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Magnetocaloric $GdMn_{1-x}Ru_xSi$ compounds with x = 0 – 1 for gas liquefaction

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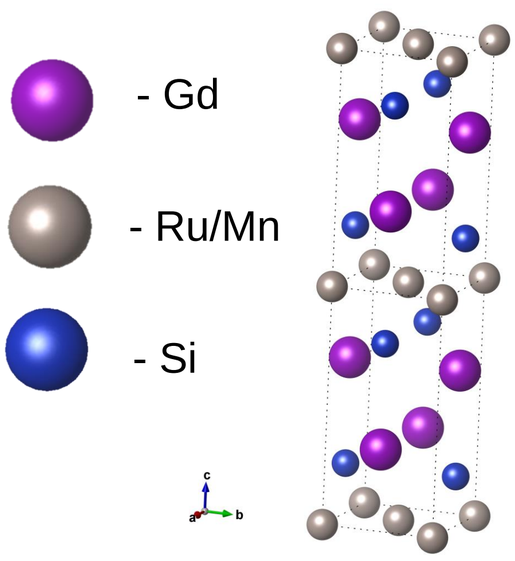
Introduction and Aim



RTX compounds, where R is a 4f metal, T and X are d- and p-elements, are of great interest due to the combination of diverse magnetic, transport and spectral properties, which are retained up to very high temperatures. For example, the GdFeSi compound demonstrates a rather large value of the magnetocaloric effect MCE, amounting to 5 J/kgK, in a field changing up to 17 kOe at the Curie temperature T_C = 130 K. Intermetallic compounds GdTMSi crystallize in a tetragonal structure of the CeFeSi type (P4/nmm), w hich is built of alternating (001) layers in the sequence: Gd-Si-2TM-Si-Gd-Gd-Si-2TM-Si-Gd (TM = Ti, V, Cr, Mn, Fe, Ru). The magnetic properties of GdTMSi compounds depend on the interatomic distances as well as TM ions types.

The aim of this study is the search for new GdTMSi compounds with MCE optimal for gas liquefaction.

Results and Discussions

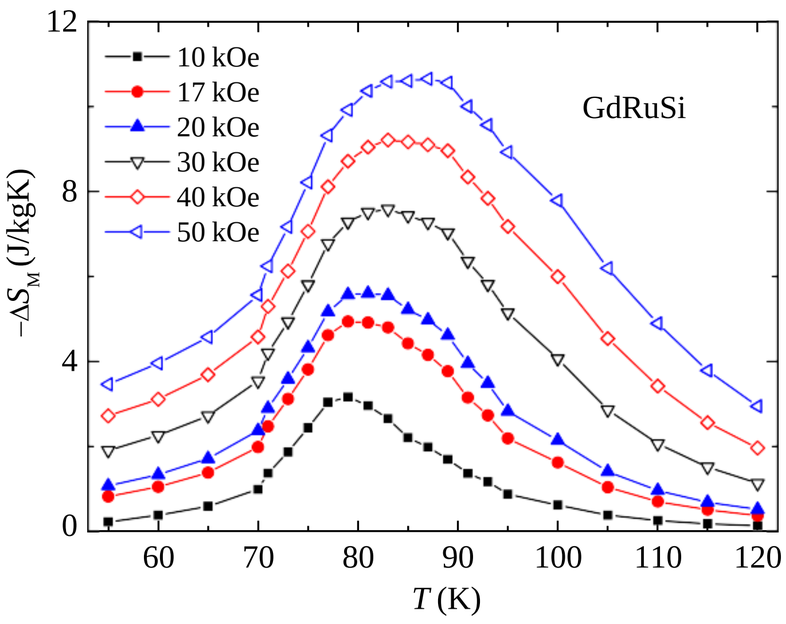


Fig. 1. Temperature dependences of the change in magnetic entropy $-\Delta S_M(T)$ in a field changing up to 10-50 kOe for GdRuSi.

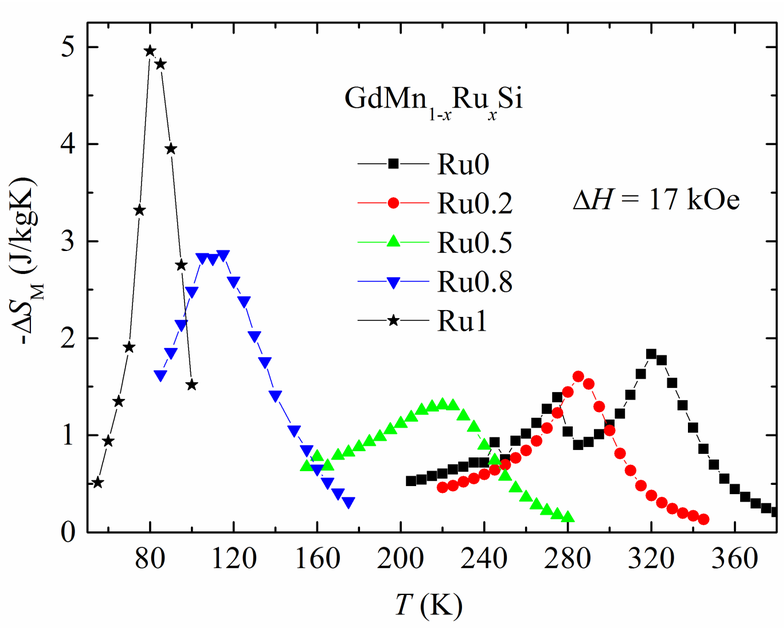


Fig. 2. Temperature dependences of the change in magnetic entropy $-\Delta S_M(T)$ for $GdMn_{1-x}Ru_xSi$, x=0-1 with a field change of 0-17 kOe.

For equiatomic compound GdRuSi, a large MCE value was found, which is 10.7 at 87 K and 4.94 J/kgK at 79 K for field changes of 0-50 and 0-17 kOe, respectively (Fig. 1). At the same time, the Curie temperature T_C for GdRuSi is 78.3 K, which is much lower than room temperature for practical application in household magnetic refrigerators. In this situation, successful doping can be a good way to increase T_C of these compounds. Also, T_C = 78.3 K and hence the maximum $-\Delta S_M(T_C)$ for GdRuSi is closer to the nitrogen liquefaction temperature of 77.4 K than T_C = 70 K for DyFeSi. Thus, GdRuSi is more effective for nitrogen liquefaction than DyFeSi. For the $GdMn_{1-x}Ru_xSi$ system, the MCE decreases with the appearance of ruthenium, and the MCE = 1.19 J/kgK is minimal for the intermediate composition x = 0.5 (Fig. 2). That is, independent magnetic ordering of the gadolinium and manganese subsystems is absent in the $GdMn_{1-x}Ru_xSi$ alloys, judging by the results of measuring the temperature dependences of magnetization and magnetocalorics.

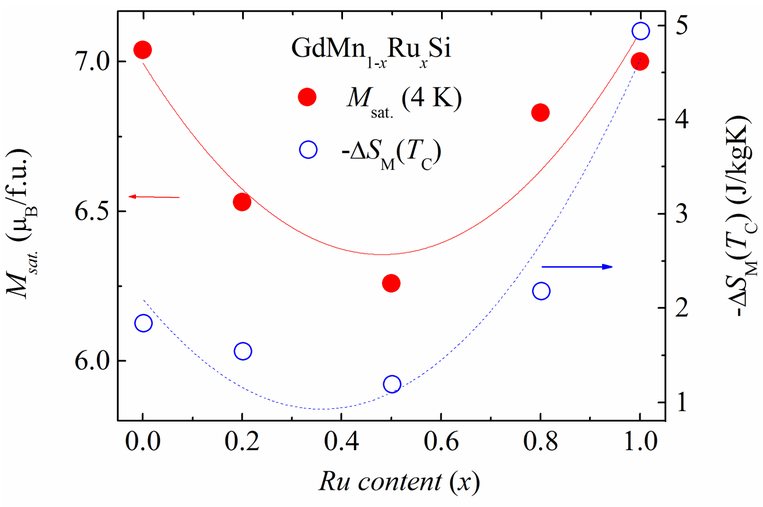


Fig. 3. Curie temperature (▲) and total DOS (□) at the Fermi level of the $GdFe_{1-x}Cr_xSi$ system versus Cr content.

The density of electron states of GdMnSi is distinguished by the presence of a pseudogap in one of the spin projections (Fig. 4). In the GdRuSi compound, more symmetrical states of Ru and Si are observed. Thus, in contrast to pure compounds with transition metals, which we studied earlier, the Fermi level passes through intense peaks. When doping this compound with Ru ions, a redistribution of states occurred, as a result of which there is no pseudogap in $GdMn_{0.5}Ru_{0.5}Si$ and the total density of states at the Fermi level has increased significantly. Partial and total magnetic moments were also calculated. Gadolinium ions have the highest magnetic moment of 7.13-7.23μ_B. Ru and Si ions are practically non-magnetic, and Mn ions have a large magnetic moment in all compounds and significantly reduce the total magnetic moment of the systems.

Electronic structure

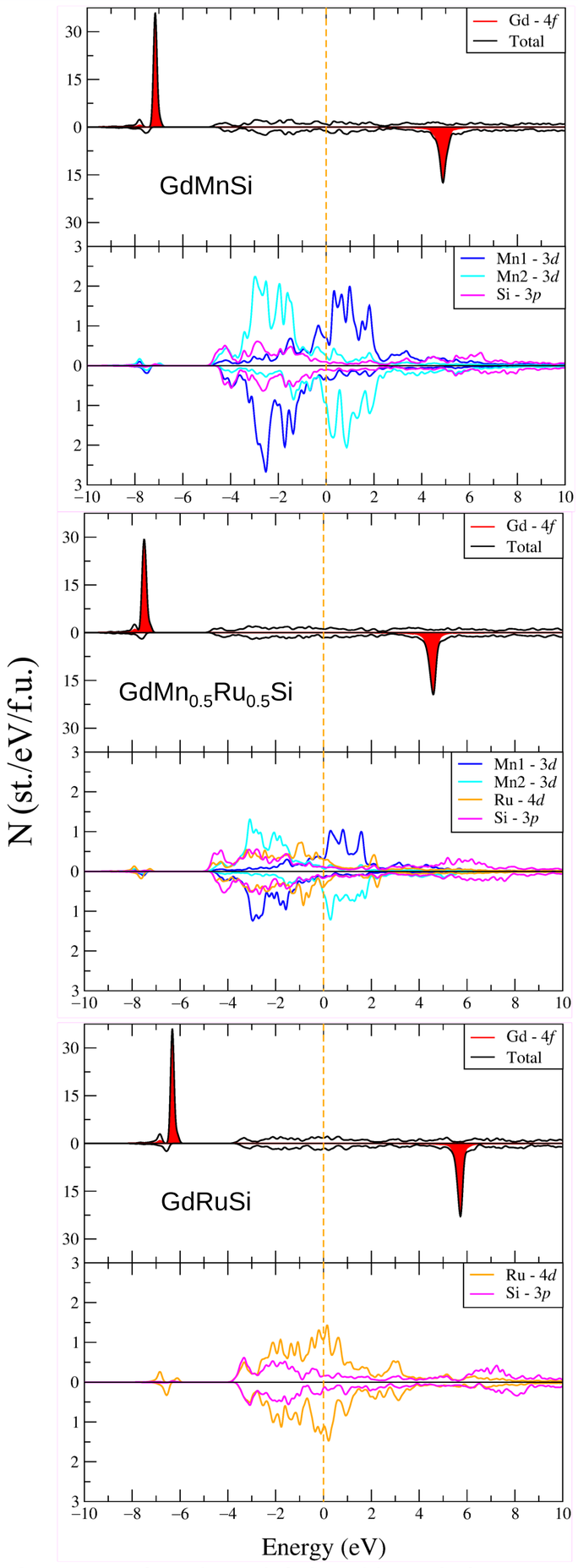


Fig. 4. Total and partial densities of electron states of N $GdMn_{1-x}Ru_xSi$. The Fermi energy E_F is shown by the dotted line at zero energy.

Conclusions

The obtained results indicate that doping of GdTMSi intermetallic compounds with transition metal ions leads to changes in the crystal lattice parameters, electronic structure, and magnetic properties of the compounds. Our calculations were carried out taking into account strong electronic correlations. The $GdMn_{1-x}Ru_xSi$ magnetic refrigeration compounds operate at temperatures from 320 K to 78.3 K, close to the nitrogen liquefaction temperature of 77.4 K. Therefore, the $GdMn_{1-x}Ru_xSi$ system, where x ranges from 0 to 1, could be of practical interest for nitrogen liquefaction, provided that these alloys are assembled into a cassette with a single large refrigerant capacity.

References an Acknowledgments

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S.P. Platonov, A.G. Kuchin, A.S. Volegov, V.S. Gaviko, R.D. Mukhachev, A.V. Lukoyanov, M.Y. Yakovleva, The $GdMn_{1-x}Ru_xSi$ compounds cassette for magnetocaloric nitrogen liquefaction. *Physica B: Condensed Matter* **685** (2024) 416060.

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