Effect of N, C and B interstitials on the structural and magnetic properties of alloys with Cu3Au-structure

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High-throughput density functional calculations are used to investigate the effect of interstitial B, C and N atoms on 21 alloys reported to crystallize in the cubic Cu3Au structure. It is shown that the interstitials can have a significant impact on the magneto-crystalline anisotropy energy (MAE), the thermodynamic stability and the magnetic ground state structure, making these alloys interesting for hard magnetic, magnetocaloric and other applications. For 29 alloy/interstitial combinations the formation of stable alloys with interstitial concentrations above 5% is expected. In Ni3Mn interstitial N induces a tetragonal distortion with substantial uniaxial MAE for realistic N concentrations. Mn3XN_x (X=Rh, Ir, Pt and Sb) are identified as alloys with strong magneto-crystalline anisotropy. For Mn3Ir we find a strong enhancement of the MAE upon N alloying in the most stable collinear ferrimagnetic state as well as in the non-collinear magnetic ground state. Mn3Ir and Mn3IrN show also interesting topological transport properties. The effect of N concentration and strain on the magnetic properties are discussed. Further, the huge impact of N on the MAE of Mn3Ir and a possible impact of interstitial N on amorphous Mn3Ir, a material that is indispensable in today’s data storage devices, are discussed at hand of the electronic structure. For Mn3Sb, non-collinear, ferrimagnetic and ferromagnetic states are very close in energy, making this material potentially interesting for magnetocaloric applications. For the investigated Mn alloys and competing phases, the determination of the magnetic ground state is essential for a reliable prediction of the phase stability.

I. INTRODUCTION

Magnetic materials with a large magnetocrystalline anisotropy energy (MAE) are an important component in a wide range of everyday devices. Permanent magnets are used, for example, in hard disks, electro motors and direct drive wind turbines. Antiferromagnetic compounds find applications in exchange biased films for data storage devices, magnetic random access memories (MRAMs) and are of interest for spintronics applications [1–3]. Present high performance magnets like Nd-Fe-B contain a considerable amount of rare-earth elements, which in the past years have been subject to strong price fluctuations. The development of alternative permanent magnets without the use of critical rare-earth elements is therefore highly important [4–6]. Basic requirements for a good permanent magnet material are a large uniaxial magnetocrystalline anisotropy energy (MAE), a large saturation magnetization and a high Curie temperature.

High-throughput calculations in the framework of density functional theory (DFT) have become an increasingly important tool for the design of new functional materials in the past years [7, 8]. In addition to increased computational resources, this is due to the development of large databases and new concepts like efficient descriptors. A number of studies have focussed on the development of improved materials for batteries [9, 10], thermoelectrics [11, 13] or photovoltaics [14].

For magnetic materials, despite their technological importance, computational high-throughput studies are scarce. Bocarsly et al. performed a screening of potential magnetocaloric materials [15], and Els"asser et al. performed a screening of potential hard magnetic rare earth lean compounds [16–18]. Other studies focussed on potential rare earth free permanent magnets [19, 22] or the prediction of new magnetic compounds [23, 20]. A complete study of the stability, magnetic properties and MAE of potential new magnetic compounds taking into account competing phases and magnetic structures was to the best of our knowledge not performed up to now.

Theoretical high-throughput descriptions of functional magnetic materials involve different energy scales like Coulomb correlations, exchange interactions or magnetocrystalline anisotropy. Antiferromagnetic (AF) order can have a strong impact on the total energy and phase stability of Mn alloys [24, 27], which are of interest for hardmagnetic [28], magnetocaloric [29] as well as spintronics applications [2]. The effect of AF interactions on the energetics has been explored in high-throughput calculations for certain classes of compounds with known magnetic structure prototypes [24, 30, 51] or on selected compounds using evolutionary algorithms [25, 32] or a symmetry based prioritization [33]. Complex (non-collinear) structures and non-ferromagnetic ground states of competing structures in a phase diagram haven’t been studied up to now, as the computational effort is easily enlarged by one order of magnitude, and often suitable magnetic structure prototypes are not known. For the calculation of the MAE, which sets an upper limit

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to the coercivity, a proper treatment of spin-orbit coupling (SOC) and a high numerical accuracy, especially a careful convergence of k-space integrals are essential. Rare earth compounds with localized 4f electrons require a careful treatment of Coulomb correlations, where accurate methods suitable for a high-throughput screening still have to be developed. While individual aspects have been carefully addressed in the literature, the complexity of interactions makes a high-throughput prediction of functional magnetic materials still highly demanding.

It has been shown in the literature that interstitials can have a strong impact on the magnetic properties, including the magneto-crystalline anisotropy energy $34$–$36$, the magnetic structure $37$ and magnetostructural phase transitions $38$ $39$. The Cu$_3$Au-structure is one of the most common structure types with more than 200 reported compounds. Ferromagnetic compounds like Fe$_3$Pt can exhibit a large magnetization, which makes them interesting for applications in exchange coupled magnets $40$ $41$. Due to the cubic symmetry they have however only a relatively small MAE. In contrast, antiferromagnetic compounds like Mn$_3$Ir can exhibit a huge magneto-crystalline anisotropy $42$, but have vanishing magnetization. This is used in exchange biased films, where giant exchange anisotropies are reported $43$ $44$. Further, Mn-rich alloys of Cu$_3$Au-prototype have attracted attention due to a large magnetostriction $45$ and Berry curvature related phenomena like a large anomalous Hall effect and a facet-dependent spin Hall effect $46$ $48$.

In this article, we investigate the effect of interstitial N, C and B atoms on the stability and magnetic properties of 21 experimentally reported compounds with cubic Cu$_3$Au-structure and the magnetic 3d elements Cr, Mn, Fe, Co, and Ni. To identify the magnetic ground state of Mn alloys, the energy of different spin structures with maximal magnetic subgroup symmetry is calculated, which according to Ref. $49$ accounts for the majority of measured magnetic structures on the Bilbao Crystallographic Server. This allows to identify also complex non-collinear (NC) ground states, which are frequently observed in Mn alloys. In total, about 1000 magnetic configurations are probed to obtain reliable energies for alloys relevant for the phase stability of binary and ternary Mn alloy systems related to this study (Fig. 1). Stable interstitial phases are identified taking into account all possible decomposition reactions. We show that interstitials can lead to a strong enhancement of the magneto-crystalline anisotropy energy in these alloys and that high-throughput DFT calculations are able to identify hard magnetic materials even in the presence of complex magnetic interactions.

II. COMPUTATIONAL DETAILS

Calculations were performed within density functional theory using an extended version of the high-throughput environment HTE $50$ $51$ employing the VASP $52$ $53$ and FPLO $54$ codes. For the present investigation, the HTE has been extended for the calculation of magnetic properties such as the investigation of different spin configurations and the calculation of the magneto-crystalline anisotropy energy (MAE). Further, an automatic detection of suitable interstitial positions was implemented.

For structure optimizations and the calculation of formation energies the VASP code $52$ $53$ has been used following a similar approach as in Ref. $51$. For the final results shown here PAW pseudopotentials with a cut-off energy $E=500$ eV and constant density of k-points corresponding e.g. to a k-mesh of 14x14x14 k-points in the full Brillouin zone (FBZ) for Mn$_3$Sb were used. For MAE calculations, the full potential local orbital method $54$ (FPLO, version 14-49) was used, employing the optimized crystal and (collinear) magnetic structure obtained in the preceding VASP calculations. In the FPLO code, accurate full potential calculations are combined with the solution of the 4-component Kohn-Sham-Dirac equation, which implicitly contains spin-orbit coupling up to all orders.

As a descriptor for hard magnetic alloys the magnetic force theorem with a moderate k-mesh (up to 20x20x20 k-points in the FBZ) was used, where starting from a selfconsistent scalar relativistic calculation, the MAE is evaluated in one-step calculations as the band energy difference between different magnetization directions. In addition, selfconsistent relativistic calculations with up to 32x32x32 k-points in the FBZ were used to check the MAE for hard magnetic alloys identified in this study. Further, the magnetic anisotropy of Mn$_3$Ir(N) and

FIG. 1. Total number (filled blue) and number of NC (shaded blue) magnetic structures calculated for binary and ternary Mn alloys in this work. Some examples for automatically generated initial spin structures are shown as well: a non-collinear structure and collinear ferrimagnetic structure for Mn$_3$Ir and a collinear ferrimagnetic structure for Mn$_3$C$_6$ (from left to right).
Mn$_3$Pt(N) in the non-collinear ground state was calculated using the VASP code employing the magnetic force theorem. Here, a cut-off energy $E=400$ eV and a k-mesh of 12x12x12 k-points in the FBZ were used.

The results have been cross checked with selfconsistent calculations including spin-orbit coupling using a cut-off energy $E=500$ eV and k-meshs of 25x25x25 k-points (Mn$_3$IrN) and 13x13x13 k-points (Mn$_3$PtN), showing a convergence of the anisotropy energy of about 1 meV.

The anomalous Hall conductivity (AHC) is determined by using the WannierTools (WT) code, which is implemented within the framework of tight binding (TB) models [55]. The maximally localized Wannier functions (MLFWs) TB models are obtained by using the Wannier90 code [56]. We considered the s, p and d orbitals for Mn and Ir atoms, and s plus p orbitals were incorporated for N, resulting in 72 and 80 MLFWs for Mn$_3$Ir and Mn$_3$IrN respectively. To evaluate the AHC, the Berry curvature integration is carried out using a uniform $k$-points mesh of 401x401x401, which is expressed as following:

$$\sigma_{xy} = -\frac{e^2}{h} \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_n f[\epsilon(\mathbf{k}) - \mu] \Omega_{n,xy}(\mathbf{k})$$

(1)

$$\Omega_{n,xy}(\mathbf{k}) = -2\text{Im} \sum_{m \neq n} \langle \psi_{km} | v_x | \psi_{km} \rangle \langle \psi_{km} | v_y | \psi_{km} \rangle \left[ \epsilon_m(\mathbf{k}) - \epsilon_n(\mathbf{k}) \right]^2$$

(2)

where $e$ is elementary charge, $\mu$ is the chemical potential, $\psi_{n/m}$ denotes the Bloch wave function with energy eigenvalue $\epsilon_{n/m}$, $v_x, v_y$ is the velocity operator along Cartesian $x/y$ direction, and $f[\epsilon(\mathbf{k}) - \mu]$ is the Fermi-Dirac distribution function. In this work, the ANC $\alpha_{xy}$ is determined by using the Mott relation, where we considered only the derivative of AHC at the Fermi level $d\sigma_{xy}/de$, which provides a quantitative measure of the ANC.

$$\alpha_{xy} = \frac{\pi^2 k_B^2 T}{3e} \frac{d\sigma_{xy}}{de} |_{\epsilon=\mu}$$

(3)

Unless otherwise mentioned, the generalized gradient approximation (GGA) in the parameterization of Perdew, Burke and Ernzerhof [58] was used. For calculations in the local spin density approximation (LSDA), which often provides a better description of the spin magnetic moments for weakly correlated systems, the parameterization of Perdew and Wang [59] is used.

For the detection of suitable interstitial positions, the unit cell is scanned on a discrete mesh (here 24x24x24 points) for sufficient empty space, taking into account the covalent radii $r_j$ of the atoms as tabulated in the Atomic Simulation Environment (ASE) [60, 61]. As condition for suitable positions for the interstitial $I$ we use $d_{\text{min}} = \min_j (d_{ij} - s(r_j + r_I)) > 0$ for all neighbouring atoms $j$, where $s=0.7$ is a scaling factor and $d_{ij}$ is the distance between the interstitial and the atom $j$. This approach is similar to the detection of empty spheres used in some electronic structure codes. Preference can be given to high symmetry positions, which was done for the present calculations. The resulting interstitial positions for the Cu$_3$Au structure are shown in Fig. 2. The interstitial concentration was varied using 2x2x2, 1x2x2, 1x1x2 and 1x1x1 supercells with one interstitial position occupied, corresponding to interstitial concentrations of 3.0, 5.9, 11.1 and 20 at.%. An additional slight orthorhombic distortion (2-5% change of the lattice parameters) of the interstitial supercells was also tested, but did not have an effect on the stable interstitial compounds discussed in this manuscript.

The calculation of convex hulls is based on about 2000 alloys (experimental structures and structures close to the convex hull reported in the Materials Project [62]) for ternary $M$-$X$-$Z$ systems ($M=$Fe, Co, Ni, Cr and Mn; $Z=$B, C, N; $X$: other element). For the competing phases, the same numerical setup as for the interstitial compounds was used. For compounds with Mn atoms, where antiferromagnetic couplings are to be expected, the magnetic ground state was determined for the parent compounds with Cu$_3$Au-structure and their stable interstitial alloys. To ensure reliable phase diagrams for the Mn systems, the magnetic ground state was also determined for the experimentally reported competing phases and some of the energetically low lying phases reported in the Materials Project. Depending on the size and symmetry of the cell, we used spin configurations compatible with the maximal magnetic subgroups with propagation vector $q=(0,0,0)$, $(1/2,1/2,0)$, $(0,0,1/2)$ or alternatively an antiferromagnetic coupling of sublattices to probe the magnetic ground state.

In case of Mn$_3$Ir with Cu$_3$Au-prototype this results (after removal of symmetry equivalent configurations) in three collinear (FM, AF and ferrimagnetic) and one non-collinear initial spin structures for $q=(0,0,0)$, which is sufficient to obtain the experimentally reported ground state. It is noteworthy that the experimentally reported magnetic structure of Mn$_3$Ir does not have maximal magnetic subgroup symmetry, but is compatible with the symmetry of the second generation of maximal subgroups (maximal subgroup $R3m'$ (#166.101) of the maximal subgroup $Pm3m'$ (#221.95)). Nevertheless, the NC initial structures relax to the correct ground state, which underpins the high efficiency of the approach. We explicitly checked that the second generation of maximal subgroups results in the same magnetic ground state. The AF coupling of sublattices corresponds to the highest...
ranked magnetic structures in the prioritization scheme proposed in Ref. 33 and is especially useful when the paramagnetic structure has a relatively large number of inequivalent magnetic atoms $N_{\text{AF}}$ which are allowed to couple AF. The number of possible magnetic configurations at this prioritization level is $2^{N_{\text{AF}}-1}$, which results for instance for Mn$_3$C$_3$ of hexagonal Cr7C3,hP80,186-prototype (with 56 Mn atoms in the unit cell and $N_{\text{AF}} = 8$) in 128 possible magnetic configurations. While this approach does not ensure that the correct magnetic ground state is found in all cases, we believe it should give a reliable basis for the prediction of the phase stability in these systems. For the remaining alloys a ferromagnetic magnetic configuration was assumed.

### III. RESULTS OF HIGH-THROUGHPUT SCREENING

#### A. Parent compounds in Cu$_3$Au-structure

Tab. I shows the calculated properties of the 21 parent compounds with Cu$_3$Au structure. The calculated lattice parameters are in good agreement with the reported experimental values, with the largest deviation of about 2.6\% for Mn$_3$Rh and Mn$_3$Pt. With exception of Mn$_3$Sb and Fe$_3$Sn the calculated formation energy is negative for all compounds, as expected from formation of alloys in nature. Mn$_3$Sb has only been recently synthesized by high pressure synthesis 57 and the Cu$_3$Au-structure of Fe$_3$Sn was reported in high pressure studies, whereas the ambient pressure phase is hexagonal, in agreement with our calculations (see Supplementary). For 18 of the 21 compounds the calculated distance from the convex hull $\Delta E_h$ is below 50 meV/atom, which has been proposed in Ref. 51 as a simple descriptor for potentially stable or meta stable alloys in high-throughput calculations. In addition to the above mentioned Mn$_3$Sb and Fe$_3$Sn alloys, only Co$_3$Al is above this criterion, which shows the largest $\Delta E_h = 124$ meV/atom among the calculated compounds. A Cu$_3$Au-structure of Co$_3$Al has been discussed in the literature, but could not be stabilized experimentally 64. This result is confirmed by our study, where only the Al rich phases AlCo, Al$_3$Co$_2$, Al$_3$Co$_4$, and Al$_3$Co$_2$ are found to be stable. A complete list of the alloys relevant for the calculation of the convex hull in Tab. I is given in the supplementary material.

For Ni$_3$X ($X=$Si, Ge, Sn) and Co$_3$Ta the Fermi energy lies in a pseudo gap or valley of the density of states (DOS), see Fig. 3 resulting in a non-magnetic or only weakly ferromagnetic ground state despite the presence of the magnetic elements Ni and Co. For Mn$_3$X ($X=$Rh, Ir, Pt and Sb) the calculated ground state is non-collinear. The antiferromagnetic coupling of the neighbouring Mn atoms can not be fully satisfied in the Cu$_3$Au structure, resulting in a frustrated triangular spin structure with compensated magnetic moments, see for instance Ref. 42 for a more detailed discussion. For X=Rh, Ir, Pt this result is in agreement with experiment 65, 66 and earlier calculations 42. In case of Mn$_3$Pt a transition from a triangular to a collinear AF magnetic structure with a doubled unit cell along $c$ was observed around 365 K in neutron scattering experiments 65, while a similar transition was not observed for Mn$_3$Rh and Mn$_3$Ir. For Mn$_3$Sb, the energies of NC, ferrimagnetic and ferromagnetic (FM) magnetic structures are within less than 35 meV/atom. Experimentally, a collinear magnetic structure with almost compensating Mn magnetic moments was deduced from neutron scattering experiments 67, while more recent Mössbauer studies suggest a triangular magnetic structure 68.

![Fig. 3. Total DOS of Co$_3$Ta and Ni$_3$Sn for the experimental lattice parameters calculated with FPLO using GGA.](image-url)

**TABLE I. Properties of parent compounds in Cu$_3$Au-structure.** Experimental ($a_{\exp}$) and calculated lattice parameters ($a_{\text{GGA}}$), the calculated formation energy $E_f$ and distance from the convex hull $\Delta E_h$ and the total magnetic moment $M$ are shown. Non-collinear magnetic structures are marked with $^*$.**

| alloy | $a_{\exp}$ [Å] | $a_{\text{GGA}}$ [Å] | $E_f$ [eV/atom] | $\Delta E_h$ [eV/atom] | $M$ [$\mu_B$/f.u.] |
|-------|----------------|----------------|----------------|----------------|----------------|
| Co$_3$Al | 3.66 | 3.57 | -0.175 | 0.124 | 3.86 |
| Co$_3$Ta | 3.65 | 3.63 | -0.241 | 0.001 | 0.02 |
| Co$_3$Ti | 3.62 | 3.60 | -0.257 | 0.000 | 2.71 |
| Fe$_3$Ga | 3.70 | 3.66 | -0.133 | 0.000 | 6.98 |
| Fe$_3$Ge | 3.68 | 3.63 | -0.099 | 0.010 | 6.37 |
| Fe$_3$Pt | 3.77 | 3.73 | -0.076 | 0.041 | 8.46 |
| Fe$_3$Sn | 3.87 | 3.82 | 0.086 | 0.088 | 6.94 |
| Mn$_3$Ir | 3.77 | 3.71 | -0.215 | 0.000 | 0.01* |
| Mn$_3$Pt | 3.84 | 3.74 | -0.251 | 0.000 | 0.00* |
| Mn$_3$Rh | 3.81 | 3.71 | -0.142 | 0.000 | 0.00* |
| Mn$_3$Sb | 4.00 | 3.91 | 0.076 | 0.086 | 0.04* |
| Ni$_3$Fe | 3.55 | 3.55 | -0.088 | 0.000 | 4.72 |
| Ni$_3$Ga | 3.58 | 3.59 | -0.289 | 0.000 | 0.79 |
| Ni$_3$Ge | 3.57 | 3.57 | -0.292 | 0.000 | 0.05 |
| Ni$_3$In | 3.75 | 3.75 | -0.052 | 0.045 | 0.69 |
| Ni$_3$Mn | 3.59 | 3.56 | -0.102 | 0.000 | 4.85 |
| Ni$_3$Si | 3.51 | 3.51 | -0.462 | 0.000 | 0.04 |
| Ni$_3$Sn | 3.74 | 3.74 | -0.188 | 0.000 | 0.06 |
| Pt$_3$Co | 3.83 | 3.88 | -0.061 | 0.000 | 2.91 |
| Pt$_3$Cr | 3.88 | 3.91 | -0.254 | 0.000 | 2.60 |
| Rh$_3$Cr | 3.79 | 3.78 | -0.122 | 0.000 | 1.24 |
The energy difference $\Delta E_{FM}$ between the magnetic ground state and a ferromagnetic state can be as large as 243 meV/atom for Mn$_3$Pt. For some of the competing phases the calculated $\Delta E_{FM}$ is of similar magnitude, while $\Delta E_{FM}$ is trivially zero for compounds with a ferromagnetic ground state. The magnetic structure can thus have a deep impact on the calculated phase stability, going beyond typical effects due to finite temperature. This is exemplarily shown in Fig. 4 for binary Mn-Ir alloys. Assuming a ferromagnetic spin alignment a hexagonal MnIr$_3$ phase, L1$_0$-MnIr and a hexagonal Mn$_3$Ir phase are on or nearly on the convex hull, in good agreement with results obtained from Materials Project [62] and the Open Quantum Materials Database OQMD [69]. In contrast, calculations with the magnetic ground state search resulted in a collinear AF and a NC triangular AF structure, respectively, in agreement with experiment [66, 70]. This is accompanied by a significant lowering of the formation energy of nearly 0.2 meV/atom. In contrast, the magnetic ground state search for the two (experimentally not observed) hexagonal phases did not have an effect on the calculated formation energy. A similar impact of the magnetic structure on the calculated phase stability as for Mn-Ir is found also in other Mn alloy systems and will be discussed in more detail elsewhere.

B. Effect of interstitials on structural and magnetic properties

Fig. 5 shows the calculated MAE (maximum energy difference between magnetization directions [001], [010], [100], [110], and [111]) for the Cu$_3$Au parent compounds and interstitial alloys with one N, C or B atom per f.u., assuming a ferromagnetic alignment of the spins. It can be seen that interstitials can induce huge MAEs up to about 2 meV/atom, one to two orders higher in magnitude than in their cubic parent compounds. N, C and B are similarly efficient in inducing these large MAEs. The largest MAEs are calculated for the 3d interstitial position, which leads to a tetragonal distortion of the lattice and therefore is promising to achieve uniaxial MAEs. There can be also a significant MAE between the [001], [110] and [111] directions of up to about 0.2 meV/atom.

FIG. 4. Calculated formation energies (red squares) and convex hull (solid red line) for the magnetic ground state of Mn-Ir alloys. For comparison the convex hull (dashed lines) and formation energies obtained assuming a ferromagnetic structure from our calculations (red circles), the Materials Project (blue diamonds) and the Open Quantum Materials Database (green triangles) are shown. Formation energies for the experimentally reported L1$_0$- and Cu$_3$Au-crystal structures obtained with the different calculation methods are plotted with filled symbols. Top: calculated magnetic groundstate structures for MnIr and Mn$_3$Ir. Note that SOC was not included here and that the spin-quantization axis has been rotated to match the results of calculations with SOC.

FIG. 5. Maximum energy difference $\Delta E_{max} = \max(E_i - E_j)$ between different magnetization directions vs. total magnetic moment $M$ (top) and maximal change of the ratio of lattice parameters $\delta = \max(a_i/a_j)$ vs. change in the formation energy $\Delta E_f$ (bottom). Data are shown for parent compounds in the Cu$_3$Au-structure (filled black squares) and interstitial structures with B (red), C (blue) and N (green). The interstitial 1b, 3d and 8g positions are marked with filled squares, circles and triangles, respectively.
for the cubic compounds with heavy 5d elements (while the energy difference between [001], [010] and [100] is zero due to symmetry). It should be noted however, that neither stability nor potential antiferromagnetic spin configurations are considered in this plot. The MAEs shown in Fig. 5 thus rather serve to illustrate the potential of interstitials in inducing large MAEs, while for the majority of the data points an experimental realization will not be possible.

This becomes also clear from the lower plot of Fig. 5 which shows the formation energy difference \( \Delta E_f \) with respect to the Cu3Au parent compound in relation to the induced structural distortion for different interstitial atoms and positions. For the majority of data points \( \Delta E_f \) is positive indicating that the interstitial alloy is unlikely to form, at least at the interstitial concentration of 20 at.% considered in the plot. Further, in most cases the 1b interstitial position, which does not break the cubic symmetry, is most stable. In most cases this means that a significant amount of the MAE is not expected. Noteworthy exceptions, where the spin structure breaks the cubic symmetry, will be discussed further below. Significant structural distortions are found for the 3d and 8g interstitial positions with negative \( \Delta E_f \) in some cases, suggesting that the interstitial alloys are likely to form. For the 8g interstitial position, which is least favourable from geometrical considerations, we often find relaxation to a structure corresponding to the 1b or 3d position, which provide a higher volume for the interstitial atom.

### C. Stable interstitial phases

Tab. II lists interstitial alloys, where the distance to the convex hull \( \Delta E_h \) is below 50 meV/atom for an interstitial concentration of more than 5 at.%, suggesting that a substantial amount of interstitial atoms can be incorporated in the parent alloy with Cu3Au-structure. This criterion is matched for 29 parent/interstitial combinations, including known antiperovskites like Mn3YN (\( X = \text{Rh, Fe, Pt and Sb} \)) [45, 71], Ni3InN [72], Fe3PtN [73], Co3AlC [74], Fe3GaN [75] and Fe3SnN [76]. Only the most stable interstitial position is listed. In some cases more than one interstitial position lowers the formation energy, suggesting a partial occupation of the interstitial sites. This is for instance the case for Ni3Mn, where the N 3d and 1b positions are close in energy and both of them fulfill the \( \Delta E_h \) criterion for 5.9 at.% N concentration.

The largest gain in formation energy with respect to the parent compound \( \Delta E_f \approx -0.3 \text{ eV/atom} \) is found for Mn3SbN, where the 'interstitial' alloy [71] was reported more than 20 years before the meta stable 'parent' alloy Mn3Sb [69]. For the majority of interstitial alloys, stability is accompanied by a negative \( \Delta E_f \). However, even a large negative \( \Delta E_f \) does not ensure formation of a stable alloy. This is the case for instance for Co3AlN and Co3TiN, where the distance from the convex hull is 0.3-0.4 eV/atom, despite \( \Delta E_f \) values of about -0.16 and -0.13 eV/atom, respectively. Vice versa, a positive \( \Delta E_f \) does not prevent the formation of a stable interstitial alloy, as in the case of Fe3GaC.

For the majority of interstitials, the 1b position is energetically preferred. The 3d position, which is especially interesting for hard magnetic applications as it breaks the cubic symmetry and leads to a tetragonal distortion of the lattice, is energetically most preferable for Ni3FeN \( x \), Ni3MnN \( x \), Co3TaN \( x \) and Co3TiN \( x \). In all cases substantial MAEs between -0.67 and 1.3 meV/cell are induced for \( x = 1 \) (20 at.% N). The largest absolute value of 1.3 meV/cell is found for Ni3MnN, albeit with an easy plane magnetization. However, for Co3TaN \( x \) and Co3TiN \( x \) only a minor amount of interstitials is expected to be incorporated due to the formation of extraordinary stable Ta-N and Ti-N phases (see Supplementary). For realistic N concentrations (11 at.% N, \( x = 0.5 \)), Ni3MnN \( x \) shows a significant uniaxial MAE around -100 \( \mu \text{eV/atom} \), com-

### TABLE II. Interstitial alloys with a distance from the convex hull \( \Delta E_h < 50 \text{ meV/atom} \). The parent compound with Cu3Au-structure, the interstitial atom and position and the highest interstitial concentration \( c_{\text{max}} \) with \( \Delta E_h < 50 \text{ meV/atom} \) (with three different values of the interstitial concentrations: 5.9, 11.1 and 20) in at.% are shown along with the calculated formation energy \( E_f \), \( \Delta E_h \) and the total magnetic moment \( M \) per atom. Non-collinear magnetic structures and antiferromagnetic/ ferrimagnetic structures are indicated with * and †, respectively.

| Parent |Interstitial | \( c_{\text{max}} \) | \( E_f \) [eV/atom] | \( \Delta E_h \) [eV/atom] | \( M \) [\( \mu_B \)/atom] |
|--------|-------------|---------------------|---------------------|---------------------|---------------------|
| Co3Al  | 1b          | 20.0                | -0.230              | 0.010               | 0.00                |
| Co3Ta  | 1b          | 11.1                | -0.272              | 0.044               | 0.00                |
| Co3Ta  | 1b          | 11.1                | -0.211              | 0.035               | 0.00                |
| Co3Ti  | 1b          | 11.1                | -0.324              | 0.043               | 0.31                |
| Co3Ti  | 1b          | 11.1                | -0.302              | 0.003               | 0.22                |
| Fe3Ga  | 1b          | 5.9                 | -0.124              | 0.047               | 1.50                |
| Fe3Ga  | 1b          | 20.0                | -0.063              | 0.044               | 0.63                |
| Fe3Ga  | 1b          | 20.0                | -0.255              | 0.016               | 1.05                |
| Fe3Ge  | 1b          | 5.9                 | -0.096              | 0.040               | 1.32†               |
| Fe3Pt  | 1b          | 20.0                | -0.263              | 0.000               | 1.63                |
| Fe3Sn  | 1b          | 20.0                | -0.104              | 0.037               | 1.02                |
| Mn3Ir  | 1b          | 20.0                | -0.359              | 0.026               | 0.00*               |
| Mn3Pt  | 1b          | 20.0                | -0.471              | 0.000               | 0.00*               |
| Mn3Rh  | 1b          | 20.0                | -0.356              | 0.000               | 0.00*               |
| Mn3Sb  | 1b          | 20.0                | -0.226              | 0.036               | 0.55†               |
| Ni3Fe  | 3d          | 5.9                 | -0.063              | 0.038               | 1.05                |
| Ni3Ga  | 1b          | 5.9                 | -0.305              | 0.014               | 0.00                |
| Ni3Ga  | 1b          | 5.9                 | -0.273              | 0.006               | 0.00                |
| Ni3Ga  | 1b          | 11.1                | -0.258              | 0.036               | 0.00                |
| Ni3In  | 1b          | 20.0                | -0.205              | 0.049               | 0.00                |
| Ni3In  | 1b          | 20.0                | -0.040              | 0.038               | 0.00                |
| Ni3In  | 1b          | 20.0                | -0.173              | 0.000               | 0.00                |
| Ni3Mn  | 1b          | 5.9                 | -0.110              | 0.041               | 1.09                |
| Ni3Mn  | 1b          | 5.9                 | -0.071              | 0.025               | 1.08                |
| Ni3Mn  | 3d          | 11.1                | -0.113              | 0.044               | 0.95                |
| Ni3Sn  | 1b          | 5.9                 | -0.196              | 0.038               | 0.00                |
| Ni3Sn  | 1b          | 5.9                 | -0.134              | 0.043               | 0.00                |
| Ni3Sn  | 1b          | 11.1                | -0.135              | 0.045               | 0.00                |
| Pt3Co  | 8g          | 11.1                | -0.069              | 0.048               | 0.59                |
parable to hcp-Co. For Ni$_3$Fe$_x$, the MAE is small ($\approx 15 \, \mu$eV/atom) and depends on the choice of supercell.

Table I lists stable alloys with large uniaxial MAE for the most stable collinear magnetic structure obtained in the preceding VASP calculations. For comparison, also results obtained within LSDA (employing the same GGA optimized structure) are shown. For both GGA and LSDA, the listed alloys show a large uniaxial MAE, but the absolute values can differ significantly. While GGA typically yields lattice parameters in better agreement with experiment, the local spin magnetic moments are often better described within LSDA. As the MAE depends on subtle details of the electronic structure, this can have a strong impact on the calculated MAE [17], as is the case here for Mn$_3$Ir, Mn$_3$PtN and Mn$_3$SbN. According to the table, Mn$_3$XN and Mn$_3$X (X=Rh, Ir, Pt and Sb) would have a huge potential for application as hard magnets, provided they could be stabilized in a ferrimagnetic magnetic structure. The largest MAE of about $-1.3 \, \text{meV/atom}$ is obtained for Mn$_3$IrN. Comparison with Tabs. I and II shows however that with exception of Mn$_3$SbN these large MAEs can not be readily used in bulk magnets, as the ground state is non-collinear with zero net magnetization. However, the strong MAE could be used in exchange coupled multilayer structures, where a hard magnetic antiferromagnetic layer is coupled to a soft magnetic layer with large magnetization and high Curie temperature. Moreover, as Mn$_3$SbN shows, a ferrimagnetic ground state may be induced either by variation of the interstitial concentration or epitaxial strain in a thin film. This will be further explored in Sec. IV.

Comparison of Tab. II and Tab. I shows some remarkable effects of the interstitials on the magnetic properties. While the ground states of Co$_3$Al is ferromagnetic, Co$_3$AlC is found to be non-magnetic. Similar, the total magnetic moment of Co$_3$Ti is strongly reduced by interstitial B, resulting in a non-magnetic ground state for (unstable) Co$_3$TiB. On the contrary, Co$_3$Ta is non-magnetic, while Co$_3$TaN would be magnetic if it could be synthesized. This shows that interstitials can be very efficient to tune magnetic transitions, which makes interstitial alloying interesting also for magnetocaloric applications.

**TABLE III. Stable alloys with large uniaxial MAE=E$_{[001]} - E_{[100]}^\dagger$.**

| alloy        | interst. | $M_{GGA}/M_{LSDA}$ [µB/atom] | MAE$_{GGA}$/MAE$_{LSDA}$ [meV/atom] |
|--------------|----------|-------------------------------|-----------------------------------|
| Mn$_3$Ir     | -        | 0.37$^\ddagger$/0.30$^\ddagger$ | 0.19/-0.43                        |
| Mn$_3$IrN    | N 1b     | 0.54$^\ddagger$/0.49$^\ddagger$ | 1.32/-1.27                        |
| Mn$_3$Pt     | -        | 0.36$^\ddagger$/0.32$^\ddagger$ | 0.09/-0.11                        |
| Mn$_3$PtN    | N 1b     | 0.63$^\ddagger$/0.57$^\ddagger$ | 0.16/-0.22                        |
| Mn$_3$Rh     | -        | 0.43$^\ddagger$/0.37$^\ddagger$ | 0.10/-0.12                        |
| Mn$_3$RhN    | N 1b     | 0.56$^\ddagger$/0.51$^\ddagger$ | 0.31/-0.29                        |
| Mn$_3$SbN    | N 1b     | 0.55$^\ddagger$/0.52$^\ddagger$ | 0.20/-0.13                        |
| Ni$_3$MnN$_{0.5}$ | N 3d | 0.96/0.90                  | 0.09/-0.10                        |

In conclusion, with Tabs. I and II shows however that with exception of Mn$_3$SbN these large MAEs can not be readily used in bulk magnets, as the ground state is non-collinear with zero net magnetization. However, the strong MAE could be used in exchange coupled multilayer structures, where a hard magnetic antiferromagnetic layer is coupled to a soft magnetic layer with large magnetization and high Curie temperature. Moreover, as Mn$_3$SbN shows, a ferrimagnetic ground state may be induced either by variation of the interstitial concentration or epitaxial strain in a thin film. This will be further explored in Sec. IV.

Comparison of Tab. II and Tab. I shows some remarkable effects of the interstitials on the magnetic properties. While the ground states of Co$_3$Al is ferromagnetic, Co$_3$AlC is found to be non-magnetic. Similar, the total magnetic moment of Co$_3$Ti is strongly reduced by interstitial B, resulting in a non-magnetic ground state for (unstable) Co$_3$TiB. On the contrary, Co$_3$Ta is non-magnetic, while Co$_3$TaN would be magnetic if it could be synthesized. This shows that interstitials can be very efficient to tune magnetic transitions, which makes interstitial alloying interesting also for magnetocaloric applications.

**IV. DISCUSSION**

In the previous section we have shown that interstitial atoms can have a strong impact on the stability, magnetic properties and MAE of compounds. While in principle huge MAEs are possible (see Fig. 5), stable compounds with the necessary properties for a good permanent magnet are rare, in line with experimental experience. Among the 63 parent/interstitial combinations investigated in this study, Ni$_3$Mn$_x$ with $x \approx 0.5$ was identified as a stable ferrimagnetic alloy with magnetic properties comparable to hcp-Co at reduced cost. Further, Mn$_3$SbN was identified as a ferrimagnet with large MAE, in agreement with a large magnetostriction observed experimentally in this compound [15]. It can be expected that an extended screening of more parent/interstitial combinations leads to even more promising candidates for hard magnetic applications.

The most striking observation in this study is however the huge MAE in Mn$_3$XN$_x$ (X=Rh, Ir, Pt, and Sb) and the strong impact of interstitial N on the MAE in these compounds, especially for Mn$_3$IrN. Fig. 6 shows the calculated DOS of Mn$_3$IrN and Mn$_3$Ir. Interstitial N induces changes in the electronic structure in the range of a few hundred meV, larger than the changes in the electronic structure due to a change of the magnetization direction from the easy [001] axis to the hard [100] axis due to spin-orbit coupling, also shown for comparison in Fig. 3. The changes in the electronic structure lead also to a change of the orbital moment anisotropy of Ir ($\Delta \mu_l = 0.046 \mu_B$ for Mn$_3$Ir; $\Delta \mu_l = 0.073 \mu_B$ for Mn$_3$IrN) and Mn ($\Delta \mu_l = 0.052 \mu_B$ for Mn$_3$Ir; $\Delta \mu_l = -0.085 \mu_B$ for Mn$_3$IrN), which is closely related to the MAE. Thus, interstitial N has an impact on the electronic structure and MAE going far beyond structural distortions, in agreement with earlier investigations [36].

Another important observation is that according to our calculations the N concentration in Mn$_3$XN$_x$ (X=Rh, Ir, Pt) can be varied in the range between 0-20 at.%, thus providing an additional tuning parameter for the MAE, see Fig. 7. In all cases the interstitial alloys are on or close to the convex hull, well below the $\Delta E_h < 50 \, \text{meV/atom}$ criterion for (meta-)stability. However, a triangular spin structure is energetically preferred over the collinear ferrimagnetic structure throughout the whole concentration range. For Mn$_3$RhN$_x$, where the N 1b and 3d position lead to a gain in formation energy, more than 30 at.% interstitial N can be incorporated into the structure based on the $\Delta E_h$ criterion. For these high N concentrations a ferrimagnetic structure with significant net magnetization around $2 \, \mu_B$/f.u. and MAE around $-1 \, \text{meV/f.u.}$ is expected based on our calculations. A partial occupation of the N 1b and 3d interstitial positions may also explain deviations from an ideal triangular spin structure.
the suitability of the band anisotropy as a descriptor in high-throughput calculations. The results show that the huge MAE is robust against variations of the c/a-ratio in epitaxially grown films, with values between -4 and -8 meV/cell for a wide range of lattice parameters. In particular, the results show that the symmetry breaking due to the ferrimagnetic spin arrangement is responsible for the huge MAE and not a slight distortion of the lattice from cubic symmetry (c/a=1). The total spin moment ranges between 2 and 4 µB/cell for c/a-ratios investigated. Thus, Mn₃IrN would be an excellent hard magnetic material if it could be stabilized in the ferrimagnetic spin configuration, which would outperform the known hard magnet L₁₀-CoPt with an MAE of about 0.5 meV/atom and a total magnetic moment of about 1.1 µB/atom. According to our VASP calculations, N has also a strong impact on the magnetic anisotropy in the NC ground state and enhances the energy difference ΔE = E₆₄₋₅₅ between the easy Γ₆₄ ground state and the Γ₅⁵ representation of the magnetic structure from about -8 meV (Mn₃Ir) to -12 meV (Mn₃IrN). Based on these results we expect that interstitial N has also a strong impact on amorphous Mn₃Ir, which is one of the state-of-the-art materials for providing exchange bias in hard magnetic films. In case of Mn₃Pt(N), ΔE changes sign from -3 meV (Mn₃Pt) to 6 meV (Mn₃PtN), so that the Γ₅⁵ representation becomes the ground state.

According to our calculations, a relatively large epitaxial strain around 5% is necessary to stabilize a ferrimagnetic ground state in Mn₃IrN at 0 K, see Fig. 1 in the Supplementary. Surprisingly, the necessary strain in Mn₃Ir is lower, despite the smaller energy difference between the NC and ferrimagnetic state in Mn₃IrN, possibly indicating that an intermediate N concentration is most suitable for a stabilization of the ferrimagnetic state. Further, we note that in the related compound Mn₃GaN calculations by Zemen et al. [78] have shown that a combination of strain and temperature leads to a stabilization of a ferrimagnetic ground state. A similar behaviour was recently also experimentally observed for a Mn₃NiN film on a SrTiO₃ substrate [79]. In passing we note also that epitaxial strain in Mn₃XN as well as reported in this compound [71].

Fig. 8 shows the calculated MAE as a function of c/a for Mn₃IrN in the ferrimagnetic spin configuration employing selfconsistent calculations with an enhanced k-mesh. First, we note that the MAE obtained from selfconsistent calculations for the bulk value is in good agreement with the MAE obtained with the force theorem in the high-throughput calculations, underlining

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**Fig. 6.** Band structure (top) of ferrimagnetic Mn₃IrN for the easy [001] axis (blue) and the hard [100] axis (red). Comparison of the DOS of ferrimagnetic Mn₃IrN (blue) and ferrimagnetic Mn₃Ir (cyan) for the easy [001] axis (bottom). For simplicity c/a in both graphs was set to 1.

**Fig. 7.** Distance from convex hull ∆Eₖ vs. N concentration for Mn₃XₙₓNₓ (X=Rh, Ir, Pt) (filled symbols: NC, plus: ferrimagnetic).

**Fig. 8.** Calculated MAE vs. c/a-ratio for Mn₃IrN. Results are shown for GGA (filled green circles) and LSDA (filled red squares). The volume was fixed to the GGA optimized bulk value.
in Mn$_3$X ($X=\text{Rh, Ir, Pt}$) is accompanied by a significant net magnetization in the NC ground state, albeit smaller than the one predicted for Mn$_3$SnN.\cite{80} This suggests that a piezomagnetic effect may be observed in a wide range of NC antiferromagnets. A more detailed analysis is beyond the scope of the present high-throughput study and left for future experimental and theoretical work.

The anomalous Hall conductivity (AHC) and anomalous Nernst coefficient (ANC) depend on the spin configuration of the system. The origin of non-vanishing AHC in $\Gamma_{4g}$ magnetic ordering has been studied based on symmetry analysis.\cite{81, 82} Fig. 9 shows the calculated band structure and anomalous Hall conductivity (AHC) of Mn$_3$Ir and Mn$_3$IrN in the $\Gamma_{4g}$ ground state. For both compounds, the calculated AHC $\sigma_{111}$ is significant, with 315 S/cm for Mn$_3$Ir and 157 S/cm for Mn$_3$IrN. This is in overall good agreement with a study by Chen et al., where a moderately smaller AHC was calculated for Mn$_3$Ir (230 S/cm)\cite{83}, but differs in sign and magnitude from a recent AHC calculation by Huyen et al. for Mn$_3$IrN (-573.3 S/cm)\cite{84}. The AHC has been shown to be highly sensitive to the choice of the magnetization direction\cite{82}, which may be a reason for this discrepancy. Based on our calculations, we predict a huge anomalous Nernst effect, with calculated ANC $\alpha_{111}$ of 6195 and -4892 S/(cm eV) for Mn$_3$Ir and Mn$_3$IrN respectively. Noteworthy, there is change in sign of ANC for Mn$_3$IrN. Both end members of the Mn$_3$IrN$_x$ (0 $\leq x \leq 1$) series show interesting transport properties. According to our thermodynamic calculations alloys with intermediate N concentrations are stable, which may be a route for further tuning of the transport properties.

FIG. 9. Calculated band structure and AHC of Mn$_3$Ir [(a) and (b)] and Mn$_3$IrN [(c) and (d)] respectively. Fig. (b) and (d) the black, red and green line correspond to the $\sigma_x$ and $\sigma_y$ and $\sigma_z$ AHC tensor.

V. SUMMARY

In summary we have shown that the interstitial elements N, C and B can have a strong impact on the magnetic properties of cubic compounds and that interstitial alloying can be a fruitful approach to enhance the magnetic properties. For reliable predictions of materials with enhanced properties, phase stability (i.e. calculation of the convex hull including all possible decomposition reactions) is essential. The high-throughput scheme introduced in this article is a unique computational tool to identify promising magnetic materials in order to guide the design of new magnetic materials.

Based on our calculations, we expect ferromagnetic Ni$_3$Mn$_x$N$_{2-x}$ (with about 11 at.\% N) and the ferrimagnetic compounds Mn$_3$SbN and Mn$_3$Rh$_x$N$_{2-x}$ (with about 30 at.\% N) to show bulk magnetic properties with sizable magnetization in the range of 0.5-1 $\mu_B$/atom and sizable uniaxial MAE in the range of 100-200 $\mu$eV/atom.

The Mn$_3$X(N) ($X=\text{Rh, Ir, Pt and Sb}$) compounds are identified as materials with strong MAE and a sizable magnetization in a collinear ferrimagnet state. The calculations also show that -- with exception of Mn$_3$SbN -- these materials can not readily be used as bulk magnets, as their ground state is a non-collinear fully compensated antiferromagnet. As a strong magnetic anisotropy is also present in the non-collinear ground state of these alloys, these materials are however of strong interest for multilayer structures where the hard magnetic antiferromagnetic layer is exchange coupled to a soft magnetic layer with large magnetization. Especially for Mn$_3$Ir, which is already used as exchange bias material in hard magnetic films, interstitial alloying with N can be a promising approach to enhance the magnetic properties of a state-of-the-art material. Further, an application of the strong MAE in the ferrimagnetic state may be possible in thin films, but requires more detailed theoretical and experimental investigations. For Mn$_3$Ir and Mn$_3$IrN we find a significant anomalous Hall effect and a huge anomalous Nernst effect. Variation of the N concentration may be a route for further optimization of the topological transport properties, as the Mn$_3$IrN$_x$ alloys share the same triangular spin arrangement. Moreover, we have shown that interstitials can also have an impact on the ground state magnetic structure and can thus be used to tune magnetic transitions for instance in magnetocaloric applications. In Mn$_3$Sb(N) the magnetic ground state changes from NC to ferrimagnetic upon alloying with N. For Mn$_3$Sb, NC, ferrimagnetic and ferromagnetic magnetic states are very close in energy, indicating that moderate fields may be sufficient to switch from a low to a high magnetization state. This makes Mn$_3$Sb based alloys potentially interesting for magnetocaloric applications, but requires further studies.

For Mn alloys, our calculations show that the assumption of a ferromagnetic spin alignment, which is currently used in high-throughput databases, is unreliable to predict formation energies and phase stability. The symme-
try based methods applied in this article to determine the magnetic ground state can pave the way to predictions of the phase stability based on DFT-high-throughput calculations for these important alloy systems, which are of interest e.g. for permanent magnet, magnetocaloric and spintronics applications.

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