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# Negative Magnetisation and Absence of Superconductivity in $\text{RFe}_4\text{Al}_8$ ( $\text{R}=\text{Lu}, \text{Yb}$ ) Compounds

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We report researches for superconductivity and the effect of negative magnetisation in the  $\text{RFe}_4\text{Al}_8$  ( $\text{R} = \text{Lu}, \text{Yb}$ ) compounds by means of magnetic measurements and microwave absorption. Contrary to the earlier reports we do not confirm the existence of any traces of superconductivity in these compounds. Instead of the superconductivity and the Meissner effect, the  $\text{YbFe}_4\text{Al}_8$  compound exhibits the effect of negative magnetisation, whereas the  $\text{LuFe}_4\text{Al}_8$  compound shows an exact antiferromagnetic behaviour.

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## 1. Introduction

The intermetallic alloys of  $\text{RFe}_4\text{Al}_8$  family (where R is the rare earth) exhibits a very wide range of behaviours. The compounds containing nonmagnetic rare-earth ions like: Y, Lu, La, Ce are antiferromagnets with the transition temperatures changing from 100 K to 200 K [1]. If the rare earth is a magnetic ion (for example Yb) then the compounds exhibit a complex magnetic ordering with the antiferromagnetic interaction between the two magnetic sublattices.

Recently, we have shown [2] that this unusual ordering can lead to the effect of negative magnetisation. This effect was found also in the non-metallic systems [3–7]. In this way we have excluded the existence of superconductivity in the  $\text{YbFe}_4\text{Al}_8$  system in which, the negative moment originates from magnetic ordering of spins and is not related to the Meissner effect. The opened question is, however, the existence of superconductivity in  $\text{LuFe}_4\text{Al}_8$  system with nonmagnetic Lu ions. To clarify the doubts about the superconducting properties of the  $\text{LuFe}_4\text{Al}_8$  compound reported earlier [8], we have performed extended investigations using magnetic and microwave absorption measurements.

## 2. Experimental

To synthesise the metallic  $\text{YbFe}_4\text{Al}_8$  compound we have applied the technique of inductive melting of a stoichiometric mixture of the metallic powders in a water-cooled boat and under an argon atmosphere. The structure of the compounds was examined using X-ray diffraction.

The magnetic measurements were carried out by means of two types of magnetometers, namely the extraction magnetometer MagLab 2000 System (Oxford Instruments Ltd.) and SQUID magnetometer MPMS DC (Quantum Design). The measurements of the magnetically modulated microwave absorption (MMMA) were performed using a Radiopan SE/X X-band (9.4 GHz) EPR spectrometer.

## 3. Results and discussion

We have found no traces of superconductivity in  $\text{YbFe}_4\text{Al}_8$  and  $\text{LuFe}_4\text{Al}_8$  compounds using microwave absorption, which is a powerful method in the detection of a small amount of a superconducting phase (results not presented here). The open symbols in Figs. 1 and 2 present the field cooled magnetisation of the compounds vs. temperature,  $M(T)$ , in a low applied magnetic field (50 Oe). The magnetisation of the compounds increases as the temperature decreases, in agreement with the Curie-Weiss law. Below the Néel temperature of the Fe subsystem equal to  $T_N = 150$  K for the  $\text{YbFe}_4\text{Al}_8$  compound and  $T_N = 90$  K for  $\text{LuFe}_4\text{Al}_8$ , the increase becomes more rapid. For  $\text{YbFe}_4\text{Al}_8$  there is a maximum of magnetisation observed at the temperature of about 100 K. Below the maximum, the

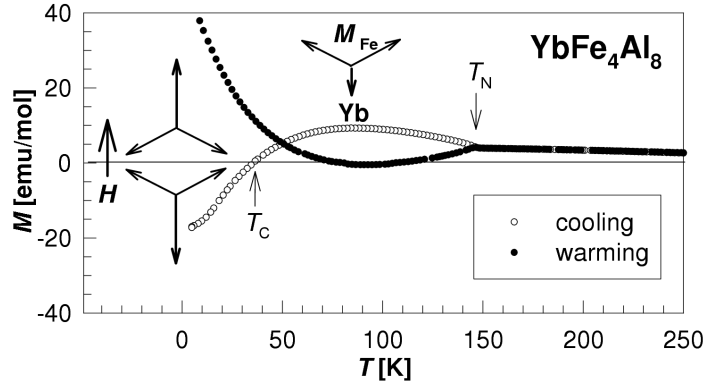


Fig. 1. Magnetisation versus temperature for the  $\text{YbFe}_4\text{Al}_8$  compound measured during warming and cooling at the applied magnetic field of 50 Oe. The  $M(T)$  curve for warming was obtained after applying a high magnetic field equal to 10 kOe for a few minutes, which reversed the configuration of spins. The arrows show the configurations of spins in different temperature ranges.  $T_N$  and  $T_c$  denote the Néel and the compensation temperature, respectively.

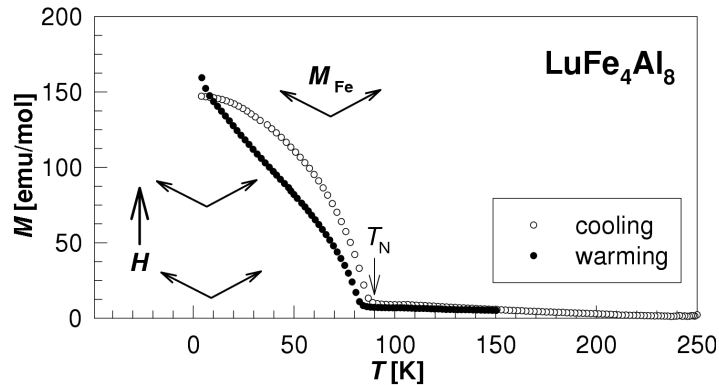


Fig. 2. Magnetisation versus temperature for the  $\text{LuFe}_4\text{Al}_8$  compound measured during warming and cooling using the same conditions and procedures like for the  $\text{YbFe}_4\text{Al}_8$  compound. This time the application of a high magnetic field does not change the configuration of spins.  $T_N$  is the Néel temperature.

magnetisation of the  $\text{YbFe}_4\text{Al}_8$  compound decreases and, for temperature lower than  $T_c = 34$  K, it changes a sign to negative. The magnetisation of the  $\text{LuFe}_4\text{Al}_8$  gradually increases and remains positive even at lowest temperatures.

The full symbols in Figs. 1 and 2 show the magnetisation of  $\text{YbFe}_4\text{Al}_8$  and  $\text{LuFe}_4\text{Al}_8$  compounds measured on warming at the same applied field of 50 Oe. This time, however, before measurements, a strong magnetic field was applied up to 10 kOe for a few minutes and at a low temperature of 4.2 K. In this way the previous, negative magnetisation of the  $\text{YbFe}_4\text{Al}_8$  system was reversed. On warming, the magnetisation decreases, and in a narrow temperature range once again becomes negative. There is not any known superconducting mechanism causing such behaviour. In the case of the  $\text{LuFe}_4\text{Al}_8$  magnetisation is always positive and the effect of negative magnetisation is absent in this compound.

This unusual behaviour we have explained earlier [2] using the model of Cooke et al. [9]. Namely, at the magnetic ordering temperature, a spontaneous ferromagnetic moment  $M_{\text{Fe}}$  due to the canting of Fe spins appears. The  $M_{\text{Fe}}$  moment dominates at high temperatures over the paramagnetic moment of Yb and is directed parallel to the applied magnetic field. The Yb moment is coupled antiparallel to the effective moment  $M_{\text{Fe}}$  and also to the applied magnetic field. As the temperature of the system decreases, the total Yb moment increases according the Curie law. Below the compensation temperature  $T_c$  the Yb component dominates over the constant component of  $M_{\text{Fe}}$  and the system starts to exhibit a negative magnetisation. In the case of  $\text{LuFe}_4\text{Al}_8$  compound below the antiferromagnetic transition there is only the effective moment  $M_{\text{Fe}}$  of the canted spins of iron. As the temperature decreases the Fe moments are better oriented along the applied magnetic field, causing an increase in the total magnetisation of the sample.

#### 4. Conclusions

In contrary to the previous report we found no evidence of superconductivity in the  $\text{YbFe}_4\text{Al}_8$  and  $\text{LuFe}_4\text{Al}_8$  compounds. The negative magnetisation, which occurs for  $\text{YbFe}_4\text{Al}_8$  at low temperature originates from antiferromagnetic interactions between the Yb rare-earth spins and the effective moment of the canted Fe spins. This mechanism is absent in the  $\text{LuFe}_4\text{Al}_8$ , in which only the antiferromagnetic behaviour due to Fe ordering is observed.

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#### References

- [1] H. Drulis, P. Gaczyński, W. Iwasieczko, W. Suski, B.Ya. Kotur, *Solid State Commun.* **123**, 391 (2002).
- [2] B. Andrzejewski, A. Kowalczyk, J.E. Frąckowiak, T. Toliński, A. Szlaferek, S. Pal, Ch. Simon, *Phys. Status Solidi B* **243**, 295 (2006).
- [3] G.J. Snyder, C.H. Booth, F. Bridges, R. Hiskes, S. DiCarolis, M.R. Beasley, T.H. Geballe, *Phys. Rev. B* **55**, 6453 (1997).
- [4] J. Hemberger, S. Lobina, H.-A. Krug von Nidda, N. Tristan, V.Yu. Ivanov, A.A. Mukhin, A.M. Balbashov, A. Loidl, *Phys. Rev. B* **70**, 024414 (2004).
- [5] K. Yoshii, A. Nakamura, Y. Ishii, Y. Morii, *J. Solid State Chem.* **162**, 84 (2001).
- [6] N. Menyuk, K. Dwight, D.G. Wickham, *Phys. Rev. Lett.* **4**, 119 (1960).
- [7] Y. Ren, T.T.M. Palstra, D.I. Khomski, A.A. Nugroho, A.A. Menowski, G.A. Sawatzky, *Phys. Rev. B* **62**, 6577 (2000).
- [8] A.M. Gurevich, N.M. Dmitriev, V.N. Eropkin, L.A. Ischchenko, N.N. Prentslau, L.V. Shylk, *Low Temp. Phys.* **25**, 10 (1999) (*Fiz. Nizk. Temp.* **25**, 15 (1999), in Russian).
- [9] A.H. Cooke, D.M. Martin, M.R. Wells, *J. Phys. C* **7**, 3133 (1974).