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*Note*

# Electrical Resistivity and Deviation from Matthiessen's Rule in Polycrystalline Lead-Doped Tin

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The results of measurements of the electrical resistivity, $\rho$, of lead-doped (weight concentrations: 0.001, 0.01, 0.1, and 1%) polycrystalline tin are presented. The experiments were performed using a comparative method with the aid of a tantalum thermomagnetic modulator applied as a null indicator. A nonlinear dependence of the residual resistivity on lead concentration was obtained. An anomalous character of $\rho(T)$ dependence was observed in the lowest-Pb concentration sample (0.001%). The deviation of the resistivity–temperature characteristics from Matthiessen's rule (DMR) was determined. The characteristics of the DMR do not show "humps" in the temperature range from 3.7 to 28 K.

**KEY WORDS:** electrical resistivity; lead-doped tin; Matthiessen's rule.

## 1. INTRODUCTION

The low-temperature electrical resistivity of metals weakly diluted with nonmagnetic admixtures is usually described by the following relation:

$$\rho_a(T, c) = \rho_0 + ET^5 + \Delta\rho \tag{1}$$

where $\rho_0$ is the residual resistivity, $c$ is the admixture concentration, $ET^5$ is the term of the Bloch–Grüneisen theory describing the electron–phonon scattering, and $\Delta\rho$ is the deviation from Matthiessen's rule (DMR).

In the case of a resistivity term originating at low temperatures from admixtures, component $\Delta\rho$ can be presented in the form [1]

$$\Delta\rho = BT^2 + CT^4 + DT^5 \tag{2}$$

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where $BT^2$ is the scattering of electrons by vibrating admixture ions, $CT^4$ is the phonon-assisted impurity scattering proposed by Klemens and co-workers [2, 3], $DT^5$ is the contribution of inelastic electron scattering by the deformed phonon spectrum, and $B$, $C$, and $D$ are constants or weak temperature-dependent functions.

Combination of Eqs. (1) and (2) leads to

$$\rho_a(c, T) = \rho_0 + BT^2 + CT^4 + (D + E)\, T^5 \tag{3}$$

Equation (3) comprises only the linear dependence of the electrical resistivity on the admixture concentration. However, in certain regions of temperatures and concentrations (0.01–1 %), a nonlinear dependence of the electrical resistivity on the concentration should be taken into account. This problem is considered in Ref. 4. The authors of Ref. 4 determined the component of electrical resistivity $\Delta\rho_T$ due to the anisotropy of scattering of electrons by phonons considering the influence of elastic scattering by admixtures. This component is nonlinearly temperature dependent [through a certain function $\eta(T)$] and concentration dependent. The function $\eta(T)$ comprises the anisotropy of electron–phonon scattering (mostly Umklapp scattering anisotropy). It displays a maximum around $\Theta_D/10$. Taking the above component into account, the expression for the total resistivity of a doped metal adopts the form

$$\rho_a(c, T) = \rho_0 + BT^2 + CT^4 + (D + E)\, T^5 + \Delta\rho_T \tag{4}$$

where $\Delta\rho_T$ is the DMR due to the nonlinear dependence of the electrical resistivity on the admixture concentration.

## 2. EXPERIMENTS

Five polycrystalline tin samples[2] of 6N purity doped with lead of 5N purity were prepared, at the following weight concentrations: Sn(6N), Sn + 0.001 % Pb, Sn + 0.01 % Pb, Sn + 0.1 % Pb, and Sn + 1 % Pb. Quantitative spectral analysis revealed the presence of the following metals.

Sn(6N): Sb, $2 \times 10^{-5}$%; Cu, $2 \times 10^{-5}$%; Fe, $1 \times 10^{-6}$%; Pb, $2 \times 10^{-5}$%; Bi, $2 \times 10^{-5}$%; Ga, $1 \times 10^{-6}$%; Zn, $2 \times 10^{-6}$%; Cr, $1 \times 10^{-6}$%; Mn, $1 \times 10^{-6}$%; Ti, $1 \times 10^{-6}$%.

Pb(5N): Bi, Fe, Mg, Cu, Sb, Zn, and Ga, $1 \times 10^{-4}$% each; In and Al, $0.5 \times 10^{-4}$% each; Ag, $1 \times 10^{-5}$%; Cd, $5 \times 10^{-6}$%.

[2] Samples were prepared at the Institute of Electronic Materials Technology, Warsaw.

The samples, enclosed in ampoules under vacuum, were annealed in an oven for 24 h at 472 K and subsequently slowly cooled to room temperature at an average rate of 4.5 K/h.

Measurements of the electrical resistance were conducted by a comparative method using a thermomagnetic tantalum modulator as a null indicator of sensitivity of $5 \times 10^{-12}$ V [5]. The experimental setup allowed for measurements with a relatively low current through the sample ($I_x = 1$ A).

Experiments were conducted in the temperature range from 3.7 to 28 K. The dimensions of the samples were length $l \cong 40$ mm (the distance between point contacts made of Wood alloy) and diameter $\phi \cong 3$ mm.

Random errors of the measurements are estimated to vary from 0.2 to 2%, depending on the temperature, while the systematic error due to uncertainties of determination of the sample geometry is estimated to be 2%.

The resistance ratio, $R_{300}/R_{4.2}$, for the particular samples was as follows: Sn(6N), 17,400; Sn + 0.001% Pb, 19,000; Sn + 0.01% Pb, 2900; Sn + 0.1% Pb, 740; and Sn + 1% Pb, 40.

## 3. RESULTS

The results of the measurements are presented in Fig. 1. The curves $\rho(T)$ run parallel to each other and the resistivity increases monotonically

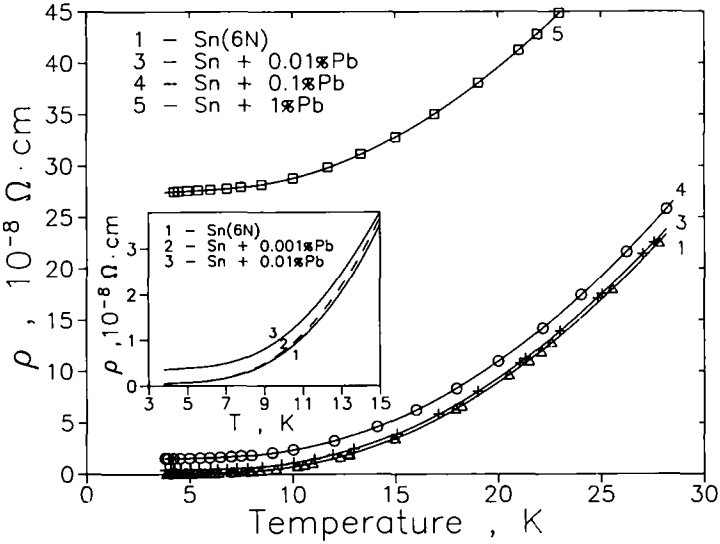


**Fig. 1.** Dependence of the electrical resistivity of lead-doped tin on temperature at various concentrations of lead admixtures.

with increases in the lead admixture concentration. For the sample with the lowest concentration of lead (Sn + 0.001 % Pb), deviation from the parallel courses of the $\rho(T)$ curves was observed (inset in Fig. 1, dashed line), and the curve for Sn + 0.001 % Pb crosses the curve for Sn + 0.01 % Pb at a temperature of about 19.5 K.

The curves were represented by fifth-order polynomials, which gave better fits to the experimental points than the representation according to the dependence given by Eq. (3). The latter representation was, however, correct for the samples Sn + 0.1 % Pb and Sn + 1 % Pb.

Coefficient $B$ of $T^2$ in Eq. (3) is $(4.39 \pm 0.79)$ and $(5.57 \pm 1) \times 10^{-11}\ \Omega \cdot \text{cm}$ for samples Sn + 0.1 % Pb and Sn + 1 % Pb, respectively. These coefficients describe quantitatively the scattering of electrons by vibrating ions of lead in polycrystalline tin.

The residual resistivity, $\rho_0$, was determined for all the samples by extrapolation of $\rho(T)$ curves to $T = 0$. A function of the form $f(T) = a + bT^2$ was used for this purpose. The results are as follows: $\rho_0 = 3.676 \times 10^{-10}$, $4.75 \times 10^{-11}$, $3.13 \times 10^{-9}$, $1.474 \times 10^{-8}$, and $2.729 \times 10^{-7}\ \Omega \cdot \text{cm}$ for the samples Sn(6N), Sn + 0.001 % Pb, Sn + 0.01 % Pb, Sn + 0.1 % Pb, and Sn + 1 % Pb, respectively.

The dependence $\rho_0 = \rho_0(c)$ (Fig. 2) is not linear ($\rho_0 = 3.81 \times 10^{-6}\, c^{0.74}$). Deviation of the residual resistivity on admixture concentration dependence from linearity was observed also by other investigators [6].

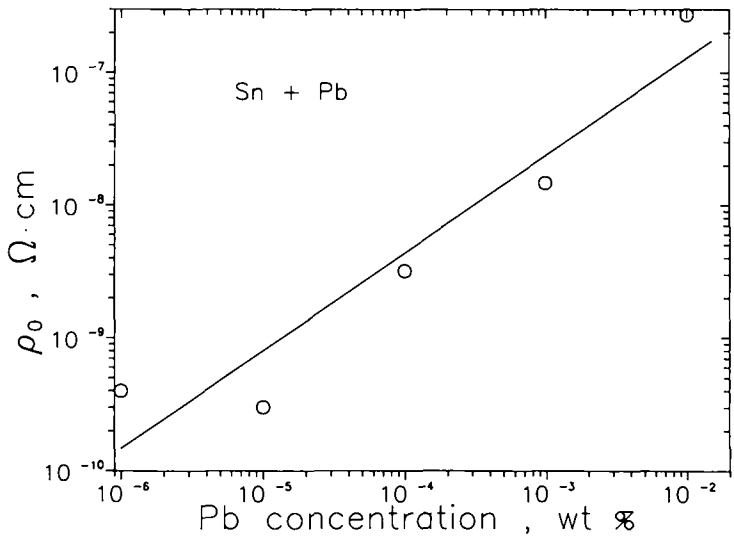


**Fig. 2.** Dependence of the residual resistivity of tin on the concentration of the lead admixture; $\rho_0 = 3.81 \times 10^{-6}\, c^{0.74}$.

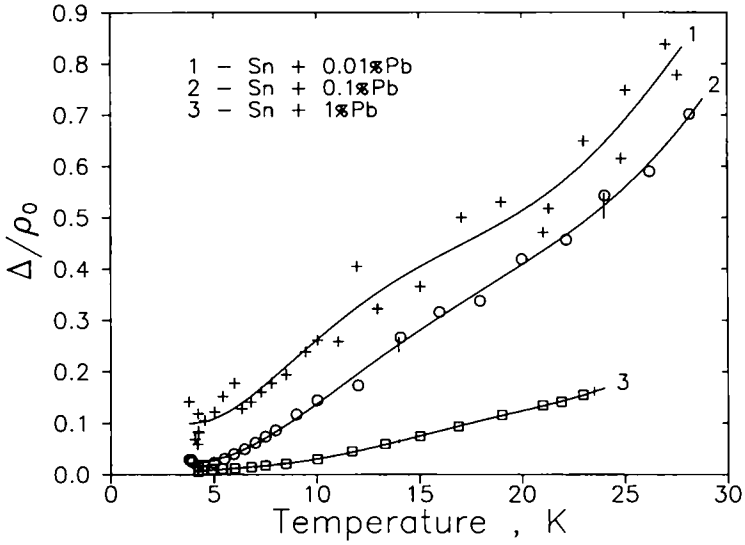


**Fig. 3.** Dependence of the DMR on temperature for tin with various concentrations of lead admixtures.

The DMR as a function of temperature (Fig. 3) in the present work does not confirm the presence of a "hump" for lead-doped tin in the temperature range 3.7–28 K. The presence of humps has been observed for several doped metals, e.g., in very dilute alloys Cu:Au [7] and Mg:Al [8, 9] and in Al:Mg, Al:Si, Al:Zn, and Al:Ag [6]. Bass [10] stated that the existence of humps for tin had not been unambiguously confirmed or rejected.

The authors of Ref. 11 noted the appearance of maxima (humps) on the total DMR curves for the thermal conductivity of aluminum. The total DMRs are the sums of corresponding deviations for "normal" and "Umklapp" scattering. Umklapp scattering leads to a decreasing total DMR, and because its role grows with increasing temperature, the appearance of maxima of the total DMR functions is possible at higher temperatures.

## 4. DISCUSSION

The experimental data of our electrical resistivity measurements on Sn:Pb are better described by fifth-order polynomials than by Eq. (3), which is based on physical grounds. Only the results from the two samples with the highest lead concentration, Sn + 0.1 % Pb and Sn + 1 % Pb, can

be described by Eq. (3) with an uncertainty approaching or smaller than the measurement uncertainty. This situation can result from the following: (a) disregarding in Eq. (3) the above-mentioned term describing the nonlinear dependence of resistivity on the admixture concentration [Eq. (4)] and (b) neglecting the scattering of electrons by grain boundaries. The latter contribution to electrical resistivity should be constant because of the comparable average size of crystallites in all the samples.

The anomaly observed by us of the curve for the sample of the lowest lead concentration (Sn + 0.001% Pb) (Fig. 1), i.e., its deviation from a parallel course, may be the result of an influence of magnetic impurities (Fe, Cr).

Such a large deviation from linearity in the $\rho_0(c)$ function (Fig. 2) indicates the essential role of the nonlinear terms which emerge from the coexistence of the inelastic scattering of electrons by phonons and the elastic scattering by admixture atoms, as discussed in Ref. 4.

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