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Coulomb gap measurement in non-compensated Si:As

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Abstract. We report resistivity measurements in the temperature range 0.15–1.2 K performed on non-compensated Si:As samples doped by ion implantation on the insulating side of the metal–insulator transition. The data show a crossover of the variable-range-hopping (VRH) resistivity dependence $\rho(T)$ from $\rho \propto \exp(T_{1/4}/T)^{1/4}$ (Mott VRH predicted behaviour) to $\rho \propto \exp(T_{1/2}/T)^{1/2}$, which demonstrates the persistence of a Coulomb gap in the density of states even in the vicinity of the critical concentration. The simultaneous observation of the $T_{1/4}$ and $T_{1/2}$ parameters allows the determination of the effective size of the gap.

Great attention has been focused in the last years on electronic transport properties of semiconductors doped near the metal–insulator transition (MIT).

The theoretical relevance of these studies lies in the peculiar effect which characterizes their electronic behaviour at low temperatures, i.e. electron localization. As a matter of fact, doped semiconductors can be considered disordered systems in which the dopant atoms are randomly dispersed in a solid matrix. The MIT is driven by the overlap of randomly distributed impurity states. The Anderson theory of localization and subsequent developments [1] give a good description of the observed behaviour of critical quantities near the transition.

In the framework of the tight-binding Anderson model it is possible to introduce the interaction between electrons essentially via the insertion in the Hamiltonian of the intra-atomic potential U (Hubbard) and of the long-range Coulomb potential [2].

The effect of Coulomb correlation in doped semiconductors, inducing anomalies in the density of states, plays an important role in determining the temperature dependence of the transport and thermodynamic properties of the material at very low temperatures.

Corrections to the electron hopping energy, due to electron–hole attraction between localized states, lead to the opening of a gap at the Fermi level [3]. Passing through the MIT from the insulating to the metallic phase, a progressive filling up of the gap occurs, leaving a dip in the density of states [4].

This effect is described by the Efros–Shklovskii (ES) theory, which predicts the development of an energy gap in the single-particle density of states (DOS) at the Fermi level μ [5], whose analytical form is

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$$N(E) \propto |E-\mu|^2 \tag{1}$$

The size of the gap is

$$\Delta = \frac{e^3 g_0^{1/2}}{\kappa^{3/2}} \tag{2}$$

where e is the elementary electronic charge, κ is the dielectric constant of the semiconductor, and g_0 is the impurity DOS calculated neglecting the Coulomb interaction.

The size of the Coulomb gap is expected to be considerably reduced from its original value (2) due to the screening effect of the impurity electrons in the vicinity of the MIT, provided that the polaron enhancement of the dielectric permittivity is taken into account.

The resistivity of the impurity system in the VRH regime (dominating at very low temperature) is a good tool for the investigation of the features of the electronic DOS.

In the framework of VRH theory [6, 7], when the hopping energy is lower than the Coulomb gap width, the assumption of a parabolic form in the DOS (1) implies a resistivity dependence with temperature of the type

$$\rho(T) = \rho_1 \exp\left[(T_{1/2}/T)^{1/2}\right] \tag{3}$$

where

$$k_B T_{1/2} = \frac{\beta_1 e^2}{\kappa\lambda} \tag{4}$$

with $\beta_1 = 2.8$ [7] and λ the localization radius of the impurity electronic state.

At higher temperatures, where the hopping energy is larger than the Coulomb gap energy but lower than the inter-impurity barrier, one should observe a typical Mott VRH behaviour as

$$\rho(T) = \rho_0 \exp\left[(T_{1/4}/T)^{1/4}\right] \tag{5}$$

where

$$k_B T_{1/4} \equiv \frac{\gamma}{g(\mu)\lambda^3}. \tag{6}$$

($g(\mu)$ is the DOS at the Fermi level, $\gamma = 18 \equiv$ constant). This behaviour is due to the fact that the DOS, outside the Coulomb gap, is only a slowly varying function of energy and can be considered constant.

In this framework, approaching the MIT one should observe a crossover of the resistivity behaviour between the functional form (3) at very low temperature to the form (5) at higher temperatures.

Both the $T^{-1/4}$ and $T^{-1/2}$ exponential dependence of $\rho(T)$ have been observed in previous experiments [8–13]. Clear evidence of the existence of a gap in the DOS has been given by Zhang *et al* in highly compensated n-type CdSe [14].

In non-compensated semiconductors the importance of the Coulomb interaction is reduced due to the enhancement of multi-electron effects. This fact manifests itself in a decrease in the size of the gap. Very precise measurements, carried out by Shafarman and Castner [10] on non-compensated Si:As samples doped near the MIT, in the temperature range 0.4–10 K, show, in the whole range where the VRH is active, a Mott behaviour (5).

In a previous paper [15] we reported evidence of the presence of a sizeable Coulomb gap Δ in ion implanted Si:P. The observed value of the Coulomb gap was $\Delta/k_B \approx 3.48$ K at a concentration of 3.5×10^{18} cm^{-3}, slightly lower than the critical concentration N_c of the MIT transition for Si:P, i.e. $N_c = 3.74\times10^{18}$ cm^{-3} [16].

In this work we report resistivity measurements in the temperature range 0.15–1.2 K of two just-insulating, non-compensated, Si:As samples, with special emphasis on the temperature range 150–250 mK.

The samples were obtained by ion implantation on single-crystal, Czochralski-grown p-type ⟨100⟩ silicon slices of 4 inch diameter and 0.5 mm thickness. The silicon substrate was boron doped at a nominal concentration of $5-7\times10^{15}$ cm^{-3}. The implantation doses have been adjusted in order to have all samples nominally doped at the critical concentration of the MIT (in Si:As the critical concentration is $N_c = (8.55\pm0.10)\times10^{18}$ cm^{-3} [10]) After ion implantation all the samples were annealed at 920 C for 30 min in N_2O_2. After contact opening and metal patterning, each sample was cut into a 1 mm × 0.5 mm dice [17].

Since the impurity profile was achieved by means of multiple implants and doubly ionized ions, a calibration of the dopant concentration was obtained from $C-V$ plots and and threshold voltage measurements (see figure 1). The complete activation of the implanted dose was checked with a four-point probe and Hall effect measurements at room temperature. The optimization of the implantation condition has been reported in [18]. Small fluctuations in doping concentration are still possible, due to non-homogeneity of the implantation process. Process tolerance was estimated to be ±2%. This can justify the slight difference in the observed behaviour of the two samples here reported.

The compensation ratio $K = N_A/N_D$ (where N_A and N_D are the acceptor and the donor concentration respectively) can amount to up to 10^{-4} due to the presence of boron impurities in the silicon substrate before doping. However, a higher compensation has to be expected because of defects induced in the silicon lattice by ion implantation. This also may lead to local deviations in K owing to the extended nature of the defects.

Low-resistance electrical and thermal links with the measurement apparatus were obtained by ultrasonic bonding of Au wires on the Al pads of the samples. The thermal coupling to the bath of the ensemble was made through a large Cu plate on which was placed a Ge NTD doped crystal calibrated thermometer. The cryogenic temperatures were obtained by using an Oxford Instruments He^{3-4} dilution refrigerator, which is able to reach temperatures as low as 10 mK. Resistivity

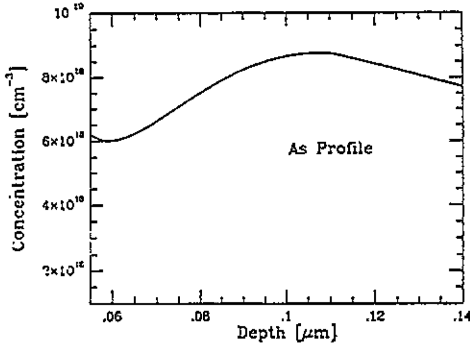

Figure 1. Concentration profile of Si:As samples as obtained from C–V plot measurement. Surface enhacement of the dopant concentration is due to impurity segregation in the oxide grown on the silicon surface during post-implant anneal.

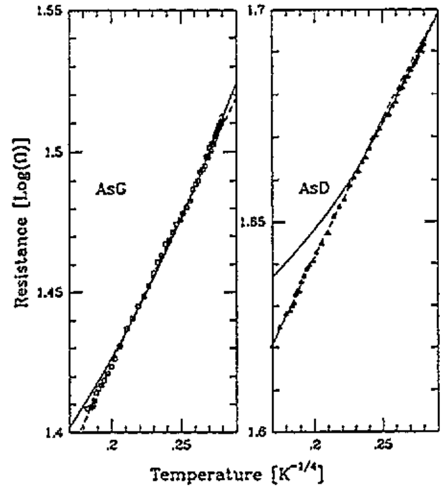

Figure 2. Resistance of the two just-insulating Si:As samples over the whole temperature range 0.150–1.2 K . The fitting lines correspond to the Mott VRH law (dashed line) and to the ES VRH (full line) behaviour, respectively.

measurements were performed at 30 Hz with an AC resistance bridge. The total power dissipation in the bolometer was kept below 10 pW in order to avoid overheating. Temperature accuracy was better than ±2% over the full range investigated.

In order to improve measurement accuracy in the temperature range 150–250 mK, the mean temperature step ΔT between the experimental points was reduced from 25 to 6 mK.

The experimental results are reported in figure 2.

The data fitting was performed following two different procedures, in order to ensure a relatively high accuracy in the determination of the functional form of the temperature dependence of the resistivity as well as the estimate of the related parameters.

One procedure is based on the determination of the logarithmic derivative of the resistivity (the sensitivity $A(T)$) defined as

$$A(T) = -\frac{\mathrm{d}(\ln \rho)}{\mathrm{d}(\ln T)} \tag{7}$$

This quantity is identically equivalent, in the VRH conduction regime, to the reduced energy W

$$W = \frac{1}{T}\frac{\mathrm{d}(\ln \rho)}{\mathrm{d}(1/T)} \tag{8}$$

which represents the dimensionless activation energy in the VRH conduction regime.

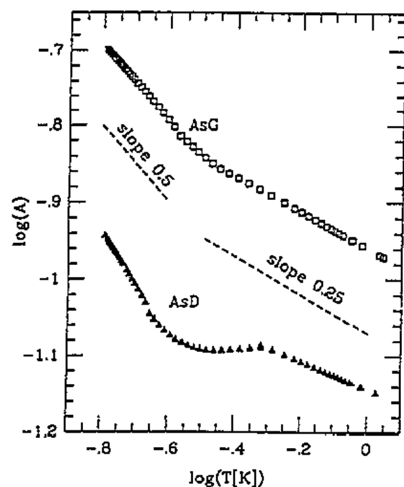

Figure 3. Logarithmic plot of sensitivity versus temperature. The slope of the linear part of the curves indicates the p parameter in the generalized VRH conduction law $\rho = \rho_0 \exp(T_p/T)^p$.

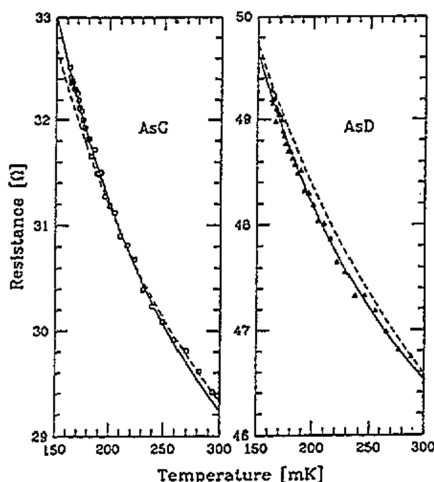

Figure 4. Resistance of the two just-insulating Si:As samples in the restricted range 0.15–0.3 K. The fitting lines correspond to the Mott VRH law (dashed line) and to the ES VRH (full line) behaviour, respectively.

The advantage of this method is that no functional dependence is assumed *a priori*, because the $\ln A$ versus $\ln T$ plot, where it follows a linear behaviour, defines the region where VRH occurs. The slope of the curve gives directly the p parameter ($\ln A = -p \ln T +$ constant). The disadvantage is that the derivative is very sensitive to any variation of the experimental points, and even small local fluctuations are amplified.

In order to overcome this drawback, we have carried out a polynomial fit of the expression $\ln(\ln \rho)$ against $\ln T$ assuming a functional dependence of the form

$$\ln(\ln \rho) = a + b(\ln T) + c(\ln T)^2 + \dots \tag{9}$$

from which we have

$$A(T) = -\ln(\rho)[b + 2c(\ln T) + \dots] \tag{10}$$

The expansion (9) was stopped at the fifth order in $\ln T$. The advantage of this method is that the double logarithmic function has the effect of smoothing the data and therefore the interpolation is much less sensitive to local fluctuations.

The reliability of this method has been checked and reported in [15]. The $A(T)$ curves of the two samples are reported in figure 3. The ρ_0, T_p and p parameters have been compared for consistency with those obtained by fitting the data with a least-square procedure to the general form of the VRH resistivity

$$\rho(T) = \rho_0 \exp\left[(T_p/T)^p\right] \tag{11}$$

where p can assume any value between 0 and 1.

The set of parameters obtained with the two different methods are more or less the same within 10% agreement in all the temperature range. The uncertainty in the VRH parameters is mainly due to the noise present in the resistivity curves, which induces some errors in the determination of the sensitivity via polynomial fit.

The linear dependence of the $\ln A(T)$ versus $\ln T$ curves of both samples above 350 mK shows that they are in a VRH condition (figure 3). Moreover the observed p values (table 1) are consistent with an electronic transport process in the Mott VRH region.

Table 1. Coulomb gap width Δ/k_B and fitting parameters T_p, p from the expression $\rho = \rho_0 \exp(T_p/T)^p$ for samples AsG and AsD doped near the MIT

Samples	n 10^{18} cm^{-3}	350–1200 mK, ΔT=25 mK T_p(mK)	p	150–250 mK, ΔT =6 mK T_p(mK)	p	Δ/k_B (mK)
AsG	$\approx$ 8.45	39.9	0.23	26.08	0.50	21
AsD	$\approx$ 8.55	6.124	0.18	9.92	0.61	13

The values assumed by the p and T_p parameters in this temperature range (table 1) are in good agreement with the one reported by Shafarman and Castner [10] for the two samples with concentration 8.41 and 8.48 $\times$ 10^{18} cm^{-3} respectively.

The small values of the $T_{1/4}$ parameter might be expected from the scaling of the Mott expression for $T_{1/4}$ with the localization length, approaching the MIT

$$T_{1/4} = [\gamma/k_B N(\mu)\lambda](1 - N/N_c)^{3\nu} \tag{12}$$

with the scaling exponent ν in the range 0.77–0.97 [10]. The observed $T_{1/4}$ values are consistent with doping concentration in the range $8.40 - 8.6 \times 10^{18}$ cm^{-3}.

The fact that we can measure such a low value of $T_{1/4}$ at temperature $T > T_{1/4}$ is due to the fact that $T_{1/4}$ is not a real activation energy, but just a weighted measure of the band width of the active states participating in the conduction. The increase of the density of states and of the localization radius near the MIT both contribute to the reduction of the width of the active energy band.

The transition in the VRH conduction behaviour can be observed by the change in the slope of the $A(T)$ curves. In fact, for both samples, $\mathrm{d}(\ln A)/\mathrm{d}(\ln T) \approx 0.5$ when $T < 250$ mK (see figure 3) according with ES VRH behaviour.

The transition can also be appreciated in the ρ versus T graph (figures 2 and 4) , where the ES VRH fitting line reproduces in a better way the data at low temperature, but fails at temperatures higher than 0.35 K.

The transition is not a masked effect due to a temperature dependence of the prefactor ρ_0 which gives a resistivity law of the type

$$\rho(T) \propto T^s \exp\left[(T_p'/T)^p\right] \tag{13}$$

Attempts to fit the data points to (13) give s-values ≈ 0.

Even in the ES VRH condition the observed values of $T_{1/2}$ are far lower than the predicted ones. However such a reduction in $T_{1/2}$ values is comparable with

the one observed near the MIT on n-type CdSe [14], n-type GaAs [19] and Si:P [15]. That is related to the divergence of both the localization radius and the dielectric permittivity approaching the metal–insulator transition.

The fact that in our samples the Mott and the ES VRH behaviour is observed simultaneously allows us to estimate quantitatively the Coulomb gap size. Combining equations (2), (4) and (6) we obtain

$$\Delta \approx k_B \left(T_{1/2}^3 / T_{1/4}\right)^{1/2} \tag{14}$$

with the assumption $g(\mu)$ = constant = g_0.

Table 1 gives the values of Δ calculated with formula (14). The inferred transition temperatures, calculated using the equation

$$\Delta / k_B = 0.24 (T_{tr} T_{1/4}^3)^{1/4}$$

from [14], are $\approx$ 100 mK for both samples. This temperature is consistent with the roughly estimated transition temperature of $\approx$ 300 mK.

In summary, from the temperature dependence of resistivity in non-compensated Si:As samples doped to the just-insulating phase of the MIT, we have observed a crossover from a Mott VRH to an ES VRH behaviour at a temperature of about 300 mK.

This leads to the conclusion that the electron–electron interaction, even if strongly reduced by multi-electron effects in the vicinity of the MIT, still affects the DOS of the impurity band giving rise to an energy gap at the Fermi level.

The fact that the same effect has not been observed in the Si:P system is probably due to the lower density of states in the phosphorous band and to the different role played by residual defects after ion implantation damage recovery [20].

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