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ABSTRACT

During MeV $^{14}N^{+}$ backscattering analysis to detect sub-monolayer heavy impurities, a "beam effect", i.e. high impurity loss was found. To clarify the situation a systematic study was done on gold evaporated films /in the thickness range of 0.5-3200 atom/nm^2/ onto silicon. Results suggest that sputtering of cascades induced by energetic nitrogen ions is responsible for the phenomenon. The sputtering yield of gold was a linear function of surface coverage in the range of 0.5-130 atom/nm^2. For thick overlayers /$\geq$ 800 atom/nm^2/ a saturation value of S $\simeq$ 0.8 gold/N^{+} was found. Between these two regions intermediate behaviour was experienced. A rough theoretical model is outlined for overlayer sputtering in the MeV energy region.

АННОТАЦИЯ

Для определения тяжелых поверхностных примесей, менее тонких, чем монослой, успешно применяется анализ, основанный на обратном рассеянии ионов $^{14}N^{+}$ в области энергии порядка МэВ. При его осуществлении наблюдается "эффект пучка", количество поверхностных примесей в ходе анализа удивительно быстро уменьшается. Для объяснения этого эффекта проводятся систематические исследования на слое золота, напыленного на кремний в области толщин 0,5-3200 ат/нм^2. Полученные результаты указывают на то, что ответственным за процесс является распыление, вызванное ионами азота с энергией порядка МэВ. Выход распыления золота в области 0,5-130 ат/нм^2 линейно зависит от толщины покрытия. В случае толстых слоев /$\geq$800 ат/нм^2/ насыщение составляет S $\simeq$ 0,8 Au/N^{+}. Показано переходное поведение между двумя областями. Разработана также теоретическая феноменологическая модель для количественного описания явления.

KIVONAT

A monorétegnél vékonyabb nehéz felületi szennyezők kimutatására igen alkalmas a MeV energiáju $^{14}N^{+}$-ionok visszaszórásán alapuló analizis. Ennek megvalósitása folyamán "nyaláb effektust" tapasztaltunk, a felületi szennyezők mennyisége az analizis során meglepően gyorsan csökkent. Az effektus megértésére szisztematikus vizsgálatokat végeztünk sziliciumra párolt aranyrétegeken a 0,5-3200 atom/nm^2 vastagságtartományban. Az eredmények arra utalnak, hogy a MeV-es energiáju nitrogén ionok okozta porlódás felelős a folyamatért. Az arany porlódási hozama a 0,5-130 atom/nm^2 vastagságtartományban a bevonat vastagságának lineáris függvénye. Vastag rétegek esetén /$\geq$ 800 atom/nm^2/ S $\simeq$ 0.8 Au/N^{+} értékü telitést találtunk. A két tartomány között átmeneti viselkedést mutattunk ki. A fentieken tul fenomenologikus elméleti modellt dolgoztunk ki a jelenség kvantitativ leirására.

1. Introduction

Rutherford backscattering /RBS/ has proved to be an effective method for surface layer analysis. It is often referred that the non-destructive character would be one of its basic advantages. Some indications existed, however, that "beam effect" could be experienced even with light ion bombardment[1]. Using MeV energy nitrogen ions, the detection limits for heavy impurities on the surface will be lowered and this seems to be the most sensitive and straightforward method to check plasma contamination in CTR. This idea was proposed by Dearnaley et al.[2]. Previous papers also emphasized that using heavier ions for RBS (C^+, O^+, N^+), radiation damage might occur both on the target and surface barrier detector.

We have employed this technique for similar purposes and during the check runs for sensitivity and reproducibility to detect sub-monolayer gold, iron, molibdenum etc. impurities on silicon, surprisingly high impurity losses were found. To clarify the nature of this "beam effect", a systematic study was made using evaporated gold films on silicon and it is suggested that sputtering of cascades initiated by energetic $^{14}N^+$ ions are responsible for this artifact

2. Experimental

To study the beam effect i.e. for sputtering measurements, gold films of several thicknesses (0.5, 3, 11, 52, 130, 310, 900, 3200 atom/nm^2) were prepared by vacuum evaporation onto 4 Ωcm chemically polished silicon single-crystals with <111> orientation. For analysis, 2 MeV $^{14}N^+$ beam from a 5 MeV Van de Graaff generator was used. As for a crucial point when absolute sputtering yields are measured, special care was taken to detect bombarding dose properly. Both the conventional current integration with electron suppression, and the monitoring of scattering yield from a 2 nm gold-covered carbon propeller were applied. Both methods were calibrated first. For this purpose two type of samples were used. Heliur. backscattering measurements were done on several spots on an approximately 1 nm thick gold film on silicon to calculate the average quantity. The lateral homogenity of this sample was about 4 %. The second way of calibration accepted the surface yield of a 40 nm gold film to be accurate and the bombarding dose was determined using tabulated yield and stopping power data from literature[3),4)].

Standard silicon surface barrier detector with resolution 13 keV for $^4He^+$ and 40 keV for $^{14}N^+$ particles was placed at 12 cm distance from the target with a collimator system. The solid angle was 1.24 msr. Measuring the area of the bombarded spot one could calculate the total nitrogen dose on

the unit area with a maximum error of 15 %.

The sputtering of surface gold was investigated by successive measurements in the dose range of 0.2-10 μC /typically 1 μC/ on the same spot with a size of 1 mm^2. Typically 1-5 nA current was applied, but sputtering yields did not show any change even for 30 nA. The number of gold atoms removed by the nitrogen bombardment was calculated from the decrease of the area of gold peak. The ratio of this quantity and total dose was regarded as the sputtering yield. For thick layers, however, the broadening of gold distribution was also used to get sputtering yield. Some but non-systematic investigations were done with Fe, Co, Ni evaporated films on silicon, too, with similar result. For control, Sb implanted silicon with 30 keV energy and 10^{15} atom/cm^2 dose was also investigated similarly. In this case no antimony loss was experienced.

The vacuum was kept during measurements at 5.10^{-5}Pa. Special care was taken for pile-up inspection and dead time correction, too.

3. Results

Fig. 1 shows nitrogen backscattering spectra taken on a sample that was initially covered by 7 atom/nm^2 gold. It can be seen the loss of gold after prolonged bombardment. The

calculated sputtering yield was $(1.0\pm0.2)\cdot10^{-2}$ Au/N^+.

Fig. 2 summarizes the results. The sputtering yield is proportional to the quantity of gold on the surface in the range of coverage between 0.5-130 atom/nm^2 with the value of $S \cong 10^{-3}$ Nt, where Nt gives the number of gold atoms in $(\frac{atom}{nm^2})$ units. For thicker layers (in the range of 900-3200 atom/nm^2) saturation was found. Here S = 0.8 Au/N^+ is a maximum value, which presumably characterizes the sputtering of "infinite" thick evaporated layer.

The experimental data suggest an intermediate region between linear and saturated part of sputtering yield. The behavior of sputtering yield as a function of surface coverage will be discussed in next paragraph, where a rough theory will be outlined for thin film sputtering in the Rutherford energy region.

Beam effect of this kind is a rather unpleasant phenomenon at medium mass ion analysis. As a next step, some attempts were done to prevent the thin film sputtering. Some measurements were repeated in worse vacuum ($3\cdot10^{-4}$ Pa) where carbon deposition onto surface could occur. In these samples at the very beginning of bombardment only a little gold loss was found with smaller sputtering yield but after 1-5 μC dose, depending on the vacuum, the area of gold peak did not show any change. So one can avoid the beam effect of nitrogen ions at ultra thin film analysis with 1-3 nm carbon evaporation onto sample.

4. A theoretical speculation of thin layer sputtering

Let us consider an X average thickness of B element on an A bulk material and bombarding this system with I^+ ions of E energy.

As a function of X both A and B will be sputtered due to cascades initiated by energetic I^+ ions. The first comprehensive theory of sputtering of elemental targets was made by P. Sigmund[5]. Even a rough model as a modification of Sigmund's theory can explain the sputtering of both bulk and overlayer atoms.

According to Sigmund's theory the sputtering yield of some K elemental material for I^+ ion bombardment can be given as:

$$S_{KI}(E) = \frac{4.2}{U_o}\alpha_{KI}\varepsilon_{nKI}(E) \quad , \qquad (1)$$

where U_o is the surface binding energy of K atoms in eV units, α_{KI} is a dimensionless constant depending on mass ratio of target and projectile atoms and follows a weak energy dependence. In the MeV energy range α_{KI} is 0.5 independently of the previous parameters[5]. The $\varepsilon_{nKI}(E)$ is the nuclear stopping cross-section in $eV \cdot nm^2$ units for given projectile-target combination and energy and it can be derived from S_{nKI} nuclear stopping-power divided by N_K atomic density. Generally, this value can be calculated from the energy deposition function

into nuclear processes, F(X,E) at X = 0 point. If the ion energy is so high that recoiled atoms lose a substantial part of their energy in electronic processes, the $S_n(E)$ is to be calculated by

$$\int_0^{T_m} d\sigma(E,T)\nu(T) \quad , \tag{2}$$

where T is the energy of recoiled atoms in the target, $\nu(T)$ function gives the part of recoiled energy left in atomic motions, $d\sigma(E,T)$ is the differential cross-section of T energy transfer for E energy ions, $T_m = \gamma_{KI}E$, where $\gamma_{KI} = 4\, M_I M_K/(M_I+M_K)^2$ is the maximum of energy transfer. Calculations of this type were made by Brice[7] and tabulated values were extrapolated for the present case.

To apply Eq. 1 to thin film sputtering, first the coverage of bulk by B atoms has to be taken into account by considering a ΔX effective depth, where sputtered atoms are coming from. According to Sigmund, this ΔX can be given as 41 atoms/nm^2, independently of all parameters. Assuming that all atoms can leave this effective thickness with the same probability, the S_{BI} sputtering yield will be proportional to a dimensionless factor:

$$C(X) = \begin{cases} \frac{X}{\Delta X} \ , & \text{if } X \leq X \ , \\ \\ 1 \ , & \text{if } X > X \ ; \end{cases} \tag{3}$$

and the S_{AI} bulk sputtering is proportional to [1-C(X)].

The U_o energy varies from U_{oAB} (the binding energy of a B atom on A surface if X ∿ 0) to U_{oB} if X > 20 atom/nm^2. As U_o values are betwenn 2-8 eV, it is reasonable to use U_{oB} for impurity sputtering and U_{oA} for bulk process.

To evaluate the nuclear stopping-power, we have to take into consideration that I^+ ions lose energy both in B and A material so $\varepsilon_{nABI}(E,X)$ should be a combination of $\varepsilon_{nAI}(E)$ and $\varepsilon_{nBI}(E)$ and the coverage of surface.

Bulk sputtering takes place only if X is so thin that nuclear stopping can be neglected in it. So

$$\varepsilon^{A}_{nABI}(E,X) \cong \varepsilon_{nAI}(E) \quad . \tag{4}$$

In the intermediate region it is assumed that a surface layer of D thickness is responsible for the overlayer sputtering. As a further simplification, we regard all cascades originating in this layer to have the same effect on the surface processes, furthermore, that the nuclear stopping is constant over this layer. If one does not distinguish between A-B and B-B type collisions, the result can be written as:

$$\varepsilon^{B}_{nABI}(E,X) \cong \begin{cases} \dfrac{\varepsilon_{nBI}(E)X+\varepsilon_{nAI}(E)(D-X)}{D}, & \text{if } X \leq D \\ \varepsilon_{nBI}(E), & \text{if } X > D \end{cases} \tag{5}$$

In an A-B type collision, however, the maximum energy transfer to the B specimen is γ_{BA} times less than that of a B-B type.

A way to take into account this effect is to multiply ε_{nAI} γ_{BA}. So Eq. 5 is modified,

$$\varepsilon^{B}_{nABI}(E,X) = \begin{cases} \dfrac{\varepsilon_{nBI}(E)X+\varepsilon_{nAI}(E)(D-X)\gamma_{BA}}{D}, & \text{if } X \leq D , \\ \varepsilon_{nBI}(E) , & \text{if } X > D . \end{cases} \tag{6}$$

Summarizing the above theoretical speculations the sputtering yields are:

$$S_{AI}(E,X) = \frac{4.2 \cdot 0.5}{U_{oA}} \varepsilon_{nAI}(E) \begin{cases} 1 - \dfrac{X}{\Delta X}, & \text{if } X \leq \Delta X , \\ 0 , & \text{if } X > \Delta X . \end{cases} \tag{7}$$

$$S_{BI}(E,X) = \frac{4.2 \cdot 0.5}{U_{oB}} \begin{cases} \dfrac{\varepsilon_{nBI}(E)X+\varepsilon_{nAI}(E)(D-X)\gamma_{BA}}{D}, & \text{if } X \leq D , \\ \varepsilon_{nBI}(E) , & \text{if } X > D , \end{cases} *$$

$$* \begin{cases} \dfrac{X}{\Delta X}, & \text{if } X \leq \Delta X , \\ 1 , & \text{if } X > \Delta X . \end{cases} \tag{8}$$

To compare our model with experimental data, the following numerical values were used:

$$N_{Au} = 59 \text{ atom/nm}^3 \quad 3)$$

$$U_{oAu} = 3.8 \text{ eV} \quad 8)$$

$$S_{nAuN^+} \text{ (2 MeV)} = 25 \text{ eV/nm} \quad 7)$$

$$S_{nSiN^+} \text{ (2 MeV)} = 2 \text{ eV/nm} \quad 7)$$

$$Y_{Au-Si} = 0.44$$

The only fitting parameter was $D = 900 \text{ atom/nm}^2$.

It can be seen that experimental points are higher with a factor of three than the solid line which represents the calculated sputtering yield. This difference presumably comes partly from the U_o energy, because it was chosen as binding energy. For evaporated layers, however, a Van der Waals adhesion is more reasonable to count with. Besides this model disregards S_{II} type sputtering[6] which may have some contribution to the sputtering yield in the Rutherford region. With all these restrictions this rather qualitative model might be a basis of a more elaborated theoretical work.

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Figure captions

Figure 1

Nitrogen backscattering spectra taken on a silicon sample covered by 7 atom/nm^2 gold. During prolonged bombardment the gold sputtering yield is $(1.0\pm0.2)\cdot10^{-2}$ Au/N^+.

Figure 2

Sputtering yield data (full points) as a function of surface coverage. Solid line represents the results of present theoretical calculations.

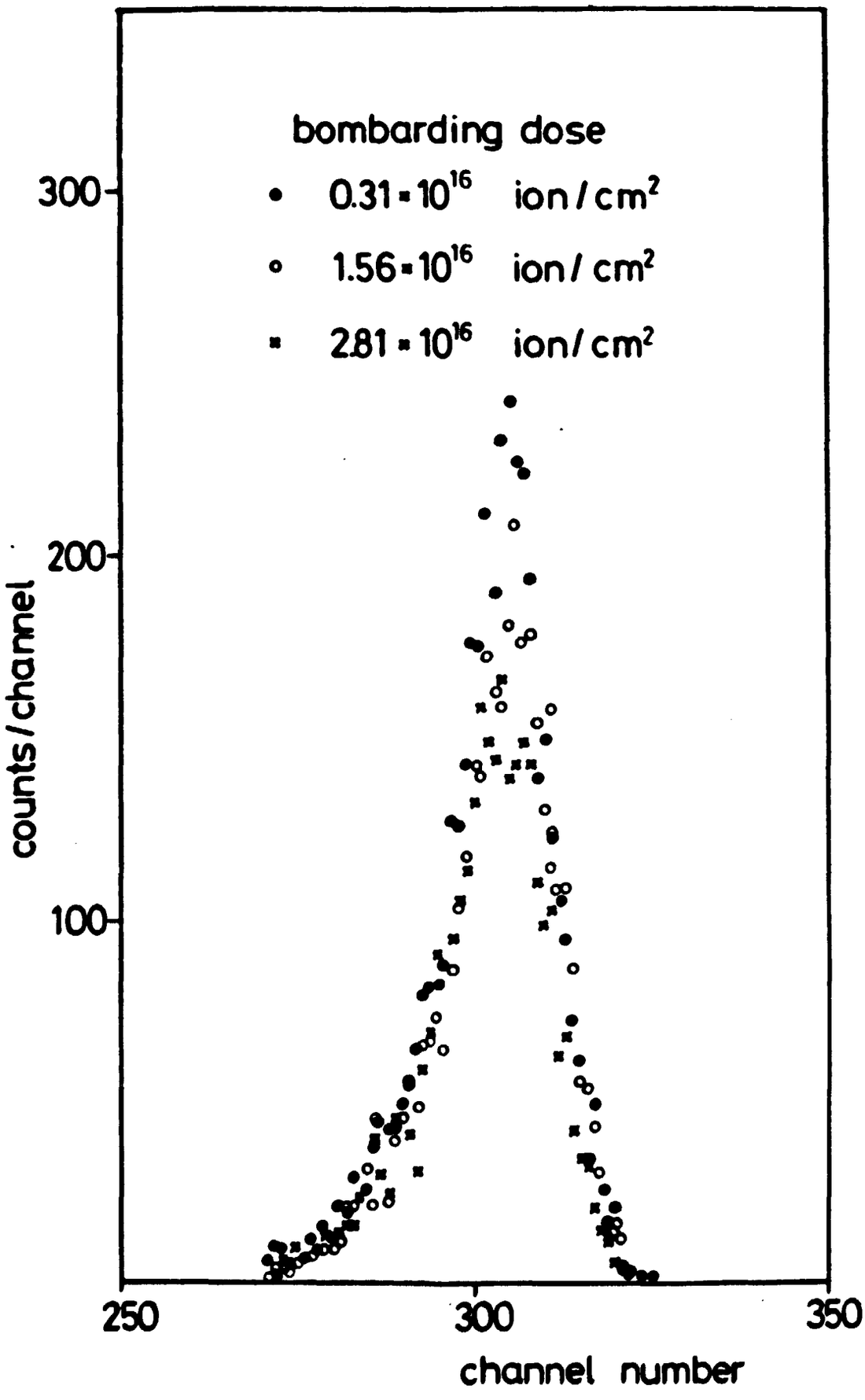
bombarding dose
• 0.31 • 10^16 ion / cm^2
o 1.56 • 10^16 ion / cm^2
× 2.81 • 10^16 ion / cm^2
300
200
100
counts / channel
250
300
350
channel number

Fig. 1.

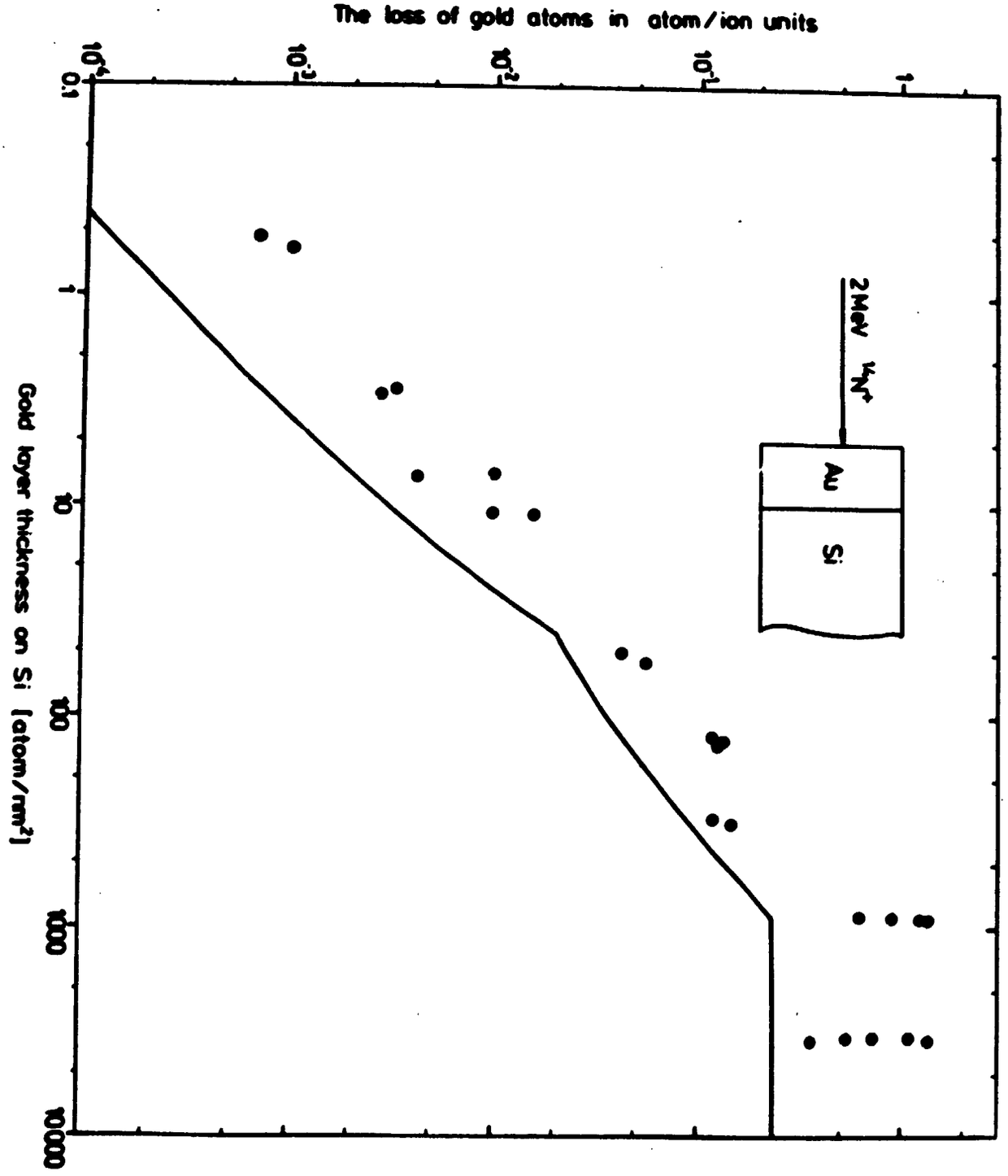
The loss of gold atoms in atom/ion units
10^{-4}
10^{-3}
10^{-2}
10^{-1}
1
0.1
1
10
100
1000
10000
Gold layer thickness on Si [atom/nm²]
2MeV $^4He^+$
Au
Si

Fig. 2.

Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Bencze Gyula
Szakmai lektor: Bakos József
Nyelvi lektor: Gyulai József
Példányszám: 515 Törzsszám: 80-533
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Budapest, 1980. szeptember hó