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HYPERFINE INTERACTIONS AND SPIN CORRELATIONS IN ErT_2Sn_2 (T = Ni, Cu) PHASES*

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The present work reports on the electrical resistivity measurements and ^{119}Sn Mößbauer effect investigations of the hyperfine interactions in novel ternary compounds $ErNi_2Sn_2$ and $ErCu_2Sn_2$. The attention was focused on the temperature region of the magnetic ordering transitions, i.e. at $T_N = 5$ K and 3.5 K, respectively. Er^{3+} spin correlations in $ErNi_2Sn_2$ are discussed in terms of their influence on resonant γ-absorption.

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1. Introduction

A wide variety of electronic and magnetic properties (some of them very particular) of the ternary intermetallic phases of a general formula RET_2X_2 (RE — rare earth or actinide element, T — *d*-transition metal and X — group IIIA through VIA *p*-element) along with the relative stability of their tetragonal structure have made those compounds to an interesting field for systematic studies of the *f*-electronic systems (e.g. [1]). Until recently systematic studies were concentrated predominantly on families of compounds with silicon and germanium as an X-element. Only recently the interest was extended onto physical properties of tin containing phases [2–4]. Nevertheless, the research efforts have been practically confined to compounds incorporating cerium as the RE-constituent as they were found to be an intriguing playground for competing of a RKKY long-range magnetic interaction and a formation of a nonmagnetic singlet ground state [5, 6]. The present work is concerned about novel members of the family of the ternary tetragonal compounds with erbium: $ErNi_2Sn_2$ and $ErCu_2Sn_2$. Magnetic susceptibilities

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of these compounds reveal regular Curie–Weiss behaviour in the temperature range 15 K to 250 K with the values of the effective magnetic moment hardly different from that of a free trivalent erbium ion [7]. It is not likely that erbium f-electrons significantly hybridize with conduction electrons and therefore these alloys may be regarded as a kind of reference (with well-localized and energetically distant from the Fermi surface $4f$-electrons) to the above-mentioned Kondo-lattice systems. A study of critical behaviour of the former compounds is of relevance to the problem whether large electron mass or critical fluctuations predominantly account for the observed anomalies of physical properties at the transition temperature in Ce-phases [5].

2. Experimental details and results

Polycrystalline samples of $ErNi_2Sn_2$ and $ErCu_2Sn_2$ compounds were prepared by the arc-melting process in an argon atmosphere. By means of the X-ray diffraction the phases were found to crystallize with the $CaBe_2Ge_2$-type of structure (space group $P4/nmm$). The samples are not free from certain degree of disorder connected with the occupation of $3d$-transition metal and tin sublattices. This may eventually lead to the occurrence of regions of the body-centred lattice ($ThCr_2Si_2$-type) rather than that of the above-mentioned primitive one. Thermal processing was not effective in shifting the equilibrium either way. The conventional four-point contact method was used for the investigation of the electrical resistivity. A drop in the temperature dependence of electrical resistance is indicative of a long range magnetic ordering which sets on in these compounds at temperatures (T_N) of 5 K ($ErNi_2Sn_2$) and 3.5 K ($ErCu_2Sn_2$). The resistivity of $ErNi_2Sn_2$ attains a local minimum under cooling at about $T_m = 8$ K followed by the resistance increase and subsequently falls down upon further cooling of the sample (Fig. 1). Further anomaly at about 2.5 K may result from a change in

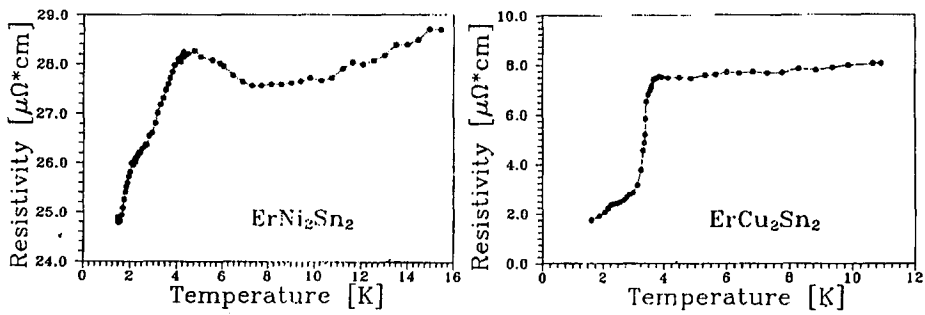

Fig. 1. Low temperature electrical resistivities of $ErNi_2Sn_2$ and $ErCu_2Sn_2$.

a magnetic structure. Taking advantage of a presence of a convenient Mößbauer isotope as a regular constituent in these compounds the resonance γ-absorption experiments with 23.8 keV radiation of ^{119}Sn have been performed down to 1.6 K. At higher temperatures ($T \gg T_N$) spectra of both compounds show a similar character with two components in agreement with the presence of tin atoms in two

crystallographically distinct sites [8]. Each of the components presents a doublet which results from the coupling of ^{119}Sn nuclear quadrupole moment with an electric field gradient originating from the distribution of charges in the crystal lattice around a given nuclear site. Energy of this interaction is further quoted in terms of a quadrupole coupling constant defined as $\mathcal{E}_Q = (1/4)e^2Qq$. The position of an individual doublet (i.e. an isomer shift IS) which is determined by electronic density at the nucleus is given relative to the $BaSnO_3$-source. At 78 K, for example, the components in the Mößbauer spectrum of $ErNi_2Sn_2$ are characterized by $\mathcal{E}_Q1 = 0.48(1)$ mm/s centred at IS1 = 1.67(2) mm/s and $\mathcal{E}_Q2 = 0.60(1)$ mm/s at IS2 = 2.04(1) mm/s, and in the case of $ErCu_2Sn_2$: $\mathcal{E}_Q1 = 0.408(5)$ mm/s at IS1 = 1.73(1) mm/s and $\mathcal{E}_Q2 = 0.37(1)$ mm/s at IS2 = 2.09(2) mm/s. In the magnetically ordered state the overall shape of $ErNi_2Sn_2$ spectrum does not change substantially while in the case of copper compound one of the spectral components splits further into two patterns. The different values of an angle between the main axis of the electric field gradient (originating from the crystal lattice) and the magnetic hyperfine field for the two above-mentioned sub-components indicate more complex, probably non-colinear spin arrangement in $ErCu_2Sn_2$. Moreover, although the Néel temperature of $ErCu_2Sn_2$ is distinctly lower than that of $ErNi_2Sn_2$ values of *transferred* hyperfine fields induced at tin nuclei (only one of the two crystallographic tin sites in $EuCu_2Sn_2$ experiences hyperfine field and not of unique value: the splitting into two sub-components is observed, cf. above) are much larger for the former compound (40(1) kOe and 24(1) kOe when extrapolated to $T = 0$) than any of the fields observed at saturation in nickel compound (the larger is 5.3(5) kOe, Fig. 2c).

3. Discussion

The use of the Mößbauer effect in the study of magnetic correlations at the critical region has two clear advantages. First, it measures locally on an atomic scale and hence it makes magnetization of an antiferromagnet an accessible quantity. Secondly, it requires no applied magnetic field which usually strongly influences the magnetization (and susceptibility). In a critical region immediately above the Néel temperature short range order effects are disclose. The degree of a directional coherence of erbium magnetic moments as a function of temperature T is probed by the hyperfine field induced at sites of tin nuclei, i.e. we assume $H_{\mathrm{hf}}(T) \propto \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle$. A two-spin correlation function $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle = \sum \boldsymbol{S}_i \cdot \boldsymbol{S}_j \exp(-\beta \mathcal{H}_{\mathrm{eff}}) / \sum \exp(-\beta \mathcal{H}_{\mathrm{eff}})$ was considered for the case it measures a degree of directional ordering of nearest-neighbour spins $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$ within the model of a two-sublattice antiferromagnet. The only interaction the two spins experience is that of Heisenberg type: $\mathcal{H}_{\mathrm{eff}} = -2J\boldsymbol{S}_1 \cdot \boldsymbol{S}_2$, while a field due to all other neighbours vanishes in a paramagnetic state. The notion of an effective 1/2-spin was further used under the assumption that at low temperatures of interest only the lowest crystal-field doublet of Er^{3+} ($^4I_{15/2}$) is eventually involved. Erbium ions are located at the positions 2(c) of the site symmetry-group $4mm$. Finally, with

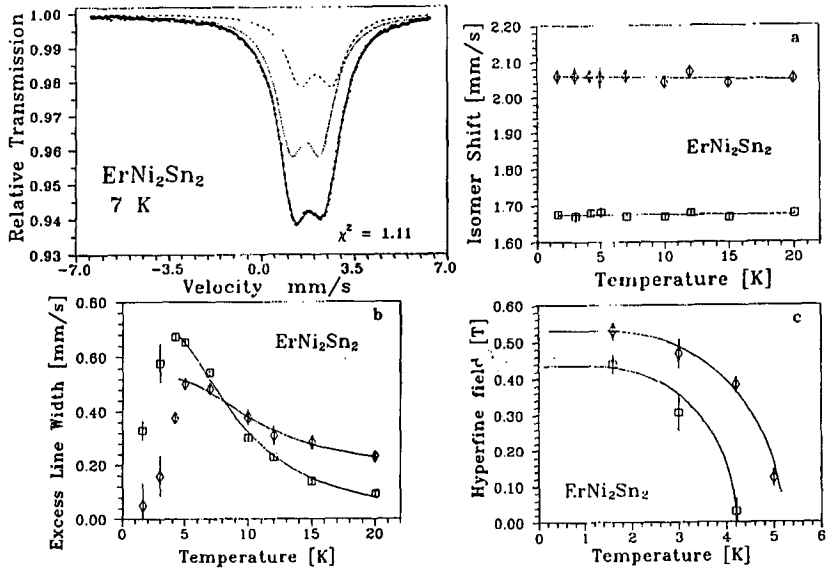

Fig. 2. ^{119}Sn Mößbauer spectrum of $ErNi_2Sn_2$ at 7 K. Temperature dependence of the nuclear hyperfine parameters of ^{119}Sn in $ErNi_2Sn_2$: (a) isomer shifts of the two components (relative to $BaSnO_3$-source), (b) excess line width $\Delta\Gamma$, (c) transferred hyperfine field vs. temperature below T_N.

the eigenvalues of Hamiltonian $\mathcal{H}_{\text{eff}}$ the two-spin correlation function is

$$\langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \rangle = -\frac{1}{4} \frac{\cosh(-\beta J) + 2\sinh(-\beta J) - \exp(\beta J)}{\cosh(-\beta J) + \exp(\beta J)},$$

where $\beta = 1/k_B T$. A process of random, isotropic reorientation (with a characteristic time τ) of the hyperfine field H_{hf} at the tin nucleus constitutes a physical background for the observed broadening $\Delta\Gamma$ of the recoilfree resonance line of the γ-absorption at temperatures close above T_N. We have adopted a simple approach in which in fast relaxation limit individual hyperfine lines preserve their lorentzian shape while an excess line width is proportional to a certain function of the relaxation time. Thus $\Delta\Gamma = Af(\tau)$, where the factor A depends on the square of the hyperfine field. In view of a number of relevant theories of the Mößbauer spectrum shape in a presence of dynamical processes (e.g. [9, 10]), the function $f(\tau)$ is linear in τ but only in a certain range of relaxation times. We have found a very weak temperature dependence of $f(\tau)$ in a region studied. Therefore while evaluating $\Delta\Gamma$ vs. T data $f(\tau)$-function was considered as a constant (cf. below). In other words the temperature variation of $\Delta\Gamma$ is mainly due to the changing value of the correlation function rather than to an evolving degree of averaging out. The fluctuation time of the Er^{3+} spin $\boldsymbol{S}$ ($< 10^{-11}$ s) is substantially shorter than the characteristic precession time of ^{119}Sn nuclear moment ($\tau_L < 10^{-7}$ s) in the effective hyperfine field $\boldsymbol{H}_{\text{hf}}$ at its site and implies that the latter quantity attains its average value which is proportional to $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle$. It involves also a spatial averaging which has to be understood as taken over a whole cluster of dimensions character-

ized by the correlation length and whose lifetime (τ_{cluster}) is definitely *longer* than the Larmor period τ_{L}. Such a long-lived (τ_{cluster}) domain gives raise to $\boldsymbol{H}_{\text{eff}}$ which is randomly oriented with respect to the local electric field gradient at ^{119}Sn. As a net effect the spectrum does not loose its symmetrical form. The smallness of the *transferred* hyperfine field $\boldsymbol{H}_{\text{hf}}$ results in a mere broadening of the absorption spectrum. The careful and consistent analysis of the data (cf. the behaviour of isomer shift vs. T in Fig. 2a) allowed one to deduce the temperature dependence of the excess spectrum widening for both components connected, respectively, with the two lattice sites of tin found in this primitive-tetragonal structure (Fig. 2b). The least-squares fit of $\Delta\Gamma \propto \langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \rangle^2$ to these data (solid line in Fig. 2b) gave the values of the *effective* erbium–erbium exchange integral of $-0.96(5)$ meV and $-1.15(11)$ meV for the two tin-positions. The difference in values reflects the *effective* character of the fitted quantities (J) and stems from a distinction in local environments of the two tin-sites. Below T_{N} the spectra were described utilizing the concept of the hyperfine Hamiltonian $\mathcal{H}_{\text{hf}}$ with the hyperfine field H_{hf} directly rather than merely considering the effect of a line broadening. $\mathcal{H}_{\text{hf}}$ gives line positions (and intensities) in the hyperfine pattern. While both magnetic dipole and electric quadrupole nuclear interactions are present, the asymmetrical spectrum shape may appear. The temperature variation below T_{N} of the *transferred* hyperfine field is shown in Fig. 2c.

4. Conclusions

Electrical resistivity measurements indicate the occurrence of a transition to the magnetically ordered state at 5 K and 3.5 K for $ErNi_2Sn_2$ and $ErCu_2Sn_2$, respectively. The observation of the hyperfine interactions of tin nuclei with the ^{119}Sn Mößbauer effect in the vicinity of the transition region allows us to conclude the occurrence of considerable Er^{3+} spin correlations to temperatures much higher than magnetic critical points as inferred from bulk measurements. The characteristic fluctuation times are too short ($\tau < 10^{-11}$ s) to influence the line shape. Nevertheless, the evolution of a short range magnetic order accounted for by the two-spin correlation function may be easily followed by observation of the temperature dependent γ-absorption line broadening.

Acknowledgments

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