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Supplementary Information to "Synthesis and characterisation of a quaternary nitride series with spin-glass behaviour: Sn_xGe_{1-x}Fe₃N"

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Supporting Figures:

X-ray diffraction pattern:



Figure S1: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ${}^{1a}(\text{Sn}_{0.040(2)}\text{Ge}_{0.960(2)}){}^{3c}(\text{Fe}_3){}^{1b}(\text{N})$ in space group $Pm\overline{3}m$ (a = 3.7935(1) Å, $R_B = 4.62\%$).



Figure S2: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ^{1a}(Sn_{0.190(2)}Ge_{0.810(2)})^{3c}(Fe₃)^{1b}(N) in space group $Pm\overline{3}m$ (a = 3.8122(1) Å, $R_B = 3.46\%$).



Figure S3: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ¹*a*(Sn_{0.34(1)}Ge_{0.63(1)}Fe_{0.03(1)})³*c*(Fe₃)¹*b*(N) in space group $Pm\overline{3}m$ (*a* = 3.8273(1) Å, *R*_B = 2.76%) with 4% of a tin side phase.



Figure S4: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ¹*a*(Sn_{0.43(1)}Ge_{0.51(1)}Fe_{0.06(1)})³*c*(Fe₃)¹*b*(N) in space group $Pm\overline{3}m$ (a = 3.8371(1) Å, $R_B = 2.74\%$) with 8% of a tin side phase.



Figure S5: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ${}^{1a}(\text{Sn}_{0.44(1)}\text{Ge}_{0.52(1)}\text{Fe}_{0.04(1)}){}^{3c}(\text{Fe}_3){}^{1b}(\text{N})$ in space group $Pm\overline{3}m$ (a = 3.8391(1) Å, $R_B = 2.56\%$).



Figure S6: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ¹*a*(Sn_{0.57(1)}Ge_{0.38(1)}Fe_{0.05(1)})³*c*(Fe₃)¹*b*(N) in space group $Pm\overline{3}m$ (a = 3.8528(1) Å, $R_B = 2.75\%$) with 7% of a tin side phase.



Figure S7: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ${}^{1a}(\text{Sn}_{0.66(1)}\text{Ge}_{0.26(1)}\text{Fe}_{0.08(1)})^{3c}(\text{Fe}_3)^{1b}(\text{N})$ in space group $Pm\overline{3}m$ (a = 3.8614(1) Å, $R_B = 2.60\%$) with 9% of a tin side phase.



Figure S8: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ^{1a}(Sn_{0.85(1)}Ge_{0.05(1)}Fe_{0.10(1)})^{3c}(Fe₃)^{1b}(N) in space group $Pm\overline{3}m$ (a = 3.8834(1) Å, $R_B = 4.88\%$) with 12% of a tin side phase.



Figure S9: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ^{1a}(Sn_{0.82(1)}Ge_{0.08(1)}Fe_{0.10(1)})^{3c}(Fe₃)^{1b}(N) in space group $Pm\overline{3}m$ (a = 3.8792(1) Å, $R_B = 3.52\%$) with 12% of a tin side phase.



Figure S10: X-ray diffraction pattern measured with Mo K α_1 radiation and Rietveld refinement of the quaternary nitride ${}^{1a}(Sn_{0.82(1)}Ge_{0.08(1)}Fe_{0.10(1)}){}^{3c}(Fe_3){}^{1b}(N)$ in space group $Pm\overline{3}m$ (a = 3.8792(1) Å, $R_B = 3.52\%$) with 12% of a tin side phase.

Magnetic measurements:



Figure S11: a and **b** Confirmation of the spin-glass behaviour for two quaternary nitrides $Sn_xGe_{1-x}Fe_3N$: Temperature dependence of the real part of the molar AC susceptibility χ'_m , measured in a frequency range $84 \le f \le 10000$ Hz. The peak temperature T_m of the χ'_m maximum depends on the frequency.



Figure S12: a–**e** Molar FC (filled symbols) and ZFC (empty symbols) DC susceptibilities for different quaternary nitrides $Sn_xGe_{1-x}Fe_3N$ as a function of temperature under probing fields $B_0 = 0.01$ T and 0.1 T. The susceptibilities show maxima at low temperatures T_m , that characterize the magnetic transitions (T_g). The FC and ZFC curves diverge below T_m , a fingerprint of the spin-glass behaviour in these compounds.