

EFFECT OF ISOTOPIC COMPOSITION ON THE ELECTRICAL CONDUCTIVITY OF CADMIUM

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The electrical conductivity of five Cd samples differing in isotopic composition is measured at temperatures between 1.2° and 300°K. The absolute resistivities at room temperature are compared. The large differences of electrical resistivity that are observed at low temperatures, where inelastic scattering on lattice vibrations is predominant, are associated with differences of the respective phonon spectra.

THE present work continues experimental investigations<sup>[1,2]</sup> that were undertaken in conjunction with recent investigations by Yu. Kaganet et al.<sup>[3-5]</sup> of several properties observed in irregular systems and associated with modifications of the phonon spectra. In<sup>[2]</sup> we analyzed peculiarities in the temperature dependence of electrical resistances exhibited by alloys of Mg with the heavy nonmagnetic elements Pb and Ag. In MgPb we observed directly how a contribution to the impurity conduction arises through deformation of the phonon spectrum even when the electron spectrum of the same system is also modified. In the present work we investigate the electrical resistivity of isotopically different samples forming a class of irregular systems where the phonon spectrum does not vary greatly as a general rule. At the same time the electron spectrum of these samples is identical. It thus becomes possible to detect directly the differences of electrical resistivity that are associated with modifications of the phonon spectra alone.

It has been shown in<sup>[5]</sup> that the electrical resistivity of metals exhibits two types of isotope effects, which differ in their quadratic and linear dependence, respectively, on isotopic mass. In the first type the effects are associated only with irregular distribution of the isotopes and the resultant residual resistivity. The linear effects are associated with modifications of the phonon spectrum as the isotopic composition of the samples is varied.

In the present work the limited purity of the samples confined us to a study of only the isotope effects that depend linearly on isotopic mass difference. We measured the electrical resistivity of five Cd samples differing in isotopic composition. Cd is a convenient metal for this kind of study; it has a large number of stable isotopes with sufficiently large mass ratios that can induce appreciable differences in the behavior of their electrical conductivity at low temperatures. No phase trans-

formations of Cd occur in the investigated temperature region.

We note that the isotope effect in electrical resistivity was studied in<sup>[6,7]</sup>, where the effect was observed for the first time in the isotopes Li<sup>6</sup> and Li<sup>7</sup> and their alloys.

EXPERIMENT

Table I gives the isotopic compositions and impurity contents of several Cd isotope mixtures that were melted together and were then extruded through a hole of ~0.8-mm diameter. Calibrated diameters were produced by drawing the wires through Pobedit dies. The resulting Cd samples were wires ~0.65 mm in diameter and ~60 mm long. The diameters and lengths (separation of the potential contacts) were measured with an IZA-2 horizontal comparator. No ellipticity was observed when the diameter of each sample was measured at more than twenty points throughout its length, in two mutually perpendicular positions. The rms error of these measurements was ±0.002 mm; the corresponding error of the diameter was ±0.3%. The separation of the potential leads was determined from reference lines marked on the wires by clamped contacts. Since the error of the length was below 0.1%, the overall error of the geometric factor *s/l* (the ratio between the cross-sectional area and length of a sample) was at most ±0.7%.

The electrical resistivity of the Cd samples was measured in a cryostat similar to that described in<sup>[8,21]</sup>. In the 1.2°--4.2°K range the samples were in contact with a helium bath. Slow warming immediately above this range was followed by more rapid heating in the 30°--80° range. Finally, in the range 80°--300°K the sample assembly was placed in thermal contact with a nitrogen bath, using helium as the heat exchanger. Control measurements were obtained in liquid hydrogen,

Table I

Number of sample	Cd	Isotopic composition, %										Chemical impurities, %												
		mav	Cd									Fe	Al	Si	Cr	Ni	C	Pb	Sb	Sn	Zn			
			106	108	110	111	112	113	114	116														
1	106	108.62	58.9	0.6	5.6	6.0	11.0	4.7	11.2	2.0	0.001	<0.001	<0.003	<0.001	0.013	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
2	110	110.2	—	—	91.5	3.2	2.2	1.0	1.7	0.4	0.001	<0.002	<0.003	<0.001	0.006	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002
3	112	112.01	0.1	0.2	0.3	0.8	96.1	1.4	1.0	0.1	0.001	<0.003	<0.003	0.001	0.002	<0.002	<0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
4	114	113.96	—	—	0.3	0.8	0.6	0.8	97.3	0.5	<0.001	<0.001	<0.003	<0.001	0.002	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
5	116	115.61	—	—	0.9	1.8	2.6	1.6	4.0	88.5	0.002	<0.002	<0.003	0.001	0.020	<0.002	<0.001	<0.001	<0.001	0.001	<0.001	<0.001	<0.001	<0.001

Table II

Cd	$\rho$ (291° K), $\mu\Omega\text{-cm}$	$\rho$ (4.2° K), $\mu\Omega\text{-cm}$	$\rho(291^\circ\text{K})/\rho(4.2^\circ\text{K})$
106	7.41 ± 0.05	0.0206 ± 0.0002	360
110	7.38	0.0180	411
112	7.42	0.0212	350
114	7.40	0.0192	386
116	7.38	0.0228	323
106	7.37 ± 0.05	0.0197 ± 0.0002	375
110	7.40	0.0180	410
112	7.41	0.0219	338
114	7.42	0.0191	389
116	7.39	0.0237	312

neon, and nitrogen that were in equilibrium with their vapor at normal pressure. These measurements were in good agreement with those obtained during the slow warming procedure.

The electrical resistivity was measured, using knife-type clamped contacts, in five samples simultaneously. For the purpose of comparing measurements performed on more than one sample of a given isotopic composition the samples were divided into two groups. A Honeywell germanium resistance-type thermometer was used for the temperature measurements in the range 1.2°–10°K and a platinum thermometer in the range 10°–300°K. The resistances of the samples and thermometers were measured with the aid of a conventional potentiometer circuit including a PMS-48 five-decade potentiometer together with an F-16 (of 10<sup>-8</sup> V/mm sensitivity) photo-compensated amplifier as a null instrument.

## EXPERIMENTAL RESULTS AND DISCUSSION

The electrical resistance of five Cd samples having different isotopic compositions was measured in the range 1.2°–300°K. Table II gives the absolute resistivities at room temperature (291°K) and at helium tem-

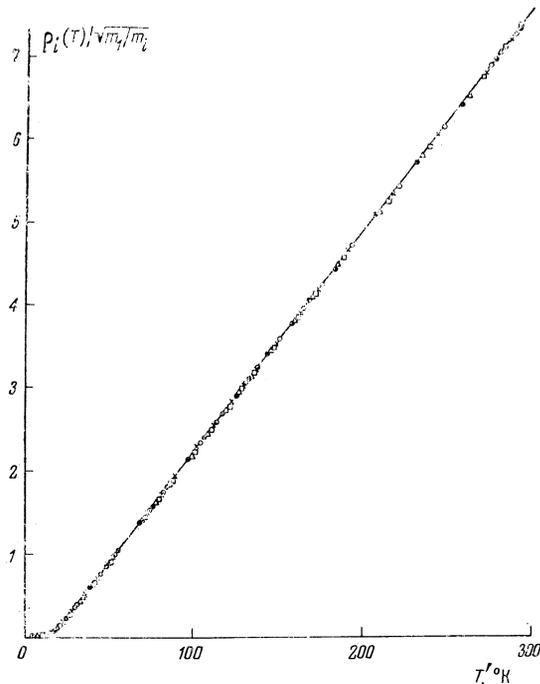


FIG. 1. Universal curve for reduced resistivity  $\rho_i(T) (m_1/m_2)^{-1/2}$  versus reduced temperature  $T'$  for Cd samples of different isotopic compositions. ● - No. 1, Δ - No. 2, □ - No. 3, X - No. 4, ○ - No. 5 (samples numbered as in Table I).

perature (4.2°K), along with the ratio of these measurements, for the two groups of samples.

Since the experimental error in R does not exceed 0.1% at the lowest temperatures and 0.01% at high temperatures, most of the error in  $\rho$  arises in measuring the geometric dimensions ( $s$ ,  $l$ ) of the samples; in the present case the given error is not more than  $\pm 0.7\%$ .

The customarily accepted resistivity of ordinary Cd is  $\rho(291^\circ\text{K}) = 7.35 \mu\Omega\text{-cm}$ , while for drawn samples it is  $7.54 \mu\Omega\text{-cm}$ .<sup>[9]</sup> Our results agree well (within experimental error limits) with the first of the two given values. Table II shows that the resistivities of the samples coincide at room temperature. This result may serve as evidence that the electronic spectrum and the electron-ion interaction are both identical for different isotopes at room temperature.

At the very lowest temperatures the effects associated with modification of the phonon spectrum are masked by electron scattering on impurities. Specifically, the Bloch temperature dependence is not observed in our samples. Also, in all cases the resistance passes through a minimum at  $\sim 2^\circ\text{K}$ , apparently because paramagnetic impurities are present. However, with increasing temperature the electron scattering on phonons begins to assume the dominant role. Thus the resistance of all samples varies as  $T^5$  in the range 10°–16°K. Since we are interested at present in effects associated with different phonon spectra depending on isotopic composition, we shall hereinafter analyze resistance measurements in the range 10°–300°K.

Our measurements show that  $\rho(T)$  for different isotopic compositions can be described by a simple universal relation,<sup>[5]</sup> depending only on the mass ratio of any two compared samples:<sup>1)</sup>

$$\rho_i(T) = \rho_1(T') \sqrt{m_1/m_i}, \quad T' = T \sqrt{m_1/m_i}. \quad (1)$$

This result is shown in Fig. 1, where the ordinate is  $\rho_i(T)/\sqrt{m_1/m_i}$  and the abscissa is the reduced temperature  $T'$ . It is here seen that the experimental results for the different isotopic compositions are actually found to lie on a single universal curve.

Figure 2 shows directly how modifications of the phonon spectrum affect the resistivity of the different samples. The experimental temperature dependence of the relative change in resistance is here shown for all the samples as fractions of the change for the sample having the smallest average mass ( $m_{av} = 108.2$ ) of Cd isotopes; the plotted curves are thus represented by

$$Y = \left( \frac{[R_i(T) - R_i(0)]/R_i(291^\circ\text{K})}{[R_1(T) - R_1(0)]/R_1(291^\circ\text{K})} - 1 \right) \cdot 100\%. \quad (2)$$

At low temperatures the differences in isotopic composition are accompanied by large differences in resistivity which increase with the corresponding difference in mass ( $m_j$ ). In the region where  $\rho \propto T^5$  holds true the ratio of two resistivities must attain a value represented by<sup>[4,5]</sup>

$$\frac{\rho_i}{\rho_1} \approx 1 + 2 \frac{\Delta m_i}{m_1}, \quad \Delta m_i = m_i - m_1. \quad (3)$$

<sup>1)</sup> We note that a similar result is derived from the Bloch-Grüneisen formula in [6], which was derived on the basis of very crude assumptions regarding the nature of the electron-phonon interaction and the phonon spectrum

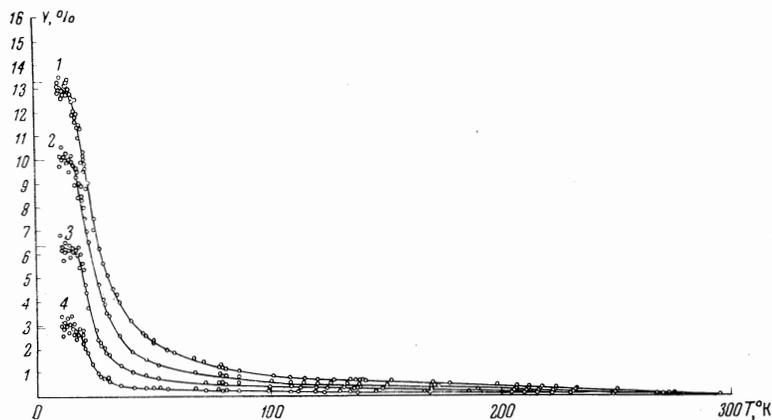


FIG. 2. Temperature dependence of the relative change in the resistivity of Cd samples having different isotopic compositions, expressed as a fraction of the relative change, for Cd samples with  $m_{av} = 108.2$ . 1 -  $m_{av} = 115.61$ , 2 -  $m_{av} = 113.96$ , 3 -  $m_{av} = 112.01$ , 4 -  $m_{av} = 110.2$ .

It is seen in Fig. 2 that the various ratio measurements agree approximately with the theoretical values, which are indicated by horizontal bars starting at the ordinate axis. Below  $20^\circ\text{K}$  the experimental values of  $Y$  are spread widely as a result of the errors incurred in determining  $R$ . Although this error in  $R$  is small ( $\sim 0.1\%$ ), the error in calculating  $Y$  becomes large ( $\sim 10\%$  at  $10^\circ$ ) because  $R(T)$  and  $R(0)$  are close at the lowest temperatures.

With increasing temperature the differences of the resistivities diminish rapidly and practically disappear at room temperature. The behavior of electrical resistance in samples of different isotopic compositions is associated physically with the fact that at low temperatures electrons collide mainly with long-wavelength phonons; these collisions are essentially inelastic. With changing isotopic composition, for example with increase of  $m_i$ , the state density increases in the low-frequency portion of the spectrum and the probability of scattering as  $T \rightarrow 0$  thus grows larger. In the high-temperature limit electron-phonon collisions become quasi-elastic. Effects associated with differences in the phonon spectra therefore disappear from the picture of electrical resistance.

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