Electrical Resistivity of Silver Films*

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(Received 21 August 1967)

The resistivities of epitaxially grown silver films have been measured as a function of temperature from 4.2 to 295°K. Both the temperature-independent residual resistivity and the temperature-dependent resistivity were found to increase with a decrease in the specimen thickness. The increase in the temperature-dependent part of the resistivity occurs in the temperature interval of 10 to 40°K. It may be interpreted as being due to low-angle electron-phonon scattering, which causes a greater increment of resistivity in a film than in a bulk specimen.

I. INTRODUCTION

THE resistivity of a thin film or wire is known to depend on the thickness or diameter whenever these dimensions become comparable to the electron mean free path. Both the residual resistivity and the temperature-dependent resistivity increase as the specimen dimensions decrease. The increase in the residual resistivity has been experimentally observed by many investigators.[1-12] Typically, the resistivity is measured as a function of the specimen dimension at a fixed temperature. Room-temperature measurements suffice for specimens of dimensions of hundreds of angstroms, whereas liquid-helium measurements are required when the specimen dimensions are of the order of several microns. Such studies may be analyzed using the Fuchs-Sondheimer theory[13,14] for films or the theories of Dingle[15] and Chambers[16] for wires to determine the electron mean free path l and specular reflection parameter p. The quantity p usually ranges from 0 to $\frac{1}{2}$, and is defined as the probability that an electron will be specularly reflected from the surface. The change in

the temperature-dependent part of the resistivity as a function of thickness has not received as much attention as the change in the residual resistivity. The first such observation was by Andrew,[3] who observed an enhancement of the resistivity of thin mercury wires and tin foils in the temperature range of 1 to 4.2°K. A similar effect over the same temperature interval was later observed by Olsen[6] in thin indium wires and by Cochran and Yaqub[8] in thin single crystals of gallium. More recently, Holwech and Jeppesen[17] have made such size-effect measurements on aluminum films over the temperature range of 1.6 to 45°K, and have observed an increase in the temperature-dependent part of the resistivity which depends on the specimen thickness. In addition, Chopra[18] has measured the temperature dependence of the resistivity of epitaxial silver and gold films from 4.2 to 300°K and has also observed size-dependent behavior.

A mechanism to account for the size-dependent increase in the temperature-dependent resistivity was given by Olsen.[6] He proposed that low-angle scattering of electrons by phonons becomes more important in a thin specimen than in a bulk specimen when the specimen thickness is of the order of the electron mean free path. This model has been treated theoretically by Blatt and Satz[19] and by Luthi and Wyder[20] for thin wires and by Azbel and Gurzhi[21] for thin films. Blatt and Satz find a size-dependent contribution to the temperature-dependent resistivity which is proportional to $T^{7/3}$. Azbel and Gurzhi predict a size-dependent additional resistivity which is zero for very low temperatures, which initially increases in proportion to T^5, and then in proportion to T^3. Finally, a temperature interval in which the additional resistivity is proportional to $\ln T$ is predicted. The data of Andrew,[3] Olsen,[6] and Cochran and Yaqub[8] on wires are in reasonable agreement with both the Blatt-Satz and the Luthi-Wyder calculations. Neither the experimental results nor the theoretical calculations, however, consider the

* Work supported in part by the Research Corporation and the U. S. Atomic Energy Commission.

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size-dependent effects occurring at temperatures above 4.2°K. Holwech and Jeppesen observe an increase of the temperature-dependent resistivity in thin films at temperatures up to 45°K. These results cannot be explained on the basis of the Fuchs-Sondheimer theory,[13,14] but also are not in accord with the predictions of the theory of Azbel and Gurzhi.[21] The results of Chopra,[18] however, seem to be largely explicable on the basis of the Azbel-Gurzhi theory.

It is the purpose of the present paper to report a series of measurements on the temperature dependence of the resistivity of epitaxially grown silver films over the temperature range of 4.2 to 295°K. These measurements were undertaken as a further test of the existence of the Olsen effect and of the validity of the Azbel-Gurzhi theory.

II. EXPERIMENTAL TECHNIQUES

The silver films were grown by epitaxial deposition of 99.9999% silver onto heated rocksalt and mica substrates in high vacuum. The substrate temperatures were 300°C, and the films were single-crystalline with {100} and {111} planes parallel to the substrate for the rocksalt and mica substrates, respectively. For purposes of isolating the size effects, a bulk silver specimen grown from 99.999% silver was also employed in the measurements. The thicknesses of the films varied from 1700 to 28 000 Å, and the resistivity ratios $\rho_{295}/\rho_{4.2}$ varied from 12 to 175; the resistivity ratio of the bulk crystal was 250.

The films and bulk specimen were mounted in a cryostat, and the resistance measured while the temperature was varied from 16 to 295°K. The resistances at the fixed temperatures of 4.2 and 77°K were also measured. The temperatures were measured with Au-0.07% Fe against copper thermocouples connected to both ends of the specimen, and the potentials were measured with a Honeywell Model 2768 potentiometer accurate to 0.01 μV. Heaters were attached to both ends of the film so that temperature gradients along the film could be minimized. The temperature measurements were generally accurate to about 0.1°K, and the resistivity measurements were accurate to 1% for the films and 8% for the bulk specimen at low temperature. In each run, the resistivities of two specimens of different thickness were measured simultaneously in order to more accurately identify the effects due only to film thickness. The resistivity of only one film was measured in the interval of 4.2 to 16°K. This measurement showed that the resistivity increased continuously and by only a small amount in this interval. Measurements in this interval are similarly unimportant for the other films.

III. EXPERIMENTAL RESULTS

Measurements were made on six silver films and one bulk silver specimen. Data will be presented on three

of the films and the bulk specimen. The remaining three films were approximately equal in thickness to the 28 000 Å film, and the results obtained with these films largely duplicate the results obtained with the 28 000-Å film. The 1700- and 28 000-Å films were deposited on rocksalt and the 7400-Å film on mica; the crystalline orientation ({100} or {111}) of the films appeared not to influence the results in a noticeable way. A plot of resistivity versus temperature is given in Fig. 1. The same data are plotted on a linear scale in Fig. 2 over the temperature interval of 4.2 to 60°K. In Fig. 3, we have plotted the temperature-dependent resistivity as a function of temperature for each film and the bulk crystal. The temperature-dependent resistivity is the total resistivity less the residual resistivity at 4.2°K. The curve for the bulk specimen shows the expected T^5 temperature-dependence of the resistivity at temperatures below about 40°K. The curves for the films show a somewhat smaller variation of resistivity with temperature in this temperature range, with the thinnest film exhibiting a T^3 temperature dependence. At temperatures above 50°K, all of the curves coincide, and the temperature dependence of the resistivity is approximately linear at room temperature. If the resistivity due to phonon scattering and the resistivity due to surface scattering were additive, Matthiessen's rule would be valid, and all of the curves of Fig. 3 would be coincident at all temperatures. The data show that Matthiessen's rule is violated, and the deviations
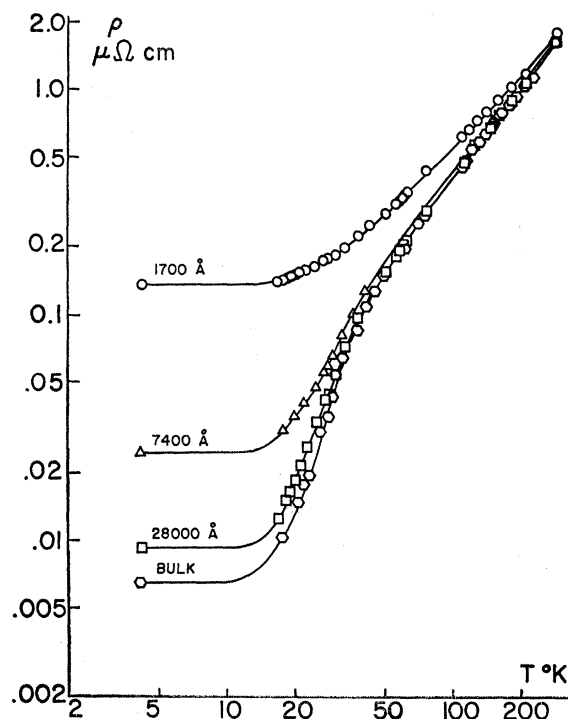

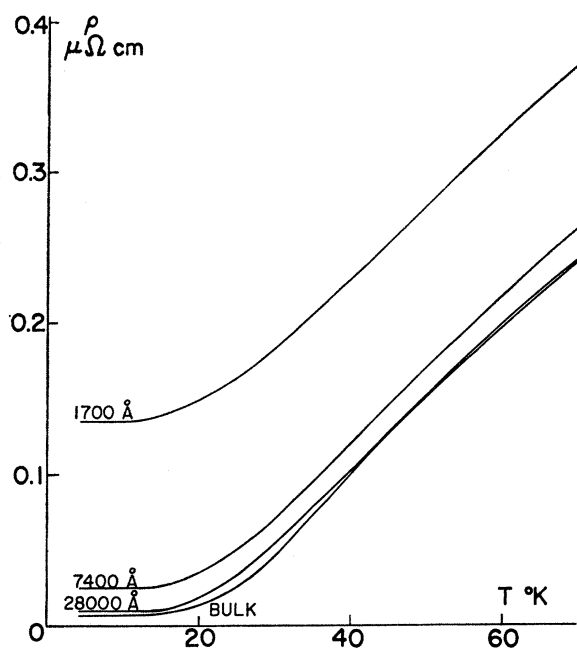FIG. 1. Total resistivity as a function of temperature.

FIG. 2. Total resistivity as a function of temperature from 4.2 to 60°K plotted on a linear scale.

from Matthiessen's rule are plotted in Fig. 4. This plot gives the difference between the temperature-dependent part of the thin-film resistivity and the temperature-
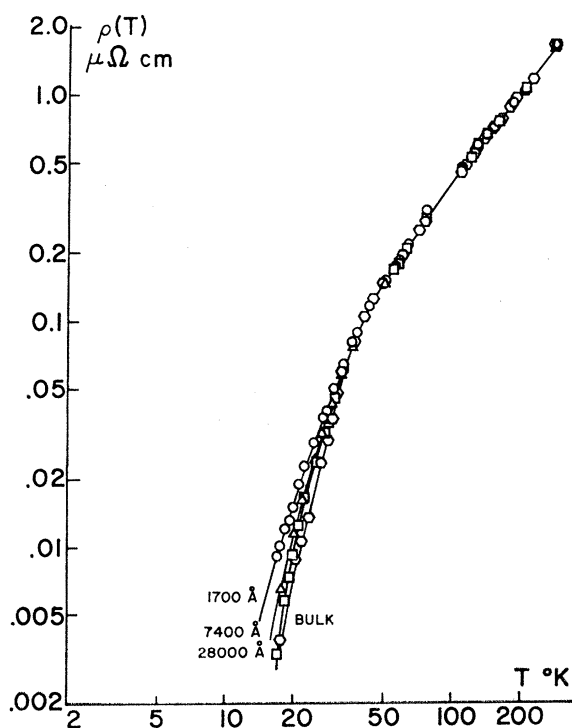

FIG. 3. Temperature-dependent part of the resistivity as a function of temperature.

dependent part of the bulk-specimen resistivity. This difference vanishes at both high and low temperatures and reaches a maximum at about 25°K for all the films.

IV. DISCUSSION

As may be seen from Fig. 1, the residual resistivity $\rho_{4.2}$ increases with a decrease in the specimen thickness. This increase, however, is larger than one would expect from the Fuchs-Sondheimer theory,[13,14] indicating that the impurity limited electron mean free path l_{ei} decreases with decreasing thickness. In a previous size-effect study[11] utilizing films grown in the same vacuum system and under the same conditions as those considered here, the quantity l_{ei} was found to be about 30μ and largly independent of the thickness. Also, the specular reflection parameter p was found to be approximately $\frac{1}{2}$. Assuming the same value of p for the films used in the present study, the quantity l_{ei} is found to be approximately 1, 6, and 12μ for the films of thicknesses 1700, 7400, and 28 000 Å, respectively. If p is assumed to be $\frac{1}{4}$, l_{ei} is calculated to be about 2, 18, and 26μ for the three films. The present experiments do not allow an unambiguous determination of p, but the values for all the films appear to lie between $\frac{1}{4}$ and $\frac{1}{2}$. Regardless of the value which is assumed for p, the electron mean free paths at 4.2°K, l_{ei}, are all substantially greater than the film thicknesses, and therefore the conditions necessary for the observation of significant size effects are present.

The effect of thickness on the temperature-dependent resistivity can be seen in Figs. 3 and 4. The temperature-dependent resistivity of the 1700-Å film deviates quite markedly from the bulk temperature-dependent resistivity when the temperature is between 10 and 40°K; the maximum deviation is about 0.012 $\mu\Omega$ cm at 25°K. This deviation represents only about 7% of the total resistivity, but more importantly, about 50% of the temperature-dependent resistivity at this temperature. The temperature-dependent resistivities of the thicker films deviate from the bulk temperature-dependent resistivities over narrower temperature intervals, and the magnitude of the deviations are about one-half as large. The temperature at which the maximum deviation occurs is about 25°K for all of the films. No special significance, however, can be attached to this temperature. What are more important are the upper and lower temperatures at which the deviation appears. According to the Olsen mechanism, the low-angle scattering events which cause surface scattering are most important. Neglecting umklapp processes, the electron-phonon scattering angles would be approximately T/Θ, where Θ is the Debye temperature. At low temperatures, T/Θ becomes very small, and the size effect should disappear. At relatively high temperatures, when the electron mean free path becomes smaller than the thickness, the size effect should also vanish. At intermediate temperatures, a size effect should be observed

FIG. 4. Difference between the temperature-dependent part of the film resistivity and the temperature-dependent part of the bulk-specimen resistivity as a function of temperature.
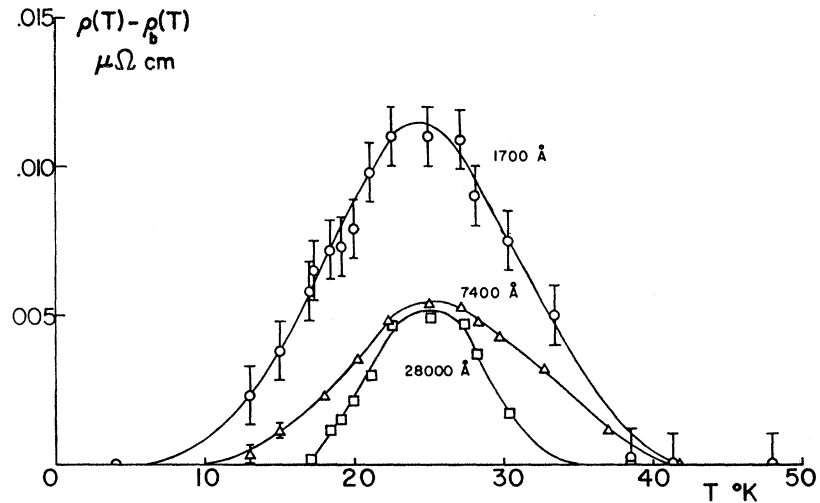
if T/Θ becomes of comparable magnitude to d/l, where d is the film thickness and l is the electron mean free path for combined electron-impurity and electron-phonon scattering, i.e., $1/l = 1/l_{ei} + 1/l_{ep}$. The results of Fig. 4 are in qualitative agreement with this model. Using a value of 226°K for the Debye temperature of silver, the quantity T/Θ does become comparable to d/l in the temperature interval of 10 to 40°K. At both higher and lower temperatures, T/Θ is much smaller than d/l.

The Fuchs-Sondheimer theory[13,14] predicts a size-dependent increase in temperature-dependent resistivity which is at least an order of magnitude smaller than the effect observed here. The Azbel-Gurzhi theory[21] predicts a rather complicated variation with temperature of the size-dependent part of the temperature-dependent resistivity. Azbel and Gurzhi, however, do not give an estimation of the magnitude of the expected effect. The experiments did not show any of the structure in the temperature dependence as predicted by Azbel and Gurzhi. Figure 4 does show, however, a size-dependent increase in the temperature-dependent resistivity at low temperatures which is roughly proportional to T^2. This compares favorably with the predictions of Blatt and Satz,[19] who predict a $T^{7/3}$ dependence. The Blatt-Satz theory, however, is applicable to thin wires and perhaps cannot be applied to thin films.

The only previous experimental observations of a size-dependent increase in the temperature-dependent part of the resistivity of films are those of Andrew on tin foils,[3] Holwech and Jeppesen on aluminum films,[17] and Chopra on gold and silver films.[18] Andrew's experiments were limited to temperatures of 1 to 4.2°K, and the magnitude of the size effects observed were much smaller than in the present experiments. Holwech and Jeppesen observed an effect which was greatest for the thinnest films, and which had its greatest magnitude between 20 and 40°K. The size-dependent resistivity increase which they observed is about eight times less than the resistivity increase observed in the present experiment. The size effects observed by Chopra with silver films are an order of magnitude larger than those observed here and furthermore exhibit a completely different temperature dependence.

V. CONCLUSIONS

A size-dependent increase in the temperature-dependent part of the resistivity of thin silver films has been observed. The increase is much larger than would be expected from the Fuchs-Sondheimer theory[13,14] and seems to be in qualitative accord with a model suggested by Olsen[6] based on the low-angle scattering of electrons by phonons. The results, however, do not compare favorably with the more detailed theory of Azbel and Gurzhi.[21]

ACKNOWLEDGMENT