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The effect of hydrogenation on the optical and electrical properties of amorphous silicon–tantalum alloys near the metal–insulator transition

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Abstract. In order to study the effect of hydrogenation on the optical and electrical properties of amorphous silicon–tantalum alloys two sets of samples (one unhydrogenated and the other hydrogenated) were prepared by sputtering. Films (about 1 μm thick) were deposited on substrates kept at room temperature in an atmosphere of pure Ar for the unhydrogenated samples and a 90% Ar–10% H_2 mixture for the hydrogenated ones. Both optical and electrical measurements were carried out on 'as-deposited' samples. Optical measurements were made with photon energies ranging from 0.5 to 3.0 eV while the electrical data were recorded over a range of temperatures from room temperature down to about 6 K. The experimental conductivity versus temperature curves were fitted to theoretical expressions corresponding to both insulating and metallic regimes, from which useful information about carrier transport in such samples is extracted. Based on the comparison between electrical and optical results and on the computer analysis of experimental data, five different methods are used to estimate the position of the metal–insulator transition in both kinds of sample. The results show a shift of about 2% to higher values of the critical tantalum concentration y_c for the hydrogenated samples in comparison with the unhydrogenated ones. It is concluded that the metal–insulator transition lies at y_c-values of about 0.15 and 0.17 in the unhydrogenated and hydrogenated samples respectively.

1. Introduction

The metal–insulator transition (MIT) in amorphous materials has attracted much attention in recent years. A comprehensive approach to this problem has been given by Mott (1990). One class of systems showing MITs consists of amorphous metal–semiconductor alloys. If, in the systems like a-$Si_{1-y}M_y$ or a-$Ge_{1-y}M_y$, the concentration y of the metal M is increased from zero, at a certain value of y the states at the Fermi level become delocalized and a transition to the metallic state occurs. This is usually taken as the position of the MIT in that system.

With regard to experimental work on the MIT, several systems have been studied, both unhydrogenated (UH) and hydrogenated (H) forms of amorphous metal–semiconductor alloys, with measurements of their optical, electrical and structural properties. Electrical properties were used to analyse the MIT in the UH films of a-$Si_{1-y}Nb_y$ (Hertel *et al* 1983) as well as in both UH and H films of a-$Si_{1-y}Au_y$ (Nishida *et al* 1983). Also, using estimates of the MIT based on electrical properties, Regan *et al* (1984) pointed out anisotropic phase separation through the transition in UH alloys of a-$Ge_{1-y}Mo_y$, a-$Ge_{1-y}Fe_y$, a-$Ge_{1-y}W_y$ and

a-$Si_{1-y}Fe_y$. In a series of papers the MIT was studied in amorphous silicon–nickel alloys: in H samples using optical properties (Bayliss *et al* 1991) and in both UH and H samples based on their electrical properties (Adkins *et al* 1994, Dammer *et al* 1993). Recently, transport (Popescu and Davis 1994), optical (Nijim *et al* 1995) and electrical (Popescu and Davis 1995) properties have been studied in amorphous hydrogenated silicon–tantalum films. In that work, attention was paid to the semiconductor behaviour of samples having tantalum concentrations on the insulating side of the MIT and far from the transition. In this paper we concentrate on the effect of hydrogenation on the optical and electrical properties of amorphous silicon–tantalum alloys near the metal–insulator transition. In order to establish the position of the MIT more accurately, many samples were prepared close to the critical metal concentration.

2. Experimental details

2.1. Sample preparation

Thin films of amorphous silicon–tantalum were prepared using a Nordiko NM 2000 sputtering system. In order to obtain a good compositional uniformity, a configuration with seven evenly spaced Ta pieces placed onto the surface of a four-inch c-Si target was used. Different values for the composition were obtained by varying, in small increments, the area of the seven tantalum pieces.

The plasma sputtering was performed by means of a 13.56 MHz r.f. generator of 1 kW power. Substrates of Corning 7059 glass for optical (in visible to UV ranges) and electrical measurements, and of Al foil for compositional analysis, were cleaned ultrasonically in acetone and then methanol before being mounted. The chamber was pumped down to a base pressure of 2×10^{-7} Torr with the help of a Meissner coil before the plasma gases (pure argon for the UH samples and a 90% Ar–10% H_2 mixture for the H films) were introduced. During deposition the temperature of the substrates was maintained close to room temperature (RT) by water cooling. The films of silicon–tantalum were prepared at a power density of 4 W cm^{-2}, for which a deposition rate of 43 pm s^{-1} was obtained. Other details on the sample preparation can be found in the references Popescu and Davis (1994), Nijim *et al* (1995), Popescu and Davis (1995).

2.2. Thickness and composition measurements

The sputtering times were chosen so that the film thicknesses were about 1 μm; this was checked by mechanical measurements (using a Talysurf).

The composition y was determined by energy-dispersive analysis with x-rays (EDAX). The values of y given in this paper refer simply to the relative atomic concentrations of tantalum and silicon: $y = \mathrm{Ta}/(\mathrm{Si} + \mathrm{Ta})$.

2.3. Optical measurements

Optical measurements were made at RT on 'as-deposited' samples with photon energies ranging from 0.5 to 3.0 eV using a double-beam Perkin–Elmer 330 spectrophotometer. In the spectral regions where coherence was important (i.e. where interference fringes were observed), the reflectance R and transmittance T were taken to be their average values between maxima and minima, after tracing the envelope of each spectrum containing such interference fringes.

Since the reflectances at the air/film, film/substrate as well as substrate/air interfaces were not separately determined, the transmittance was approximated (Nijim *et al* 1995) by the expression

$$T \approx (1 - R)\exp(-\alpha d) \tag{1}$$

where d is the film thickness (as determined by Talysurf measurements) and R is the reflectance at the front surface. The value taken for $(1 - R)$ was that when the transmittance ceases to change at long wavelengths.

Using the average values of reflectance and transmittance corresponding to each energy and taking the concentration of Ta incorporated in the films as a parameter, the dependencies of the absorption coefficient, α, on the photon energy, $h\nu$, were determined.

2.4. Electrical measurements

For measurements of electrical conductivity, the samples were inserted into an Oxford Instruments CF200 continuous-flow cryostat to allow recording of conductivity data over a range of temperatures from RT down to about 6 K.

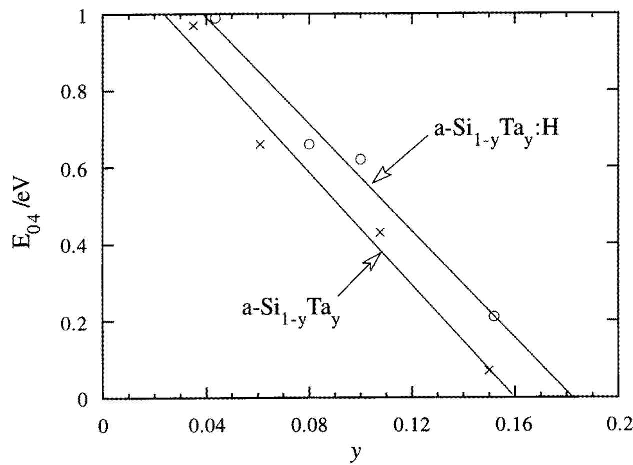

Figure 1. Plots of E_{04}-values against y for UH and H amorphous silicon- tantalum films. The dependencies clearly show a shift to higher values of y for the H samples in comparison with the UH samples.

The films containing a high proportion of metal were sufficiently conducting to allow alternating current techniques to be used. An EG&G 5208 lock-in amplifier provided the current (from its internal oscillator) and measured the sample resistance at a specific frequency of 82 Hz, by which method the noise was greatly reduced. Some of the other films, with lower metallic content, were highly insulating, and it was necessary to use a direct current technique. A Keithley 602 electrometer provided a small direct current to allow measurements of very high film resistances, up to 10^{12} Ω. For very high resistance values, the measurements were also limited by the time constant of the circuit. In order to extend the measurement range to lower temperatures an interdigitated electrode configuration was used. For all the samples the data points were recorded automatically by computer during cooling and warming to provide the very dense data sets necessary for good fitting to theoretical models.

3. Experimental results

3.1. Optical data

The values of the optical gap E_{04} were obtained by extracting from the dependencies of α versus $h\nu$ the photon energy values at which $\alpha = 10^4$ cm^{-1} (Nijim *et al* 1995). The extraction of E_{04}-values was possible only for samples with low Ta concentrations. For the others (having compositions near the MIT) these values were obtained by extrapolating the dependencies α versus $h\nu$ until α reaches the value of 10^4 cm^{-1}. This explains the higher errors appearing in the estimation of E_{04} near the MIT. The values of E_{04} (for UH and H samples respectively) are plotted versus y in figure 1.

3.2. Electrical data

Over the whole temperature range, conductivities varied by several orders of magnitude in the case of the films with the least metal content, whereas for the metallic films the changes were only a few per cent. The range of the data obtained was limited either by the minimum temperature which could be obtained in the cryostat (about 6 K), or, at a higher temperature, by the resistance becoming too large to be measured for the least-conducting samples. The master plots for both UH and H conductivity data versus temperature (on double-logarithmic scales) are displayed in figure 2.

4. Discussion

4.1. Comparison between optical and electrical data

Popescu and Davis (1994) and Nijim *et al* (1995) observed that the dependency of E_{04} versus y is non-linear for low Ta concentrations, but it becomes almost linear for y having values near the transition. For this reason, straight lines are used to represent the best fits for the data points shown in figure 1. The values of the E_{04}-gap, as determined from the optical measurements, clearly show a shift to higher values of y for the H samples in comparison with the UH samples. The dependencies of E_{04} on y close to the critical value imply critical compositions y_c of about 0.16 and 0.18 (with states localized for concentrations $y < y_c$ and extended for $y > y_c$). These compositions are taken as the position of the MIT in the UH and H alloys respectively. (The previous estimates of 0.15–0.16 reported by Popescu and Davis (1994) and Nijim *et al* (1995) for UH a-$Si_{1-y}Ta_y$ can be explained as due to the lack of samples with compositions near that of the MIT.)

The electrical measurements show that, for the same value of y, the dependencies of log σ on log T lie at lower conductivities for the H samples in comparison with the UH samples (see figure 2). The effect of hydrogenation has been explained in terms of saturation of dangling bonds preventing electron exchange with the metal bands (Dammer *et al* 1993). Plotting the conductivity at 20 K versus y for both sets of films, the curve for H samples clearly lies below (or to the right of) that for the UH samples (figure 3). In order to estimate the critical concentration y_c corresponding to the UH and H samples of silicon–nickel alloys, Dammer *et al* (1993) drew a horizontal line at a value of conductivity equal to 5×10^3 Ω^{-1} m^{-1} and took intersection points of this line as being the positions of y_c in the UH and H samples respectively. The results obtained by computer analysis of the experimental conductivity versus temperature curves for our system indicated that the UH sample with $y = 0.15$ lies slightly on the insulating side of the metal–insulator transition. Assuming that the MIT takes place in the H samples at the same conductivity, a clear shift

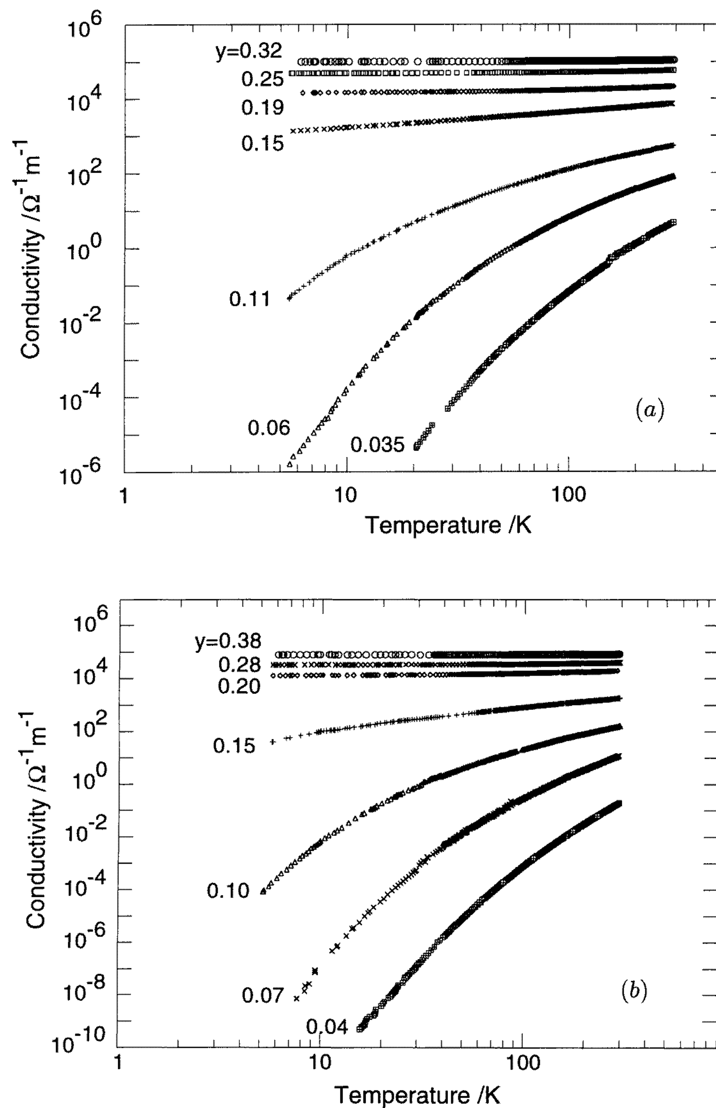

Figure 2. Master plots of conductivity versus temperature for amorphous silicon–tantalum alloys: (a) UH samples; (b) H samples. Tantalum percentages are indicated.

of y_c of about 2% is obtained by hydrogenation. There is an acceptable agreement between the values resulting from this method and those obtained by optical methods.

4.2. Computer analysis of experimental conductivity versus temperature curves

The conductivity data for each film were fitted according to whether the behaviour was predominantly insulating or metallic.

In the insulating regime, the data were consistent with conduction by variable-range

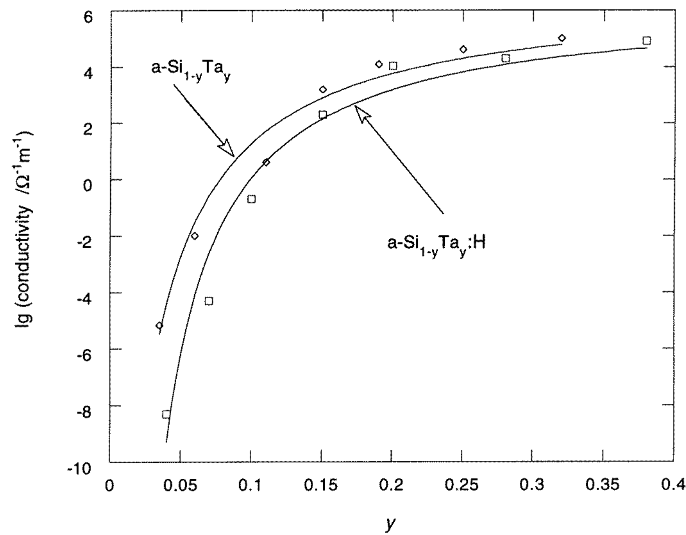

Figure 3. Conductivity at 20 K versus composition for both sets of amorphous silicon–tantalum films. Curves of the same shape are fitted to both sets of data and there is a clear shift to lower conductivities (or higher metal concentrations) for the H samples.

hopping, and were fitted to the following formula:

$$\sigma = \sigma_0 \exp[-(T_0/T)^x]. \tag{2}$$

For a constant density of states a value of $x = 0.25$ would be expected (Mott 1967), but the existence of a Coulomb gap would result in a value of 0.5, at least at lower temperatures (Efros and Shklovskii 1975).

Presuming that the system is three dimensional, the extraction of the parameters can be carried out using the formulae introduced by Adkins (1989). So, assuming $x = 0.5$ in equation (2), it was possible to obtain the temperature-dependent optimum hop energy of the variable-range hopping process, using

$$W_{\mathrm{opt}}(T) = 0.5k_{\mathrm{B}}(T_0/T)^{1/2} \tag{3}$$

where k_{B} is Boltzmann's constant and T is the temperature at which W_{opt} is to be calculated. It is interesting to observe that equation (3) contains no material parameters. Other quantities depend on the relative permittivity, ε_r. So, taking into account that in three dimensions, conforming to the theory of Efros and Shklovskii, the density of states is given by

$$g(E) \approx 2000\frac{\varepsilon_r^3\varepsilon_0^3}{e^6}E^2 \equiv g_2E^2 \tag{4}$$

(where ε_0 is the permittivity of vacuum), it is possible to obtain the formula of the mean tunnelling exponent:

$$\alpha = k_{\mathrm{B}}T_0(\pi g_2)^{1/3}/10.5. \tag{5}$$

From equations (3) and (4), it is observed that, for a given T_0, α is proportional to ε_r. Adkins (1989) also deduced the temperature-dependent optimum hopping distance:

$$R_{\mathrm{opt}} = 0.25\alpha^{-1}(T_0/T)^{1/2} \tag{6}$$

in which case R_{opt} is inversely proportional to ε_r.

Popescu and Davis (1994) analysed transport phenomena in a-$Si_{1-y}Ta_y$:H samples for a temperature range from about 130 K to beyond RT and their analyses (made by hand) showed that the results appeared to be consistent with the Mott $T^{-1/4}$-law (corresponding to variable-range hopping in a constant density of states). More recently, Popescu *et al* (1995) carried out a computer analysis of data taken down to much lower temperatures for the a-$Si_{1-y}Ta_y$:H system. They found that, at temperatures close to RT, the hopping exponent x does have values close to 0.25 but it increased steadily with decreasing temperature. In comparison with the results on amorphous germanium–chromium reported by Heinrich *et al* (1995), where a sharp change of the hopping exponent was observed, the changes in the present case were gradual and occurred over the whole temperature range, although the rate of change increased at lower temperatures. For these reasons, Popescu *et al* (1995) concentrated on analysis of the hopping transport at low temperatures.

Table 1. Conductivity and hopping parameters for the insulating samples of amorphous silicon–tantalum alloys. The values of x are average values obtained from a fit to equation (2) over the whole temperature range; x was observed to change from about 0.25 at RT to about 0.5 at lower temperatures. The other parameters were obtained from fits after fixing $x = 0.5$ and using a temperature range below T_c for which the values of x were close to 0.5.

y	x	T^* /K	T_0 /K	σ_0 /Ω^{-1} m^{-1}	R_{opt}(20 K) /10^{-9} m	$W_{opt}(T^*)$ /10^{-21} J	g_0 /10^{43} J^{-1} m^{-3}	α^{-1} /10^{-9} m
a-$Si_{1-y}Ta_y$								
0.035	0.31	100	6500	180	5.9	2.5	710	1.3
0.06	0.41	45	2300	600	10	1.5	110	3.9
0.11	0.35	45	500	900	16	0.93	43	9.4
a-$Si_{1-y}Ta_y$:H								
0.04	0.28	30	8000	1.7	5.3	2.8	260	1.1
0.07	0.37	30	4000	60	7.5	2.0	140	2.1
0.1	0.4	30	1300	600	13	1.1	44	6.6
0.15	0.16	30	70	1300	57	0.26	2.4	120

Concerning the samples studied here, which were prepared with compositions nearer that of the metal–insulator transition, their behaviour was found to be quite similar to that of the samples far from transition analysed by Popescu *et al* (1995). The values of x were not constant at 0.25 for any temperature range (but increased continuously with decreasing temperature) and it was decided to concentrate on the range of temperatures over which x was close to 0.5. Having observed the variation of x from 0.25 at RT to 0.5 at lower temperatures, we determined two temperatures: T^* at which x was 0.375 (half-way between the two regimes) and T_c, the temperature below which the behaviour starts to be dominated by the Coulomb gap. The former marks the onset of domination of the Coulomb gap. The latter defines a highest temperature at which we may expect the Efros–Shklovskii (ES) expressions to be quantitatively valid. From the fit in this range, the parameters T_0 and σ_0 in equation (2) were extracted; these are displayed in table 1.

It is expected that ES hopping would occur only at temperatures significantly less than T_0 (Efros and Shklovskii 1984) and this is consistent with the large values of T_0 obtained. At lower metal concentrations, the larger values of T_0 imply that ES hopping extends up to higher temperatures.

The sample of a-$Si_{1-y}Ta_y$:H with $y = 0.15$ obeyed the hopping form poorly, as evident from the anomalous value of x, but it did not fit the formulae for the metallic regime either.

It is postulated that it lies in a transition regime, although the small positive curvature in its log σ–log T form implies that it lies slightly on the insulating side.

The values of R_{opt}, calculated at 20 K, were of the order of several nanometres, as is displayed in table 1. The value $R_{opt} = 13$ nm obtained for our sample of a-$Si_{1-y}Ta_y$:H with 10% Ta is the same as that reported by Abkemeier *et al* (1992) for a sample of a-$Si_{1-y}Ni_y$:H containing 8% Ni. The mean Ta–Ta distance was estimated, and the optimum hopping distance was found to be equivalent to about 40 Ta–Ta distances (increasing when the Ta concentration in the samples is increased). In this case our system might support the explanation that the wavefunctions of the mobile electrons are delocalized over clusters of Ta atoms and that the hopping could take place between these clusters.

The optimum hop energy, W_{opt}, was calculated at T^* (as an estimate of the width of the Coulomb gap); the decrease towards the MIT is assumed to mirror the closure of the Coulomb gap at the MIT and this occurred in UH and H systems at values of y_c of 0.14 and 0.16 respectively, determined by extrapolating the data to zero optimum hopping energy.

The background density of states, g_0, can be estimated under the assumption of a constant relative permittivity as the density of states calculated from the Coulomb gap parabola at an energy of $k_B T^*$. The value of T^* was approximately constant near the MIT (but increased for very low metal concentrations) in both the H and UH systems. If the relative permittivity increases towards the MIT in these systems, it must do so sharply over a small range of concentrations near to that of the MIT, otherwise there must be considerable changes in the background density of states (Popescu *et al* 1995).

As expected, the scale of localization, α^{-1}, increases towards the MIT and this can also be used to estimate the critical concentration y_c. Plotting α versus y, values of 0.15 and 0.16 were obtained for y_c corresponding to the UH and H samples respectively, taking the transition to be where $\alpha \to 0$. The parameters extracted are summarized in table 1.

Table 2. Parameters corresponding to metallic and transition samples of amorphous silicon–tantalum alloys. Results obtained by fitting to the form $\sigma = \sigma(0) + aT^{1/2} + bT^p$ are shown. An initial fit gave an estimate for the value of p, which came close to unity in all cases. The other values were obtained after fixing $p = 1$.

y	$\sigma(0)$ / Ω^{-1} m^{-1}	a / Ω^{-1} m^{-1} K$^{-1/2}$	b / Ω^{-1} m^{-1} K^{-1}	p
a-$Si_{1-y}Ta_y$				
0.15	400	390	0.80	1.1
0.19	14 000	120	17	1.1
0.25	49 000	−130	38	0.84
0.32	100 000	−260	42	1.0
a-$Si_{1-y}Ta_y$:H				
0.20	12 000	210	14	0.88
0.24	22 000	42	22	0.97
0.28	33 000	−40	27	0.97
0.33	54 000	−140	34	0.92

The more highly conducting films were fitted to the following formula:

$$\sigma = \sigma(0) + aT^{1/2} + bT^p \tag{7}$$

which has terms for temperature-independent scattering, electron–electron interactions and quantum interference effects respectively. The first term represents the conductivity at zero temperature, and gives another estimate of the metal–insulator transition by extrapolating to

the concentration at which $\sigma(0) \rightarrow 0$. It is relatively insensitive to the method of fitting. By this method, values of 0.16 and 0.19 were obtained for the MIT in the UH and H systems respectively. The fitted parameters are given in table 2.

The value of p was approximately equal to unity for all of the UH and H samples, indicating that the inelastic scattering was mainly due to electron–electron scattering. The negative sign in the electron–electron interaction term a has occasionally been seen at high metallic concentrations in similar systems (see, for instance, Abkemeier *et al* 1992). Here, the sign reverses as it becomes more positive towards the MIT. The coefficient b can be seen to decrease towards the MIT, weak localization becoming less significant relative to electron–electron interactions and temperature-independent effects.

Table 3. Values of the critical concentration y_c obtained by different methods for both UH and H samples of amorphous silicon–tantalum alloys.

	Method				
	Closure of E_{04}	Conductivity at 20 K versus y	Closure of Coulomb gap	Extent of localization	Conductivity at 0 K
a-$Si_{1-y}Ta_y$	0.16	0.15	0.14	0.14	0.16
a-$Si_{1-y}Ta_y$:H	0.18	0.17	0.16	0.15	0.19

In table 3 we summarize the estimates of the critical concentration y_c obtained in all the ways described above for both UH and H samples. As can be seen, the five methods lead to results which are in good agreement. A shift in the y_c-values (of about 2% to higher tantalum concentrations) is indicated for the H samples in comparison with the UH samples. With regard to the errors which appeared during these estimates, they were higher for the UH samples (due to the lack of samples having compositions near the metal–insulator transition) but low enough to enable us to be confident of the shift between UH and H samples. Using an average of the critical tantalum concentrations shown in the table, it is possible to estimate y_c-values of 0.15 and 0.17 in the UH and H alloys respectively. The shift of 2% obtained in our system is 3–4 times smaller than that observed by Dammer *et al* (1993) in a-$Si_{1-y}Ni_y$.

5. Conclusions

The effect of hydrogenation on the electronic properties of amorphous silicon–tantalum alloys has been studied by investigating their optical and electrical properties. Optical data were used only as a comparison for the electrical results, so attention has been paid mainly to the electrical behaviour of the samples in both the insulating and metallic regimes. Experimental conductivity versus temperature results were fitted to theoretical expressions and useful information about transport parameters was obtained. For the insulating samples, decreasing the temperature from RT down to 6 K, both $T^{-1/4}$- and $T^{-1/2}$-behaviour was observed. In the case of the metallic samples, the results indicated the presence of inelastic scattering due to electron–electron scattering and weak-localization effects.

Several methods have been used to estimate the position of the MIT. They yielded similar values for the critical concentration y_c. The results also show a clear shift (of about 2%) to higher values of y_c for the H samples in comparison with the UH ones. The observed shift in y_c has been ascribed to the saturation of dangling bonds (in the case of

the H samples). This prevents electron donation from the tantalum states to the dangling bonds moving the Fermi level into a region with a greater density of states, thus preventing an increase in the conductivity. We conclude that the MIT lies at y_c-values of about 0.15 and 0.17 in the UH and H samples respectively.

Acknowledgments

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