Magnetocrystalline anisotropy and exchange probed by high-field anomalous Hall effect in fully compensated half-metallic Mn$_2$Ru$_x$Ga thin films

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Magnetotransport is investigated in thin films of the half-metallic ferrimagnet Mn$_2$Ru$_x$Ga in pulsed magnetic fields of up to 58 T. A nonvanishing Hall signal is observed over a broad temperature range, spanning the compensation temperature (155 K), where the net magnetic moment is strictly zero, the anomalous Hall conductivity is 6673 Ω$^{-1}$ m$^{-1}$, and the coercivity exceeds 9 T. Molecular field modeling is used to determine the intra- and intersublattice exchange constants, and from the spin-flop transition we infer the anisotropy of the electrically active sublattice to be 216 kJ m$^{-3}$ and predict the magnetic resonance frequencies. Exchange and anisotropy are comparable and hard-axis applied magnetic fields result in a tilting of the magnetic moments from their collinear ground state. Our analysis is applicable to collinear ferrimagnetic half-metal systems.

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Thin films with ultrahigh magnetic anisotropy fields exhibit magnetic resonances in the range of hundreds of GHz [1–3] which is promising for future telecommunications applications. Spin-transfer-driven nano-oscillators (STNOs), working on the principle of angular momentum transfer from a spin-polarized current to a small magnetic element [4,5], have achieved output powers of several µW and frequency tunabilities of ~GHz mA$^{-1}$ [6,7], useful for wireless data transmission [8]. Output frequencies of STNOs based on standard transition-metal-based ferromagnets, such as CoFeB, or cubic Heusler alloys such as Co$_2$Fe$_0$$_1$$_4$Mn$_0$$_6$Si are in the low GHz range [9–13].

Certain Heusler alloys [14,15] are a suitable choice for achieving much higher output frequencies, aimed at enabling communication networks beyond 5G [16]. The Mn$_{3-x}$Ga family contains two Mn sublattices which are antiferromagnetically coupled in a ferrimagnetic structure [14]. They have low net magnetization, $M_{\text{net}}$, and high effective magnetic anisotropy, $K_{\text{eff}}$, with anisotropy fields of $\mu_0H_K = 2K_{\text{eff}}/M_{\text{net}}$ exceeding 18 T [17,18], which results in resonance frequencies two orders of magnitude higher [1,2] than CoFeB. Furthermore, the magnetic properties of these ferrimagnetic alloys can be tuned easily with composition [19–21]. Mn$_{3-x}$Ga films have shown tunable resonance frequencies between 200 and 360 GHz by variation of the alloy stoichiometry and magnetic anisotropy field [2].

Here, we focus on the fully compensated half-metallic Heusler compound Mn$_2$Ru$_x$Ga (MRG) [20–25]. Films of MRG were first shown experimentally [20] and subsequently confirmed by density functional theory (DFT) calculations [25] to exhibit a spin gap at $E_F$. The material crystallizes in the cubic space group $F43m$. Mn on the 4a and 4c sites are antiferromagnetically coupled, while those on the same sites are ferromagnetically coupled. The crystal structure is shown in Fig. 1(a). The Ga is on the 4b sites and Ru occupies a fraction of the 4d sites [20]. We will discuss Mn on the 4a and 4c sites by referring to the Mn$_{4a}$ and Mn$_{4c}$ sublattices. By changing the Ru concentration, the magnetic properties of the Mn$_{4c}$ sublattice are altered, while those of the Mn$_{4a}$ sublattice remain relatively stable [21]. Thin films grown on MgO have an out-of-plane magnetic easy axis due to biaxial strain induced by the substrate during growth [23]. Unlike the uncompensated tetragonal $D_{022}$ Mn$_{3-x}$Ga family of alloys, MRG has a compensation temperature, $T_{\text{comp}}$, where there is no net magnetization [20,21]. Nonetheless, there is nonvanishing tunnel magnetoresistance [22], spin Hall angle [23], and magneto-optical Kerr effect [24], which all arise from the Mn$_{4c}$ sublattice. The occupied electronic states originating from the Mn$_{4a}$ sublattice lie below the spin gap [25].

The electrical transport on MRG reported to date [22,23] can be explained using the model shown in Figs. 1(b) and 1(c) where the direction of spin polarization is governed by the direction of the Mn$_{4c}$ sublattice and not Mn$_{4a}$ or $M_{\text{net}}$. Here, we make use of the dominant influence of a single sublattice on the electron transport to study the magnetism of a compensated half metal at compensation, and evaluate the exchange and anisotropy energies.
The longitudinal magnetoresistance up to 58 T also does not exceed 1% (not shown). The divergence in coercivity [black circles in Fig. 2(e)] is expected at \( T_{\text{comp}} \) because the anisotropy field in uniaxial magnets is \( \mu_0 H_K = 2 K_{\text{eff}}/M_{\text{eff}} \), where \( K_{\text{eff}} \) is the effective anisotropy energy and \( M_{\text{net}} \) is the net magnetization. The anisotropy field is an upper limit on coercivity. The temperature dependence of the spin-flop field, \( \mu_0 H_{sf} \), is also plotted in Fig. 2(e) (red squares). If both substrates contributed equally to the effect, the sum should fall to zero at \( T_{\text{comp}} \).

We refer to the model presented in Figs. 1(b) and 1(c) to explain the behavior shown in Fig. 2(f). Figure 1(b) shows the \( \text{Mn}_{4\text{a}} \) and \( \text{Mn}_{4\text{c}} \) sublattice moments and the net magnetic moment in the case of an applied field \( \mu_0 H \) along the easy axis of MRG. Below \( T_{\text{comp}} \), the \( \text{Mn}_{4\text{a}} \) moment (green arrow) outweighs that of \( \text{Mn}_{4\text{c}} \) (blue arrow), and \( M_{\text{net}} \) (orange arrow) is parallel to the \( \text{Mn}_{4\text{c}} \) sublattice. At \( T_{\text{comp}} \), \( M_{\text{net}} \) is zero but the directions of the sublattice moments have not changed with respect to \( \mu_0 H \). Above \( T_{\text{comp}} \), \( \mu_0 H \) causes a reversal of \( M_{\text{net}} \) (provided it exceeds \( \mu_0 H_{sf} \)). Here, the \( \text{Mn}_{4\text{a}} \) sublattice has a larger moment than \( \text{Mn}_{4\text{c}} \) and \( M_{\text{net}} \) will be in the same direction as the \( \text{Mn}_{4\text{a}} \) moment. Due to the antiferromagnetic alignment of both sublattices the moment on \( \text{Mn}_{4\text{a}} \) is parallel (antiparallel) to \( \mu_0 H \) below (above) \( T_{\text{comp}} \).

In the absence of an applied field [Fig. 1(c)], the direction of \( M_{\text{net}} \) will reverse on crossing \( T_{\text{comp}} \) due to the different temperature dependences of the sublattice moments. However, the net sublattice moments only change in magnitude, and not direction. The uniaxial anisotropy provided by the slight substrate-induced distortion of the cubic cell [20] provides directional stability along the \( z \) axis. Therefore, crossing \( T_{\text{comp}} \) in the absence of applied field, we expect no change in the sign of \( \sigma_{xy} \), nor should it vanish. The \( \text{Mn}_{4\text{a}} \) sublattice dominates the electron transport and determines the spin direction of the available states at \( E_F \), while the \( \text{Mn}_{4\text{c}} \) states form the spin gap.

\[
\sigma_{xy}(H) = \frac{\rho_{xy}}{\rho_{xx}}(H) = \frac{\rho_{xy}}{\rho_{xx}}(0)
\]

shown in Fig. 2(b) is small (<1%), as expected for a half metal [29]. Pulsed field measurements in Fig. 2(c) show that, close to \( T_{\text{comp}} \), \( \mu_0 H_{sf} \) exceeds 9 T and that MRG exhibits a spin-flop transition at higher fields, indicated in the figure by the gray arrows. The derivative of selected curves of \( \sigma_{xy} \) versus applied field [Fig. 2(d)] shows up the spin-flop field, especially at lower temperatures. We note that the longitudinal magnetoresistance up to 58 T also does not exceed 1% (not shown). The divergence in coercivity [black circles in Fig. 2(e)] is expected at \( T_{\text{comp}} \) because the anisotropy field in uniaxial magnets is \( \mu_0 H_K = 2 K_{\text{eff}}/M_{\text{eff}} \), where \( K_{\text{eff}} \) is the effective anisotropy energy and \( M_{\text{net}} \) is the net magnetization. The anisotropy field is an upper limit on coercivity. The temperature dependence of the spin-flop field, \( \mu_0 H_{sf} \), is also plotted in Fig. 2(e) (red squares).
FIG. 2. (a) AHC loops up to 6.5 T for Mn$_2$Ru$_{0.61}$Ga around the compensation temperature (155 K). Loops are offset vertically for clarity. (b) Magnetoresistance loops recorded at the same time as the data in (a). Loops are offset vertically for clarity. (c) AHC loops up to 58 T, where the spin-flop transition is indicated by the grey arrows. The linear slope is due to the ordinary Hall effect. Loops are offset vertically for clarity. (d) Derivative of the selected data in (c) clearly highlighting the spin flop. (e) $\mu_0H_c$ (black circles) and $\mu_0H_{sf}$ (red squares) as a function of temperature. The divergence of the coercivity is expected at $T_{comp}$ since $M_{net}=0$ and $K_{eff} \neq 0$. (f) Temperature dependence of the remanent Hall conductivity when saturated at 10 K in negative (solid line) and positive (dashed line) applied field. The black open (solid) circles record the remanent Hall resistivity after the application of 6.5 T (58 T).

The results of a molecular field model [33] based on two sublattices are presented in Fig. 3. The molecular field $H^i$ experienced by each sublattice is given by

\begin{align}
H_{4a}^i &= n_{4a-4a}M_{4a} + n_{4a-4c}M_{4c} + H,
\end{align}

(1)

\begin{align}
H_{4c}^i &= n_{4a-4c}M_{4a} + n_{4c-4c}M_{4c} + H,
\end{align}

(2)

where $n_{4a-4a}$ and $n_{4c-4c}$ are the intralayer exchange constants and $n_{4a-4c}$ is the interlayer exchange constant. $M_{4a}$ and $M_{4c}$ are the magnetizations of the 4$a$ and 4$c$ sublattices. $H$ is the externally applied magnetic field. The moments within the Mn$_{4a}$ and Mn$_{4c}$ sublattices are ferromagnetically coupled and hence $n_{4a-4a}$ and $n_{4c-4c}$ are both positive. The two sublattices couple antiferromagnetically and therefore $n_{4a-4c}$ is negative.

The equations are solved numerically for both temperature and applied field dependences to obtain the projection of both sublattice magnetizations along the $z$ axis, $M_{z-\alpha} = M_\alpha \cos \theta_\alpha$, where $\alpha = 4a, 4c$. In the absence of an applied field, $\theta = 0$, therefore $M_{z-\alpha}$ reduces simply to $M_\alpha$.

The model parameters are given in Table I. Based on previous x-ray magnetic circular dichroism (XMCD) measurements [21] as well as DFT calculations [25] we take values of 547 and 585 kA m$^{-1}$ for the magnetizations on the 4$a$ and 4$c$ sublattices, respectively. The values of $n_{4a-4a}$, $n_{4c-4c}$, and $n_{4a-4c}$ are fitted to reproduce $T_{comp}$ and the Curie temperature, $T_C$. The temperature dependences of $M_{z-4a}$ (blue line), $M_{z-4c}$ (green line), and $M_{net}$ (orange line) with $n_{4a-4a} = 1150$, $n_{4c-4c} = 400$, and $n_{4a-4c} = -485$ are shown in Fig. 3(a). In order to numerically obtain the temperature dependence in
The ratio is almost constant with no significant linear background.

The zero applied field, a strong field of 60 T is used to set the direction of \( M_{\text{net}} \) and then reduced to zero, so the sublattice moments reverse at \( T_{\text{comp}} = 155 \text{ K} \) as in the experiment. \( T_C \) is 625 K. \( M_{\text{net}} \) varies from 38 kA m\(^{-1}\) at 10 K to a maximum of 97 kA m\(^{-1}\) at 512 K, close to \( T_C \).

Figure 3(b) shows the measured AHC (circles), along with \( |\sigma_{xy}| \) (green line) from the molecular field model. It can be seen clearly that \( \sigma_{xy} \) follows the temperature dependence of \( M_{-4c} \) below \( T_{\text{comp}} \) and not \( M_{\text{net}} \). As a further step, we plot \( |\sigma_{xy}| \) from the molecular field model (orange line) with \( |\sigma_{xy}(T) - \sigma_{xy}\text{-comp}| \) (trangles). As \( \sigma_{xy} \) is proportional only to \( M_{dc} \) and at compensation \( M_{dc} = M_{4c} \), subtracting the value of \( \sigma_{xy} \) at \( T_{\text{comp}} \) \( (|\sigma_{xy}(T) - \sigma_{xy}\text{-comp}|) \) gives an approximate indication of how \( M_{\text{net}} \) behaves with temperature. Even though this ignores the weak \( M_{4c} \) temperature dependence, the trend of \( M_{\text{net}} \) follows \( |\sigma_{xy}(T) - \sigma_{xy}\text{-comp}| \), showing that \( \sigma_{xy} \) is a reflection of \( M_{dc} \) and not \( M_{\text{net}} \). The inset in Fig. 3(b) shows both \( M_{-4c} \) and \( M_{-4c} \) with the experimentally obtained \( \sigma_{xy} \), and shows that \( \sigma_{xy} \) more closely follows \( M_{-4c} \). The relative decrease of \( M_{-4c} \) from 10 to 300 K, ~40%, is more than double that of \( M_{-4c} \), in line with previously reported XMCD measurements [21]. Figure 3(c) shows the ratio of \( \sigma_{xy} \) to \( M_{-4c} \) (green dotted line), \( M_{-4d} \) (blue dotted line), and \( M_{\text{net}} \) (inset). Linear fitting of \( \sigma_{xy}/M_{-4d} \) (solid green line) and \( \sigma_{xy}/M_{-4c} \) (solid blue line) shows that \( \sigma_{xy}/M_{-4c} \) remains constant over the measured temperature range, and is equal to 0.0136 \( \Omega^{-1} \text{m} A^{-1} \text{m} \), similar to what has been reported for other itinerant ferromagnetic systems [34,35]. The linear slope for \( \sigma_{xy}/M_{-4d} \) and the divergence of \( \sigma_{xy}/M_{\text{net}} \) shows that \( \sigma_{xy} \) reflects neither of these two quantities.

A recent study has shown via \textit{ab initio} calculations that this must be the case for a fully compensated half-metallic ferrimagnetic system [36], although previous reports on bulk films found \( \rho_{xy} \), and hence \( \sigma_{xy} \), falling to zero at \( T_{\text{comp}} \) [37].

For the evaluation of the magnetic anisotropy we use the initial low-field change of \( \sigma_{xy} \) vs \( \mu_0 H_x \) and extrapolate to zero and obtain \( K_4 \) (not shown). The values obtained vary from 100 to 250 kJ m\(^{-3}\) over the entire data range. We also calculate the anisotropy directly from the spin-flop transition \( H_{sf} = \sqrt{2|H_K H_{sf}^2|} \), where \( H_K \) is the sublattice anisotropy field and \( H_{sf}^2 \) is the exchange field, the first term in Eq. (2). The anisotropy field \( H_K \) is related to the sublattice anisotropy energy \( K_{4c} \).

A comparison between the experiment and the model at 220 K for both \( \mu_0 H_z \) and \( \mu_0 H_c \) is shown in Fig. 4. The

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**TABLE I.** Initial parameters input to the molecular field model according to Eqs. (1) and (2). \( M_{4c} \), \( M_{-4c} \), and \( K_{4c} \) are the magnetizations and uniaxial anisotropies on the 4c, 4c sublattices. \( n_{4a-4d} \) and \( n_{4c-4c} \) are the intralayer exchange constants. \( n_{4a-4c} \) is the interlayer exchange constant. Derived parameters are outputs of the molecular field model.

| Parameter          | Value          | Derived Parameter          | Value          |
|--------------------|----------------|---------------------------|----------------|
| \( M_{4c} \) (0 K)  | 547 kA m\(^{-1}\) | \( M_{\text{net}} \) (0 K) | 585 kA m\(^{-1}\) |
| \( M_{-4c} \) (0 K)| 547 kA m\(^{-1}\) | \( n_{1a-1a} \) | 400 kJ m\(^{-3}\) |
| \( K_{4c} \)       | 0 kJ m\(^{-3}\)   | \( n_{4a-4c} \) | −485 kJ m\(^{-3}\) |
| \( K_{4c} \)       | 216 kJ m\(^{-3}\) | \( n_{4a-4c} \) | 395 kA m\(^{-1}\) |
| \( M_{\text{net}} \) (10 K) | 38 kA m\(^{-1}\) | \( T_C \) | 625 K |
| \( M_{\text{net}} \) (max.) | 97 kA m\(^{-1}\) | \( T_{\text{comp}} \) | 155 K |
This has the effect of increasing (decreasing) $H_d$ above (below) $T_{\text{comp}}$. While this improves the match between $\sigma_{xy}$ vs $\mu_0 H_x$ below $T_{\text{comp}}$, it worsens the match of $\sigma_{xy}$ vs $\mu_0 H_x$ at all temperatures. This and the slight discrepancies between the model and experiment when a low value of $K_{4c}$ is used (Fig. 4) indicate that additional anisotropies, likely cubic, in MRG, as well as antisymmetric exchange (Dzyaloshinskii-Moriya interaction) should be taken into account.

We have shown that the uniaxial molecular field model reproduces the main characteristics of the experimental data and we confirm the relationship $\sigma_{xy} \propto M_d \cos \theta_{M_d}$. Knowing $H_K$ and $H_{ex}^{\text{cs}}$, we can predict the frequencies of the anisotropy, $f_{\text{anis}} = \gamma \mu_0 H_K$, and the exchange, $f_{\text{exch}} = \gamma \mu_0 \sqrt{2H_K H_{ex}^{\text{cs}}} = \gamma \mu_0 H_{sf}$, magnetic resonance modes, where $\gamma = 28.02$ GHz T$^{-1}$ [39]. At 220 K, $\mu_0 H_d = 26$ T and $\mu_0 H_{ex}^{\text{cs}} = n_{d}H_d$, $M_d = 294$ T, therefore $\mu_0 H_K = 1.15$ T and the resonances are $f_{\text{anis}} = 32$ GHz and $f_{\text{exch}} = 729$ GHz.

In conclusion, $\sigma_{xy}$ for fully compensated half-metallic ferrimagnetic alloys follows the relevant sublattice magnetization $M_d \cos \theta_{M_d}$ and not $M_{\text{net}} \cos \theta_{M_d}$. High-field magnetotransport and molecular field modeling allows the determination of the anisotropy and exchange constants provided the half-metallic material is collinear. Mn$_2$Ru$_2$Ge behaves magnetically as an antiferromagnet and electrically as a highly spin-polarized ferromagnet: It is capable of operation in the THz regime and its transport behavior is governed by the Mn$_{4c}$ sublattice. The immediate, technologically relevant, implication of these results is that spin-transfer torque effects in compensated ferrimagnetic half metals will be governed by a single sublattice.

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