$L2_1$ -atomic order and spin-polarization in Co₂MnZ (Z = Ge, Sn) Heusler thin films

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Half-metallic Co₂-based Heusler alloys have attracted much interest for spintronic applications because of their predicted 100% spin-polarization (*P*) and high Curie temperature (*T*_C), which are expected to enhance the performance of spintronic devices. In fact, several experimental studies have already demonstrated their effectiveness in enhancing the giant-magnetoresistance, tunnel-magnetoresistance and spin-accumulation effects. Despite large magnetoresistance (MR) ratio observed in Heusler-based devices, structural disorder is still one of the remaining issues which lowers the spin-polarization. Therefore, a high degree of structural ordering is necessary to realize the half-metallicity. An enhanced MR has been reported in various devices using Co₂-based Heusler alloys such as Co₂MnSi and Co₂FeGe_{0.5}Ga_{0.5} by promoting the structural order ($B2 \rightarrow L2_1$ -ordering) by annealing at high temperature (> 500 °C). For various applications, however, applicable maximum annealing temperature is limited, e.g., less than 300 °C is required for a magnetic read head for HDD because of the temperature tolerance of the NiFe shield. Therefore, it is desirable to search for other Heusler alloys which crystallize in $L2_1$ -order below 300 °C. The present work is motivated by the Okubo *et al.* [1]'s report, where the $L2_1$ to *B2*-order transition temperature of Co₂MnZ (*Z* = Ge, Sn) alloys is found to be above 1500 K; and hence $L2_1$ -ordering is expected to appear even by annealing at relatively low temperature.

Epitaxial Co₂Mn*Z* (*Z* = Ge, Sn) (30 nm) and Co₂FeGe_{0.5}Ga_{0.5} (50 nm) films were grown on MgO (001) single crystal substrate using ultra high vacuum magnetron sputtering at room temperature and subsequently annealed *in-situ* at $T_{ann} = 200-700$ °C to promote the Heusler ordering. Here, Co₂Mn*Z* (*Z* = Ge, Sn) films were grown on Co₅₀Fe₅₀ (3 nm) buffer layer. The degree of *L*₂₁-ordering (*S*_{L21}) as a function of T_{ann} of Co₂Mn*Z* (*Z* = Ge, Sn), Co₂FeGe_{0.5}Ga_{0.5} and Co₂MnSi (Ref. [2]) films is displayed in Fig. 1(a). In case of Co₂FeGe_{0.5}Ga_{0.5} and Co₂MnSi, *L*₂₁-ordering was observed above 500 °C; whereas that of Co₂Mn*Z* (*Z* = Ge, Sn) films appeared even in as-deposited and varies systematically with increasing T_{ann} . Non-local spin-valve (NLSV) devices were micro-fabricated to estimate the spin-polarization *P* of Co₂Mn*Z* (*Z* = Ge, Sn) and Co₂FeGe_{0.5}Ga_{0.5} films by measuring the spin-accumulation signal in Cu channel. A systematic variation in *P* with increasing T_{ann} was observed in these alloys [Fig. 1(b)]. At $T_{ann} = 300$ °C, the spin-polarization *P* of Co₂MnSn and Co₂FeGe_{0.5}Ga_{0.5}Ga_{0.5} were found to be very close (~0.56) whereas that of Co₂MnGe film was higher (~0.67) which would be due to higher degree of *L*₂₁-ordering. These results suggest that Co₂MnGe alloy might be a better ferromagnetic electrode for practical applications.



Figure 1. Annealing temperature dependence of (a). degree of $L2_1$ -ordereing $(S_{L21}),$ and (b). bulk spin-polarization (P)measured in NLSV devices. S_{L21} exceeding 1 is expected to be caused by off-stoichiometry in films.

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