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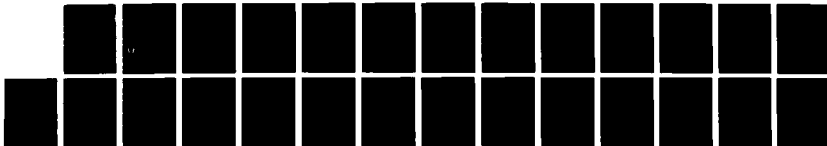
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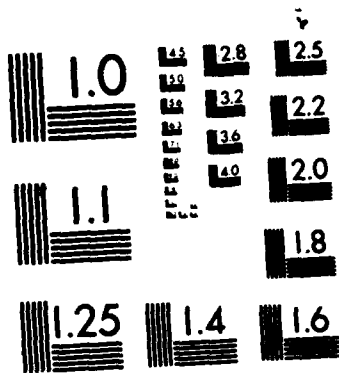
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TECHNICAL REPORT ARCCB-TR-86026

# PRESSURE DEPENDENCE OF ELECTRICAL RESISTIVITY FOR MAGNESIUM-BASED AMORPHOUS ALLOYS

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AUGUST 1986



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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number)<br>Measured pressure coefficients of resistance and the corresponding pressure coefficients of resistivity, estimated using "law of mixtures" isothermal compressibility values, are reported for a series of magnesium-based amorphous alloys. The pressure coefficients correlate with the thermopower and the resistivity. When combined with results for non-magnetic amorphous alloys containing transition metals, there are suggestions of universal correlations between the pressure coefficients and the electrical resistivity. Data are discussed in the framework of the diffraction model. |                                            |                                                                                                                       |

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## INTRODUCTION

The temperature coefficient of resistivity (TCR) in amorphous transition-metal alloys exhibits a strong correlation with the resistivity  $\rho$ : the TCR decreases with increasing  $\rho$  and changes its sign from a positive to a negative value as  $\rho$  exceeds about 150  $\mu\Omega\text{cm}$ . This empirical law is often referred to as the Mooij correlation (ref 1). A number of investigators have argued that the reversal of the TCR sign could be explained on the basis of the Ziman theory originally developed for simple liquid metals. Matsuda and Mizutani (ref 2) studied an interrelation between the TCR and  $\rho$  for a number of simple amorphous alloys, which are defined as those having the Fermi level in the sp-conduction band. Here the electron-transport properties are dominated by the sp-conduction electrons. Matsuda and Mizutani revealed that the TCR versus  $\rho$  data for these simple amorphous alloys again fall on a master curve, which is entirely separated from the Mooij correlation curve for the transition-metal alloys. In this regard Mizutani (ref 3) emphasized the need for classifying amorphous alloys into different groups, in order to facilitate the discussion of electron-transport properties. According to this classification, the simple metallic glasses are designated as group 5, whereas the non-magnetic amorphous alloys containing transition metals and rare earths are designated as group 4 alloys. Typical examples of the former are Mg-Zn, Ca-Mg, Ag-Cu-Ge, and those in the latter Cu-Ti, Cu-Zr, etc. Mizutani and Matsuda (ref 4)

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<sup>1</sup>J. H. Mooij, Phys. Stat. Sol., Vol. A17, 1973, pp. 521-530.

<sup>2</sup>T. Matsuda and U. Mizutani, Solid State Commun., Vol. 44, 1982, pp. 145-149.

<sup>3</sup>U. Mizutani, Prog. Mat. Sci., Vol. 28, 1983, pp. 97-228.

<sup>4</sup>U. Mizutani and T. Matsuda, Proc. of Int. Conf. on Rapidly Quenched Metals, (S. Steeb and H. Warlimont, eds.), 1985, pp. 1035-1038.

noted that a negative TCR in group 5 alloys occurs when  $\rho$  exceeds about 60  $\mu\Omega\text{cm}$  and that not only the TCR against  $\rho$  correlation, but also the overall temperature dependence of resistivity can be explained in terms of the generalized Faber-Ziman theory, i.e., the extension of the original Faber-Ziman theory to low temperatures (for example, see Reference 5). In contrast to the apparent success of analyzing the group 5 data, the interpretation of the Mooij correlation for group 4 amorphous alloys remains a subject of controversy.

The temperature dependence of the electrical resistivity is generally measured at constant pressure, while most theoretical studies have been made at constant volume. Hafner (ref 6) pointed out that thermal-expansion effects are significant in determining the temperature dependence of resistivity at constant pressure in an amorphous  $\text{Mg}_{70}\text{Zn}_{30}$  alloy. Meisel and Cote (ref 7) incorporated thermal-expansion effects into their electron-transport calculations based on the generalized Faber-Ziman theory; excellent agreement with the experimental isobaric  $\rho$ -T curve was obtained when a value of the pressure coefficient of resistance (PCR) near that measured here for the Mg-Zn-based alloys was assumed. These theoretical calculations demonstrate the importance of thermal-expansion effects in the temperature dependence of the resistivity.

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<sup>5</sup>P. J. Cote and L. V. Meisel, in Glassy Metals I, (H. J. Guntherodt and H. Beck, eds.), Springer Verlag, Heidelberg, Vol. 46, 1981, pp. 141-166.

<sup>6</sup>J. Hafner, J. Non-Cryst. Solids, Vol. 69, 1985, pp. 325-346.

<sup>7</sup>L. V. Meisel and P. J. Cote, Phys. Rev., Vol. B31, 1985, pp. 4872-4878.

Besides being essential to the interpretation of the isobaric  $\rho$  versus  $T$  behavior, the pressure dependence of the electrical resistivity itself is important in providing insights into the scattering mechanism in amorphous alloys. To date, PCR data have been essentially limited to group 4 amorphous alloys. The data for group 5 are sparse. The present report is focused on a more systematic study of the pressure dependence of resistivity for a large number of group 5 amorphous alloys for which the data can, in principle, be discussed within the framework of the generalized Faber-Ziman theory.

#### EXPERIMENTAL PROCEDURES AND RESULTS

The following alloy systems were studied:  $Mg_{70}Zn_{30-x}M_x$  ( $M = Cu, Ga, \text{ and } Sn$ ) and  $Mg_{100-x}(GaAl)_x$ . All alloy ingots were prepared by induction melting the constituent elements (99.998% Cu, 99.95% Mg, 99.999% Zn, 99.999% Ga, 99.999% Al, and 99.999% Sn) in a helium atmosphere. Amorphous ribbons were fabricated by using a single-roll spinning-wheel apparatus in a reduced argon atmosphere. The formation of a single amorphous phase was confirmed by ordinary x-ray diffraction.

The electrical resistivity was measured at room temperature using the standard four-probe technique. Hydrostatic pressures up to 8 kbar were obtained in a Bridgman-Birch type cell with a 50-50 mixture of pentane and isopentane as the pressure transmitting medium. The resistivity increased almost linearly with increasing pressure in all the alloys studied. The value of the pressure coefficient of resistance (PCR), defined as  $(d \ln R / dP)_{T=300K}$  where  $R$  is the resistance and the derivative is evaluated near  $P = 0$ , is therefore positive for all samples studied.

Frequently the pressure coefficient of resistivity  $(d\ln\rho/dP)_T$  is reported rather than the directly measured PCR. The relationship between them is given by

$$\begin{aligned}\text{PCR} &\equiv (d\ln R/dP)_T = (d\ln\rho/dP)_T + (d\ln(L/A)/dP)_T \\ &= (d\ln\rho/dP)_T + (3B_T)^{-1}\end{aligned}$$

where  $B_T$  is the bulk modulus. Thus, to obtain  $(d\ln\rho/dP)_T$  one subtracts  $(3B_T)^{-1}$  from the PCR. For example, if one assumes that  $B_T$  is approximately 450 kbar (from the law of mixtures) for the Mg-based alloys studied here, then one subtracts approximately  $0.75 \times 10^{-3} \text{ kbar}^{-1}$  from the PCR to obtain  $(d\ln\rho/dP)_T$ . Since  $B_T$  is not generally known for amorphous metals, the directly determined PCR's are reported here.

The results are summarized in Table 1. The only previously determined PCR for group 5 alloys is for  $\text{Mg}_{70}\text{Zn}_{30}$ . In 1982 Fritsch et al. (ref 8) calculated a PCR value (including the  $(3B_T)^{-1}$  correction) of about one-half that suggested by the present data. It may be noted, however, that the PCR value calculated in 1985 by Hafner (ref 6) for  $\text{Mg}_{70}\text{Zn}_{30}$  is in good agreement with the present data.

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<sup>6</sup>J. Hafner, J. Non-Cryst. Solids, Vol. 69, 1985, pp. 325-346.

<sup>8</sup>G. Fritsch, J. Willer, A. Wildermuth, and E. Luescher, J. Phys., Vol. F12, 1982, pp. 2965-2974.

TABLE 1. MEASURED VALUES OF  $10^3$  KBAR  $\cdot$  PCR (i.e.,  $10^3$  KBAR  $\cdot \left. \frac{\partial \ln R}{\partial P} \right|_T$ )  
FOR THE MAGNESIUM-BASED AMORPHOUS METALS

| $Mg_{0.7}Zn_{0.3-x}X_x$ Alloys |      |      |      |      |      |      |
|--------------------------------|------|------|------|------|------|------|
| $x \backslash X$               | 0.02 | 0.04 | 0.06 | 0.10 | 0.15 | 0.20 |
| Ga                             | 4.49 | 4.50 | 3.97 | 4.00 | 3.53 | 2.73 |
| Sn                             | 4.00 | 3.88 | 3.40 |      |      |      |
| Cu                             |      | 3.88 | 4.26 |      |      |      |
| $Mg_{1-x}(GaAl)_x$             |      |      |      |      |      |      |
| $x$                            | 0.30 | 0.36 | 0.42 | 0.48 |      |      |
|                                | 2.18 | 1.90 | 1.89 | 1.38 |      |      |

## DISCUSSION

As noted in the Introduction, the electron-transport properties of group 5 amorphous alloys may be discussed in the context of the generalized Faber-Ziman theory. The PCR in amorphous metals is essentially determined by the elastic scattering part of the resistivity. Thus, incorporation of refinements in the treatment of electron-phonon interaction (e.g., Pippard-Ziman saturation) or distinctions between structure factors (e.g., between x-ray, geometric, or resistivity structure factor) are not warranted. Therefore, we represent the resistivity by the ordinary Ziman expression

$$\rho = \frac{3\pi\Omega}{4e^2\hbar v_F^2 k_F^4} \int_0^{2k_F} a(K) |U(K)|^2 K^3 dK \quad (1)$$

where  $a(K)$  is the structure factor (in regard to the PCR, one can take  $a(K)$  as the ordinary geometrical structure factor),  $U(K)$  is the scattering-matrix element or pseudopotential,  $\Omega$  is the volume,  $v_F$  is the Fermi velocity, and  $k_F$  is the Fermi momentum. Differentiation of Eq. (1) with respect to the volume  $\Omega$  yields the following relation:

$$\left( \frac{d \ln \rho}{d \ln \Omega} \right)_T = \frac{2}{3} \xi - 1 + \frac{\langle \Omega (\partial |U|^2 / \partial \Omega) a \rangle}{\langle |U|^2 a \rangle} + \frac{\langle |U|^2 (\partial a(K) / \partial \Omega_T) \rangle}{\langle |U|^2 a \rangle} \quad (2)$$

where  $\xi$  is the thermoelectric parameter defined as

$$\xi = -E_F (\partial \ln \rho / \partial E)_{E=E_F} \quad (3)$$

The term  $\xi$  arises from the volume dependence of reciprocal-space quantities like  $k_F$ . The value of  $d \ln \rho / d \ln \Omega$  can be derived from the measured PCR through the relation

$$(d \ln \rho / d \ln \Omega)_T = -B_T \times (\text{PCR}) + 1/3 \quad (4)$$

Unfortunately,  $B_T$  is not available for the present alloys and, thus, is roughly estimated by the "law of mixtures" using the values of pure constituent elements (refs 7,9).

In Figure 1, the resulting values of  $d\ln\rho/d\ln\Omega$  are plotted against the thermoelectric parameter  $\xi$ , which has been measured by Matsuda and Mizutani (ref 10) for all samples studied in these experiments. The corresponding data for liquid alkali metals are also included (ref 11). It can be seen that each set of data falls near a straight line having a slope of about 2/3. This is consistent with Eq. (2), with the combination of its last two terms approximately constant, and lends support to the validity of the discussion based on the Faber-Ziman theory for these simple amorphous alloys. The exciting point is the difference in the intercept of the line for alkali liquids and that for the Mg-based amorphous alloys: the one for liquid metals is about 3 and the other for the Mg-based amorphous alloys is about -1.5. As discussed by Faber (ref 11), the major contribution to the intercept for non-transition metals results from the last term in Eq. (2), i.e., the volume dependence of the structure factor. If the volume dependence of the pseudopotential is ignored as was assumed by Faber, a negative intercept of -1.5 for the present amorphous alloys indicates a small negative value for the

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<sup>7</sup>L. V. Meisel and P. J. Cote, Phys. Rev., Vol. B31, 1985, pp. 4872-4878.

<sup>9</sup>K. A. Gschneider, Solid State Physics, (F. Seitz and D. Turnbull, eds.), Academic Press, New York, 1964, pp. 298-313.

<sup>10</sup>T. Matsuda and U. Mizutani, submitted to J. Phys. F., 1985.

<sup>11</sup>T. E. Faber, Introduction to the Theory of Liquid Metals, Cambridge at the University Press, 1972, pp. 339-343.

last term. Of course, the corresponding value for liquid alkali metals is positive and large under these assumptions.

Faber suggested that when the Fermi momentum  $2k_F$  is to the left of the main peak of the structure factor as in monovalent liquids, the last term in Eq. (2) is roughly 4. This is consistent with the data for alkali liquids. He also considered the situation where  $2k_F$  falls to the right of the peak, and predicted a negative value for this term. Indeed, Cheung and Ashcroft (ref 12) deduced theoretically a positive PCR or perhaps a negative  $d\ln\rho/d\ln\Omega$  for polyvalent liquids Mg, Al, and Sn. The present results may be discussed in a similar manner: the last term in Eq. (2) for all Mg-based amorphous alloys studied here (for which  $2k_F$  falls slightly to the right of the main peak in the structure factor) is expected to be small, negative, and approximately constant, so that the variation of  $d\ln\rho/d\ln\Omega$  is determined essentially by the thermoelectric parameter as observed.

Figure 2 shows the relationship between PCR and  $\rho$  for the Mg-based group 5 alloys, along with group 4 data in the literature (refs 8,13-16). The value of PCR in group 4 alloys, which are characterized by high resistivities in the

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<sup>8</sup>G. Fritsch, J. Willer, A. Wildermuth, and E. Luescher, J. Phys., Vol. F12, 1982, pp. 2965-2974.

<sup>12</sup>J. Cheung and N. W. Ashcroft, Phys. Rev., Vol. B18, 1978, pp. 559-568.

<sup>13</sup>R. W. Cochrane, J. O. Strom-Olsen, J. P. Rebouillat, and A. Blanchard, Solid State Commun., Vol. 35, 1980, pp. 199-202.

<sup>14</sup>P. J. Cote and L. V. Meisel, Phys. Rev., Vol. B25, 1982, pp. 2138-2143.

<sup>15</sup>E. Luescher, J. Willer, and G. Fritsch, J. Non-Cryst. Solids, Vols. 61 and 62, 1984, pp. 1109-1114.

<sup>16</sup>J. Destry, M. El Amrani, and R. W. Cochrane, Phys. Rev., Vol. B31, 1985, pp. 2499-2501.

range 150-300  $\mu\Omega\text{cm}$ , is always negative and falls in the narrow range 0 to  $2 \times 10^{-3}/\text{kbar}^{-1}$ . In contrast, the value of PCR for the Mg-based group 5 amorphous alloys studied here is positive; however, it tends to decrease in magnitude as the resistivity increases and it seems to connect smoothly to the data for the high-resistivity group 4 alloys. As suggested by Cote and Maisel (ref 14), a correlation seems to exist between PCR and  $\rho$ . Moreover, we may say that a "Mooij-like" correlation exists in the PCR against  $\rho$  plots: a master curve is formed by the combination of results for the Mg-based group 5 alloys with those for group 4. Corresponding to variation from positive to negative TCR as  $\rho$  increases, the variation of the PCR is from positive and relatively large values in the group 5 Mg-based amorphous metals to negative and small values in the group 4 alloys as  $\rho$  increases. It is of interest to ascertain whether this correlation has some physical meaning or is merely accidental.

It is also interesting to show the present results in the form of  $d\ln\rho/d\ln\Omega$  against  $\rho$  graph and to compare this with the data for some typical liquid simple metals (ref 11). The data for the group 4 amorphous alloys are omitted. Here we find that the value of  $d\ln\rho/d\ln\Omega$  for the Mg-based group 5 amorphous alloys is always negative and can be fitted accurately to a straight line with a positive slope. Although the liquid data shown in Figure 3 exhibit positive  $d\ln\rho/d\ln\Omega$  values distributed over a wide range of resistivity, a negative value is also expected to occur in polyvalent liquids

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<sup>11</sup>T. E. Faber, Introduction to the Theory of Liquid Metals, Cambridge at the University Press, 1972, pp. 339-343.

<sup>14</sup>P. J. Cote and L. V. Maisel, Phys. Rev., Vol. B25, 1982, pp. 2138-2143.

like Mg, Al, and Sn (ref 12). Thus, no immediate correlation can be drawn between  $d\ln\rho/d\ln\Omega$  and  $\rho$  in the liquid metals. At this stage it is, therefore, not clear if the observed correlation in Mg-based amorphous alloys has some particular meaning. It may be worthwhile mentioning that the value of  $d\ln\rho/d\ln\Omega$  for pure crystalline simple metals, for which phonon scattering rather than elastic scattering is the major contributor to the resistivity, generally ranges from +3 to +6.

Before concluding, we must give one example which suggests that the systematic decrease in PCR with increasing  $\rho$  observed in the Mg-based amorphous metals is not a universal feature even among the group 5 amorphous alloys. Although the PCR of the amorphous Ca-Mg alloys has not been determined, there are good reasons to expect that it will be inconsistent with our master curves. Mizutani and Matsuda (ref 4) have determined that  $\rho = 44\mu\Omega$  cm,  $\text{TCR} = +0.84 \times 10^{-4}/\text{K}^{-1}$ , and the thermoelectric parameter  $\xi = -5$  for amorphous  $\text{Ca}_{70}\text{Mg}_{30}$ . If the measured value of  $\xi$  of -5 is accepted, then the master curve for Mg-based alloys in Figure 1 leads us to the value of  $d\ln\rho/d\ln\Omega$  of approximately -5. However, if the generalized Faber-Ziman theory incorporating the thermal-expansion effect, as given by Meisel and Cote (ref 7), is to describe electrical transport in Ca-Mg alloys, then a small or positive value of  $d\ln\rho/d\ln\Omega$  will be required. That is, the value of  $d\ln\rho/d\ln\Omega$

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<sup>4</sup>U. Mizutani and T. Matsuda, Proc. of Int. Conf. on Rapidly Quenched Metals, (S. Steeb and H. Warlimont, eds.), 1985, pp. 1035-1038.

<sup>7</sup>L. V. Meisel and P. J. Cote, Phys. Rev., Vol. B31, 1985, pp. 4872-4878.

<sup>12</sup>J. Cheung and N. W. Ashcroft, Phys. Rev., Vol. B18, 1978, pp. 559-568.

and the corresponding PCR required for a description of the temperature dependence of  $\rho$  lie far from the "master curve". Indeed, Hafner (ref 6) has predicted an approximately vanishing PCR for amorphous  $\text{Ca}_{70}\text{Mg}_{30}$  in a first principles pseudopotential calculation with no adjustable parameters. He also obtained a small positive TCR for this alloy in accord with his data and the measurements of Mizutani and Matsuda (ref 4) alluded to above.

Thus, the correlations seen in Figures 1 to 3 may be specific to the Mg-rich amorphous alloys studied here, for which the partial structure factors and scattering potentials do not differ drastically from one another. Further studies, particularly in the Ca-based amorphous alloys, are needed to clarify this interesting point.

#### SUMMARY AND CONCLUSIONS

The pressure coefficients of resistance PCR for a series of Mg-based amorphous metals have been determined. The corresponding values of  $d\ln\rho/d\ln\Omega$  were deduced employing "law of mixtures" values for the isothermal compressibility. The values of  $d\ln\rho/d\ln\Omega$  were shown to correlate with the thermoelectric parameter  $\xi$ . Both the PCR and  $d\ln\rho/d\ln\Omega$  were shown to correlate with  $\rho$ . A master curve was deduced which described the variation of the PCR with  $\rho$  in the Mg-based amorphous alloys and the group 4 amorphous alloys. However, results were cited which suggest that the master curve description will not hold when all group 5 amorphous metals are included and

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<sup>4</sup>U. Mizutani and T. Matsuda, Proc. of Int. Conf. on Rapidly Quenched Metals, (S. Steeb and H. Warlimont, eds.), 1985, pp. 1035-1038.

<sup>6</sup>J. Hafner, J. Non-Cryst. Solids, Vol. 69, 1985, pp. 325-346.

that an envelope description, as suggested earlier by Cote and Meisel (ref 14), may be required to correlate all the group 4 and group 5 amorphous metals.

The values of the PCR or of  $d\ln\rho/d\ln\Omega$  have important consequences in regard to the consistency of theory and experiment of electrical transport. The present results, in particular, yield significant thermal-expansion contributions to  $\rho$  which dramatically improve agreement between generalized Faber-Ziman theory and experiment.

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<sup>14</sup>P. J. Cote and L. V. Meisel, Phys. Rev., Vol. B25, 1982, pp. 2138-2143.

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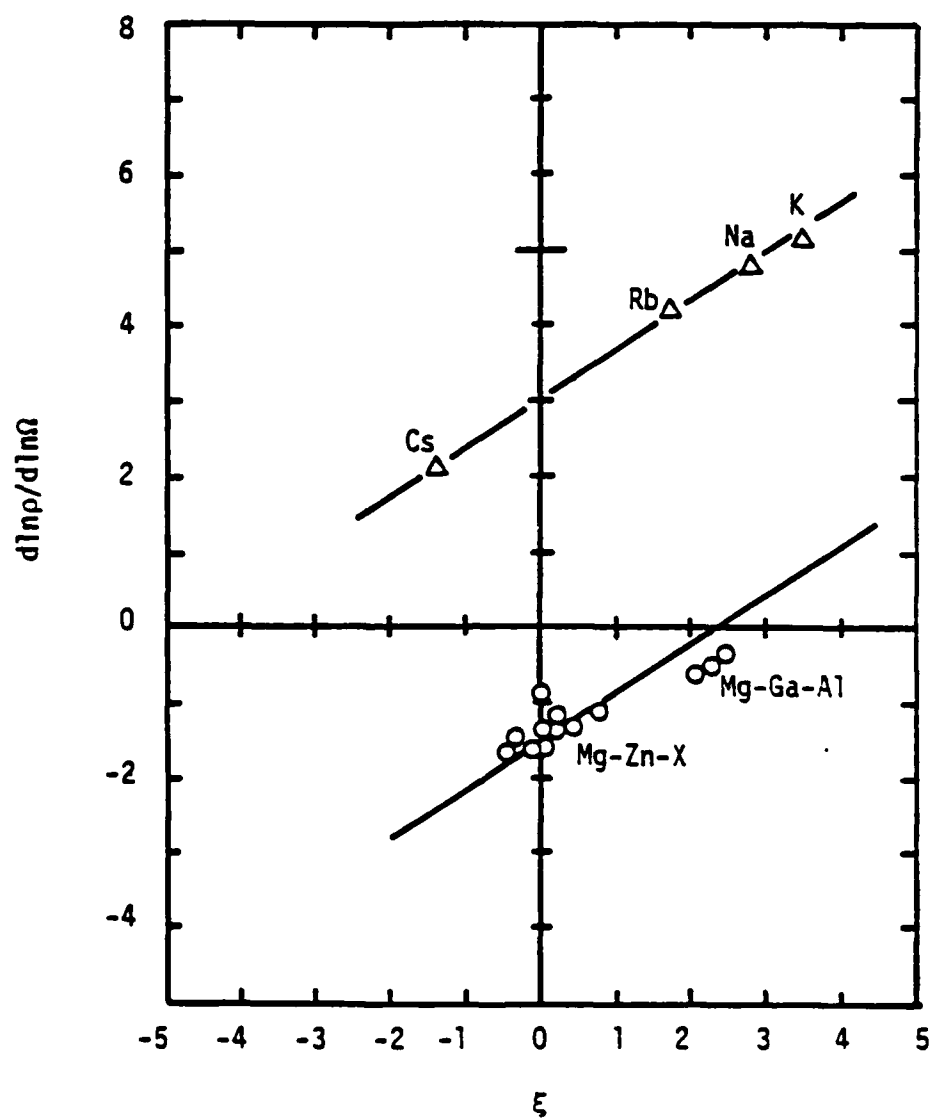


Figure 1. Plot of  $d\ln\rho/d\ln\Omega$  against  $\xi$  for liquid alkali metals (open triangles) and simple amorphous alloys (open circles).

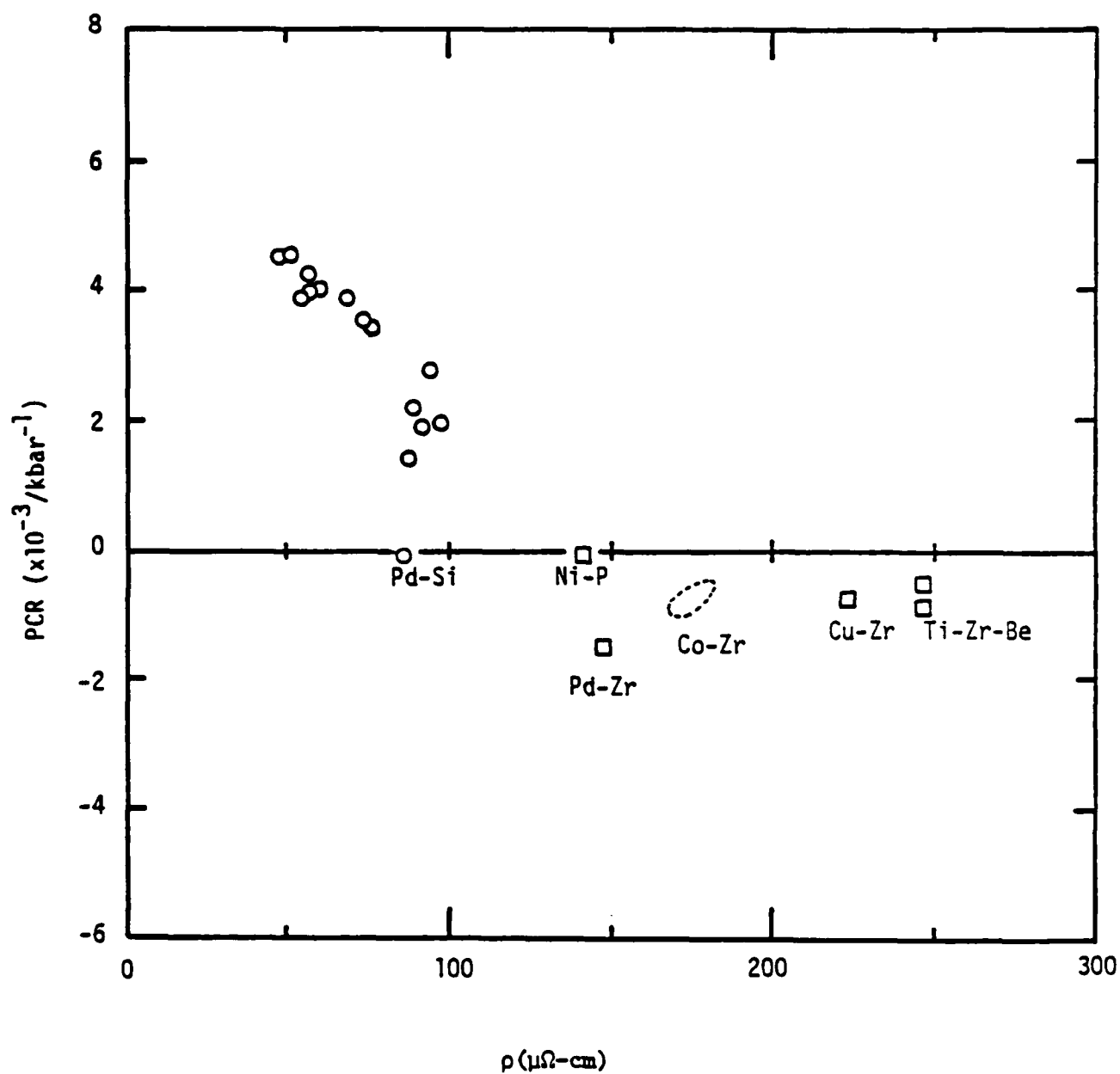


Figure 2. Plot of PCR against  $\rho$  for Mg-based alloys and for various group 5 alloys (labelled points).

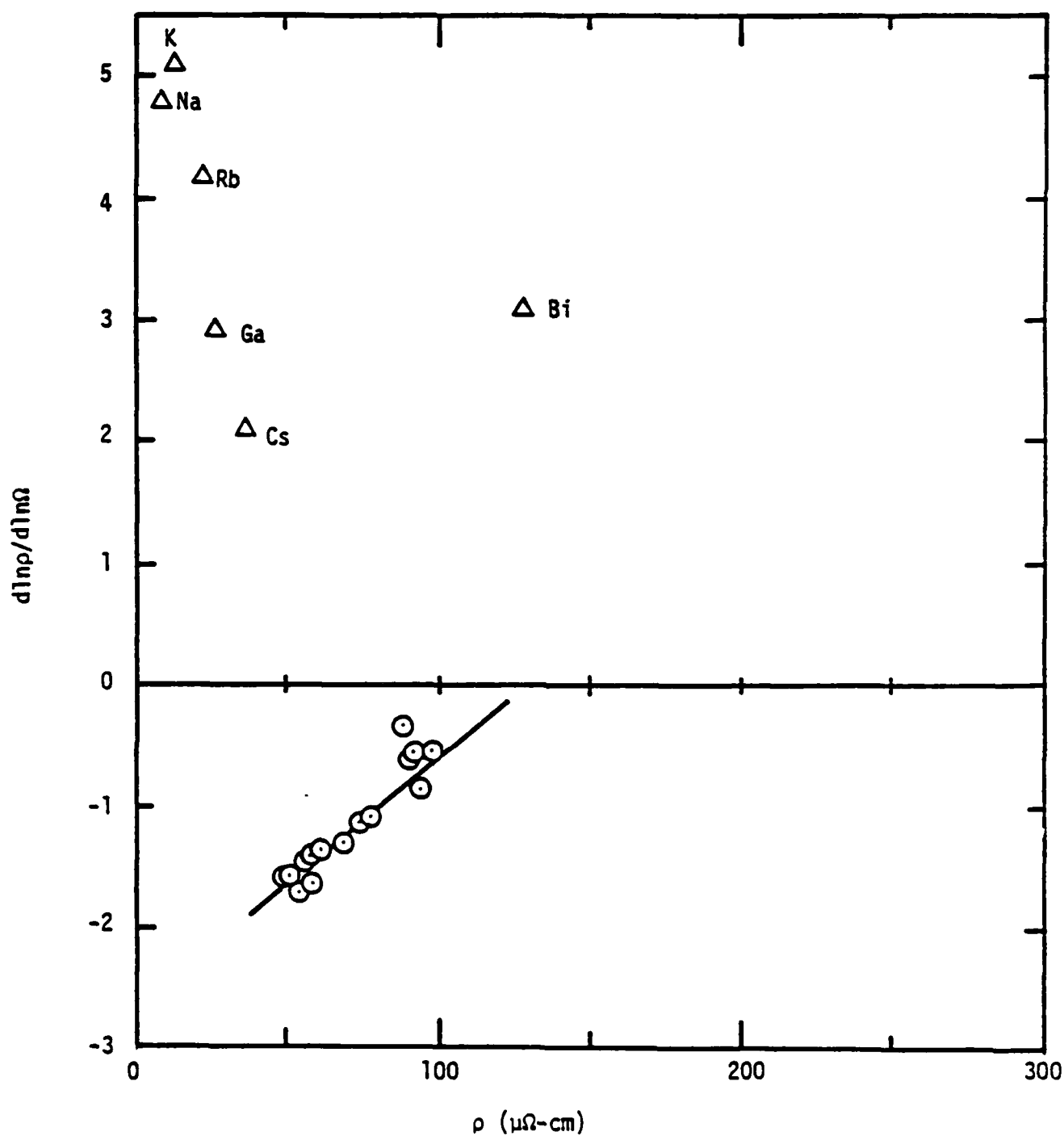


Figure 3. Plot of  $d\ln\rho/d\ln\Omega$  against  $\rho$  (open triangles) show the spread of values for liquid metals and (open circles) show the correlation for Mg-based amorphous alloys.

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