° 結合のみが現れることを見いだしている。さらに、$Co_2MnSi/Cr/Co_2MnSi$において偏極中性子反射率測定を行うことにより、2つのCo_2MnSi層の磁化がほぼ90° の相対角度を有していることを直接的に観測している。

第4章では、Cr以外の中間層としてVおよびAgを用いた積層構造を作製し、その層間交換結合を評価している。Cr以外の非強磁性層を用いた場合では、180° 結合も90° 結合も現れず、大きな90° 結合の起源がCrの有する反強磁性に起因することを示唆している。

第5章では、Co-Mn-Si/Cr/Co-Mn-Siの積層構造における層間交換結合のCo組成依存性について述べている。Co組成を増大させることによって90° 結合の寄与が小さくなること、およびCo組成が73 at.%を超えると、それまで消失していた180° 結合が現れるという結果を得た。これは90° 結合がB2規則度と関係していることを示す重要な知見である。

第6章では、フルホイスラー合金と比較するため、2元合金のCoFeとCrを用いた積層構造における層間交換結合を調べ、CoFeの組成依存性およびアニール温度依存性について述べている。CoFeがas-depositedの不規則状態においては180° 結合が支配的であるが、アニールしCoFeがB2規則化した場合においては90° 結合の寄与が増大することを見いだしている。この結果は、第5章におけるCo-Mn-Siの結果と同様に、90° 結合とB2規則度の関係性を示すものである。

第7章は考察である。90° 結合を説明する既存の全ての理論モデルについて詳細に検討し、それらでは本研究の実験結果を完全には説明できないことを述べている。また、界面におけるCrとCoおよびCrとMn（Fe）との交換結合、強磁性層におけるCoとMn（Fe）のB2規則化、およびCr層の反強磁性が組み合わさった特異な状況において生じる磁気的なフラストレーションが重要であることを示唆している。

第8章は結論である。

以上要するに本論文は、フルホイスラー合金系積層構造における層間交換結合について、系統的かつ詳細な研究を行い、その特異性を明らかにするとともに、起源解明のための重要な知見を得たもので、スピントロニクスや磁性薄膜に関わる材料物性学の発展に寄与するところが少なくない。

よって，本論文は博士（工学）の学位論文として合格と認める。