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Doping-induced metal-insulator transition in aluminum-doped 4*H* silicon carbide

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We report an experimental determination of the doping-induced metal-insulator transition in aluminum-doped 4*H* silicon carbide. Low temperature transport measurements down to 360 mK and temperature dependent Raman experiments down to 5 K, together with secondary ion mass spectroscopy profiling, suggest a critical aluminum concentration lying between 6.4 and 8.7 $\times 10^{20}$ cm^{-3} for the metal-insulator transition in these epilayers grown by the vapor-liquid-solid technique. Preliminary indications of a superconducting transition in the metallic sample are presented. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2885081]

SiC is a promising semiconducting material for manufacturing high-power, high-temperature, and high-speed electronic devices, in particular, the 4*H* polytype. In order to minimize on-state losses in SiC devices, highly doped *p*-type layers are necessary to reduce the Ohmic contact resistance of devices below 1×10^{-5} Ω cm^2. Aluminum is the usual shallow acceptor dopant in SiC crystals and epilayers with an activation energy of about 191 meV (from photoluminescence studies[1]) or around 205 meV (from electronic transport studies[2]). Very high doping concentrations are needed in order to obtain low resistivity material. Aluminum implantation is commonly used to prepare highly doped samples but problems related to the annealing of the crystal damage and the electrical activation of acceptor atoms in SiC still remain.[3,4] As an alternative, vapor-liquid-solid (VLS) mechanism in Al–Si melt has been shown to be an appropriate method for growing high quality epitaxial 4*H*-SiC layers with a high amount of aluminum.[5] Besides the technological relevance of heavily doped SiC, the study of doping-induced insulator to metal transitions in wide band gap semiconductors is interesting in its own sight. Persson *et al.* have studied theoretically the metal-insulator transition in *p*-type SiC polytypes.[6] Hereby, Mott's original model, an extended Mott–Hubbard model, and a model based on the total energy of the metallic and nonmetallic phases have been used and led to values of the critical aluminum concentration in the same range, with an upper limit of 2.7×10^{20} cm^{-3}.[6] The electrical transport properties of highly doped 4*H*-SiC samples prepared by aluminum implantation have been reported for samples with n_{Al} up to 2×10^{21} cm^{-3} but no metallic behavior could be found (finite activation energy of the conductivity at low temperatures).[2,7–9] Recently, renewed interest came up for the metal-insulator transition in *n* type SiC polytypes.[10] In this paper, we report on the experimental observation of the metal-insulator transition in highly aluminum-doped 4*H*-SiC polytype.

The Al-doped epitaxial 4*H*-SiC layers were grown by a VLS mechanism in an Al–Si melt at low temperatures (1100 °C) on n^+ doped Si face 4*H*-SiC (0001) substrate, 8° off oriented toward [11$\bar{2}$0]. The growth conditions have been described elsewhere in detail.[11] In the following, VLS93 and VLS98 are the names of the insulating (3×4 mm^2) and the metallic (2×3 mm^2) layers, respectively. Visible micro-Raman backscattering studies have been performed at room temperature using a HeNe cw laser (632.8 nm) and a $\times 100$ objective under confocal conditions. The setup configuration of the laser and the grating led to a mostly parallel scattering geometry. Low temperature measurements were obtained at about 5 K using a He flow cryostat and a long working distance at $\times 50$ objective. A liquid nitrogen cooled charged-coupled device was used for signal collection. Electrical resistivity measurements were performed using a Quantum Design physical properties measurements system from room temperature down to 360 mK, with a four terminal configuration for the contacts (no mesa structure and no passivation process). The aluminum concentration n_{Al} has been deduced from secondary ion mass spectroscopy (SIMS) depth profiles.

In Fig. 1, one can see the SIMS profile of two samples grown by the VLS mechanism with aluminum concentrations of about $(3.4 – 6.4) \times 10^{20}$ cm^{-3} for sample VLS93 and of about 8.7×10^{20} cm^{-3} for sample VLS98. The uniformity of doping is an indication of the high quality of the epilayers grown by the VLS mechanism, as previously reported using Raman spectroscopy and transmission electron microscopy.[5] One should note that such a high doping level is difficult to be reached by standard chemical-vapor deposition or by ion implantation without any degradation of the layer quality. The onset of the SIMS profile of sample VLS93 seen in Fig. 1 is only an artifact of the measurement. Due to the nonuniformity of the SIMS profile of sample VLS93, the uncertainty on the resulting thickness value (1.7 and 4.0 μm for samples VLS98 and VLS93, respectively) led us to refer to sheet resistance values in the following.
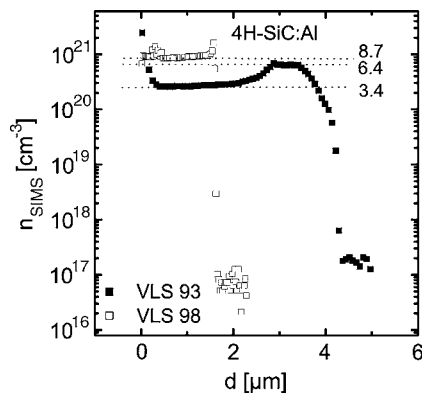
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FIG. 1. SIMS profiles for highly aluminum-doped $4H$-SiC samples VLS93 and VLS98, with aluminum concentrations of about $(3.4–6.4)\times10^{20}$ cm^{-3} for sample VLS93 and of about 8.7×10^{20} cm^{-3} for sample VLS98.

The temperature dependence of the sheet resistance down to 360 mK for the two different aluminum-doped $4H$-SiC samples is shown in Figs. 2(b) and 2(c), respectively. One clearly sees that sample VLS93 is following an insulating behavior while sample VLS98 follows a metallic behavior $d\rho/dT<0$. Assuming a doping efficiency of 100%, the critical concentration of aluminum for the doping-induced metal-insulator transition lies in the range between 3.4 and 8.7×10^{20} cm^{-3} as determined from SIMS. In Fig. 2(a), the doping dependence of the activation energy E_{act} of the aluminum level in $4H$-SiC is shown. Experimental values of Ivanov *et al.*,[12] Matsuura *et al.*,[13] and Pernot *et al.*[2] are included. The theoretical concentration dependence following $E_{act}=E_0-\alpha\times n_{Al}^{1/3}$ with $E_0=220$ meV and $\alpha=2.32\times10^{-5}$ meV cm is indicated. As mentioned, Persson *et al.* predicted a critical value of aluminum concentration of 2.7×10^{20} cm^{-3},[6] already questioned by the experimental findings of several groups.[2,7–9] Matsuura *et al.*[13] and Pernot *et al.*[2] calculated values for α of about 1.9×10^{-5} and 1.7×10^{-5} meV cm, respectively. The upper limit of 8.7×10^{20} cm^{-3} for the aluminum concentration determined in this work can be used to calculate a lower limit of $\alpha=2.32\times10^{-5}$ meV cm.

As shown in Fig. 2(b), the sample VLS93 follows an insulating behavior. Attention should be paied to the logarithmic scale for the sheet resistance, indicating an increase of about three orders of magnitude down to low temperatures. No general fitting in terms of hopping conduction or temperature activated behavior was possible, and the conductance of sample VLS93 showing a remarkable linear temperature dependence over the whole temperature range is probably a result of the nonuniformity of the carrier concentration as indicated by the SIMS profile in Fig. 1.

Figure 2(c) shows the temperature dependence of the sheet resistance for the aluminum-doped $4H$-SiC samples VLS98, showing a clear metallic behavior $d\rho/dT<0$. The increase in resistance at low temperatures can be explained by localization effects and electron-electron interactions typical for disordered metals (see Ref. 14 and references herein), leading to

$$\sigma_T = \sigma_0 + A\times T^{1/2} + B\times T^{p/2}. \qquad (1)$$

The appearance of a drop in resistance shown in Fig. 2(c) at very low temperatures up to about 7 K was verified several times and may indicate the onset of a superconducting transition, probably masked by the high disorder and
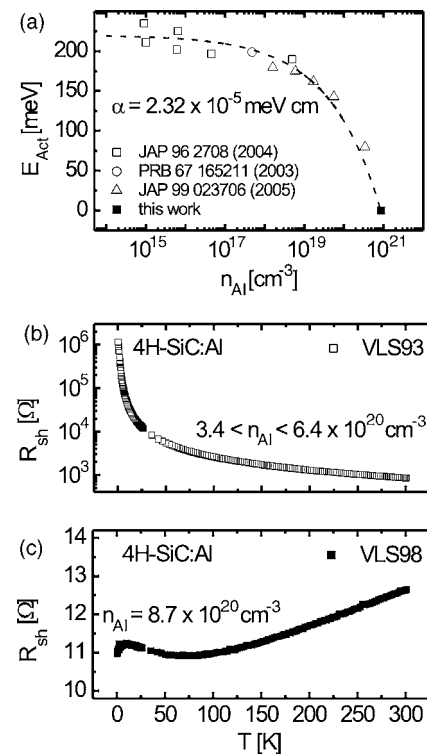

FIG. 2. (a) Doping dependence of the activation energy E_{act} on the aluminum level in $4H$-SiC. Experimental values of Ivanov *et al.* (Ref. 12) Matsuura *et al.* (Ref. 13) and Pernot *et al.* (Ref. 2) are included. The dashed line represents the theoretical concentration dependence following $E_{act}=E_0-\alpha\times n_{Al}^{1/3}$ with $E_0=220$ meV and $\alpha=2.32\times10^{-5}$ meV cm; (b) Temperature dependence of the sheet resistance (logarithmic scale) for VLS93, showing a clear insulating behaviour. (c) Temperature dependence of the sheet resistance (linear scale) of sample VLS98. A metallic behaviour $d\rho/dT<0$ is clearly seen. The appearance of a drop in resistance at very low temperatures probably indicates the onset of a superconducting transition. Attention should be payed to the different scales of the resistance.

inhomogeneity of the sample. Indeed, superconductivity was found recently in boron-doped SiC with a critical temperature of $T_c\approx1.4$ K.[15] A similar behavior of the temperature dependence of the resistivity was seen in heavily boron-doped silicon,[16] as well as heavily boron-doped diamond.[17]

Raman spectroscopy clearly identifies the SiC polytype to be $4H$-SiC.[18] Distortion and asymmetry of Raman bands occur when scattering by a discrete phonon state interferes with a broad continuum of electronic states.[19] This effect, called Fano interference or resonance, was first observed in SiC by Colwell and Klein.[20] The Fano resonance seen around the central TO phonon line at 776 cm^{-1} is a clear indication of a high free carrier concentration.[18] Figure 3 shows the Raman spectra (633 nm) of the two aluminum-doped $4H$-SiC samples VLS93 and VLS98 taken at room temperature and at around 5 K, respectively. One clearly sees that the Fano resonance persists in the case of the sample VLS98 down to 5 K, therefore, following a clear metallic behavior. In contrast to this, the Fano resonance seen in sample VLS93 had completely disappeared at around 5 K. This indicates a strong decrease in the number of free carriers with decreasing temperature as expected for an insulating sample.

In conclusion, we reported on the experimental findings of the metal-insulator transition in aluminum-doped $4H$-SiC samples grown from a VLS mechanism. Temperature dependent conductivity and Raman experiments clearly indicate
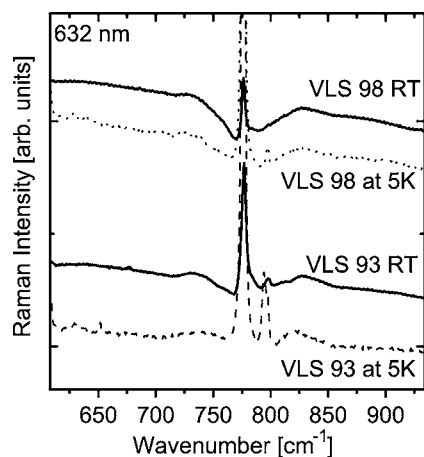
FIG. 3. Comparison of Raman spectra (632 nm) taken at room temperature and 4 K for two aluminum-doped samples VLS93 and VLS98. All samples show a clear Fano resonance at room temperature. Only for sample VLS98 does the Fano resonance persist down to 5 K, indicating no significant change in the carrier concentration and, therefore, suggesting metallic behavior.

the threshold of the critical aluminum concentration to lie in the range $(6.4-8.7) \times 10^{20}$ cm^{-3} as determined from SIMS.

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