# THERMOPOWER IN RARE EARTH INTERMETALLIC COMPOUNDS AND THE VALIDITY OF s-f SCATTERING MODELS (*)

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*ABSTRACT* — The s-f scattering models of Kasuya and Abel'skii — Irkhin are tested against experimental data on the thermopower of several rare-earth intermetallic compounds (Gd Zn, Gd Cd, Tb Zn). Good agreement is obtained for Gd Zn but not for Tb Zn; for Gd Cd the present data are inconclusive.

The s-f interaction of strength G between conduction electrons (spin $\vec{s}$) and localized ionic spins ($\vec{S}_i$) in rare earth systems,

$$G \; \delta(\vec{r}-\vec{R}_i) \; \vec{s}\,.\,\vec{S}_i \tag{1}$$

changes sign for conduction electrons with spin up or down, making the corresponding relaxation times different when a net magnetic polarization $\vec{M}$ exists in the system ($T < T_c$; $T_c$ = Curie point). Pronounced anomalies then arise in the thermopower Q, as was first shown by Kasuya [1]. For temperatures not too low compared to $T_c$, Kasuya expression can be written:

$$\begin{aligned} Q(T) = & (k^2 \pi^2 / 3 e E_F) T + \\ & + (2k(g-1)\zeta / e E_F)\, G \langle J_z \rangle \cdot (1 - e^{-x})/(1 + e^{-x}) \quad , \end{aligned} \tag{2}$$

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$<J_z>$ being the z-axis expectation value of the total angular momentum $\vec{J}$, $g$ the Landé factor, $x = 3[J(J+1)]^{-1}.(T_c/T).<J_z>$, $\zeta$ the fraction of magnetic ions in the system, $E_F$ the Fermi level, and k the Boltzmann constant. The first term in $Q(T)$ is the normal diffusion electronic contribution, and the second term characterizes the magnetic anomaly, with a magnitude controlled by the parameter $(G/E_F)$.

The shape of the anomaly depends on the sign of G, which rises (or lowers) the energy of each electron group (↑ or ↓), making electrons to flow preferentially to one (or the other) of the thermoelectric junctions. The thermopower measurements can be used to obtain the sign of G, in contrast to the case of the electrical resistivity ρ [1] which depends on the square of G.

Fig. 1 summarizes the different types of Q-anomalies expected in ferromagnetic systems near $T_c$, according to the possible signs of e and G:

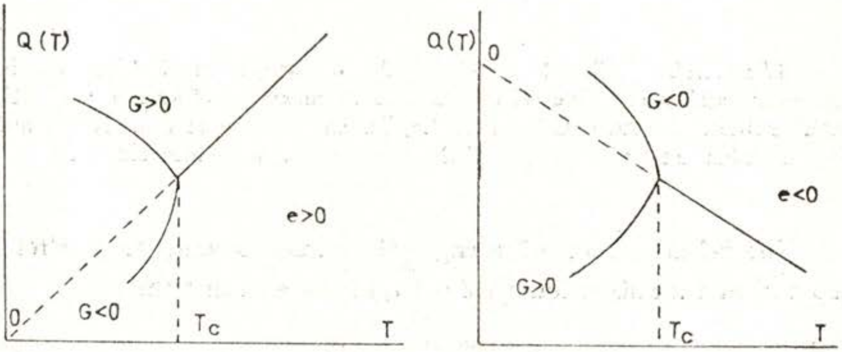


Fig. 1 — Different types of thermoelectric power (Q) anomalies expected in ferromagnetic systems near $T_c$, according to the possible signs of *e* and *G*.

Kasuya model has been improved by Abel'skii-Irkhin [2], through inclusion of electron-phonon scattering and more accurate calculations. The following mean-field expression for Q(T) near or above $T_c$ is obtained:

$$Q(T) = A_1 T . F_1(t, z; J, \gamma) + A_2 z . F_2(t, z; J, \gamma) \qquad (3)$$

where $A_1 = \text{const}(kT/E_F)$, $A_2 = \text{const}(G/E_F)\zeta J$, $t = T/T_c$, $z = M(T)/M(0)$ and $F_1$, $F_2$ are simple functions of t, z, J, γ, with $\gamma = (\rho_s/\beta T_c)[J(J+1)]^{-1}$, where $\rho_s$, $\beta$ T are the total spin and phonon resistivities, respectively.

The second term in (3), which vanishes above $T_c$, becomes the dominant magnetic contribution below $T_c$. The first term is a diffusion-like contribution which entirely determines $Q(T)$ in the paramagnetic phase.

Expressions (2) and (3) describe reasonably well the shape of $Q(T)$ in ferromagnets, but quantitative tests in rare earth systems are rather scarce, and practically limited to the case of Gd [1,3]. For this reason we report now on a detailed investigation of the validity of the s-f model to describe the Q-anomalies in rare earth ferromagnets. To avoid the complexities of uniaxial crystals, we have chosen a series of intermetallic compounds with the simple CsCl structure, of the type RM, where R = heavy rare earth, M = Zn, Cd. Here we select three of these compounds (GdZn, TbZn, GdCd), one presenting a very *large* Q-anomaly (TbZn), the other a *moderate* one (GdZn) and the last one presenting only a *small* Q-anomaly near $T_c$ (GdCd).

Fig. 2 shows $Q(T)$ for TbZn and GdZn:

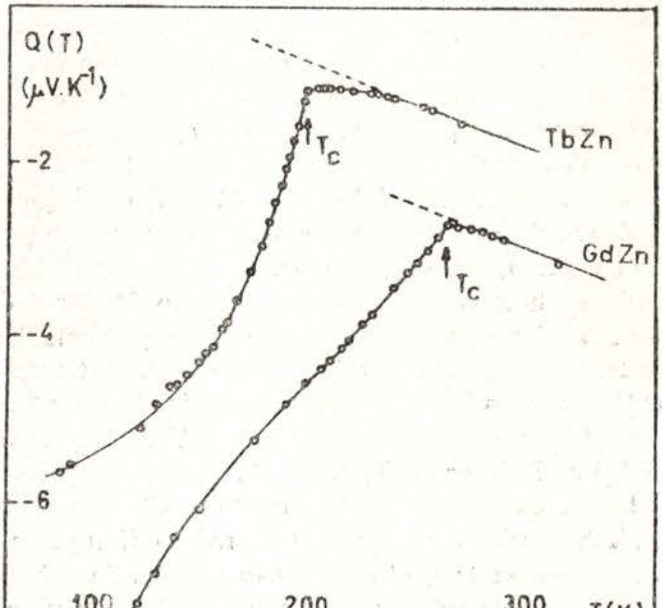


Fig. 2 — Temperature dependence of thermoelectric power (Q (T)) for TbZn and GdZn.

In the paramagnetic phase Q has a negative slope ($e<0$), approaching gradually a quasi-linear behaviour (a small quadratic term is present), as expected from (3) in terms of a diffusion-like electronic contribution to Q:

$$Q_{para} = (k^2 \pi^2 / 3 e E_F) T (3/2 + r_1 \beta T / \rho + r_2 \rho_s / \rho) \qquad (4)$$

where $r_1$, $r_2$ characterize the energy dependence of the electron relaxation times for phonon and spin scattering $\tau \propto E^{r_1}, E^{r_2}$ respectively. Of course, the experimental curve bends progressively as T approaches $T_c$ from above, due to the increasing role of the critical fluctuations, which are not accounted for in the above (mean-field) models.

For numerical calculations we use the standard values $r_1=3/2$, $r_2=-1/2$; $J=7/2$, $g=2$ (Gd); $J=6$, $g=3/2$ (Tb); $\zeta=1/2$. From accurate electrical resistivity studies we obtained $\beta=0.136\ \mu\Omega$. cm. $K^{-1}$, $\rho_s=52.5\ \mu\Omega$. cm, $T_c=265$ K for GdZn, and $\beta=0.119\ \mu\Omega$. cm $K^{-1}$, $\rho_s=36.3\ \mu\Omega$ cm, $T_c=199.6$ K for TbZn. [4].

From eq(4) we then obtain $E_F=4.4$, $6.9$ eV for GdZn and TbZn respectively. Since Q gets more negative below $T_c$ (and $e<0$, we also conclude that G is positive both in TbZn and GdZn.

We are now in a position to test eq. (3) in the ferromagnetic phase. For GdZn, a good fit to our experimental data can be obtained if one assumes $G=0.18$ eV, as shown in Fig. 3 (heavy line; the broken line shows the paramagnetic-like line, as given by eq. (2).

The G value obtained appears quite reasonable and fairly close to the quoted values for pure Gd (0.17 – 0.20 eV) [5]. It gives a ratio $G/E_F \sim 0.04$, which is of the same order of magnitude as previously obtained for Gd from Q(T) measurements [3].

For TbZn, however, it is not possible to fit our Q(T) data using 'reasonable' values for G ($\sim 10^{-1}$ eV); they would lead to a much smaller anomaly than found experimentally. Within the assumption that Q-anomalies below $T_c$ are entirely due to the s-f exchange interactions [1,2] one would require $G \sim 7$eV to match the steep decrease of Q in TbZn below $T_c$ ($G/E_F \sim 1$).

An alternative approach is, of course, to admit the possible dominance of factors which are not considered in Kasuya and Abel'skii-Irkhin models. In fact, other mechanisms besides phonon and normal spin scattering might be operative in TbZn, causing $Q_{exp}$ to decrease more sharply below $T_c$. An interesting case is the possible effect of quadrupole ordering below $T_c$ [6] and of the tetragonal lattice distorsion which arises at the onset of the ferromagnetic phase in TbZn [7]. Both features are absent in GdZn, which might justify the good agreement between experiment and theory found in this compound. Effects due to the possible non-s character of the conduction band in TbZn should also be carefully investigated.

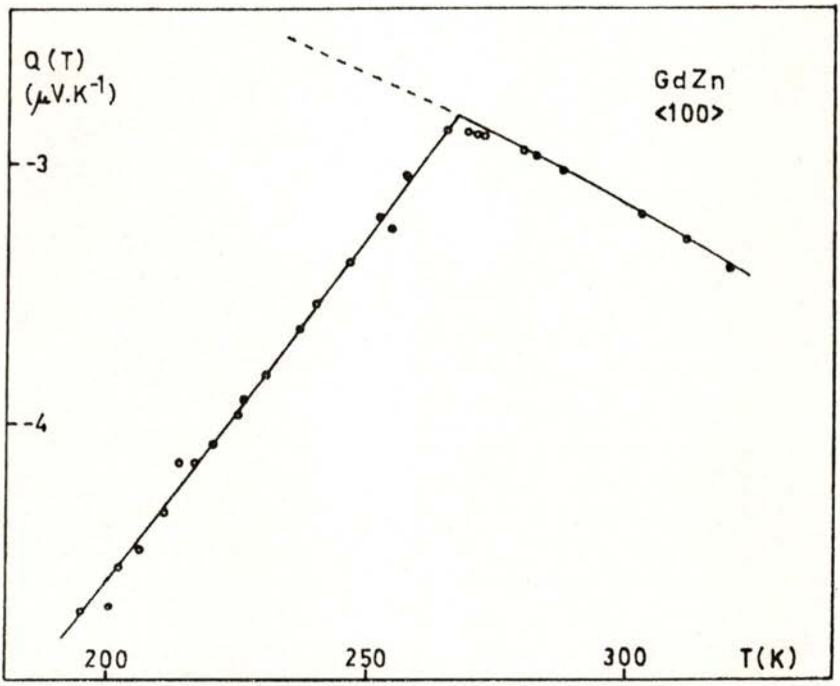


Fig. 3 — Fitting of the experimental data of thermoelectric power (Q) for GdZn $< 100 >$ to equation (2), assuming $G = 0.18$ eV.

Finally we show in Fig. 4 the temperature dependence of the thermoelectric power for GdCd.

The anomaly is now much smaller than in previous samples, although with the same qualitative features as in GdZn and TbZn, since $dQ/dT < 0$ in the paramagnetic state, and below $T_c$ we have Q depressed with respect to the linear extrapolation from the paramagnetic state [8]. However, due to the smallness of the anomaly in this sample and the existence of a second transition below $T_c$ [8], work is still in progress for the detailed analysis in this sample.

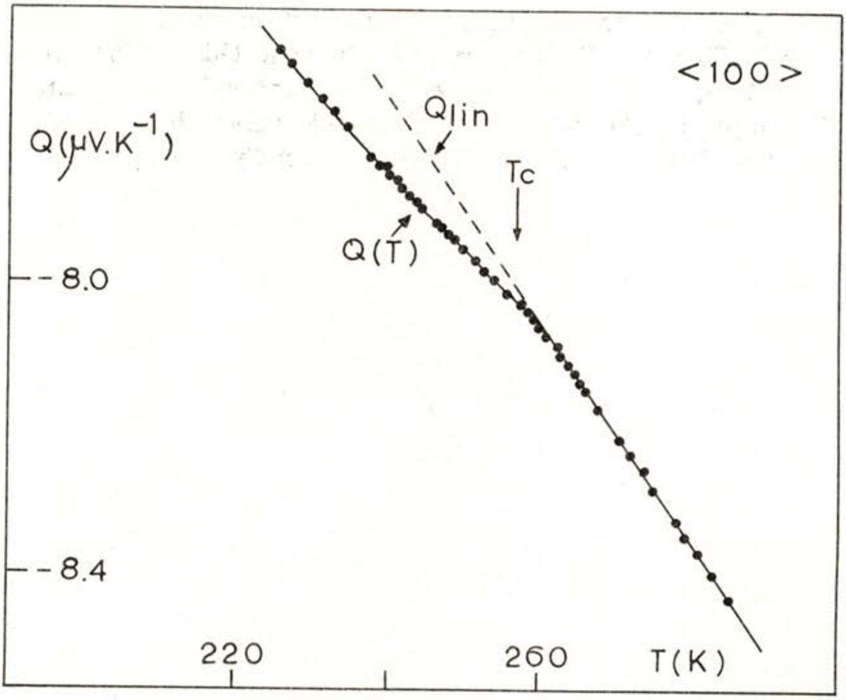


Fig. 4 — Temperature dependence of thermoelectric power Q (T) for GdCd near $T_c$.

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