ELECTRICAL RESISTIVITIES OF METALLIC MUTLILAYERS.

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ABSTRACT

We calculated the resistivity and TCR of multilayers by the model analogous with the Fuchs-Sondheimer theory and compared the calculated dependence of resistivity and TCR on bilayer thickness with the experimental results. The bilayer thickness dependence of Au/Pd, Al/Ag, Mo/Ta, Pd/Co and Ag/Co was governed by the interface scatterings. All the conduction electrons were scattered diffusely at the interfaces in the multilayers which have fcc (111) and bcc (110) planes parallel to the surface. The slope of the increase of resistivity of Nb/Cu, Mo/Al, Nb/Al and Cu/Ta with the inverse bilayer thickness was very large and negative TCR was observed in these multilayers. The large increase of resistivity and negative TCR could be due to the scattering at the grain boundaries.

INTRODUCTION

A lot of studies of metallic multilayered films have been performed and several novel properties were reported. For example, perpendicular magnetic anisotropy in noble-metal/Cobalt multilayers is a subject of great technological interest. Recently, giant magnetoresistance effects have been found in magnetic layered structures with antiferromagnetic couplings[1]. When the antiferromagnetically coupled adjacent layers are brought into parallel alignment by an external magnetic field, the resistance drops. In some cases this decreases is over 40%[2]. The existence of this giant magnetoresistance in multilayers is promising for applications to magnetoresistance sensors.

Most of the novel properties arose from the introducing interfaces in metallic multilayers. The electrical transport properties of metallic multilayered films have expected to have a particular behavior because of electron scattering at the interfaces. Several resistivity measurements have been performed on metallic multilayers[3–16]. But only a few theoretical calculations on this subject have appeared[17–20] and quantitative comparison experimental results with the theories has not been made.

Here we present a simple theoretical model which reproduced dependence of resistivity and temperature coefficient of resistance (TCR) of multilayers on bilayer thickness. Furthermore, the parameters obtained from the comparison the experimental data with the model calculation give some general features of electrical conduction in metallic multilayers.

CALCULATION MODEL

We calculated the resistivity and TCR of multilayers by a model analogous with the Fuchs-Sondheimer theory[21,22] which have been applied to calculate the resistivity of thin films. The details of the calculation are published in a separate paper[23]. We only give the outline of the calculation model here.

The multilayered films were assumed to have only two different metals, alternatively grown on each other with constant layer thickness d_1 and d_2, where $\Lambda = d_1 + d_2$ was called bilayer thickness.

Films extend infinitely in the $x-y$ plane and the stacking direction is parallel to z axis. The Boltzmann equation in each layer has the form

$$\vec{v} \cdot grad_r g - \frac{e\vec{E}}{m^*} grad_v f^0 = -\frac{g}{\tau} . \tag{1}$$

We introduced the transmission parameter t and the reflection parameter r to take electron scattering at interfaces into account. For simplicity of calculation electrons assumed to have the same probability for specular transmission from metal 1 to metal 2 and from metal 2 to metal 1. The reflection coefficients for the metal 1 and 2 were also assumed to be the same. Taking the conservation of electrons at the interfaces into account as a boundary condition, we could solve the Boltzmann equation in each layer. The electrical conductivities of metal 1 and metal 2 are

$$\begin{aligned}\frac{\sigma_1}{\sigma_1^0} &= 1 + Q_1 \\ &= 1 + \frac{3}{2k_1} \int_0^1 F_1(\xi - \xi^3) \left\{1 - \exp\left(-\frac{k_1}{\xi}\right)\right\} d\xi ,\end{aligned} \tag{2}$$

and

$$\begin{aligned}\frac{\sigma_2}{\sigma_2^0} &= 1 + Q_2 \\ &= 1 + \frac{3}{2k_2} \int_0^1 F_2(\xi - \xi^3) \left\{1 - \exp\left(-\frac{k_2}{\xi}\right)\right\} d\xi ,\end{aligned} \tag{3}$$

where

$$\begin{aligned}F_1 &= -\frac{1}{G}\Bigg[(1 - r - t2C_{21})\left\{1 + r\exp\left(-\frac{k_1}{\xi_1}\right)\right\} \\ &\quad + (1 - r - tC_{12})tC_{21}\exp\left(-\frac{k_2}{\xi_2}\right) \\ &\quad + r\{tC_{21}(1 - tC_{12}) - r(1 - r)\}\exp\left(-2\frac{k_2}{\xi_2}\right) \\ &\quad + tC_{21}\{r(1 - r) - tC_{12}(1 - tC_{21})\}\exp\left(-\frac{k_1}{\xi_1} - \frac{k_2}{\xi_2}\right) \\ &\quad - (t^2 - r - 2)\left\{tC21(1 - tC_{12}) - r(1 - r)\right\}\exp\left(-\frac{k_1}{\xi_1} - 2\frac{k_2}{\xi_2}\right)\Bigg] ,\end{aligned} \tag{4}$$

$$\begin{aligned}F_2 &= -\frac{1}{G}\Bigg[(1 - r - tC_{12})\left\{1 + r\exp\left(-\frac{k_2}{\xi_2}\right)\right\} \\ &\quad + (1 - r - tC_{21})tC_{12}\exp\left(-\frac{k_1}{\xi_1}\right) \\ &\quad + r\{tC_{12}(1 - tC_{21}) - r(1 - r)\}\exp\left(-2\frac{k_1}{\xi_1}\right) \\ &\quad + tC_{12}\{r(1 - r) - tC_{21}(1 - tC_{12})\}\exp\left(-\frac{k_1}{\xi_1} - \frac{k_2}{\xi_2}\right) \\ &\quad - (t^2 - r^2)\{tC12(1 - tC_{21}) - r(1 - r)\}\exp\left(-2\frac{k_1}{\xi_1} - \frac{k_2}{\xi_2}\right)\Bigg] ,\end{aligned} \tag{5}$$

$$\begin{aligned}G &= 1 - r^2\exp\left(-2\frac{k_1}{\xi_1}\right) - r^2\exp\left(-2\frac{k_2}{\xi_2}\right) \\ &\quad - 2t^2\exp\left(-\frac{k_1}{\xi_1} - \frac{k_2}{\xi_2}\right) \\ &\quad + (t^2 - r^2)^2\exp\left(-2\frac{k_1}{\xi_1} - 2\frac{k_2}{\xi_2}\right) ,\end{aligned} \tag{6}$$

$$C_{12} = \frac{\tau_1 m_1^*}{\tau_2 m_2^*} = \frac{1}{C_{21}}, \tag{7}$$

$$k_i = \frac{d_i}{\lambda_i}\,(i = 1,2), \tag{8}$$

where σ_1^0 and σ_2^0 are the conductivities of the infinitely thick film (having the same structure) of metal 1 and metal 2, respectively. These σ_1^0 and σ_2^0 would not have the same values for the ideal single crystals. Because the scatterings of conduction electrons at grain boundaries and impurities and other defects increase the resistivity of the films. Q_1 and Q_2 describe the size effects of the multilayered film.

The resistivity σ_m and ρ_m of the whole multilayer can be calculated by

$$\sigma_m = \frac{1}{\rho_m} = \frac{d_1\sigma_1 + d_2\sigma_2}{d_1 + d_2}. \tag{9}$$

In the calculation of temperature coefficient of resistance (TCR) it was assumed that the change of the thickness of the film by thermal expansion and the effects of the strain arose from the difference of the coefficient of thermal expansion between the film and the substrate could be negligible.

Within a free electron model, the product of the resistivity rho and the mean free path λ of a bulk metal is constant and depends only on the density of the conduction electrons. So the temperature coefficient of resistance (TCR) beta of the metal is

$$\beta = \frac{1}{\rho}\frac{d\rho}{dT} = -\frac{1}{\sigma}\frac{d\sigma}{dT} = -\frac{1}{\lambda}\frac{d\lambda}{dT}. \tag{10}$$

When the conductivity of the layer of metal 1 σ_1 is given by the equation (2), TCR of the layer of metal 1 is

$$\beta_1 = -\frac{1}{\sigma_1}\frac{d\sigma_1}{dT} = \beta_1^0 - \frac{1}{1+Q_1}\frac{dQ_1}{dT}, \tag{11}$$

where β_1^0 is TCR of the infinitely thick film of metal1. Consequently

$$\beta_1 = \beta_1^0 - \frac{1}{1+Q_1}\frac{\partial Q_1}{\partial k_1}k_1\beta_1^0 + \frac{\partial Q_1}{\partial k_2}k_2\beta_2^0 \tag{12}$$

and

$$\beta_2 = \beta_2^0 - \frac{1}{1+Q_2}\frac{\partial Q_2}{\partial k_1}k_1\beta_1^0 + \frac{\partial Q_2}{\partial k_2}k_2\beta_2^0. \tag{13}$$

TCR of the multilayer β_m is given by

$$\beta_m = \left(\frac{d_1\sigma_1\beta_1 + d_2\sigma_2\beta_2}{d_1\sigma_1 + d_2\sigma_2}\right). \tag{14}$$

RESULTS AND DISCUSSIONS

A number of resistivity measurements have been performed on metallic multilayers. We have calculated the bilayer thickness dependence of resistivity and TCR of the several kinds of multilayered films and have made the quantitative comparisons the experimental results with the calculations.

The resistivities ρ_1^0, ρ_2^0, the mean free paths λ_1, λ_2, the temperature coefficients of resistance β_1^0, β_2^0 of the very thick films of metal 1 and metal 2 should be known to calculate the resistivity and TCR of the multilayer. We also have need for the values of electron mass m^* and the Fermi velocity

Table I: The parameters used for the calculation.

	V_F $(10^8 cm/s)$	m^*/m	$\rho^0\beta^0$ $10^{-3}\mu\Omega cm K^{-1}$	$\rho^0\lambda$ $\mu\Omega cm Å$
Al	1.32	1.13	9.9614	545
Ti	0.30	1.30	187.64	580
Co	0.30	4.38	21.067	293
Ni	0.3	5.0	30.974	350
Cu	1.13	1.34	6.12	741
Nb	0.63	3.22	53.0	412
Mo	0.81	1.93	22.615	679
Pd	0.35	6.06	38.233	370
Ag	1.45	0.932	5.8407	917
Ta	0.65	2.93	45.767	460
Au	1.32	1.025	7.6545	920

V_f for metal 1 and metal 2. The parameters used for the calculation are listed in Table I. The values of the Fermi velocity were obtained from the results of the calculation by Papaconstantpoulus[24]. The values of electron mass were estimated by the nearly free electron model to reproduce the density of state, Fermi velocity, Fermi energy and plasma energy of the metal. The products $\rho^0\lambda$ were calculated from the Fermi velocity and the plasma frequency of the bulk metal by

$$\rho^0\lambda = \frac{mV_F}{ne^2} = \frac{4\pi V_F}{\Omega_p^2} \tag{15}$$

We assumed that the product $\rho^0\beta^0$ is the same value for the ideal bulk metal, $\rho^0\beta^0 = \rho^b\beta^b$. The bulk values ρ^b and β^b were obtained from the data collected by Meaden[25]. The resistivities of the infinitely thick film of metal1 and metal2, ρ_1^0 and ρ_2^0, the transmission parameter t and the reflection coefficient r were used as the adjustable parameters.

The parameters obtained from the fitting calculations are listed in Table II. The results of the calculations for two kinds of Au/Pd multilayer[6,13], Al/Ag[14] and Mo/Ta[10] reproduce well the bilayer thickness dependence of resistivity and TCR of the multilayers. Figure 1 shows the experimental and calculated dependence of in-plane resistivity and TCR of Au/Pd synthesized by Carcia et al.[6] on inverse bilayer thickness $1/\Lambda$. The transmission parameter t was 0.94 and the reflection coefficient r was 0.05 for the Au/Pd multilayer. Very few electrons are scattered diffusely at the interfaces in the Au/Pd. The resistivity of the Pd layer was extraordinarily high in stead of the interface scatterings.

We were also able to fit the results of the calculation to the experimental data of the Au/Pd multilayers deposited by de Vries et al[13]. The parameters of this Au/Pd obtained from the fitting calculations differed from those of the Au/Pd by Carcia et al. The difference of the parameters would be due to the difference of the deposition method between the two Au/Pd. Carcia et al. synthesized Au/Pd by r.f. spattering and de Vries et al. used a method of the vacuum evaporation.

The observed TCR of the Pd/Co multilayers prepared by Carcia et al.[8] lie on the calculated line of the bilayer thickness dependence. On the contrary the observed values of the resistivity scattered. Commonly the value of TCR have a smaller amount of error than that of the resistivity. Because of TCR do not include the absolute value of the resistivity and is a ratio of the temperature change. It seems that the absolute values of the resistivity of the Pd/Co multilayer by Carcia et al. have a large amount of error.

The calculated bilayer thickness dependence of resistivity and TCR of Mo/Cu were well fitted to the experimental results[12] (Fig. 2). In this Mo/Cu multilayer both the specular transmission

Table II: the specular transmission probability, specular reflection probability and mean free path provide the best fit to exprimental resuls.

	Ref.	t	r		ρ^0 $\mu\Omega cm$	λ Å
Au/Pd	[6]	0.94	0.05	Au	1.37	671
				Pd	174	2.13
Au/Pd	[13]	0.20	0.43	Au	2.94	313
				Pd	17.8	20.8
Al/Ag	[14]	1.0	0.0	Al	90.7	6.0
				Ag	5.78	159
Mo/Ta	[10]	0.64	0.36	Mo	16.7	40.6
				Ta	116	3.98
Ag/Co	[11]	0.09	0.35	Ag	3.64	252
				Co	10.3	28.4
Pd/Co	[8]	0.0	0.42	Pd	19.1	19.3
				Co	21.9	13.4
Al/Ni	[26]	0.0	0.0	Al	27.3	20.0
				Ni	121	2.47
Cu/Ni	[15]	0.0	0.0	Cu	10.36	71.5
				Ni	140	2.50
Mo/Ni	[7]	0.0	0.0	Mo	24.4	27.8
				Ni	140	2.5
Nb/Ti	[3]	0.0	0.0	Nb	144.4	2.85
				Ti	59.3	9.78
Nb/Cu	[4]	0.0	0.0	Nb	144.4	2.85
				Cu	13.3	55.7
Nb/Cu	[5]	0.0	0.0	Nb	144.4	2.85
				Cu	10.5	70.8
Nb/Al	[9]	0.0	0.0	Nb	144.4	2.85
				Al	11.0	49.7
Cu/Ta	[15]	0.0	0.0	Cu	4.1	183
				Ta	461.4	2.85
Mo/Cu	[12]	0.0	0.0	Cu	8.97	82.6
				Mo	64.0	10.6
Mo/Al	[16]	0.0	0.0	Mo	16.9	40.1
$\Lambda > 50$Å				Al	33.4	16.3
Mo/Al	[16]	0.0	0.0	Mo	76.1	8.93
$\Lambda < 50$Å				Al	190.56	2.86

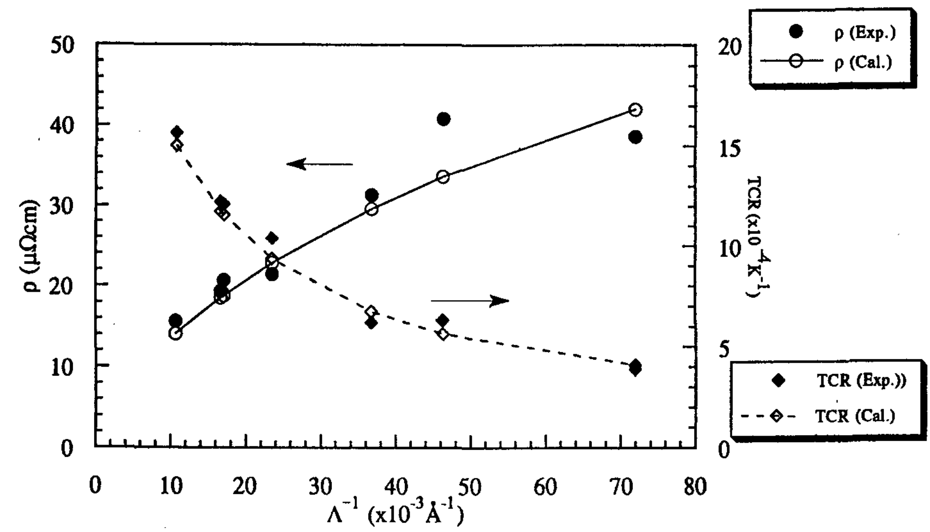

Figure 1: The bilayer thickness dependence of resistivity and TCR of the Au/Pd multilayer. The solid circles and the solid triangles are the observed values replotted from [6]. The open circles and the open triangles are the best fit to the observed data.

probability and the specular reflection probability became zero in the result. In other words all the conduction electrons were scattered diffusely at the interfaces. The multilayers which have fcc (111) and bcc (110) planes parallel to the surface, such as Mo/Al and Nb/Cu, had the same tendency to Mo/Cu. Namely they have no probability of the specular transmission and reflection.

The multilayered films which were constituted by Ni and another metal showed high values of the resistivities and TCR of the multilayers decreased drastically as the bilayer thickness decreased. Figure 3 shows the experimental and calculated dependence of the resistivity of the Mo/Ni multilayers[7] on inverse bilayer thickness and the relationship between TCR and the bilayer thickness is shown in Fig. 4. The results of the calculations could not reproduce the sudden decrease of

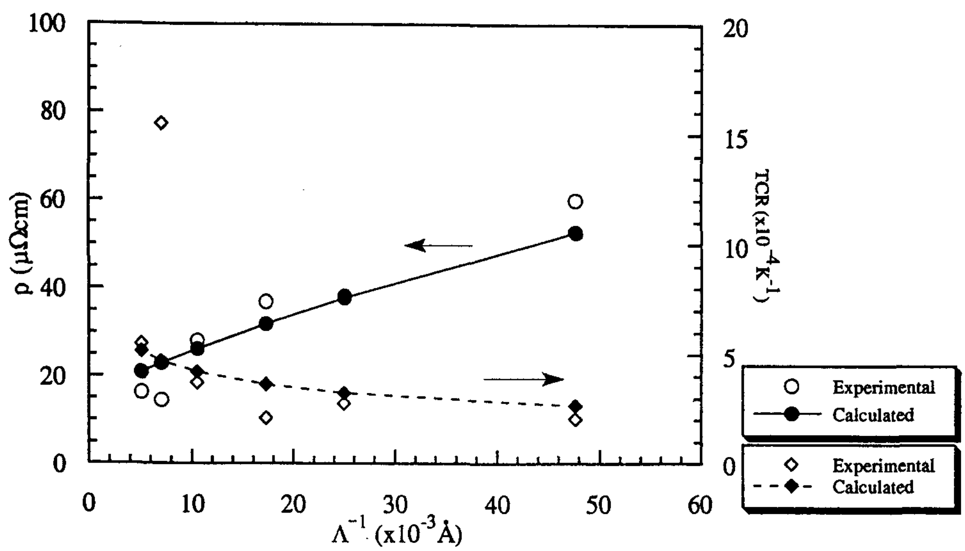

Figure 2: The bilayer thickness dependence of resistivity and TCR of Mo/Cu prepared by a metal-MBE method.

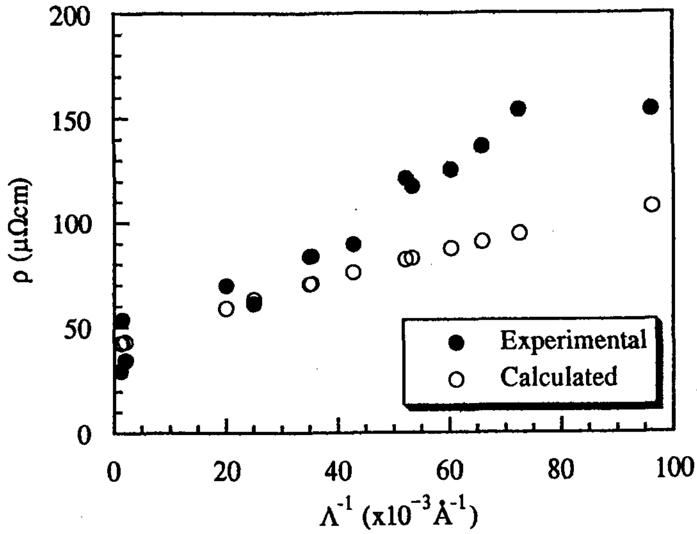

Figure 3: The bilayer thickness dependence of resistivity of Mo/Ni. The experimental data were replotted from [7].

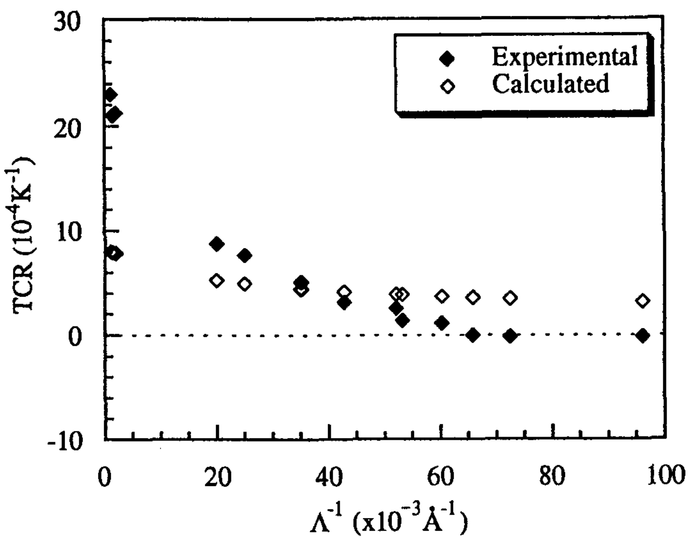

Figure 4: Dependence of TCR of Mo/Ni[7] on the inverse bilayer thickness.

the TCR.

Figures 5 and 6 describe the dependence of resistivity and TCR of Nb/Cu synthesized by Werner et al.[5] on inverse bilayer thickness. The resistivity of the Nb/Cu multilayer varies inversely with the bilayer thickness. The slope of the increase of resistivity with the inverse bilayer thickness was so large that the calculation for Nb/Cu could not reproduce the experimental results. The steep increases of the resistivity similar to the Nb/Cu were observed in the Nb/Al[9], Cu/Ta[15] and the other Nb/Cu prepared by Lowe et al.[4]. The calculated dependence of the resistivity and TCR of these multilayers on the inverse bilayer thickness were not able to fit the experimental dependence.

The TCR of the Nb/Cu at the shortest multilayer wavelength had a negative value. This negative TCR could not be explained by the model in this paper. Alloys, thin films and amorphous metals with high residual resistivities tend to have low or negative TCR. Mooji collected some data and found the correlation between TCR and the resistivity[27]. It was pointed out that the TCR transition form positive to negative value occurred in the resistivity range from 100 to 160$\mu\Omega cm$ which the mean free path became comparable to the lattice spacing. The resistivity of Nb/Cu with a negative TCR was about 130 $\mu\Omega cm$ and satisfied with Mooji's correlation.

The negative TCR was also observed in Nb/Al, Mo/Ni and Mo/Al. The resistivity and TCR of these multilayers have also satisfied with the Mooji's correlation. We had calculated the resistivity and TCR of the Mo/Al multilayers[16] by a simple diffraction model taking the negative temperature dependence of resistance due to the Debye-Waller factor in account[28].

Figures 7 and 8 showed the observed and calculated dependence of resistivity and TCR of Mo/Al on inverse bilayer thickness. The negative TCR of Mo/Al could be explained as follows; the grain size of Mo/Al multilayer became small as the bilayer thickness decreased. Therefore, the residual resistivity largely increased by the grain boundary scattering. The large resistivity reduced the positive phonon contribution and enhanced the negative temperature dependence described by

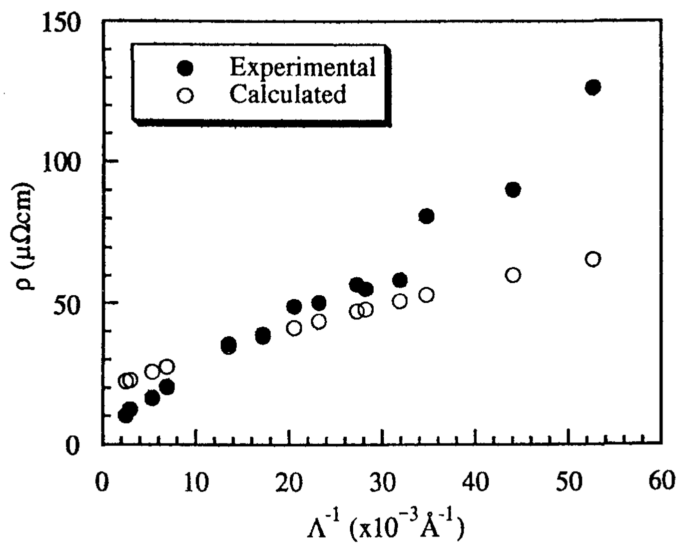

Figure 5: The bilayer thickness dependence of resistivity Nb/Cu synthesized by Schuller et al.[5]

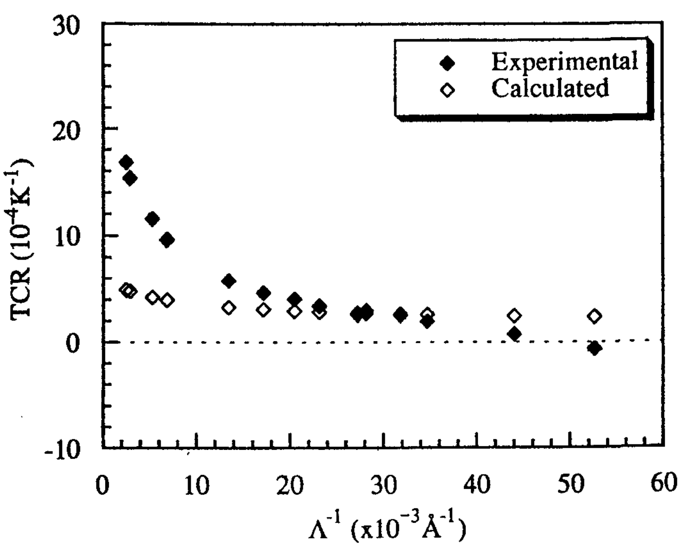

Figure 6: Dependence of TCR of the Nb/Cu multilayers[5] on the inverse bilayer thickness.

the Debye-Waller factor.

CONDLUSIIONS

We calculated the resistivity and TCR of multilayers by the model analogous with the Fuchs-Sondheimer theory and compared the calculated dependence of resistivity and TCR on bilayer thickness with the experimental results. The results of the calculations for two kinds of Au/Pd multilayer, Al/Ag, Mo/Ta, Co/Pd, Ag/Co and Mo/Cu reproduced well the bilayer thickness dependence. In other words, the bilayer thickness dependence of these multilayers is governed by the

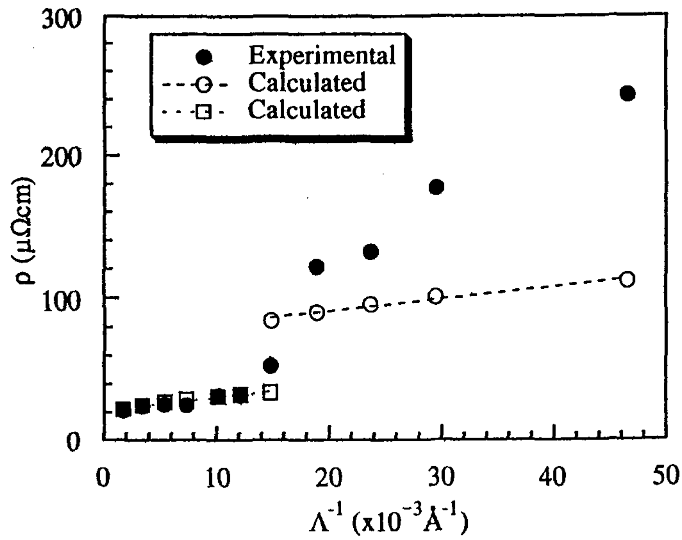

Figure 7: The bilayer thickness dependence of resistivity of Mo/Al[16]. The resistivities of Mo/Al were calculated by a simple diffraction model taking the negative temperature dependence of resistance due to the Debye-Waller factor in account.

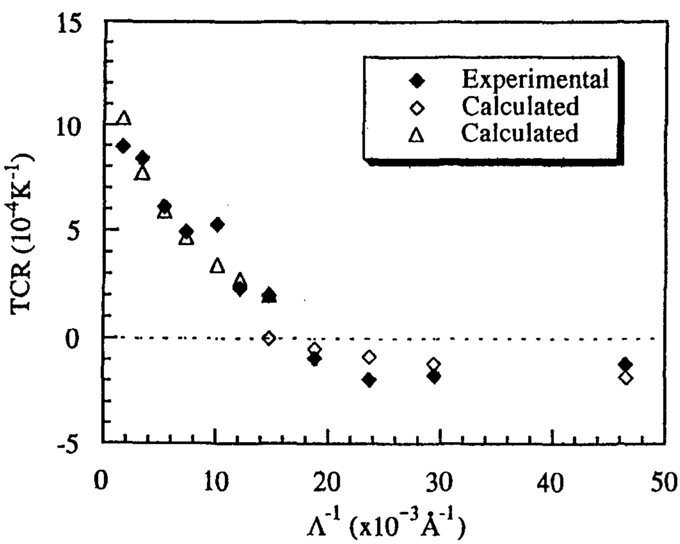

Figure 8: The experimental and calculated dependence of TCR of Mo/Al on the inverse bilayer thickness.

interface scatterings. All the conduction electrons were scattered diffusely at the interfaces in the multilayers which have fcc (111) and bcc (110) planes parallel to the surface. TCR of the multilayered films constituted by Ni and another metal decreased drastically as the bilayer thickness decreased. The calculation could not fit this sudden decrease of the TCR. The slope of the increase of resistivity of Nb/Cu, Mo/Al, Nb/Al and Cu/Ta with the inverse bilayer thickness was so large that the calculation could not reproduce the experimental results. The negative TCR was observed in these multilayers at very short bilayer thickness. The large increase of resistivity and negative TCR could be due to the scattering at the grain boundaries.

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