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Microstructure of Al/GaAs Interface Studied by Slow Positron Beam

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The interface models were founded, and the expressions of the relation between Doppler broadening S-parameter and the implanted energy of positrons were got based on the diffusion equation of positrons. Interfaces formed between Al overlayer and the (110) surface of GaAs were studied with slow positron beam. We found the system of Al/GaAs interfaces could be well described by the perfectly absorbing linear interface model. From the results fitted by this model, we obtained the relation between the *S*-parameter of the interface and the annealing temperatures or the overlayer thickness. The microstructure of interfaces and its dynamical characters were discussed.

1. INTRODUCTION

Because of the importance of interfaces in electronics and materials science, the physical properties of interfaces have been studied extensively. Many techniques, such as Auger Electron Spectroscopy, Transmission Electron Microscopy and Rutherford Backscattering etc., have been used

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to detect the microstructure of interfaces. However, the non-destructive measurement of the interface remains to be a difficult problem. Positron annihilation technique (PAT) is a well-established means to research the type and the concentration of defects in solids due to the high sensitivity of positron annihilation features to the sampled area by positrons. But positrons from radioactive isotopes used in the conventional PAT are emitted over a broad distribution of energy. It is very difficult to use these positrons for the microstructure investigation of the heterogeneous structure system, for example, the interfaces near a surface, the ion implantation region, various strain layers and so on. A slow positron beam consists of monoenergetic positrons, which energy can be varied by adjusting the different accelerating potential. Positrons with different energy could be implanted into the different depth of solid. Measuring the energy spectrum of the annihilation photons could give the depth profile of the defects [1,2]. Comparing with other techniques, slow positron beam is a unique method with the high defect sensitivity and non-destructions.

In this paper, the Al/GaAs series of samples with different Al overlayer thickness and various annealing temperatures were studied by a probe of slow positron beam. We give up the qualitative analysis for positron energy dependence of S parameters used in the previous researches and attempt to find a new, more quantitative method directly by solving the positron diffusion equation with several interface model. The results show that the perfectly absorbing linear interface model can reproduce the experimental data and could be used to reveal the dynamical characteristics of the interfaces of Al/GaAs.

2. EXPERIMENT

The experiment was performed using the slow positron beam facility at USTC in Hefei [3]. Fast positrons from a ^{22}Na source with a energy of several hundred keV were emitted to a well-annealed W (tungsten)-moderator. The slow positrons remitted from the moderator surface were focused, accelerated and constituted a monoenergetic positron beam (FWHM <2 eV) which would be transported to the target chamber. The beam intensity was 2×10^4/s. The voltage of the sample support was supplied by a 30 kV stable voltage source with which the positron energy could be adjusted in broad region from 0 to 20 keV. The vacuum in the target chamber was below 2×10^{-4} Pa. The detection system consisted of a high pure Ge detector (ORTEC Model GEM-10175) and a IBM PC/XT computer multichannel analyzer. The energy resolution (FWHM) is 1.12 keV for 514 keV γ-ray of ^{85}Sr.

The samples of Al/GaAs were prepared by University of Hong Kong. The substrates are undoped single-crystals of GaAs with (110) surfaces. In order to prevent the surface from serious oxidation the substrates were passivated by HN_4 before overlaying [4]. The Al films were deposited by means of resistivity evaporation with a rate about 30-50 A/s. The purity of Al was 99.999%. The vacuum during the deposition was about 1×10^{-4} Pa. The samples had different overlayer thicknesses or annealing temperatures. The annealing was performed under a forming gas (80% N_2, 20% H_2). The annealing time was 30 min for all annealed samples.

The Doppler broadening technique of the annihilation radiation was adopted. Owing to the different contribution of the annihilating pairs with different momentum to the shape of the energy spectra at 511 keV, various shape parameters could be defined to describe the shape feature of the measured spectrum. In terms of the variation in these shape parameter the problems of defects, phase transition etc. could be investigated. The S parameter of Doppler broadening spectrum is defined as following:

$$S = \int_{-a}^{+a} C(E)\mathrm{d}E \Big/ \int_{-\infty}^{+\infty} C(E)\mathrm{d}E, \tag{1}$$

where $C(E)$ is the measured spectrum with the background subtracted, $(-a, a)$ is a energy region in

the coordinate axis with original point at the peak energy 511 keV, here a is 1 keV. The variation of S-parameter is mainly influenced by the relative counts around the peak, so it provides the information about the positron annihilating with the lower momentum electrons. At the defect region, the electron density decreases and the density of the core electrons with higher momentum decreases more rapidly than others. Because the defects, especially the vacancy-type defects, have a certain ability trapping positrons, the contribution of low momentum electrons to the energy spectrum increases at the defect region and the S parameter was raised. Considering the statistical precision, the total counts were 1×10^5 for each spectrum in this experiment.

3. LINEAR INTERFACE MODELS

When a positron enters into a solid, it is thermalized during a very short period (~ 1ps). At this time the positron distribution is called the implantation profile.

The researchers of BNL in USA simulated the behavior of positrons implanted into a electronic gas with the Monte Carlo method and got the positron implantation profile as following:

$$P(z,E) = \frac{m}{z_0}\left(\frac{z}{z_0}\right)^{m-1}\exp\left[-(z/z_0)^m\right], \tag{2}$$

$$z_0 = \bar{z}/\Gamma\left(\frac{1}{m}+1\right), \tag{3}$$

$$\bar{z} = \frac{\alpha}{\rho}\cdot E^n, \tag{4}$$

where ρ is the density of the material, E is the incident positron energy, $\Gamma(1/m+1)$ is the Γ function, α, n and m are the constants:

$$\alpha = 4.00 \times 10^2 \mathrm{g/cm^2 \cdot keV^n} \tag{5}$$

$$n = 1.6; \tag{6}$$

$$m = 2. \tag{7}$$

The propagation of thermalized positrons can be described by a diffusion equation [1,2]. From the Fick second law and considering the various correction terms, the positron diffusion equation is of the form:

$$\frac{\partial n(z)}{\partial t} = D\frac{\partial^2 n(z)}{\partial z^2} - \frac{n(z)}{\tau_{\mathrm{eff}}} + \dot{n}_0(z), \tag{8}$$

where $n(z)$ is the positron density, D is the positron diffusion coefficient, τ_{eff} is the effective lifetime of the free diffusing positrons and $\dot{n}_0(z)$ is the deposited rate of thermalized positrons, that is, the positron density arriving at z in a unit time.

1) Consider a homogeneous, semi-infinite system. The expression of the S parameter could be obtained after solving Eq.(8):

$$S = S_{\mathrm{B}} + (S_{\mathrm{S}} - S_{\mathrm{B}})\left\{1 - \frac{\sqrt{\pi}}{2}\cdot\frac{z_0}{L_{\mathrm{eff}}}e^{\left(\frac{z_0}{2L_{\mathrm{eff}}}\right)^2}\mathrm{erfc}\left(\frac{z_0}{2L_{\mathrm{eff}}}\right)\right\}, \tag{9}$$

where $L_{\mathrm{eff}} = (D\tau_{\mathrm{eff}})^{1/2}$ is the effective diffusing length of positrons; S_{B} and S_{S} are the bulk S parameter

and the surface S parameter, respectively. The result of Eq.(9) is the basis of investigating interfaces or other systems. From this expression we can get the surface S parameter S_S, the bulk S parameter S_B and the effective diffusion length of positrons L_{eff} in various homogeneous materials to facilitate studying the microstructures of complicated systems.

2) The freely diffusing linear interface model

In order to reveal the microstructure character of the interface near surface, we consider a bilayer system (see Fig. 1) and assume the interface between the two materials is an ideal contact, namely, there is no any interface effect disturbing the free diffusion of positrons.

Then we can get the S parameter from the diffusion Eq.(8):

$$\begin{aligned}
S = S_S \cdot \{ & \frac{e^{T/L_O}}{2Q} \cdot I(0, \frac{T}{z_0} \cdot \frac{z_0}{2L_O}) (\frac{D_B}{D_O} \cdot \frac{1}{L_B} + \frac{1}{L_O}) \\
& - \frac{e^{-T/L_O}}{2Q} \cdot I(0, \frac{T}{z_0} \cdot - \frac{z_0}{2L_O}) \cdot (\frac{D_B}{D_O} \cdot \frac{1}{L_B} - \frac{1}{L_O}) \\
& + \frac{e^{T/L_B}}{2Q} \cdot I(\frac{T}{z_0}, \infty, \frac{z_0}{2L_B}) \cdot \frac{2}{L_O} \} \\
& + S_O \cdot \{ I(0, \frac{T}{z_0}, 0) \\
& + \frac{I(0, \frac{T}{z_0}, \frac{z_0}{zL_O})}{2Q} \cdot [\frac{D_B}{D_O} \cdot \frac{1}{L_B} \cdot (1 - e^{\frac{T}{L_O}}) - e^{\frac{T}{L_O}} \cdot \frac{1}{L_O}] \\
& + \frac{I(0, \frac{T}{z_0}, - \frac{z_0}{zL_O})}{2Q} \cdot [\frac{D_B}{D_O} \cdot \frac{1}{L_B} \cdot (e^{-\frac{T}{L_O}} - 1) - e^{-\frac{T}{L_O}} \cdot \frac{1}{L_O}] \\
& + \frac{I(\frac{T}{z_0}, \infty, \frac{z_0}{2L_B})}{2Q} \cdot 2e^{\frac{T}{L_B}} \cdot [\frac{1}{L_O} ch(\frac{T}{L_O}) - \frac{1}{L_O}] \} \\
& + S_B \cdot \{ I(\frac{T}{z_0}, \infty, 0) \\
& + \frac{D_B}{D_O} \cdot \frac{1}{L_B} \cdot \frac{[I(0, \frac{T}{z_0}, - \frac{z_0}{2L_O}) - I(0, \frac{T}{z_0}, \frac{z_0}{2L_O})]}{2Q} \\
& - \frac{e^{\frac{T}{L_B}}}{Q} \cdot I(\frac{T}{z_0}, \infty, \frac{z_0}{2L_B}) \cdot \frac{1}{L_O} \cdot ch(\frac{T}{L_O}) \},
\end{aligned} \tag{10}$$

where

$$I(a,b,c) \equiv \int_a^b 2y e^{-(2cy+y^2)} dy; \tag{11}$$

$$Q = \frac{D_B}{D_O} \cdot \frac{1}{L_B} sh(\frac{T}{L_O}) + \frac{1}{L_O} ch(\frac{T}{L_O}). \tag{12}$$

Here we denote parameters of the surface, of the overlayer and the of substrate by subscripts S, O and B, respectively.

3) Perfectly absorbing linear interface model

Here we consider another bilayer structure. In this case we assume the interface is a perfect

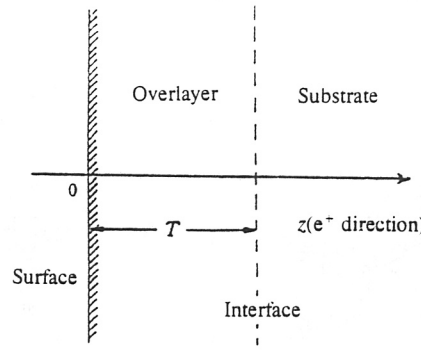

Fig. 1
Sketch of the freely diffusing linear interface model. The thickness of the overlayer is T and the thickness of the interface layer is neglected.

absorber of positrons. This is suitable for the case there are a large number of defects at the interface. In the same way, the expression of the S parameter could be obtained from the diffusion Eq.(8):

$$
\begin{aligned}
S = & S_S \cdot \{\frac{1}{2\mathrm{sh}(\frac{T}{L_O})}[e^{\frac{T}{L_O}} I(0,\frac{T}{z_0},\frac{z_0}{2L_O}) - e^{-\frac{T}{L_O}} I(0,\frac{T}{z_0},-\frac{z_0}{2L_O})]\} \\
& + S_O \cdot \{I(0,\frac{T}{z_0},0) + \frac{(1-e^{\frac{T}{L_O}})}{2\mathrm{sh}(\frac{T}{L_O})} \cdot I(0,\frac{T}{z_0}\cdot\frac{z_0}{2L_O}) \\
& + \frac{(e^{-\frac{T}{L_O}}-1)}{2\mathrm{sh}(\frac{T}{L_O})} I(0,\frac{T}{z_0},-\frac{z_0}{2L_O})\} \\
& + S_I \cdot \{\frac{1}{2\mathrm{sh}(\frac{T}{L_O})} \cdot [I(0,\frac{T}{z_0},-\frac{z_0}{2L_O}) - I(0,\frac{T}{z_0},\frac{z_0}{2L_O})] \\
& + e^{\frac{T}{L_B}} \cdot I(\frac{T}{z_0},\infty,\frac{z_0}{2L_B})\} \\
& + S_B \cdot \{e^{-(\frac{T}{z_0})^2} - e^{\frac{T}{L_B}} \cdot I(\frac{T}{z_0},\infty,\frac{z_0}{2L_B})\},
\end{aligned}
\tag{13}
$$

where the parameters of the interface are denoted by subscript I.

From diffusion equation we have obtained the expressions of the relations between S parameter and implanted energy of positrons for the two interface models. Comparing these models with the results of experiment, we could understand some microstructure characteristics of the interfaces.

4. RESULTS AND DISCUSSION

4.1 Comparison of the Two Linear Interface Models with Experiments

In order to observe the sensitivity of S parameter to the existence of interfaces, we compare the freely diffusing linear interface model and the perfectly absorbing linear interface model with the

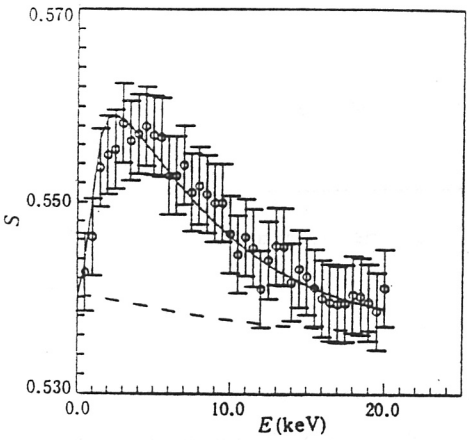

Fig. 2
Results of 300 Å Al/GaAs (unannealed).

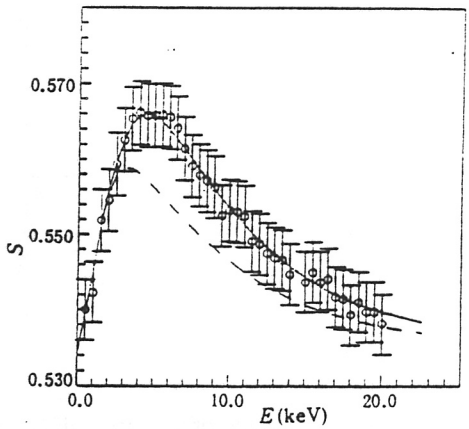

Fig. 3
Results of 1500 Å Al/GaAs (400°C annealed).

experiment results. Fig.2 shows the results of the unannealed sample of 300 Å Al/GaAs. The solid line is the result fitted by the perfectly absorbing linear interface model and the dashed line is the calculated result with the freely diffusing linear interface model. Obviously the experiment data are consistent with the prediction of the perfectly absorbing interface model and far away from the prediction of the freely diffusing interface model. The situation for the annealed samples is also the same. Fig.3 is the results of the annealed sample at 400°C of 1500 Å Al/GaAs.

So we could draw a conclusion that the Al/GaAs interfaces could be well described with the perfectly absorbing linear interface model, and we could analyze the microstructure of Al/GaAs interfaces with this model.

4.2 Microstructure Analysis of Al/GaAs Interface by Slow Positron Beam

In order make the S parameter analysis to be more rational, we must have a primary understanding for the basic feature of Al/GaAs system. In the process of preparing the samples, the surfaces of GaAs were bombarded with Al atoms and were damaged. The damaged extent increases with the Al atoms deposited increases. Meanwhile the thickening of the overlayer is also a reconstruction process of the overlayer. The structure for a thinner overlayer is looser than that for a thicker overlayer. Annealing will also make the overlayer reconstructed and recover the defects in it. The replacement reaction takes place very easily between Al and GaAs [5,6]:

(14)

When the annealing temperature is over 200-250°C, the reaction proceeds violently. A more stable phase $Al_xGa_{1-x}As$ and a wandering state of Ga are produced. Al and Ga all belong to the main group III and have similar properties of atom structures, such as atomic valence, ion radius, etc.. So if the structure of the Al overlayer near the interface is loose, Ga might fill in the vacancies or other defects in the Al overlayer. If the structure of the Al overlayer near the interface is compact, a large number of defects might be generated in the vicinity of the interface by the wandering Ga diffusing. In face, the two motion mechanisms appear synchronously. At this time, the main defects around the interface are V_{Al} or V_{Ga}.

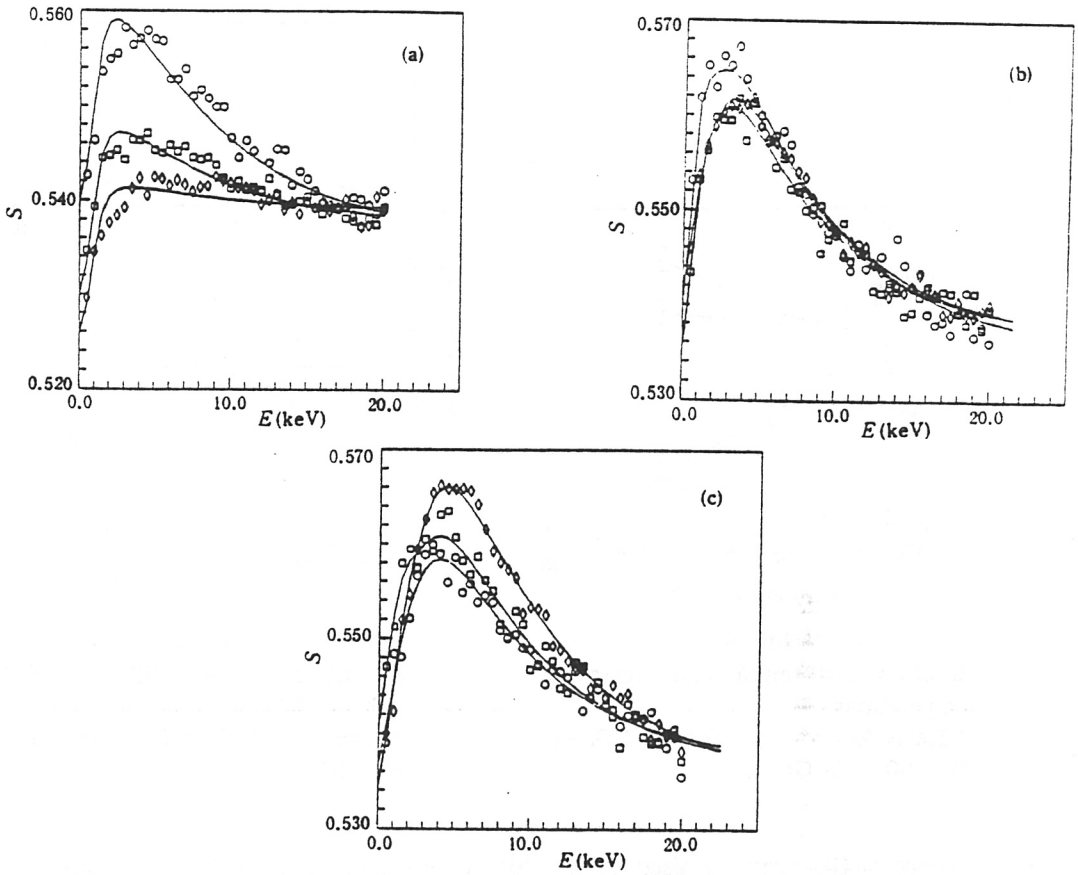

Fig. 4

(a) Results of 300 Å Al/GaAs with different annealing temperatures. (b) Results of 1000 Å Al/GaAs with different annealing temperatures. (c) Results of 1500 Å Al/GaAs with different annealing temperatures. ○ unannealed; □ 200°C annealed; ◇ 400°C annealed; The solid lines is the fitting results with the perfectly absorbing linear interface model.

Fig.4 is the results of Al/GaAs with the three different thicknesses or different annealed temperatures. The solid lines in figure are the fitting curves using the perfectly absorbing linear interface model. The obtained interface S parameter, S_I, can be seen in Fig.5. S_I decreased for the overlayer thickness T = 300 Å and S_I increased for T = 1500 Å with the increasing annealing temperature. While T = 1000 Å, S_I has a dropping tendency when the annealing temperature is 200°C and has a raising tendency when the sample is annealed at 400°C.

From the former analysis, the variation of S_I could be explained as follows. The structure of the 300 Å Al overlayer is looser, so in the process of annealing, the main variations of the samples are the generation of $Al_xGa_{1-x}As$ and the reconstruction or the recovery of the defects in the overlayer. Therefore the value of S_I descend as the annealing temperature increasing. For the case of T = 1000 Å, the overlayer is compact and the recovery of Ga atoms with the defects in it will be rarely happened. Thus S_I slightly decreases when annealing temperature is 200°C and slightly increases when the samples is annealed at 400°C. Comparing with the case of T = 300 Å, many new defects are formed for T = 1000 Å case. When T = 1500 Å the overlayer is so compact that the generating

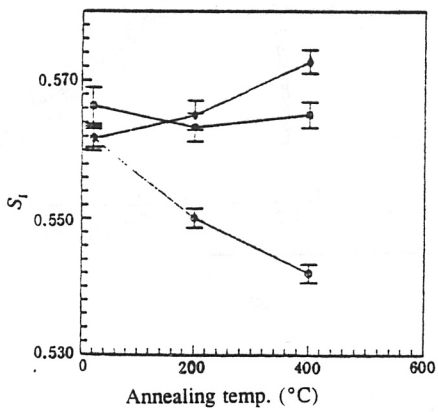

Fig. 5
Relation between S_I and annealing temperatures.
○ 300 Å Al/GaAs; □ 1000 Å Al/GaAs;
◇ 1500 Å Al/GaAs.

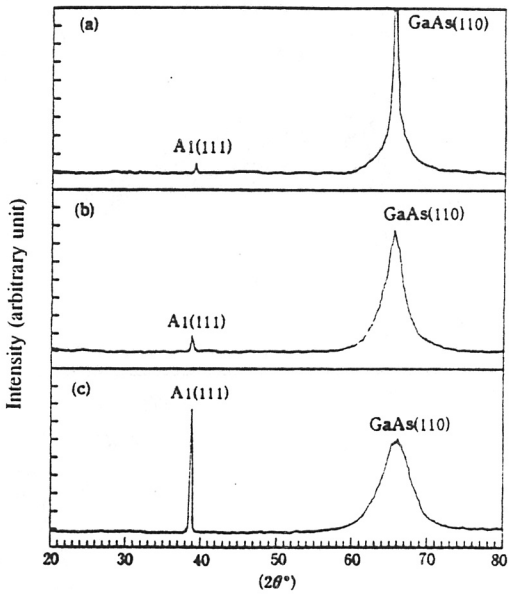

Fig. 6
The pattern of X-ray diffraction for 1000 Å Al/GaAs.
(a) unannealed; (b) 200°C annealed;
(c) 400°C annealed.

possibility of the Ga vacancy is raised greatly which leads to S_I increasing with the increasing annealing temperature. The above analysis indicates that the S parameter is more sensitive to V_{Ga} or V_{Al} at Al/GaAs interface, namely, V_{Ga} and V_{Al} have a stronger ability to trap positrons. As to the defects in the substrate GaAs, only V_{Ga} could trap positrons because of the selectivity of positrons to defects [4,5]. The diffusion of Al into GaAs can generate various defects, e.g., V_{Ga}, V_{As}, As_{Ga}, Ga_{As}, etc., but V_{Ga} could be replaced by following reaction [4]:

$$V_{Ga} + As \longrightarrow As_{Ga} + V_{As}. \tag{15}$$

Nevertheless the concentration of V_{Ga} is too small which cannot be clearly identified in statistical precision. The results show that most of the defects trapping positrons concentrate near the interface. So it is reasonable to analyze the Al/GaAs interfaces with the perfectly absorbing linear interface model.

4.3 Support of X-Ray Diffraction to Analysis of *S* Parameter

In order to verify the reliability of the analysis of S parameter based on positron annihilation technique, we have checked microstructure of the sample Al/GaAs with T = 1000 Å at different annealing temperature by X-rays diffraction. The results are indicated in Fig.6. From the figure we can find the structure of the overlayer Al was reconstructed when the sample was annealed. The overlayer is orientated along (111) direction in the unannealed sample. The annealing at 400°C improves the orientation degree. Meanwhile it could be found that the lattice of the substrate near the

interface has been distorted. So the discussions of S parameter in this paper are reasonable.

5. CONCLUSIONS

The interface models were founded, and the analytical expressions of the relation between Doppler broadening S parameter and the implanted energy of positrons, $S(E)$, have been got based on the diffusion equation of positrons. $S(E)$ was measured for Al/GaAs with different overlayer thicknesses and different annealing temperatures. Fitting the experimental results with the expressions, we found the system of Al/GaAs interfaces can be well described by the perfectly absorbing linear interface model. From the results fitted by this model, we obtained the relation between the S parameter of the interface and the annealing temperatures or the overlayer thickness. This reveals to some extent the microstructure of interfaces and its dynamical characters.

From Fig.4(a), we can find a certain difference between the prediction of the perfectly absorbing linear interface model and the experiment data for Al/GaAs samples with T = 300 Å when the implanted energy of positrons was 1.5 to 3.5 keV. So the interface model needs further improvement in future.

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