Although high-tunnelling spin polarization has been observed in soft, ferromagnetic, and predicted for hard, ferrimagnetic Heusler materials, there has been no experimental observation to date of high-tunnelling magnetoresistance in the latter. Here we report the preparation of highly textured, polycrystalline Mn$_3$Ge films on amorphous substrates, with very high magnetic anisotropy fields exceeding 7 T, making them technologically relevant. However, the small and negative tunnelling magnetoresistance that we find is attributed to predominant tunnelling from the lower moment Mn–Ge termination layers that are oppositely magnetized to the higher moment Mn–Mn layers. The net spin polarization of the current reflects the different proportions of the two distinct termination layers and their associated tunnelling matrix elements that result from inevitable atomic scale roughness. We show that by engineering the spin polarization of the two termination layers to be of the same sign, even though these layers are oppositely magnetized, high-tunnelling magnetoresistance is possible.
Key to the successful development of magnetic random access memory (MRAM), one of the most promising emerging non-volatile memory technologies today, are new magnetic materials for the magnetic tunnel junction (MTJ) memory elements that have sufficient stability against thermal fluctuations to sustain deeply scaled devices. The most promising magnetic materials to date are considered to be magnetic alloys formed from Co, Fe and B, in conjunction with MgO(001) tunnel barriers. The magnetic electrodes must possess sufficient perpendicular magnetic anisotropy (PMA) that their magnetizations lie perpendicular to the plane of the MTJ device, since this allows for reduced currents to switch the magnetization of the electrode that forms the memory layer of the device using spin torque. The PMA of Co–Fe–B layers arises from the interfaces between these layers and the tunnel barrier and/or the underlayer on which the Co–Fe–B layer is deposited. Thus, these layers must be made sufficiently thin that the interface PMA overcomes the demagnetization energy that arises from the magnetic volume and increases in proportion with the magnetic volume of the Co–Fe–B layer. In practice, this means that the PMA is too weak to overcome thermal fluctuations when the device has a critical dimension $< \sim 20$ nm in size, since the thickness of the magnetic layer has to be below that required to maintain its moment perpendicular, and, below that needed to switch the magnetic layer with reasonable current densities. Magnetic materials in which the PMA is derived from volume magnetocrystalline anisotropy are then needed. One of the most promising class of such materials are the Heusler alloys—compounds having the chemical formula $X_2YZ$ or $X'X''YZ$, wherein $X$, $X'$, $X''$ and $Y$ are transition metals, or lanthanides (rare-earth metals), and $Z$ is from a main group metal. Some of these compounds are ferromagnetic or ferrimagnetic depending on the exchange interaction between the magnetic moments on the $X$ and/or $Y$ sites. Moreover, while the parent Heusler compounds are cubic and exhibit weak or no significant magnetic anisotropy, the structure of some of these compounds is found to be tetragonally distorted: due to this distortion the magnetization exhibited by these compounds may be aligned along the tetragonal axis. Thus, thin films formed from such materials may exhibit PMA due to a magnetocrystalline anisotropy associated with the tetragonally distorted structure. Some examples of such tetragonal Heusler compounds are Mn$_3_x$Ga (ref. 6) and Mn$_x$Ge (ref. 7).

Thin films of these materials have been shown to exhibit large PMA but, to date, all work on these materials has involved films that are grown epitaxially on single crystalline substrates such as SrTiO$_3$(001) or MgO(001) using seed layers formed from a variety of materials but preferably Cr or Pt (refs 7–11). Such single crystalline substrates are not useful for MRAM applications in which the MTJs must be deposited on wires formed from polycrystalline copper, which may be covered with other layers that are also polycrystalline or amorphous. Here we show that thin films of ferrimagnetic Mn$_x$Ge with giant PMA can be grown on amorphous substrates (Si(001)/SiO$_2$) using underlayers formed from TaN/IrMn$_3$ (TI). The magnetic properties of these films are comparable or superior to films that we have grown under similar conditions on single crystal MgO(001) substrates using epitaxial Cr(001) underlayers. Very high magnetic anisotropy fields exceeding 7T are found. However, the tunnelling magnetoresistance (TMR) is negative and much smaller than theoretical predictions for tunnel junction devices formed with MgO tunnel barriers. We attribute this to predominant tunnelling from the lower moment Mn–Ge termination layers that are oppositely magnetized to the higher moment Mn–Mn layers. The net spin polarization of the current reflects the different proportions of the two distinct termination layers and their associated tunnelling matrix elements at the tunnel barrier interface, that result from inevitable atomic scale roughness. We have confirmed that a second perpendicularly magnetized Heusler compound, Mn$_2$CuSb, which we identified by computational materials discovery methods, similarly displays small TMR, but that the sign of the spin polarization of each of the termination layers is opposite to that of their magnetization, in contrast to Mn$_3$Ge. Therefore, the low TMR that we have found in Mn$_3$Ge and Mn$_2$CuSb, which we attribute to termination layer compensation, is not an inherent property of ferrimagnetic Heuslers. The spin polarization of the two termination layers can be engineered to be of the same sign even though these layers are oppositely magnetized, therefore providing a path to low magnetization electrodes with high TMR for high density spin transfer torque MRAM applications.

Results

Growth of highly textured Mn$_3$Ge on amorphous substrates.

Highly textured, polycrystalline and tetragonal Mn$_3$Ge films were grown by either ion-beam deposition or d.c.-magnetron sputtering in an ultra-high vacuum chamber with a base pressure of $\sim 4 \times 10^{-10}$ torr on Si(001) substrates covered with 250 Å of amorphous SiO$_2$. We find that seed layers formed from bilayers of TaN/IrMn$_3$ that are first deposited on the SiO$_2$ induce (001) textured Mn$_3$Ge films that are tetragonally distorted. IrMn$_3$ is known to have a L1$_2$ structure that is cubic, and which matches one of the sublattices of the structure that Mn$_3$Ge is known to form. Moreover, the lattice mismatch between IrMn$_3$ and Mn$_3$Ge is $<1\%$ (ref. 12). The TaN layer that is grown by reactive sputtering, promotes the growth of (001) textured IrMn$_3$, since fcc-IrMn$_3$ favours the $(111)$ out-of-plane orientation when grown directly on amorphous SiO$_2$ surface (Supplementary Fig. 1 and Supplementary Note 1). However, in contrast to theoretical predictions of giant values of TMR for MTJs using Mn$_3$Ge electrodes we find much smaller values experimentally, which we attribute to compensation in the tunnelling spin current polarization from atomic layer variations of the electrode surface termination at the tunnel barrier interface. We propose that this is an inevitable consequence of ferrimagnets with layer-by-layer alternation of magnetization, when the spin polarization of these layers compensates each other. We confirm this conjecture by preparing MTJs using another ferrimagnetic Heusler compound, Mn$_3$CuSb, which also shows high PMA but yet very small TMR. The structural and magnetic properties of the Mn$_3$Ge films depend sensitively on its composition and atomic order. The latter is strongly influenced by the deposition temperature and subsequent anneal conditions, which also affect the smoothness of the Mn$_3$Ge film. To achieve optimal MTJ performance the electrode must be atomically smooth. We find that the root mean square (r.m.s.) roughness of the Mn$_3$Ge film, $r_{\text{rms}}$, measured using atomic force microscopy, increases significantly when the growth temperature ($T_G$) exceeds modest temperatures of just $\sim 200$ °C, but higher growth temperatures are needed to sustain the Heusler structure, as measured from X-ray diffraction (Supplementary Fig. 2 and Supplementary Note 2). Thus, we find that an optimal growth method, which includes a three-step process for the Mn$_3$Ge electrode, in which an initial 20 Å Mn$_3$Ge layer is grown at 450 °C, followed by a thicker Mn$_3$Ge layer deposited at $T_G = 150$ °C, with a final in situ anneal at 450 °C for 1–2 h in vacuum, gives smooth films ($r_{\text{rms}} \sim 3\AA$) with high PMA. During the annealing step of Mn$_3$Ge films, there is a substantial inter-diffusion between IrMn$_3$ and Mn$_3$Ge layers (Supplementary Fig. 3), which causes deterioration of the magnetic properties of Mn$_3$Ge. We discovered that this
interdiffusion can be prevented by using a thin 10–20 Å TaN barrier between IrMn3 and Mn3Ge layers, as revealed by electron energy loss spectroscopy measurements (Supplementary Note 3). Thus, the preferred underlayer is formed from TaN/IrMn3/TaN (TIT). We note that a single TaN underlayer gives much poorer quality Mn3Ge layers.

Figure 1 compares the magnetic properties of Mn3Ge films with varying thickness grown on amorphous substrates using the three-step process and on a crystalline MgO(001) substrate with a Cr seed layer (MC). Excellent PMA is observed in all cases but the highest coercive and anisotropy fields are found for structures grown on the TIT underlayer. Coercive fields of 6T and anisotropy fields exceeding 7T are found. Figure 1c summarizes the magnetic moment \( m \), coercive field \( H_C \) and uniaxial magnetic anisotropy \( K_U \) for these films. Values of \( m \) for Mn3Ge films grown on TIT underlayers are close to those theoretically predicted for bulk Mn3Ge (ref. 15), but \( m \) is significantly lowered by \( 15–35\% \) for Mn3Ge films grown using Ti underlayers or a MC single crystal substrate. For the latter film, we also find that the magnetic anisotropy \( K_U \) is substantially lower: we attribute this to the large lattice mismatch (\( \sim 7\% \)) between Cr and Mn3Ge.

**Figure 1 | Mn3Ge Heusler films with giant perpendicular magnetic anisotropy.** (a) Magnetization versus field hysteresis loops of Mn3Ge films grown on Si(001)/SiO2 substrate with Ti (magenta) and TIT (blue) underlayers and MgO(001) substrate with MC (orange) underlayers. Ti (TIT) have the following structure: Si(001)/SiO2/200 Å TaN/200 Å IrMn3 (10 Å TaN) while MC has the following structure: MgO(001)/20 Å MgO/400 Å Cr. For the TIT films, the thickness of the Mn3Ge layer, deposited using the three-step process, was varied. Out of plane (in-plane) \( M \) vs. \( H \) loops are shown as solid lines (open squares). (b) and (d) are schematics of the MTJ structures grown on Si(001)/SiO2 and MgO(001) substrates, respectively. In some cases a TaN diffusion barrier layer was used as indicated by a dashed line. For characterization of structural, topographical and magnetic properties of the Mn3Ge films, a 30 Å TaN film was used as a capping layer instead of the upper layers shown within the dotted lines. (c) Magnetic moment \( m \), coercive field \( H_C \) (solid triangles) and uniaxial magnetic anisotropy constant \( K_U \) (empty squares), extrapolated from Fig. 1a, versus Mn3Ge thickness. The green straight line shows the calculated moment of bulk D022-Mn3Ge (ref. 15). As shown in Fig. 1a, the Mn3Ge magnetization cannot be saturated in-plane using the available magnetic field (7 T); thus, \( H_{eff} \) is a lower bound.
Figure 2 | Characteristics of Mn₃Ge-based magnetic tunnel junctions. (a) TMR versus $H$ (perpendicular to the device) measured at 300 K (smaller squares) and 3 K (bigger squares) for MTJ devices grown using TI (magenta) and TIT (blue) underlayers. For the TIT junction, two sets of data were measured at 3 K (blue bigger squares) after cooling down the device from 300 K in a magnetic field of $+9$ T and $-9$ T, respectively. These data are mirror images of each other, as can be seen in the figure. All the other measurements were performed without field cooling. (b) Temperature dependence of TMR, and $R_{AP}$ and $R_{P}$. (c) MgO thickness dependence of TMR (open symbols) and $R_{AP}$ product (solid symbols), averaged over >20 devices. Solid and dashed lines are guides to the eye for $R_{AP}$ and TMR, respectively. $R_{AP}$ scales exponentially with barrier thickness. (d) HRTEM image of an MTJ device ~27 nm in size, with the structure: Si/250 Å SiO₂/200 Å TaN/200 Å IrMn₃/300 Å Mn₃Ge/15 Å MgO/15 Å CoFeB/50 Å Ta/50 Å Ru. HRTEM, high-resolution transmission electron microscopy.

For a given MTJ device $R_{AP}$ barely changes, while $R_{P}$ increases monotonically as $T$ decreases, resulting in higher TMR at low temperatures (Fig. 2b). These properties, as well as the dependence of the resistance-area product $R_{AP}$ and TMR on the barrier thickness (Fig. 2c) are characteristic of a high quality tunnel barrier. A cross-sectional high-resolution transmission electron microscopy image of a typical MTJ device with a width of 27 nm, shown in Fig. 2d, illustrates the high quality of the structure and the device patterning.

Figure 2a compares TMR versus perpendicular magnetic field $H$ measured at 300 K (smaller squares) and 3 K (larger squares) for patterned MTJ devices (1×2 $\mu$m²) using TI and TIT underlayers. In each case high applied magnetic fields ($\pm9$ T) are needed to align the magnetic moments of the Mn₃Ge and CoFeB layers parallel to each other (P state) because of the giant uniaxial anisotropy of Mn₃Ge. The junction resistance is higher in the P state compared with the antiparallel (AP) state, obtained when the CoFeB moment switches close to zero field. Thus, the TMR ($[(R_{AP} - R_{P})/R_{AP}] \times 100$) is negative with values of ~$-35\%$ at 300 K and ~$-74\%$ at 3 K, where $R_{AP}$ and $R_{P}$ are the junction resistances in the P and AP states, respectively. These are the highest values of TMR reported to date in perpendicularly magnetized MTJ devices using a tetragonally distorted Heusler compound as a magnetic electrode. Nonetheless, these values are much smaller than those predicted by density functional theory (DFT) calculations, as discussed below.

For a given MTJ device $R_{AP}$ barely changes, while $R_{P}$ increases monotonically as $T$ decreases, resulting in higher TMR at low temperatures (Fig. 2b). These properties, as well as the dependence of the resistance-area product $R_{AP}$ and TMR on the barrier thickness (Fig. 2c) are characteristic of a high quality tunnel barrier. A cross-sectional high-resolution transmission electron microscopy image of a typical MTJ device with a width of 27 nm, shown in Fig. 2d, illustrates the high quality of the structure and the device patterning.

Notwithstanding the exceptionally high PMA values exhibited by polycrystalline Mn₃Ge films the surprisingly low TMR values lessen their potential importance for MTJ devices. We attribute the low TMR, as discussed below, to their ferrimagnetic structure. This limitation could be overcome by identifying Heusler compounds that display high PMA, and which are ferromagnetic.

**Discovery of tetragonal Heuslers by computational methods.** To identify potential candidate tetragonal materials we used computational materials discovery methods to calculate the structure and electronic properties of several hundred Heusler compounds (with $X = Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd$; $Y = Sc, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Os, Ir, Pt$; and $Z = Al, Si, Ga, Ge, In, Sn, Sb$) for both the regular and inverse structures and for ferri- and ferromagnetic configurations. Surprisingly, ~40% of these compounds are calculated to be tetragonal in their ground state. Rank ordering these tetragonal compounds according to the combination of the energy difference between the tetragonal and cubic structures and that between the inverse and regular structures, we identified ~30 possible candidate tetragonal materials, where the average energy difference exceeded ~0.3 eV per formula unit. Of these, we prepared eight in thin film form using the same buffer layers and deposition conditions as discussed above. Half of these compounds showed a tetragonal structure and two showed excellent PMA properties, namely, Mn₃CuSb and Rh₂CoSb. In the other cases, we rationalize that the theoretical predictions were not fulfilled due to atomic disorder on the X and Y sites. This is particularly the case where X and Y have similar chemical properties or atomic size.

Similar magnetic properties were found for Mn₃CuSb and Rh₂CoSb films grown using either TI underlayers (Supplementary Fig. 4 and Supplementary Note 4) or an MC crystalline substrate but the latter films were typically smoother and better suited for MTJ studies. Results are shown in Fig. 3 for Mn₃CuSb. Square magnetic hysteresis loops consistent with PMA are found. The calculated lowest energy configuration of Mn₃CuSb is a tetragonal ferromagnetic regular Heusler with a moment of 5.4 $\mu_B$ per formula unit (Table 1). Although we find that the deposited films are tetragonal (Supplementary Fig. 5 and Supplementary Note 5),
in agreement with the calculations, the measured $c/a$ ratio is much smaller in the films ($c_i/\sqrt{2}a_i \approx 1.1$) than that predicted for a fully ordered Mn$_2$CuSb bulk compound ($c_i/\sqrt{2}a_i \approx 1.4$), and thus only slightly distorted from the cubic phase. Moreover, the magnetization of the films is much too low—$\sim 0.4 \mu_B$ per formula unit—to be consistent with the predicted ferromagnetic state, as is the TMR, as we discuss below. We rationalize these findings by the likelihood of chemical disorder within the deposited films. We note that the calculations predict that for the cubic phase the structure will be the inverse structure with a ferrimagnetic ordering, with a low moment (Table 1).

MTJ devices were prepared from Mn$_2$CuSb films with the layer stack shown in Fig. 3d and the magnetic hysteresis loop shown in Fig. 3b. The latter shows clear independent switching of the free layer (CoFeB) and the reference layer (Mn$_2$CuSb) but TMR curves measured on $1 \times 2 \mu$m$^2$ size devices show only very small TMR values of $\sim -1\%$ (Fig. 3c).

**Termination-layer dependent tunnelling spin polarization.** We performed ab initio calculations of the electronic structure and transport properties of Mn$_3$Ge/MgO/Fe and Mn$_2$CuSb/MgO/Fe MTJs (see Methods for details of the calculations). Note that we use bcc Fe rather than CoFeB that is used in the experiments to simplify the calculations. The TMR for Mn$_3$Ge/MgO/Fe MTJs is shown in Fig. 4a as function of the number of MgO layers, $N_{\text{MgO}}$, for $N_{\text{MgO}} \geq 2$. Two definitions of TMR are shown: $(T_P - T_{\text{AP}})/\min(T_P, T_{\text{AP}})$, that can vary from $-\infty$ to $\infty$ (that we use for the experimental data) and $(T_P - T_{\text{AP}})/\max(T_P, T_{\text{AP}})$, that can vary from $-1$ to $1$. Here $T_P$ and $T_{\text{AP}}$ are the transmission functions (calculated at zero bias voltage) corresponding to the P and AP states, respectively. The most important result is that the TMR depends sensitively on the atomic configuration of the termination layer in the Mn$_3$Ge adjacent to the MgO tunnel barrier. The calculation gives magnetic moments of $+4.08$ and $-3.12 \mu_B$ for the Mn–Mn and Mn–Ge termination layers, respectively. The TMR has opposite signs for the two termination layers, Mn–Mn and Mn–Ge, increasing in magnitude with $N_{\text{MgO}}$ for Mn–Mn but decreasing in magnitude for Mn–Ge. These results can be understood from the layer dependent, spin-dependent density of states calculated for the bulk electronic structure of Mn$_3$Ge, and the well understood symmetry spin-filtering properties of the MgO/Fe interface. The spin polarization of the native termination layers is negative in both cases but much bigger for Mn–Ge (see Supplementary Fig. 6 and Supplementary Note 6 for more details). A Brillouin zone filtering effect$^{14}$ arising from the Mn$_3$Ge/MgO interface tends to make the TMR positive for both terminations as the MgO layer thickness is increased. Thus, the TMR is negative for Mn–Ge but positive for Mn–Mn for intermediate MgO thicknesses that are of interest experimentally. The balance between the native spin polarization and the Brillouin zone filtering not only accounts for the dependence of the TMR on $N_{\text{MgO}}$ and the termination layer but also the bias voltage (Supplementary Fig. 7 and Supplementary Note 7).

These calculations give a natural explanation for the low TMR values found experimentally using Mn$_3$Ge electrodes. Even though the Mn$_3$Ge/MgO interface is very smooth (Fig. 2d) inevitably there will be atomic scale fluctuations in the morphology of the Mn$_3$Ge layer that gives rise to regions with Mn–Mn and Mn–Ge terminations, due to the fundamental underlying structure of the Heusler compound (see illustration in Fig. 4b). The simplest way to model such fluctuations is to average the transmission functions over the different terminations (separately for P and AP states), assuming that the MgO thickness is the same across the device. The TMR

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**Table 1 | Calculated magnetic and structural properties of Mn$_2$CuSb.**

| Heusler structure | Coupling of the magnetic moments of two Mn atoms | $E_{\text{tot}}$ in cubic phase (eV) | Lattice constant $a_i$ in cubic phase (Å) | Magnetic moment in cubic phase ($\mu_B$) | $E_{\text{tot}}$ in tetragonal phase (eV) | Lattice constant $a_i$ in tetragonal phase (Å) | Lattice constant $c_i$ in tetragonal phase (Å) | Magnetic moment in tetragonal phase ($\mu_B$) |
|------------------|----------------------------------------|-----------------------|-----------------------------|----------------------------------|------------------|-----------------------------|------------------------|-------------------|
| Regular          | ferro                                  | $-25.11$              | 6.23                        | 6.4                              | $-25.569$        | 3.86                        | 8.1                    | 7.80               |
| Regular          | ferro                                  | $-25.107$             | 6.28                        | 0                                | No stable solution found. |
| Inverse          | ferro                                  | $-24.840$             | 6.12                        | 4.3                              | $-25.292$        | 3.83                        | 8.19                   | 6.1                |
| Inverse          | ferro                                  | $-25.176$             | 6.22                        | 1.0                              | $-25.272$        | 3.95                        | 7.82                   | 0.4                |

DFT, density functional theory; $E_{\text{tot}}$, total energy. Calculated by the DFT-based VASP$^{20}$ program: lattice parameters and magnetic moments for different configurations of Mn$_2$CuSb Heusler alloy ($T$ = 0 K).
calculated from this simple model with an assumption of equal areas occupied by Mn–Ge and Mn–Mn terminations is shown in Fig. 4a. The calculations give a negative TMR since both $T_P$ and $T_{AP}$ for the Mn–Ge termination are larger than those for the Mn–Mn termination (for all $N_{MgO}$ considered—ranging from 2 to 12). The negative TMR is consistent with our experimental measurements. Note that due to the large 10.5% lattice mismatch between MgO and Mn$_3$Ge the Brillouin zone filtering effect, which critically depends on the existence of the well-defined 2D Brillouin zone of the Mn$_3$Ge/MgO interface, is likely more suppressed in actual devices as compared with the large negative spin polarization effect that is less sensitive to the existence of the 2D Brillouin zone. Thus, both ideal crystal theoretical simulations (Fig. 4a) and non-ideal crystal arguments predict negative TMR for the Mn$_3$Ge/MgO/Fe system, in agreement with experimental results (at low temperatures the TMR = $-75\%$ for $\sim 27\text{ Å}$ thick MgO: Fig. 2b). We note that the surface energy of the different terminations will very likely not be identical. This could lead to the formation of a more complex surface structure but kinetic considerations will constrain the surface beyond that of the equilibrium structure.

The calculated TMR versus $N_{MgO}$ for Mn$_2$CuSb/MgO/Fe MTJs, where the Mn$_2$CuSb has a tetragonal ferrimagnetic inverse Heusler structure, with Mn–Sb and Mn–Cu terminations at the MgO interface are compared in Fig. 4c. Note that the calculated magnetic moment of the Mn–Sb layer ($3.2\mu_B$) is larger than that for the Mn–Cu layer ($-2.8\mu_B$). Interestingly, the tunnelling spin polarization from the native termination layers, that is, in the limit of $N_{MgO} \rightarrow 0$, is of the opposite sign to the magnetic moment direction of these layers, whereas for Mn$_3$Ge the spin polarization is negative for both terminations (Supplementary Fig. 8 and Supplementary Note 8). But for the range of MgO thicknesses of $2 < N_{MgO} \leq 12$, tunnelling spin polarization is parallel and antiparallel, to the magnetic moment of the respective termination layer for Mn$_3$Ge and Mn$_2$CuSb, respectively. Moreover, there is no significant Brillouin zone filtering effect for Mn$_2$CuSb/MgO. In this case the dependence of the TMR on $N_{MgO}$ is due to the symmetry spin filtering effect from MgO/Fe, as can be seen from Fig. 4c by comparing the calculated TMR for Mn$_2$CuSb/MgO/Fe, for both terminations, with that for Fe/MgO/Fe. For the Mn$_2$CuSb/MgO/Fe MTJ a negative TMR is predicted for the termination layer with the largest magnetic moment (Mn–Sb) whereas for Mn$_3$Ge/
MgO/Fe is the opposite case. The TMR calculated for an MTJ with equal areas of Mn–Cu and Mn–Sb terminations is shown in Fig. 4c. The resulting TMR is significantly reduced due to the cancellation of contributions from the different terminations and has an overall negative sign (for $N_{390} > 2$). The negative sign of TMR and its small value agrees with our experimental findings.

**Discussion**

In summary, we have shown a method to grow highly textured ferrimagnetic Heusler films with large perpendicular anisotropy on amorphous substrates, thereby opening a path to their potential use for many applications such as magnetic recording media and ferrimagnetic Heusler films with large perpendicular anisotropy on MgO. In summary, we have shown a method to grow highly textured ferrimagnetic Heusler films with large perpendicular anisotropy on MgO (100) tunnel barriers. Nat. Mater. 9, 826–867 (2010).

**Methods**

Mn$_3$Ge and Mn$_2$CuSb films were deposited by d.c.-magnetron sputtering or ion-beam deposition at temperatures that were varied from ambient to 550 °C. Ta$_x$N$_y$ films were deposited with different Ar/N$_2$ ratios by reactive magnetron sputtering from a Ta target, and 200 Å IrMn$_3$ films were deposited using ion beam sputtering from an IrMn$_3$ target. Ta$_x$N$_y$ and IrMn$_3$ layers were grown at room temperature. Film compositions were measured by Rutherford back scattering measurements. X-ray diffraction measurements were carried out using a Bruker GADDS or a Bruker D8 Discover system. For the Ar/N$_2$ ratios within the range 95/5–75/25, all IrMn$_3$ films on Ta were grown with the local density approximation of DFT for the exchange-correlation energy.

**Magnetic tunnel junction fabrication and characterization.** MTJ devices were encapsulated in Al$_2$O$_3$. Electrical contacts were formed from 50 Å Ru/650 Å Au. TMR of the patterned devices was measured using a Quantum Design DynaCool. Magnetic properties were measured at 300 K using a Quantum Design Superconducting Quantum Interference Device (SQUID-VSM) magnetometer. Time-resolved transmission electron microscopy and electron energy loss spectroscopy studies were made using a JEOL ARM 200F with a Cold-FEG source operated at 200 keV. Magnetic properties were measured at 300 K using a quantum design superconducting quantum interference device vibrating sample magnetometer (SQUID-VSM) in magnetic fields of up to +7 T in both in-plane and out-of-plane directions. Uniaxial magnetic anisotropy $K_A$ values were calculated from $K_A = H_{sat} = M_s(2 + 2ME(5ME(\text{the effective magnetic field and } M_s \text{ the saturation magnetization)$.}}}$

**Calculation details.** The electronic structure and transmission functions of Mn$_3$Ge/MgO/Fe, Mn$_2$CuSb/MgO/Fe and Fe/MgO/Fe MTJs were calculated using a tight-binding linear muffin-tin orbital method in the atomic sphere approximation with the local density approximation of DFT for the exchange-correlation energy. For the Mn$_3$Ge/MgO/Fe MTJ, the in-plane lattice constant was fixed to the experimental lattice constant $a$ of bulk tetragonal Mn$_3$Ge ($a = 3.816$ Å and $c = 7.261$ Å (ref. 19)). For Mn$_2$CuSb/MgO/Fe the in-plane lattice constant was fixed to the calculated lattice constant $a$ of the Mn$_2$CuSb tetragonal inverse ferrimagnetic phase $a = 3.95$ Å (Table 1). Relaxed positions of atoms at the Mn$_3$Ge/MgO and Mn$_2$CuSb/MgO interfaces (for all possible terminations) were determined using the VASP molecular dynamic program. The O-top configuration was found to be the most stable configuration (as compared with Mg-top and hollow) for both terminations at the Mn$_3$Ge/MgO interface (in agreement with ref. 14), and for both terminations at the Mn$_2$CuSb/MgO interface. For Fe/MgO interface the atomic positions from ref. 21 were used.

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**Author contributions**

J.I. and Y.F. prepared the films and performed the experiments. DFT calculations were performed by S.V.F. S.S.P. conceived of the study. All authors discussed the results, contributed to the analysis of the data and wrote the paper.

**Additional information**

Supplementary Information accompanies this paper at http://www.nature.com/naturecommunications

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