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MAGNETIC STRUCTURES AND SUPERCONDUCTIVITY IN $(\text{Sn}_{1-x}\text{Er}_x)(1)\text{Er}(2)_4\text{Rh}_6\text{Sn}_{18}$ WITH $x = 0, 0.42$ AND 0.61

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Abstract. – The superconducting and magnetic states exhibited by the title compounds are investigated by low temperature a.c. susceptibility and neutron diffraction measurements. The coherence length of the magnetic short range order is shown to be of drastic importance for the coexistence of both states.

The rare earth metal-rhodium-stannide series was discovered in 1980 [1], and it was found that the erbium compound can exhibit reentrant superconductivity. Further studies, including the determination of its crystal structure [2], have shown that the low temperature physical properties of this compound strongly depend on its chemical composition. The chemical formula of erbium rhodium stannide (hereafter denoted as Er-Rh-Sn) can be written as

The crystal structure can be described as formed by a three-dimensional network of corner sharing RhSn_6 trigonal prisms which generates sites occupied by the $\text{Er}(2)$ atoms 10-coordinated by $\text{Sn}(2)$, and cuboctahedral sites occupied by a mixing of $\text{Er}(1)$ and $\text{Sn}(1)$ atoms. The chemical composition, and consequently the low temperature properties of Er-Rh-Sn can be modified by changing the substitution ratio x on this site. For instance, when $x \approx 0.6$, this compound undergoes a magnetic transition, when $x \approx 0.4$, it is a reentrant superconductor, and when $x \approx 0$ the superconducting state is not destroyed by a magnetic ordering [3]. We have undertaken a systematic study of the evolution with x of the low temperature behaviour of Er-Rh-Sn by a.c. susceptibility and single crystal neutron diffraction measurements.

As the single crystals (≈ 50 mg) are grown in a tin flux, it is not possible to control very accurately the x value, which has to be determined *a posteriori*. Thus, for each crystal studied, we have cut a small part of it, ground it to a sphere of radius ≈ 0.1 mm and measured on a CAD4 diffractometer a convenient set of reflexions most sensitive to the x value and not affected by twinning (because belonging to the cubic supercell).

Refinement of only x , with all other structural parameters fixed to the values previously found by complete structural determination, allows a relatively fast and accurate determination of its value. The x value found by this method and by complete structure refinement of the same sample are in good agreement. The a.c. susceptibility of these crystals was measured in a dilution cryostat with low applied fields ranging from 3 mOe to 0.3 Oe. The results are summarized in figure 1. At constant applied field, T_m increases and T_c

Fig. 1. – Low temperature a.c. susceptibility curves with different applied fields, for three single crystals with $x = 0, 0.42$ and 0.57 . Inset: "Phase diagram" showing the evolution of the superconducting and magnetic transition temperatures versus x and the a.c. applied field.

decreases with increasing x , up to the disappearance of the superconducting state for $x = 0.6$. For a given x , T_m increases and T_c decreases with increasing applied field. The $x = 0$ sample is particularly interesting, since the superconducting state appearing at 1.32 K for a 30 mOe applied field, is not destroyed by the presence of a magnetic transition at 0.34 K. This magnetic transition is very sensitive to the applied field, but the sample susceptibility remains always diamagnetic in the applied field range investigated. Magnetization and resistivity measurements have shown that a ferromagnetic state indeed exists in this sample, with zero resistivity [4]. Thus, the magnetic ordering and the superconducting states coexist for all temperatures below $T_m = 0.34$ K.

The magnetic structures of the $x = 0, 0.42$ and 0.61 compounds were investigated by single crystal neutron diffraction on the D15 instrument at the I.L.L. equipped with a dilution cryostat. The solution of the magnetic structure for $x = 0.42$ has been previously described [5]. The same procedure was applied to the two other samples: due to the small number of magnetic reflexions, symmetry constraints deduced from group representation theory were applied to the erbium moments. The symmetry constraints which gave the best agreement for the $x = 0.42$ compound were also applied for the refinements of the $x = 0$ and 0.61 ones: the Er(1) moments are parallel to the c axis, while the Er(2) moments are not colinear but have a resultant parallel to the c axis. For the three samples, it appears that the Er(2) moments are always oriented along (or almost along) the direction of the pseudo-threefold axes of the Er(2) sites, and that the magnetic resultant of the Er(2) and Er(1) sublattices are opposite in direction. The values of the total "ferromagnetic" resultants are 32(4), 14(5) and 34(3) μ_B per unit cell for the $x = 0, 0.42$ and 0.61 samples, respectively (the unit cell contains between 32 and 37 Er atoms). The relatively high value obtained for the $x = 0$ sample is due to the fact that the resultant of the Er(2) sublattice is not partly compensated by that of the Er(1) sublattice, as it is the case for the two other samples. For the three samples, the magnetic contributions appeared significantly broader than the nuclear peaks. This broadening was proved to be due to short range magnetic ordering by observation of magnetic diffuse streaks on low temperature white beam neutron diffraction photographs in Laue geometry [6]. Magnetic contributions have been obtained by subtraction of the low and high temperature profiles measured in the D15 experiments for the three samples. It is obvious that the width of the reflexions decreases with increasing x , and that, for the $x = 0.61$ sample two peaks of different width contribute to the total magnetic scattering, probably due to the two different Er sublattices (Fig. 2). From the analysis of these line shapes, qualitative correlation lengths for the magnetic ordering can be obtained,

Fig. 2. — Magnetic line profiles showing the width of the magnetic contributions for the $x = 0.61$ sample. Two Gaussians of different widths must be introduced to fit the experimental points.

namely ≈ 40 Å for the $x = 0$ compound and ≈ 60 Å for the $x = 0.42$ compound and the short range component of the $x = 0.61$ compound, while the second component is quasi long range, since the line width in this case is almost that of the nuclear peak. These results indicate that the correlation length of the magnetic order may be the relevant parameter to understand the evolution of the low temperature physical properties of the Er-Rh-Sn compound. For $x \approx 0$, the magnetic correlation length (mcl) would be too small for the superconducting state to be destroyed, because its correlation length is much longer (≈ 1000 Å). When x increases, mcl increases and the superconducting state could not coexist with the magnetic one. On further increase of x , a long range magnetic order is established in the sample, which would prevent the existence of a superconducting state.

The recent observation of a spin glass behaviour in the Ho-Rh-Sn isotypic compound by low temperature dynamic susceptibility measurements [7] could be related to our results with the Er-Rh-Sn compound. We have carried out a low temperature neutron diffraction experiment on a Ho-Rh-Sn compound. The preliminary results indicate that its magnetic structure is very similar to that of Er-Rh-Sn, and also involves short range magnetic ordering. The comparison of the data obtained for both compounds could help to understand the interactions between magnetism and superconductivity in this class of compounds.

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