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The concentration dependence of the electrical resistivity, the Hall coefficient and the superconducting transition temperature of Pb-In thin films

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Abstract. Hall coefficient and electrical resistivity values are reported for Pb-In solid solution thin films ($\approx$ 170 nm). It is found that $R_{\rm H}$ varies from the small positive value of pure Pb to negative values, reaching its maximum value (negative) at a concentration of about 80 at.% In. The change from positive to negative occurs at a concentration of about 16 at.% In. The superconducting transition temperature as a function of concentration was also measured for Pb rich samples.

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Introduction

Several authors have determined the Hall coefficient, $R_{\rm H}$, of In rich In-Pb solid solutions. Sasaki [1] reported results for bulk samples as a function of pressure. Merriam [2] measured the superconducting transition temperature, T_c, as a function of indium concentration in lead and Schiozaki [3] determined $R_{\rm H}$ as a function of concentration. The above mentioned results are on bulk samples. Takano and Sato [4] carried out Hall and resistivity measurements at room temperature on rolled samples over a wide range of concentration values.

In this work we have made Hall measurements on thin film samples of Pb-In solid solutions ranging from pure Pb to pure In. Pb has a small positive Hall coefficient while that of In is also small but negative at room temperature. This allows us to make samples with positive and negative values of $R_{\rm H}$. The change in sign occurs at a concentration of about Pb 16 at.% In at room temperature. Linde and Rapp [5] and more recently, Saenz and Rapp [6], have considered an empirical relationship between $R_{\rm H}$ and T_c for non transition metals and their alloys. They conclude that a high T_c is best obtained when $R_{\rm H}$ approaches zero from either positive or negative values, while Chapnik [7] suggests that it is favored by a positive and small value of $R_{\rm H}$. The Pb-In system is a good one to test the validity of these ideas.

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Pb at.% In	Thickness (nm)	Resistivity ($10^{-7}\Omega$ cm)		Hall coefficient (10^{-11}m^3/AS)	
		300 K	77 K	300 K	77 K
0.0	164	2.50	—	1.3	—
7.9	161	2.51	0.73	0.61	—
11.6	170	2.52	0.72	0.57	—
23.5	165	3.04	1.08	−0.84	—
27.8	150	—	—	−1.1	−1.9
30.2	171	3.31	—	−0.80	−3.0
40.1	178	3.35	1.42	−2.3	−4.1
45.1	162	—	—	−2.6	−5.2
57.8	164	3.91	2.16	−4.4	−8.7
66.7	169	4.36	2.73	−4.8	−8.6
75.1	202	4.88	2.72	−5.1	−9.7
88.0	165	5.09	3.47	−5.0	−8.2
95.6	180	2.90	—	−3.4	−4.9
100.0	182	1.18	0.292	−0.19	1.4

TABLE I. Parameters for Pb In samples.

Experimental procedures

The thin film samples were prepared by rapidly evaporating, by means of an electron beam gun, previously prepared ingots of Pb-In alloys in the desired concentrations. The starting materials were nominally 99.999% pure Pb and 99.9% In. Cleaned glass slides with deposited copper electrodes for current and electrical resistance measurements were used as substrates. The thickness of the samples was determined by means of a commercial crystal quartz monitor. Samples were typically 1.477 cm long by 0.94 mm wide. The dimensions were determined with the use of a travelling microscope suited with a scale.

Electrical resistance measurements were carried out using the usual four probe DC method, the Hall voltages being measured with a nanovoltmeter using deposited Hall contacts of the same material as that of the sample under study. Permutations of current and magnetic field were made in order to minimize undesired voltages. The measured Hall voltage was linear in magnetic field up to 0.8 T for these samples and 0.8 T was used to determine all the reported $R_{\rm H}$ values.

To guarantee thermal stability, the low temperature measurements were made immersing the samples directly in liquid nitrogen in a dewar placed between the pole pieces of the magnet.

The superconducting transition temperature was determined by monitoring the inductance change in a small coil placed in contact with the sample as the temperature was varied in a liquid helium cryostat. The temperature was measured with a diode thermometer.

Table I shows thickness, composition, Hall coefficient and resistivity values for all samples. Errors are less than 5% for Hall data and less than 1% for resistivity data. Uncertainties in concentration are of the order of 0.1 at.% and of 0.05 K in temperature.

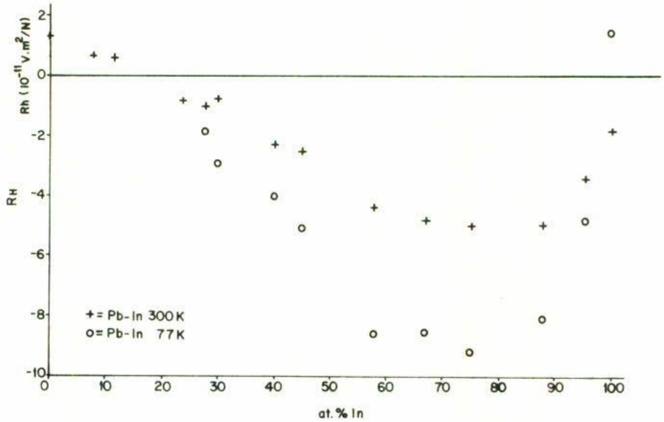

FIGURE 1. R_H as a function of In concentration for Pb-In alloys, both at room temperature and at 77 K.

Results

Figure 1 shows the variation of R_H as a function of concentration for measurements at room temperature and at 77 K. At room temperature R_H passes through zero for concentrations of about 16 at.% In in Pb. For 77 K, R_H is so small that the Hall voltages could not be measured for indium concentrations in lead smaller than 25 at.%. Room temperature values are negative for all concentrations larger than this, while the 77 K values become positive at about Pb 98 at.% In.

Another feature to be noticed in this figure is that, for small solute concentrations, both at the Pb-rich end and at the In-rich end, R_H at room temperature and at 77 K have similar values, while at intermediate concentrations of both metals, the values of R_H diverge, the largest difference appearing at a concentration of about Pb 75 at.% In.

In Figure 2, electrical resistivity values are plotted as a function of concentration. As would be expected, ρ increases as In is added to Pb. This parameter then begins to decrease to the value corresponding to that of pure In for concentrations higher than about Pb 80 at.% In when the conductivity is dominated by an indium rich phase.

The normalized superconducting transition temperature for Pb-rich samples are shown in Figure 3 as a function of alloy concentration. Changes are small and for concentrations higher than about 8 at.% In the ratio decreases with increasing In content.

Discussion

The concentration dependence of R_H and ρ shown by Figures 1 and 2 may be interpreted as having its origin in both the decrease of valence electrons per atom

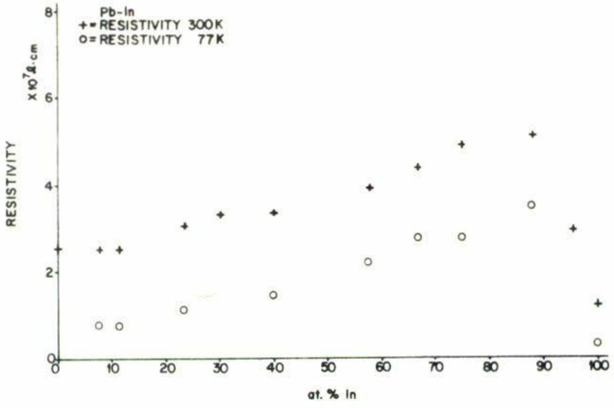

FIGURE 2. Electrical resistivity as a function of In concentration for Pb-In alloys, both at room temperature and at 77 K.

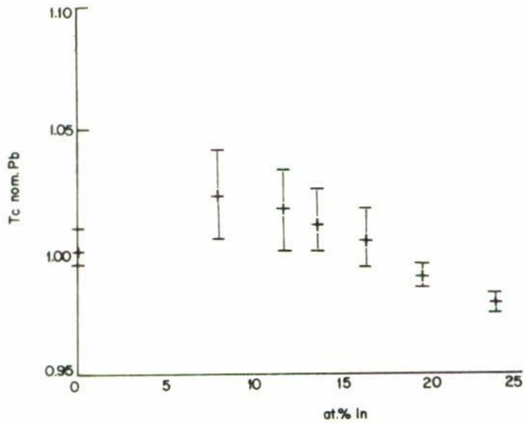

FIGURE 3. Normalized superconducting transition temperature as a function of the atomic percent of In in Pb. The spreads shown are transition widths, not experimental errors.

and the increase of scattering by solute atoms. The addition of trivalent atoms into the matrix of tetravalent Pb tends to shift it from its hole-like conducting properties to a more electron-like conducting mechanism.

In terms of the usual two band conduction model in the low field approximation, $R_{\rm H}$ may be written as

$$R_{\rm H} = \frac{n_{\rm h}\mu_{\rm h}^2 - n_{\rm e}\mu_{\rm e}^2}{e(n_{\rm h}\mu_{\rm h} + n_{\rm e}\mu_{\rm e})^2} \tag{1}$$

where e is the magnitude of the electronic charge, μ the corresponding mobility and n the carrier density.

The change in sign of R_{H} can then be attributed to the solute atoms affecting the hole mobility more strongly than the electron mobility [3], as well as changes in the relative values of n_{h} and n_{e}. As the temperature is lowered, impurity scattering is enhanced over phonon scattering making R_{H} more negative for the higher solute concentration samples.

As indicated in Figure 3, the superconducting transition temperature, T_c, does not change significantly as a function of concentration for the Pb-rich alloys, but some alloys show T_c higher than that of pure Pb. Our data show a trend following Linde's correlation. There is a maximum in T_c at around 8 at.% and a zero in the Hall data at 300 K around 15 at.%, then T_c deceases and R_{H} becomes more negative. It seems, however, that at least for this system, to assume equal mobilities would be an oversimplification. A model considering different mobilities would be more realistic. Therefore we cannot conclusively rule out Chapnik's hypotheses.

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Resumen. Se presentan los valores medidos del coeficiente de Hall y de la resistividad eléctrica para películas delgadas de aleaciones de Pb-In ($\approx$ 170 nm). R_{H} varía desde el pequeño valor positivo correspondiente al Pb puro pasando a valores negativos y alcanzando el mínimo a concentraciones cerca de 80 at.% In. El cambio de signo se da alrededor de 16 at. % In. También se presentan los datos de la temperatura de transición a superconductividad como función de la concentración para aleaciones ricas en Pb.