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Electrical Resistivity and Its Temperature Dependence of Al-base Quasicrystalline and Crystalline Alloys*

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Synopsis

Al-Mn and Al-Cr quasicrystalline and Al-Mn crystalline alloys were prepared by melt-quenching and their electrical resistivities were studied. The quenched samples exhibited an extremely high resistivity because of nonperiodic potential scattering and resonance scattering of the Fermi electrons. The temperature dependence of electrical resistivity of Al-22.5%Mn quasicrystalline was tried to fit by several models, and it was found that the plots of $\ell n T$ (from 4.2 to 20 K) and T^2 (from 20 to 60 K) showed a straight line. The electrical resistivity of Al_6Mn crystalline alloy showed a linear temperature dependence over wide high temperatures, but that of Al_4Mn crystalline alloy showed a deviation from the linearlity. The values of ρ of the quasicrystalline alloys were larger than those of the crystalline counterparts.

I. Introduction

Recently, Schechtman et al. have demonstrated that Al-14 at%Mn quenched alloy exhibits six fivefold symmetries in the electron diffraction pattern$^{1)}$. Such an unusual discovery conflicts with conventional crystallographic theorem in the solid state physics. Therefore, the structural study of the quasicrystalline alloys has attracted the attention of many researchers, and various models of the structure have been proposed in order to explain the fivefold symmetry 2-6). However, the study on the magnetic and electrical properties is not so extensive. More recently, the present authors have reported

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that the magnetic moments of Al-Mn quasicrystalline and crystalline alloys are not so large (from 0.28 to 1.52 μ_B) in the composition range between 14.3 and 22.5 at%Mn⁷⁾, compared with the values of other many alloys containing Mn such as Heusler-type amorphous alloys⁸⁾. The electrical resistivity of the same alloys is extremely large and its temperature coefficient is significantly small⁷,⁹⁾ in the similar manner as those of various kinds of amorphous alloys¹⁰⁾. In order to investigate the physical properties of Al-base quasicrystals in more detail, the systematic study of electrical and magnetic properties of these alloys has been performed by us.

The present paper gives the results and discussion of the magnitude of electrical resistivity and its temperature dependence of Al-Mn, Al-V and Al-Cr quasicrystalline alloys because it is expected that the electrical properties are closely correlated with the structure and electronic state in these alloys. Their magnetic moment and the spin glass behavior will be reported elsewhere¹¹⁾.

II. Experimental

Al-14.3, 20 and 22.5 at%Mn, Al-14.3 at%V and Al-15.4 at%Cr mother alloys were prepared by arc melting in an argon atmosphere using Al (99.99 wt%) and Mn (99.9 wt%), V (99.9 wt%) and Cr (99.8 wt% in purity) as starting materials. The quasicrystalline alloys were obtained by rapid quenching in an argon atmosphere from the melt using a single roller apparatus and the crystalline alloys were obtained by annealing these quenched samples at 973 K for 1 hr. The X-ray diffraction experiment for the quenched and annealed alloys was made using Cu-K_α radiation at room temperature. The temperature dependence of electrical resistivity was measured from 4.2 K to 250 K by a conventional four terminal method. The sample thickness was measured with a micrometer.

III. Results and discussion

Figure 1 shows the temperature dependence of electrical resistivity of Al-Mn quenched alloys, together with the data of Al_6Mn, Al_4Mn and Al-22.5 at%Mn crystallized alloys. The heat-treatment was made at 97.3 K because the transformation temperatures to Al_6Mn and Al_4Mn of Al-14.3 at%Mn and Al-20 at%Mn alloys are about 720 K and 870 K, respectively⁷⁾. It has been demonstrated that the quenched alloys composed of mainly a quasicrystalline phase have extremely high resistivity in a wide temperature range⁷,⁹⁾. Especially, Al-22.5

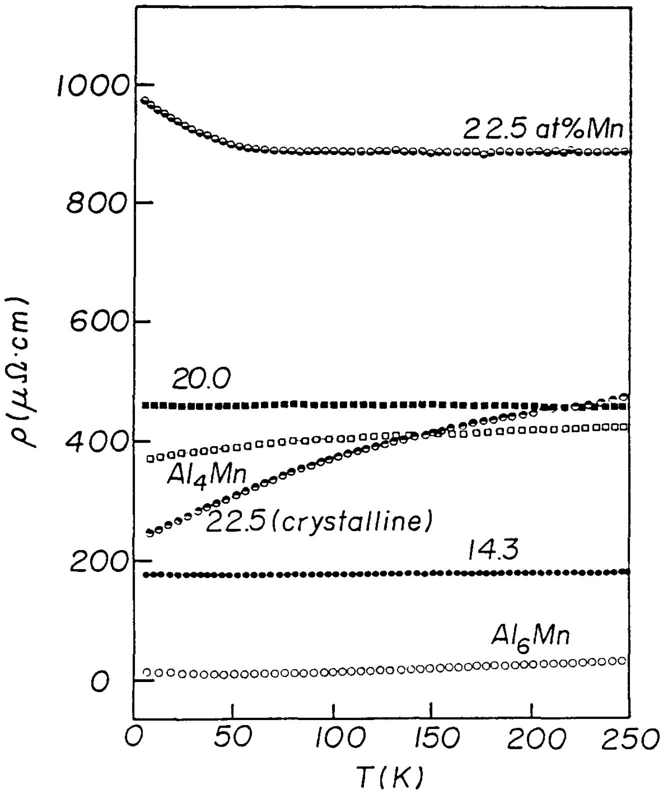

Fig. 1 Temperature dependence of electrical resistivity of Al-Mn quenched alloys, together with the data of Al_6Mn, Al_4Mn and Al-22.5 at%Mn crystallized alloys.

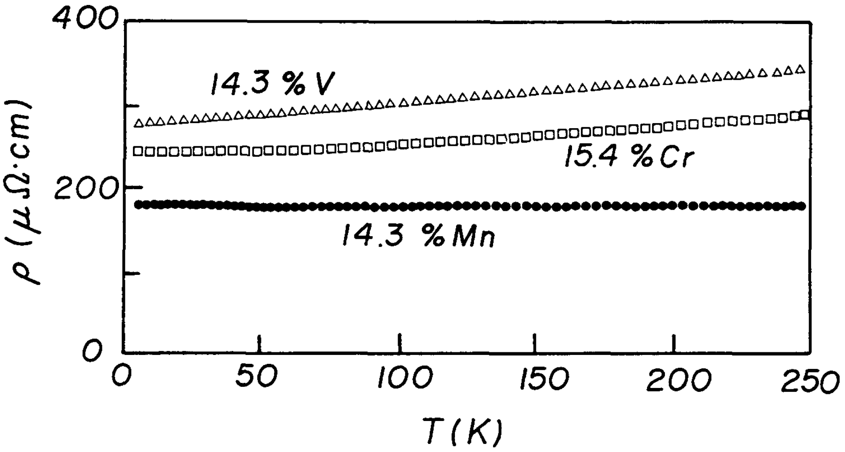

Fig. 2 Temperature dependence of electrical resistivity of Al-15.4 at%Cr, Al-14.3 at%Mn and Al-14.3 at%V quenched alloys.

at%Mn alloy exhibits a negative temperature coefficient with a high absolute value at low temperatures as shown in the figure. The extreme high value of resistivity would be explained by taking into consideration of the resonance state of the Fermi electrons and the non-periodic potential$^{7)}$. That is, the observed value of resistivity ρ would be decomposed into two components due to structural and s-d resonance scatterings and given by

$$\rho = \rho_{structure} + \rho_{s-d} .$$

As is well known, the resistivity of amorphous alloys having a non-periodic potential exhibits a very high value$^{10)}$. Therefore it is expected that the same situation occurs in the quasicrystalline alloys. According to Friedel's model$^{12)}$, on the other hand, a resonance state of the Fermi electrons with the d-state of Mn should take place when Mn atoms are dissolved in Al because the center of the virtual bound state is very close to the Fermi level, resulting in a high electrical resistivity. From this model, it is expected that the values of Al-Cr and Al-V alloys are also very high. In order to confirm this fact, in the present study, the electrical resistivity data of Al-15.4 at%Cr quenched alloy is shown in Fig. 2, together with those of Al-14.3 at%V and Al-14.3 at%Mn quenched alloys$^{9)}$. As is expected, these values are comparable with one another at a very high level. However, quantitative analysis is not possible because these alloys contain a small amount of Al crystalline phase. The model mentioned above also predicts that the value of Al-Fe alloy is relatively low, comparing with those of three kinds of alloys mentioned above. It would be interesting to note that Ca-Al amorphous alloys which have d-band character states at the Fermi level also exhibit a very high resistivity$^{13,14)}$, comparing to the value of Ca-Mg amorphous alloy having a simple band character$^{14)}$.

Going back to Fig. 1, the temperature dependence of the crystalline samples annealed at 973 K exhibits a different behavior, that is, Al_6Mn crystallized alloy shows a linear dependence in a wide temperature range, and the magnitude of resistivity becomes very small. On the other hand, other alloys of the high Mn concentration exhibit a deviation from the linearity, indicating the so-called "saturation". The former dependence is a common behavior associated with the phonon scattering. In this mechanism, the scattering probability depends on the mean square amplitude of the lattice vibrations and it is proportional to the temperature in high temperature range. The latter behavior is often observed in d-band metals and high resistive metals as well. The downward turn deviating from the linearity seems to approach to the Mott maximum metallic resistivity associated with a very short electron mean-free-path15). However, recent theoretical consideration on the scaling theory denies this explanation16). Allen has reviewed the theories of the resistivity saturation, and pointed out that there are several kinds of models for the explanation of such a saturation17). A similar behavior occurs in Co_2Y enhanced paramagnetic compound, and it is attributed to the scattering of the electrons by spin fluctuation18). Although the present study does not provide a conclusive result, it would be considered that s-d scattering process19) appears in the present alloy system, and this process is not dominant around 14.3 at% but becomes remarkably strong with increasing Mn content, resulting in a positive curvature in the temperature dependence curve. In Al-Mn alloy system, the concentration dependence of resistivity at 4.2 K drastically increases above 20 at%Mn due to the disappearance of Al crystalline phase9). Therefore, the discussion on the samples below 20 at%Mn is physically somewhat meaningless. Then, in the present paper, the temperature dependence of electrical resistivity of Al-22.5 at%Mn quasicrystalline alloy is discussed hereafter.

In the present study, the thickness of specimens has been meassured with a micrometer. However, strictly speaking, the thickness is often over-estimated due to a surface roughness of the specimen20). In the case of metallic ribbon samples, if we reduce about 20∿30%21), the magnitude of resistivity is in good agreement with the value of the sputtering films. Therefore, the magnitude of ρ of Al-Mn alloys would be smaller than the values shown in the figure. As seen from Fig. 1, this alloy exhibits a negative temperature dependence in a wide temperature range. Several kinds of models have been proposed in order to explain such behavior. The variable range hopping model22) and the percolation theory23) give a linear relationship between $\ell n\ \rho$ and $1/T^{1/4}$, and as is well known $\ell n\ \rho$ vs. $1/T$ plot shows a linear relation in semiconductors. The former relations are often observed in highly disordered materials. Figure 3 shows $\ell n\ \rho$ vs. $1/T^{1/4}$ plot for Al-22.5 at%Mn quenched alloy, but a straight line is not obtained. Similar inadequate result is obtained for $\ell n\ \rho$ vs. $1/T$ plot. Therefore, it is clear that such conduction mechanism does not occur in the present alloy.

Recently, the increase of resistivity at low temperatures has been often observed in various kinds of amorphous alloys for not only dilute but also concentrated ferromagnetic alloys. This phenomenon is

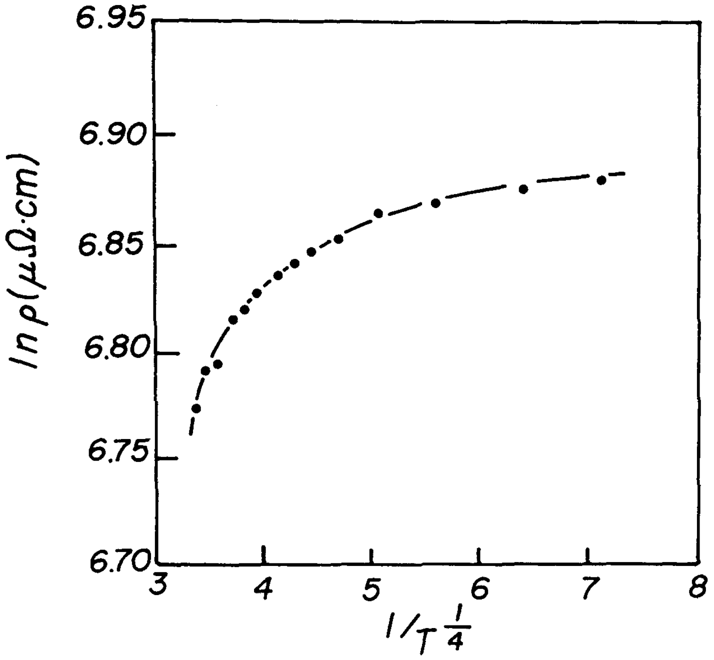

Fig. 3 The plot of $\ln\rho$ vs. $1/T^{1/4}$ for Al-22.5 at%Mn quenched alloy.

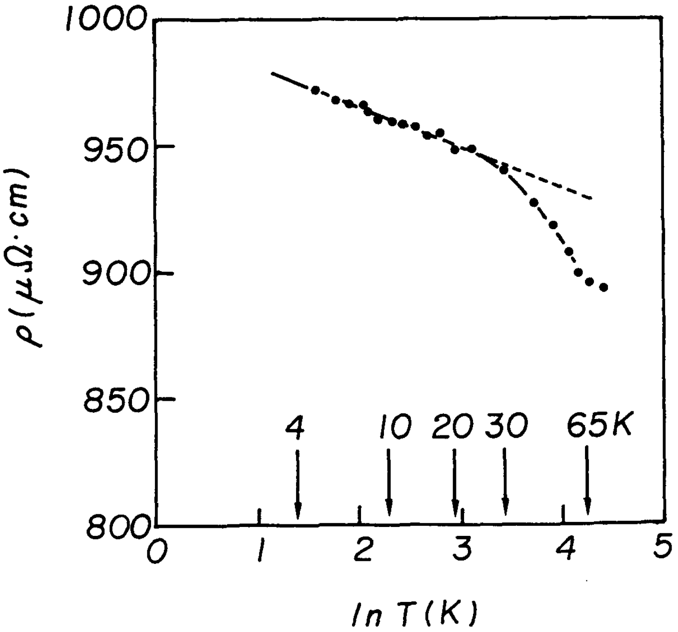

Fig. 4 The plot of ρ vs. $\ln T$ for Al-22.5 at%Mn quenched alloy.

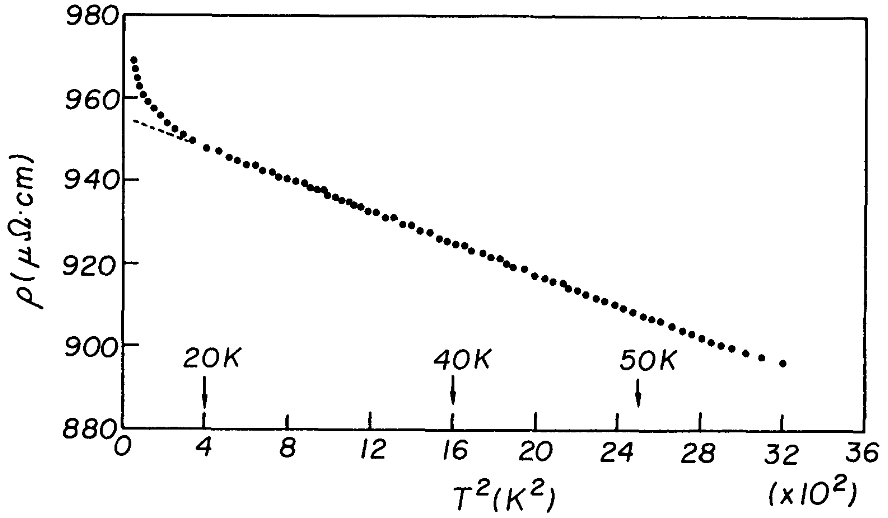

Fig. 5 The plot of ρ vs. T^2 for Al-22.5 at%Mn quenched alloy.

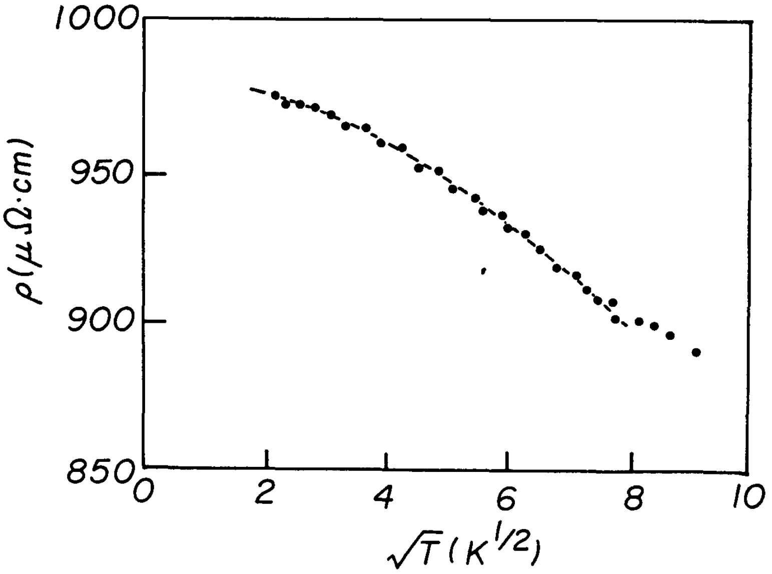

Fig. 6 The plot of ρ vs.$\sqrt{T}$ for Al-22.5 at%Mn quenched alloy.

also explained by several kinds of models, and many alloys exhibit the ℓnT or $\sqrt{T}$ dependence at low temperatures. The ℓnT dependence is explained by the Kondo effect[24)] or the two level tunneling model[25)], and the $\sqrt{T}$ dependence is associated with the Anderson localization model[16,26)] or the electron-electron interaction model[27)]. The ℓnT plot is given in Fig. 4 and the linear relationship is observed below about 20 K. Above 20 K, on the other hand, the T^2 plot gives a straight line in a wide temperature range as shown in Fig. 5. Such ℓnT plus T^2 dependences are observed in many amorphous alloys such as Ni-Pd-P[28)], Nb-Ni[29)] and Cu-Zr alloys[30)]. The T^2 dependence is expected from the extended Ziman model[31)] or the Mott s-d scattering model[32)]. Finally, the temperature dependence of resistivity is plotted as a function of $\sqrt{T}$. The result shows slightly conves upward as shown in Fig. 6, hence, the fitting is not so good as compared with T^2 and ℓnT plots. Therefore, it is concluded that dependence of ρ of Al-22.5 at%Mn quasicrystalline alloy is plotted most suitably by ℓnT (below 20 K) and T^2 (above 20 K). However, the distinct conclusion which mechanism fits to the temperature dependence can not be drawn from the present data. The measurements such as magnetoresistance, Hall effect and ultrasonic attenuation would give good information to decide which of several models mentioned above is more closely related to the experimental result.

Summary

Electrical resistivity and its temperature dependence of Al-Mn, Al-15.4 at%Cr and Al-14.3 at%V quasicrystalline and Al_6Mn and Al_4Mn crystalline alloys have been investigated. The reason why the electrical resistivity is high in the quasicrystalline alloys and the positive curvature of the resistivity occurs in the crystalline alloys have been discussed. The four kinds of plots of the temperature dependence of electrical resistivity ρ of Al-22.5 at%Mn quasicrystalline alloy have been tried using the existing models. Main results are summarized as follows:

(1) The magnitude of the electrical resistivity of the present samples is extremely high due to the quasicrystalline structure and the resonance state of Al with the transition metals such as Mn, Cr and V.

(2) The deviation from linearity in the ρ vs T plot of crystalline alloys is observed in the high concentration of Mn.

(3) The temperature dependence of Al-22.5at%Mn quasicrystalline alloy

is fitted linearly by ℓnT (below 20 K) and T^2 (from 20 K to 60 K).

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