Thermal emf and topological electronic transition in the $Mo_{1-x}Re_x$ system

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A giant local anomaly has been found in the thermal emf of the $Mo_{1-x}Re_x$ system at $x = 0.11$. This anomaly is evidence of a topological electronic transition in this system. The temperature dependence of the amplitude of this anomaly has been determined.

It has recently been established that the change in the structure of the Fermi surface of a metal which occurs as an impurity concentration is varied or as pressure is applied—a topological electronic transition—is accompanied by a giant anomaly in the thermal emf (Refs. 1–3 and the bibliographies there). We thus have a new opportunity for studying the electronic structure of a metal near the Fermi surface.

For the present study we selected molybdenum, a metal with a bcc lattice whose electronic structure has been studied in detail (see the review by Kaganov *et al.*[4]). As the impurity we used rhenium, which forms a substitutional solid solution in Mo at atomic concentrations up to $x \approx 0.3$. The lattice constant decreases negligibly (by $\sim 0.5\%$ at $x = 0.25$) as the rhenium is added.

Each Re atom contributes an extra conduction electron. This event is evidently accompanied by an increase in the Fermi energy ϵ_F:

$$\Delta\epsilon_F = x\nu^{-1}, \tag{1}$$

where $\nu = 0.8$–1.0 states/(atom·eV) (Ref. 4) is the density of electron states at the Fermi level. The atomic weights of Mo and Re differ by a factor of two, and the Debye temperature decreases from 470 K for pure Mo to 350 K in the alloy $Mo_{0.7}Re_{0.3}$. The temperature of the superconducting transition, T_c, in the system $Mo_{1-x}Re_x$ varies from 0.92 to 11 K as x is increased from 0 to 0.3, so that the rhenium concentration in a sample can be determined by measuring T_c.

The samples were cut from single crystals grown by vacuum smelting of bars of a mixture of ultrapure molybdenum and rhenium. The composition of the samples was determined from their specific weight and by activation analysis and corrected on the basis of the dependence of T_c and RRR of the samples on the Re concentration found previously. Analysis of the results of the determination of x by the various methods showed that the possible systematic error in the determination does not exceed $\sim 0.1x$ anywhere in the range measured. The width of the superconducting transition is 0.04–0.2 K for the overwhelming majority of the samples, implying that they are homogeneous in composition.

Experimentally, in the interval from T_c to 11 K, the resistivity, the thermal

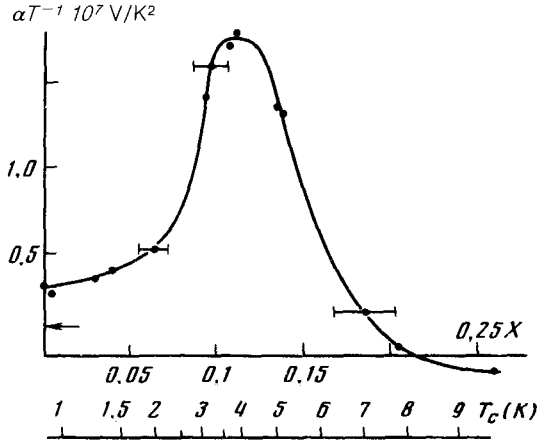

FIG. 1. Thermal emf of the $Mo_{1-x}Re_x$ system versus the Re concentration, x (for increasing T_c of the samples). The arrow shows the value of $\alpha_0 T^{-1}$ for pure Mo at room temperature.[8]

conductivity, and the thermal emf α are determined by a comparison method with SQUIDs as null detectors (Ref. 6, for example).

For molybdenum ($RRR = 2\times10^3$, the resistivity is described by $\rho = \rho_0 + 1.39\times10^{-12}\,T^2$, where the second term is apparently due to electron-electron scattering. The thermal emf of all the samples is determined by the diffusion component. The value of αT^{-1} varies only insignificantly with the temperature. For pure molybdenum, the decrease in αT^{-1} is $\sim 15\%$, while that for $Mo_{0.9}Re_{0.1}$ is $\sim 3\%$. The clearly defined local maximum in the dependence $\alpha T^{-1}(x)$ at $x = 0.11$ ($T_c = 3.65$ K), where αt^{-1} increases by a factor of nearly ten (Fig. 1), is evidently evidence of a topological electronic transition in the system $Mo_{1-x}Re_x$. Using (1) and $x = 0.11$ and $\nu = 0.9$ (in order of magnitude), we find that the transition results from an increase in ϵ_F of the molybdenum by $\simeq 0.12$ eV.

With what sort of change in the structure of the Fermi surface could we relate this transition? An increase in ϵ_F should evidently be accompanied by a decrease in the hole elements of the Fermi surface. But an increase in ϵ_F by 0.12 eV would lead, according to the data of Ref. 7, to a decrease in the cross section of the hole ellipsoids—the smallest hole parts of the Fermi surface—of only $\sim 12\%$. The transition clearly results from a different change in the structure of the Fermi surface. An increase in ϵ_F might give rise to new electron parts of the Fermi surface if the minimum of some unfilled electron part lay close to ϵ_F. This is precisely the situation which we have in molybdenum, where, according to the band structure found by calculations, the closest minimum of the unfilled electron band is $\Delta\epsilon \sim 0.2$ eV above the Fermi surface.[4] This value of $\Delta\epsilon$ is close to that found experimentally, and the transition observed in the $Mo_{1-x}Re_x$ system at $x = 0.11$ is in fact a consequence of the formation of a new electron part of the Fermi surface at point G on the NH axis (see Ref. 4 for the notation). The possibility of a topological transition of this sort at $x \sim 0.1$ has been discussed previously[5] on the basis of data on the changes in T_c in the $Mo_{1-x}Re_x$ system.

An asymmetry of a local maximum in the thermal emf is known to reflect the

nature of the topological transition. However, it is difficult to carry out a reliable analysis of the nature of the anomaly in our case since it is not clear how the thermal emf would vary in the absence of the transition near $x = 0.11$ (the size of the "base" α_0 under the anomaly is not clear). The reason is that in the electronic structure of molybdenum, according to the calculated spectrum, we would expect several topological transitions near ϵ_F, specifically, at $\Delta\epsilon = -0.1 - 0.2$ eV (the appearance of electron lenses) and at $\Delta\epsilon \sim 0.8$ eV (the disappearance of whole ellipsoids and the simultaneous appearance of new electron surfaces). These transitions lie near the energy interval which we studied, $\Delta\epsilon = 0 - 0.3$ eV, and the ledges of the corresponding anomalies in the thermal emf could substantially change the shape of the base. In evaluating the relative magnitude of the anomaly, $\alpha_{x=0.11}\alpha_0^{-1}$, we used the value $\alpha_0 T^{-1} = 1.7 \times 10^{-8}$ V/K^2 found from measurements of the thermal emf of molybdenum at[8] $T \gtrsim 200$ K, where the anomalies have become negligible.

The temperature dependence of the anomaly was determined in additional experiments over the range 4–350 K, where we measured the thermal emf of a sample with $x = 0.11$ with respect to that of a sample with $x = 0.2$:

$$\Delta\left(\frac{\alpha}{T}\right) = \left(\frac{\alpha}{T}\right)_{x=0.11} - \left(\frac{\alpha}{T}\right)_{x=0.2} \tag{2}$$

The temperature gradient was measured over the range 5–60 K with the help of carbon thermometers, while above 77 K it was measured with a copper-constantan thermocouple; it was found to be ~ 0.05 K at 4.8 K and 3–6 K at $T > 60$ K. These measurements reveal the temperature interval in which $\Delta(\alpha T^{-1})$ remains constant (0–30 K) and the region in which $\Delta(\alpha T^{-1})$ falls off monotonically with increasing temperature, $T \gtrsim 30$ K. The magnitude of the anomaly in the first region is determined by the finite carrier lifetime τ (or $\Gamma = \hbar\tau^{-1}$) while that in the second region is determined by the thermal width (here $T > \Gamma$). From the curve of $\Delta(\alpha/T)(T)$ (Fig. 2) we find $\Gamma \simeq 30$ K.

According to the theory of Refs. 9 and 10, in the region $T < \Gamma$ the size of the anomaly would be

$$\Delta\alpha \sim \alpha_0 \cdot 0.25(\epsilon_F \Gamma^{-1})^{1/2} \tag{3}$$

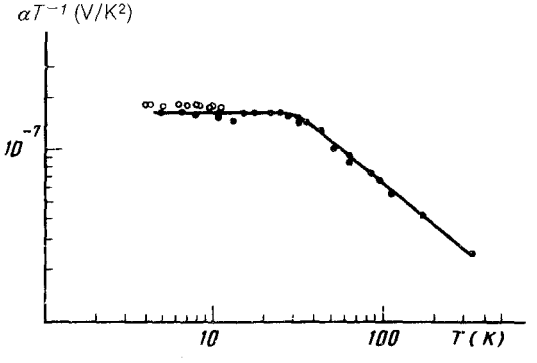

FIG. 2. Anomaly in the thermal emf versus the temperature. ●—thermal emf of a sample with $x = 0.11$ with respect to that of a sample $x = 0.2$, for which the value of αT^{-1} is 0.038×10^{-7} V/K^2; ○—absolute differential thermal emf of a sample with $x = 0.11$.

Substituting the numerical values $\epsilon_F = 6.5$ eV (Ref. 4) and $\Gamma = 30$ K, we find $\Delta\alpha = 12\alpha_0$, in agreement with the value found experimentally[1)] (Fig. 1). At temperatures $T \gtrsim \Gamma$, the maximum on the curve in Fig. 1 should shift along the concentration axis with increasing T (Ref. 10), decreasing in amplitude in proportion to $T^{-0.5}$. Experimentally, we find $\Delta(\alpha T^{-1}) \sim T^{-(0.8)}$ ($T > 30$ K). The probable reason for this result is that the point $x = 0.11$ may fall on a steep slope of the anomaly as the maximum shifts.

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[1)]The width of the feature at the level of $0.5\alpha_0$ is 700 ± 200 K. According to the estimates of Refs. 2 and 9, the half-width of the structural feature should not exceed $\approx 7\,\Gamma$, i.e., ≈ 200 K. The discrepancy between these two values probably means that relation (1) is approximate.

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