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Weak localization and interaction effects in amorphous Ta/Ge multilayers

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Abstract. We have measured the in-plane resistivity of amorphous Ta/Ge multilayers with individual layer thicknesses of between 0.4 and 34 nm from 1.5 to 300 K. Conduction occurs principally through the Ta layers, allowing us to study the effects of reduced dimensions in a structurally disordered metal. At high temperatures the negative temperature coefficient of resistance can be described in terms of the destruction of quantum interference by phonons. At lower temperatures a turnover to a positive temperature coefficient of resistance is seen, suggesting that spin–orbit scattering becomes important. At still lower temperatures the resistance is dominated by electron–electron interaction effects and we have observed a transition from two-dimensional to three-dimensional behaviour as the Ta layer thickness is increased.

1. Introduction

Over the past decade it has been suggested that the range of resistivities and temperature coefficients of resistivity (TCRs) observed at room temperature in high-resistivity disordered metallic systems is a result of the interplay between conventional electron transport (the Boltzmann conductivity) and the effect of quantum interference (often referred to as weak localization) (see Imry 1980, Kaveh and Mott 1982, Kaiser 1987). Despite the wealth of qualitative observations supporting this idea (see for example Howson and Gallagher 1988), there remain few quantitative studies of the temperature dependence of the resistivity at high temperatures (100–300 K). The low-temperature resistivity in disordered systems, on the other hand, has been studied intensively (for reviews see Bergmann 1984, Howson and Gallagher 1988). These studies have tended to focus on the simple metals rather than the more complicated transition metals.

Against this background we present the results of a study of electron conduction in films of varying thicknesses of the disordered transition metal Ta, fabricated in the form of vapour deposited Ta/Ge multilayers. In particular, we discuss a quantitative study of the resistivity of disordered Ta at high temperatures assuming weak localization effects dominate and we present the low-temperature results, which show a number of interesting features. In both temperature regimes we see strong variation in the form of the resistivity as a function of both the Ta and the Ge layer thicknesses.

2. Experimental details

Although conduction in pure amorphous metals has been studied intensively at low temperatures the study of such metals at higher temperatures is often precluded owing

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to crystallization of the amorphous structure. We have chosen to study Ta, in which we see no evidence of crystallization up to over 300 K in line with the results of Nestell *et al* (1982) and Kumar and Trodahl (1993). The Ta is fabricated in the form of Ta/Ge multilayers (in which the Ge layers are amorphous) in order to make samples with lower more easily measured total resistances as well as making the samples more mechanically robust and the Ta layers less prone to oxidation than a single Ta film. In addition to these benefits depositing Ta onto the amorphous Ge causes the Ta to form electrically continuous films at thicknesses of only a few atomic layers with the important consequence that dimensionality effects associated with the layer thickness can be investigated. (That ultrathin conductive films can be prepared by initially depositing an underlayer of amorphous Ge was first reported by Strongin *et al* (1970).)

The multilayers were prepared in a high-vacuum system by vapour deposition onto ambient temperature glass substrates, which were alternately rotated through vapour streams of Ta and of Ge. The deposition system is described in detail elsewhere (Williams *et al* 1988). The total thickness of the films is of the order of 100 nm. Transmission electron microscopy (TEM) of microtomed cross-sections of the multilayers similar to that in figure 1 showed the layers to be uniform and continuous with no evidence of the formation of islands or crystallites.

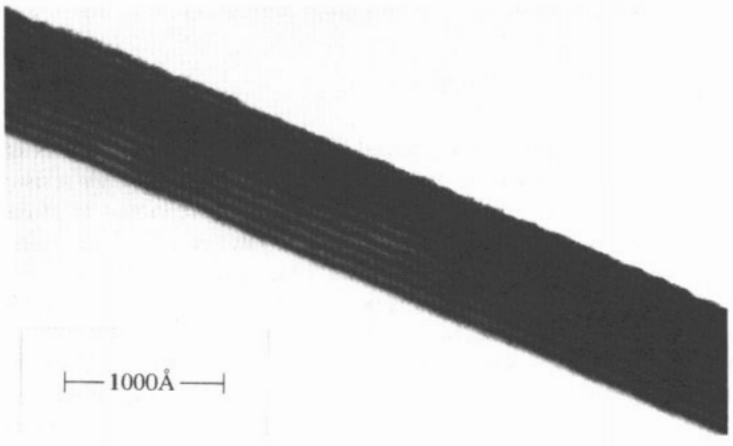

Figure 1. TEM bright field image of sample with Ta layers approximately 3 nm thick and Ge layers approximately 7 nm thick. The darker layers are Ta.

Layer thicknesses in atoms per unit area were determined by Rutherford backscattering spectrometry (RBS) using multilayer samples. The results correlated well with multiple-beam interferometry performed on films formed on substrates exposed to only one vapour stream during the deposition. Unless a layer thickness in units of length is explicitly required we will quote thickness in units of atoms per unit area because it is this quantity that is directly measured, and because a measure of the thickness carries little meaning in ultrathin films. As a rough guide, the layer thickness $a_{Ta,Ge}$ in nanometres is related to the areal density $N_{Ta,Ge}$ in units of 10^{15} atoms cm^{-2} by $a_{Ta} \approx 0.22N_{Ta}$ and $a_{Ge} \approx 0.23N_{Ge}$.

The in-plane resistance of a rectangle approximately 2 mm×6 mm was measured using a four-point probe method, the network being scribed through the entire thickness of the film. The uncertainty in the dimensions of the rectangular section of the film (measured

with a travelling microscope) contribute less than 5% uncertainty to the final measurement of the sheet resistance of the sample. Before making contacts to the samples, a small hole is scratched through the film to ensure that conduction occurs purely in the plane of the layers. To measure the very small changes in the resistance as a function of temperature accurately, the resistance was measured with a precision of better than 10 ppm and, in addition, thermal gradients were minimized by allowing the samples to warm to room temperature over a period of several days.

The resistance of the multilayers with thick Ge layers is interpreted as that of n thin isolated Ta films connected in parallel since the resistivity of amorphous Ge is at least seven orders of magnitude larger than that of the multilayers in the temperature range investigated and tunnelling across thick Ge layers is not expected (Ruggiero *et al* (1982) give the tunnelling length in amorphous Ge as ~ 0.8 nm).

Sample ageing was found not to alter the temperature dependence of the resistance over a period of six months. As an added precaution samples were stored under vacuum and the resistance measurements made soon after fabrication.

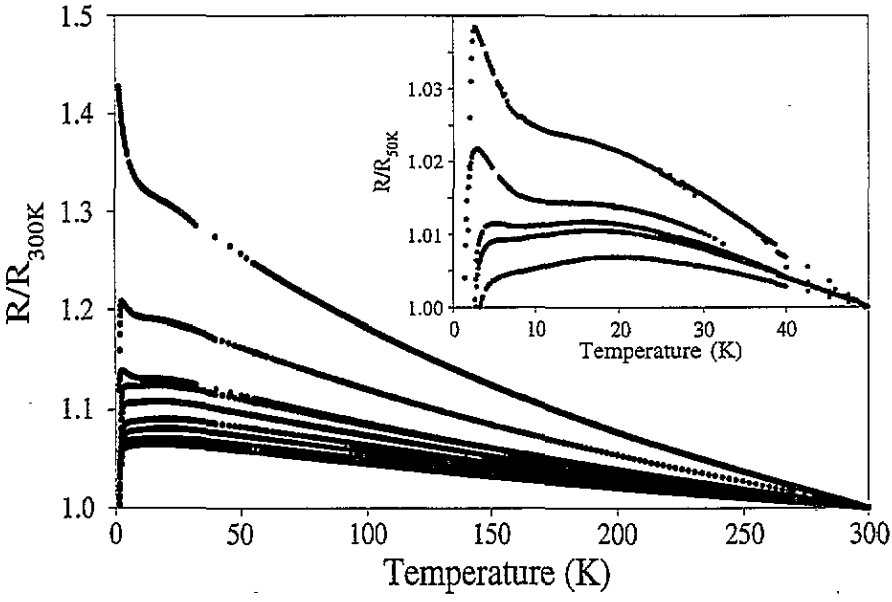

Figure 2. Resistance normalized to 300 K for multilayers with Ta layers (in units of 10^{15} atoms cm^{-2}) of (top curve to bottom curve): 2.7 ± 0.2, 5.1 ± 0.3, 6.0 ± 0.3, 7.7 ± 0.5, 9.7 ± 0.6, 11.2 ± 0.7, 17 ± 1, 21 ± 1 and 46 ± 3. The inset shows the low-temperature resistance normalized to 50 K for (top curve to bottom curve in units of 10^{15} atoms cm^{-2}): 5.1 ± 0.3, 6.0 ± 0.3, 7.7 ± 0.5, 9.7 ± 0.6 and 11.2 ± 0.7. The experimental points are plotted in each case.

3. Results and discussion

The normalized resistance of the multilayers as a function of temperature depends in a systematic way on the Ta layer thickness. A selection of the results for different Ta layer thicknesses are shown in figure 2. Down to 20 K, the resistance of all the samples increases

as the temperature is lowered, the fractional increase being greater for thinner Ta layers. Below 20 K, the resistance of thicker layers (> 5 nm) rises to a maximum and then slowly decreases. At very low temperatures (< 0.5 K) a rapid transition to a superconducting state is seen. Results relating to the superconducting transition will be presented in a separate publication. The behaviour of the resistance for Ta layers of intermediate thickness (1–4 nm—see the inset of figure 2) is more complicated. A maximum in the resistance is still evident at about 15–20 K but a second feature at low temperature (less than about 6 K) causes a minimum below which the resistance increases with the lowering of the temperature. As the Ta layers are made thinner this second feature grows in importance, eventually dominating the low-temperature resistance. For the very thinnest Ta layers studied (~ 0.5 nm) the resistance increases with falling temperature over the entire temperature range.

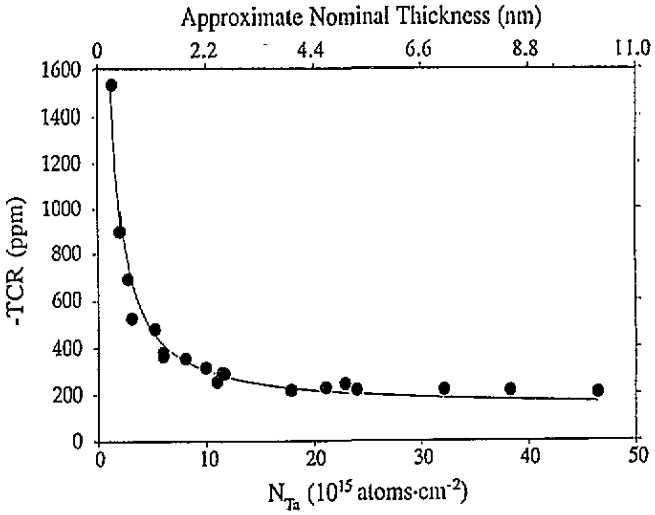

Figure 3. Temperature coefficient of resistance (TCR) at 300 K as a function of Ta layer thickness. Solid curve is $\rho \propto 1/a_{Ta} + \text{const}$.

We now turn to a more quantitative analysis of the high-temperature (100–300 K) results. The resistivity of the Ta channels is of the order of 200 $\mu\Omega$ cm while the room temperature coefficient of resistance (figure 3) shows a clear dependence on layer thickness. We believe that the variation in the TCR can be explained within the theory of weak localization.

In the presence of disorder the Boltzmann conductivity is reduced by an amount

$$\delta\sigma = (e^2/2\pi^2\hbar)/\sqrt{D\tau_{in}} \tag{1}$$

(the 'weak-localization' correction) where D is the diffusion constant and τ_{in}^{-1} is the inelastic scattering rate (see Abrahams *et al* 1979, Anderson *et al* 1979). The weak-localization correction to the Boltzmann conductivity becomes important when the distance between scattering sites is comparable to the electron wavelength, as is the case in high-resistivity metallic glasses. In this case the interference of scattered partial waves leads to constructive interference or an 'echo' in the backscattered direction that reduces the conductivity below the Boltzmann value. Inelastic scattering events, which become more

probable as the temperature is increased, destroy the phase coherence, which gives rise to coherent backscattering. As a result the echo in the backscattered direction is progressively destroyed as the temperature is raised, and the conductivity increases. The temperature dependence of the weak-localization correction arises from the temperature dependence of the inelastic scattering rate, which is generally assumed to be of the form

$$\tau_{in}^{-1}(T) = \beta T^{p}. \tag{2}$$

The magnitude, β, and temperature exponent, p, of the inelastic scattering rate depend on the scattering mechanism, the degree of disorder, the temperature and the sample dimensionality.

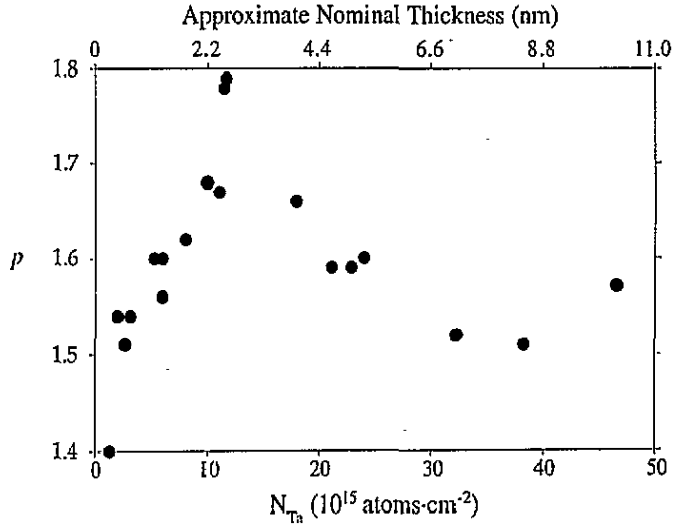

Figure 4. Value of p (the temperature exponent of the phase breaking rate) at high temperature.

Howson (1984) and Howson and Greig (1986) assumed that normal conduction processes (that is, the Boltzmann conductivity) give only a small contribution to the temperature dependence of the conductivity. Using this approach, we find that for all the samples the temperature variation of the conductivity from 100–300 K is well fitted by (1) with an inelastic scattering rate given by (2). From these fits we find $p \sim 1.6$, $\beta/D \sim 0.8 \times 10^{15}$ K^{-p} s^{-1} and $\beta \sim 90 \times 10^{9}$ K^{-p} s^{-1} (taking the free electron value for the diffusion constant for Ta, $D = 1.2 \times 10^{-4}$ m^2 s^{-1}). Interestingly, p is a function of the Ta layer thickness (see figure 4).

At high temperatures, the dominant inelastic scattering mechanism is expected to be electron–phonon scattering. Provided the system is only weakly disordered (that is, the characteristic length scale for phonons, q_{ph}^{-1}, is smaller than the mean free path, l) the system is said to be 'clean' and p should be equal to unity, the familiar result for the crystalline state. In the opposite or 'dirty' limit ($q_{ph}^{-1} > l$) Chakravarty and Schmid (1986) give $p = 2$ for scattering from transverse phonons and $p = 3$ for scattering from longitudinal phonons. A free electron estimate for Ta suggests, however, that the scattering rate from longitudinal phonons will be much smaller in our samples than that from transverse phonons.

For the samples in this study the inverse phonon wave vector is estimated to be of the order of a few tenths of a nanometre while the mean free path is limited by the

interatomic spacing, hence $q_{ph}^{-1} \sim l$ and the level of disorder is such that the samples are in neither the clean nor dirty limit as regards electron–phonon scattering. It is therefore not unreasonable that the value of p should lie between the values predicted in each of these limits. Furthermore, the value of β is of the correct order of magnitude for electron–phonon scattering in either limit.

We now turn to the low-temperature results. It is probable that the turnover to a positive TCR just below 20 K marks a transition to weak antilocalization due to spin–orbit scattering (Bergmann 1982). Still lower in temperature, electron–electron interaction effects (Al'tshuler and Aronov 1979a, b, Al'tshuler *et al* 1980) appear to become important, causing the return to a positive TCR in samples with thinner layers. That this feature is largest for thin Ta layers, and not seen at all for thick layers, is attributed to the strong dependence of interaction effects on the sample thickness. A transition from a strong logarithmic dependence of the conductivity on temperature to a weaker square root dependence is expected to occur when the Ta layer thickness is of the order of the diffusion length $L_T = \sqrt{Dh/k_B T}$. For our samples the diffusion length in the temperature range 2–5 K is of the order of 20 nm. We find that the conductivity of multilayers with Ta layers thinner than about 3 nm is logarithmic between 2 and 5 K as expected. The measured values of the coefficients of logarithmic variation decrease as the Ta layer thickness increases but in all cases they are of the same order of magnitude as the theoretical predictions. We believe that the decrease in the magnitudes of the coefficients of logarithmic variation as the Ta layers are made thicker is predominantly a result of the electron–electron interaction correction becoming weaker as electron–electron interaction effects approach a transition from two- to three-dimensional behaviour.

4. Conclusion

We have described measurements of the variation in the resistance of Ta/Ge multilayers as a function of temperature as well as Ta layer thickness. At high temperatures (100–300 K) the temperature dependence of the resistance is interpreted in terms of quantum interference effects in the Ta channels and information about the governing parameter (the inelastic scattering rate τ_{in}^{-1}) has been extracted from the data. We find τ_{in}^{-1} varies at a rate in between a linear and a quadratic dependence. At low temperatures, we see behaviour consistent with weak-localization effects in the presence of strong spin–orbit scattering together with electron–electron interaction effects.

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