E14-2002-270

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THE SPUTTERING OF THE DEFORMED GOLD UNDER IRRADIATION WITH KRYPTON SWIFT HEAVY IONS

Submitted to «Ultramicroscopy»

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INTRODUCTION

Research on the influence of irradiation with swift heavy ions at high inelastic energy loss range began to develop very intensively during last 15-20 years practically at most heavy ion accelerator centers. As is well known, the problems of ion track creation were connected with two old models: a coulomb explosion model and a thermal spike (thermal peak) one. With the development of some new methods for the study of irradiated surfaces (such as scanning tunneling (STM) and atomic force microscopy (AFM)) the possibilities of irradiated surface investigations gained a new life.

Research on the sputtering of coarse-grained metals by heavy ions in the inelastic energy loss range was induced by the development of acceleration and storage requirements for high-intensity heavy ion drivers and collected rings [1]. The sputtering yields of annealed coarse-grained gold with high-energy ^{238}U and ^{84}Kr ions have been measured experimentally [2-6], and experimental data on the sputtering yields of Au, Zr and Ti annealed metallic foils by Au swift ions were obtained recently [4].

It is very significantly and interesting to research the interaction of swift heavy ions with metals and alloys. There is opinion that in metals it is impossible to obtain the structure of so-called “ion track” with swift heavy ion irradiation and there is nothing any the inelastic energy loss influence of heavy ions on the damage creation too. This point of view is connected with the opinion that impossible to obtain the temperature around ion trajectory which is comparable with the metal melting point or evaporation temperature because the hot electrons have quite enough time (about $t_e \sim 10^{-14}$ s) for energy dissipation. The characteristic time for electron-phonon interaction is about $t_i \sim 10^{-12}$ s; so $t_i >> t_e$ it means that electron-atom interaction time much more than the electron-electron interaction time. As a result the thermal spike model cannot work here and the damage can be produced by elastic energy loss only.

The recent experimental results showed that the picture of swift heavy ion interaction with matter is a much more complicated.

The purpose of this article is to verify these models and to study the role of each models in the surface structure evolution. We carried out the research on surface structure changes during ion irradiation for the model material – the cold-deformed gold sample.

EXPERIMENTAL RESULTS AND MODELS OF GOLD SPUTTERING UNDER IRRADIATION WITH KRYPTON IONS

The sputtering yields for various gold samples under the irradiation with fission fragments (ff) of ^{252}Cf source are presented in Table 1 (see review [7]). The gold samples were prepared in various structural states (the initial structures were the following: the grains with the diameters of 50-80 Å, 100-200 Å, 300-500 Å, "flat island" with the size of about 1000 Å and hot annealed foil with the thickness of 20 μm and annealed polycrystalline samples with the thickness of 0.2 mm [7]).

Table 1. The sputtering yields S_{ex} (atom/ff) of gold targets with the various structure under the irradiation by fission fragments with the inelastic energy losses $(dE/dx)_{inel} \approx 2.0$-$2.3$ keV/Å. Here S_{IG} and $S_{C.T}$ are the calculated values in the isolated grain model [8, 9] and the cascade sputtering model [1, 10] respectively

Surface structure	Diameter 50-80Å	Diameter 100-200Å	Diameter 300-500Å	Flat island ~1000Å	Annealed foil, 20 μm	Annealed polycrystal 0.2 mm
S_{ex}, atom/ff	4000 ±600	1100 ±180	42±13	10±4	15±5	3±1
S_{IG}[8,9], $S_{C.T}$[10]	2300[8,9]	2100[8,9]	2.4[10]	4[10]	4[10]	4[10]

The sputtering yields for the fine-grained gold samples (with grain size about 50-200 Å) are very high (>10^3 atom/ff). Such high value is proportional to the atom number in grain. Also it was shown that the grain is sputtered totally if the grain diameter was less than critical one. The sputtering of fine-grained coatings on the substrate can be explained by disengagement of atomic clusters or whole grains [7, 8]. So the thermal spike model is realized and the sputtering takes place on the evaporation mechanism for the gold samples with the fine grains.

The sputtering coefficients for annealed gold foils irradiated with ^{238}U, ^{196}Au and ^{86}Kr swift heavy ions are presented in Table 2 [2–4]. As one can see, the experimental values of the sputtering coefficient are S_{ex}≈1-12 atom/ion. The Au sputtering coefficients calculated with the help of the cascade sputtering model ($S_{C.T}$), differ substantial from experimental values, especially for irradiation with ^{238}U and ^{196}Au ions with high level of inelastic energy losses. The projected ranges of ^{238}U, ^{196}Au and ^{86}Kr ions in Au, the elastic cross-section near the surface σ_d and inelastic energy losses $(dE/dx)_{inel}$ are presented in Table 3.

Table 2. The experimental sputtering yield in the inelastic energy loss range for the gold polycrystalline targets irradiated with ^{238}U, ^{196}Au and ^{86}Kr heavy ions

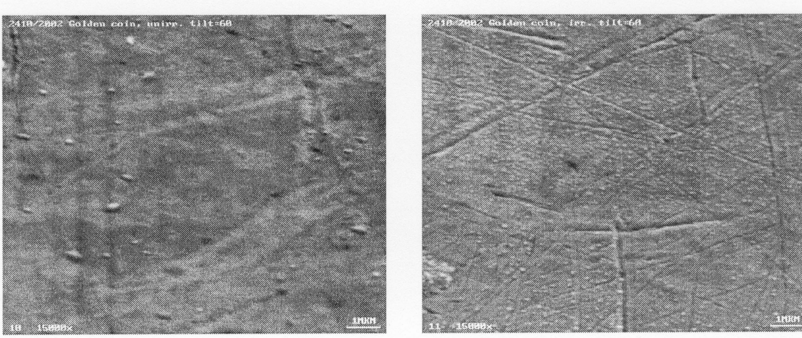
Ion/*Target*	Energy, MeV	Sputtering yield experiment, S_{ex}, atom/ion	Sputtering yield, cascade theory, $S_{C.T.}$, atom/ion	Inelastic energy loss, $(dE/dx)_{inel}$, keV/Å
^{238}U/^{196}*Au*	1400	12 ± 2	≤1	9.82
^{196}Au/^{196}*Au*	230	9.3 ± 0.9	~3	5.52
^{84}Kr /^{196}*Au*	200	1.0 ± 0.2	≤1	3.28

At the calculations of displacement cross-section under elastic ion collisions with Au atom, σ_d, the threshold energy was taken as E_d=20 eV (see Table 3).

Table 3. The calculating values of projected ranges (R_p), elastic cross-section near the surface (σ_d), and inelastic energy loss $(dE/dx)_{inel}$ for the ^{238}U, ^{196}Au and ^{86}Kr heavy ions in Au

Ion/*Target*	Energy, MeV	Projected range R_p, μm	Inelastic energy loss, $(dE/dx)_{inel}$, keV/Å	Elastic cross-section, σ_d, dpa·cm^2/ion
^{238}U/^{196}*Au*	1400	21.26	9.82	$3.7\cdot10^{-16}$
^{196}Au/^{196}*Au*	230	7.65	5.5	$1.1\cdot10^{-15}$
^{84}Kr/^{196}*Au*	253	10.78	3.33	$1.65\cdot10^{-16}$

As one can see, the elastic cross-section of the Au atom displacement, σ_d, has the highest value when the annealed gold thick foil is irradiated with ^{196}Au ion, but the sputtering coefficient S_{ex} is less in comparison with irradiation with ^{238}U ions: 9.3 atom/ion and 12 atom/ion, respectively. This difference between the sputtering coefficients cannot be explained by using only elastic collisions. It is necessary to take into account also the inelastic energy loss and the temperature effects (thermal spike model). The inelastic energy loss $(dE/dx)_{inel}$ for ^{238}U are more than those for ^{196}Au ion irradiation by a factor of ~1.8.



a *b*

Fig.1. SEM-structure of the gold surface in initial state (*a*) and after irradiation with 253 MeV ^{84}Kr ions up to a fluence of 2×10^{14} ion/cm^2 (*b*). One can see the grain boundaries on the surface. The gold purity is 99.0 %.

Then the gold samples underwent cold deformation to create high concentration of dislocations as a drawing defects (the gold purity was 99.9 %) were irradiated with 253 MeV ^{84}Kr ions up to the fluences $(Ft)_1=10^{14}$ and $(Ft)_2=2\times10^{14}$ ion/cm^2. The mean ion flux was $F=3.7\times10^9$ ion/(cm$^2\cdot$s) during irradiation. The images of good polished initial (*a*) and irradiated with swift heavy ions up to the fluence of $(Ft)_2=2\times10^{14}$ ion/cm^2 (*b*) surfaces are presented in Fig.1. The images were obtained with the help of scanning electron microscope (SEM). It is very easy seen the grain boundaries on the gold surface before irradiation. The depth of grain boundaries is not large before the irradiation (see Fig.1*a*), while after irradiation the gold has the distinct outlined grain boundaries. This irradiation was carried out with the use of the set-up described recently [11, 12].

The STM-images of initial (*a, c*) and irradiated with ^{84}Kr ions (*b, d*) gold surface structures are presented in Fig.2. The maximum differences between the surface top and bottom (the height of relief) are 30.87 nm (*a*) and 25.79 nm (*c*) for unirradiated surface parts and 8.66 nm (*b*) and 7.54 nm (*d*) for irradiated ones. The surface of gold samples begins to straighten and becomes smooth one under the irradiation because the inhomogeneities of reliefs are sputtered more effectively. The average differences of relief heights obtained by averaging of large number of images for initial (*a*) and irradiated with ^{84}Kr ions up to fluence $(Ft)_1=10^{14}$ ion/cm^2 (*b*) gold surfaces are $H_a\cong27.19$ nm and $H_b\cong11.66$ nm respectively [13]. The approximating number of atoms evaporated (sputtered) from the gold surface under irradiation with ^{84}Kr ions has been estimated by simple expression:

$$N_{SAu} \sim (H_a - H_b)\times N_{Au} = 9.17\times10^{16}\ \text{atom/cm}^2, \qquad (1)$$

where N_{Au}=5.903×10^{22} atom/cm^3 is the atom number per 1 cm^3. So the sputtering coefficient (or most probably, the evaporation coefficient) has an approximating value S_{ex}≅N_{Au}/$(F·t)_1$=9.17×10^2 atom/ion. Comparison between the sputtering/evaporating coefficients for annealed (the first case) and cold deformed (the second case) gold samples irradiated with Kr ions shows that they differ by a factor of ≈900 [9].

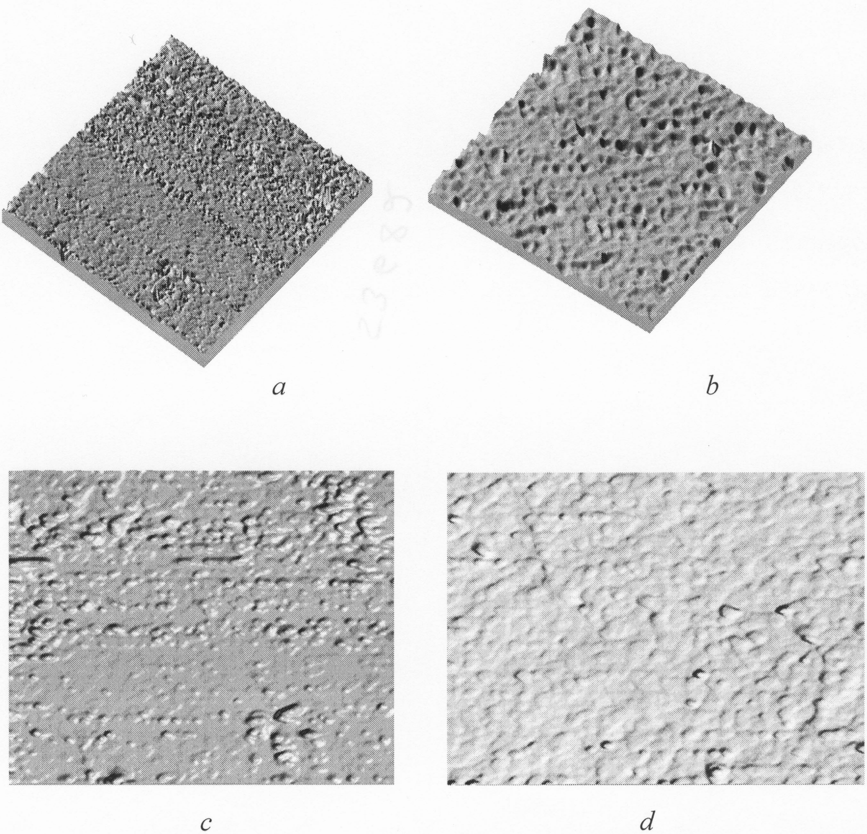


Fig.2. Surface srtuctures of gold samples (STM) before irradiation (*a*, *c*) and after irradiation with ^{86}Kr ions up to fluence of 10^{14} ion/cm^2 (*b*, *d*). The scanning areas and the relief heights have the following values: *a*) 600 nm×600 nm× 30.87 nm; *b*) 556 nm×556 nm×8.66 nm; *c*) 600 nm×600 nm×25.79 nm; *d*) 1200 nm×1200 nm×7.54 nm

Such a high value of the evaporating coefficient in the case of cold-deformed gold can be explained only by the processes of gold atom evaporation from the surface during the Kr ion passing through the surface. This means that the temperature in the volume around swift heavy ion projected range in the cold-deformed gold samples was higher than the melting point and the evaporation temperature. So the thermal spike model must be valid in this case. The approximating temperature in the volume around the ion track may be calculated using the equation from refs. [14, 15]:

$$T_{tr}(r,t) = S_{inel}/(4\cdot\pi\cdot\chi_L\cdot t)\cdot\exp\{(-C_i\cdot r^2)/(4\cdot\chi_L\cdot t)\} + T_{initial}, \quad (2)$$

here $T_{initial}$ is the irradiation temperature (room temperature). To calculate the temperature we used the following parameter values: heat conductivity χ_L=270 W/(m·K) at temperature T=1000 K and heat capacity C_i=3069 kJ/(m^3·K) at temperature T=1500 K. So, we took into account the parameter value for a high temperature.

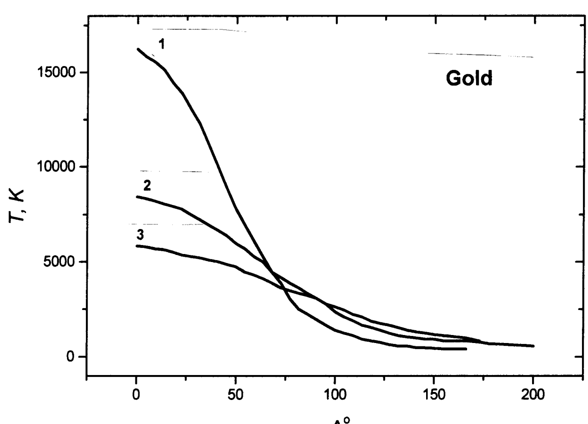


Fig.3. The temperature as a function of the distance from ion track axis for the various time: $t_1 = 1\times10^{-13}$ s, $t_2 = 2\times10^{-13}$ s and $t_3 = 3\times10^{-13}$ s

Fig.3 shows the temperature as a function of the distance from beam trajectory axis for various times: $t_1=1\times10^{-13}$ s (*1*); $t_2=2\times10^{-13}$ s (*2*) and $t_3=3\times10^{-13}$ s (3). The curves *1–3* were obtained using expression (2) [14, 15]. As one can see, the temperatures at the area around the track axis with the radius about 50-75 Å during the time up to 10^{-12} s higher than the melting point (1336 K) and the evaporation temperature (3150 K) for gold. Also the final temperature on the track axis may be estimated with the use of the expression from ref. [16]

$$T_{tr}(0)=\{4\cdot h\cdot S^2\cdot N\cdot\sigma_0\cdot\alpha\cdot r_0^2\cdot T_0^{1/2}\{(T_0/\varepsilon_F)^{1.5}-1\}/(9\cdot a\cdot\beta)\}^{1/2}+T_{initial}. \quad (3)$$

Here $\sigma_0=2\cdot\pi\cdot a_0^2$, a_0 is the Bohr radius, S - the acoustic velocity, N - the atom density for target material, r_0 - the initial radius of excited electrons, T_0 - the initial electron temperature in the excited area, a - the lattice constant, α and β are constants (for the $Z>20$ they are equal to 0.05-0.1 eV), ε_F - the Fermi energy, $T_{initial}$ is the initial sample temperature (about room temperature). The initial electron temperature T_0 can be calculated with the use the expression [16]:

$$T_0=[(dE/dx)_{inel}/(\pi\cdot r_0^2\cdot N)/(1.5\alpha+\beta)]^{1/2}. \quad (4)$$

Using this expression, it is possible to estimate the temperature in the ion track area. The temperature on the track axis T_{tr} estimated by means of expression (4) is higher than the melting point and the evaporation temperature too. Practically the same order of temperature values ($T_{tr}>T_{melt}$) follows from the more simple expression, than expressions (2) and (3), for temperature calculations presented at [17, 18]

$$T_{tr}=S_{inel}(x)/(\pi\cdot R_{tr}^2\cdot C_i\cdot\rho_i)+T_{initial}. \quad (5)$$

So, according to expression (5) the temperature on the ion track axis will be T_{tr}~2.2×10^4 K. The parameters for calculations were C_i=0.159 J/(g·K) at T=1500 K, $R_{tr}\approx50$ Å, ρ_i=19.32 g/cm^3. One can see that the temperatures on the axis of ion trajectory in gold calculated with the expressions (2), (3) and (5) have the same order of magnitude.

Besides from the pressure (as very easy to estimate) in the area around the hot ion track at the temperature $T_{tr}\sim10^4$ K (without changes of volume) must be approximately 10^2 Kbar (see [19, p.249]). So the target atoms can be thrown out from the surface by high pressure, too.

The time and distance dependence of temperature around ion track is calculated using the expressions from some publications [20-22]. The temperature peak (thermal spike) model was used for description of radiation phenomena under irradiation with swift heavy ions of solids (e.g. [20]). This model calculations is based on earlier publications [21, 22]. The expressions which were obtained at these works have the form

$$C_e\cdot(\mathrm{d}T_e/\mathrm{d}t) = 1/r\,\mathrm{d}(r\chi_e\cdot\mathrm{d}T_e/\mathrm{d}r)/\mathrm{d}r - \alpha\cdot(T_e - T_i) + A(\rho, t), \qquad (6a)$$

$$C_i\,(\mathrm{d}T_i/\mathrm{d}t) = 1/r\,\mathrm{d}(r\chi_i\cdot\mathrm{d}T_e/\mathrm{d}r)/\mathrm{d}r + \alpha\cdot(T_e - T_i). \qquad (6b)$$

Here C_e and C_i are specific capacities for electrons and lattice atoms, and χ_e and χ_i are electronic and lattice thermal conductivities, respectively; C and χ are temperature dependent parameters; α is the constant of electron-phonon interaction. The electron temperature can be estimate by means of expressions [21]:

$$T_e(\rho) = 4(\mathrm{d}E/\mathrm{d}x)_{inel}/(\pi\cdot C_e\cdot\rho_e^2)\cdot\exp(-\rho/\rho_e^2). \qquad (7)$$

If parameters C_e, C_i, χ_e and χ_i do not depend on temperature, the solution of system (6) can be written in the form [21]

$$T_i(\rho,t) = \alpha\,(\mathrm{d}E/\mathrm{d}x)_{inel}/(2\cdot\pi\cdot C_e\cdot C_i)\cdot \int \{\exp(-\omega_1 t)-\exp(-\omega_2 t)\}\times$$

$$\times\exp(-k\rho_e)^2\cdot J_0(k\rho)k\mathrm{d}k/(\omega_2 - \omega_1), \qquad (8)$$

the parameters ω_2 and ω_1 are presented in ref. [21].

The using of the expressions (2), (3), (5), (6) and (8) allows one to estimate the temperature on the axis of Kr ion track in gold. The temperature of the ion track near the axis, T_{tr}, is higher than the melting point and the evaporation temperature for this polycrystalline gold. It follows from all these expressions. So, in this material the evaporation (sputtering) processes must take place and the sputtering yield (or evaporation coefficient) have to be very high.

Under passing of heavy ion through the solids around its trajectory will be formed the cylindrical volume with excited electrons. The radius of this volume is r_0~10 Å, and initial electron gas temperature is $E_0\approx$20–40 eV [16]. The relaxation of excited electrons will be due to electron-electron collisions (electron thermal conductivity χ_e) and electron-lattice interactions, which can lead to the heating of lattice atoms. The characteristic time for energy transfer from hot electrons excited by flight ion to lattice atoms is $\tau_h\approx 10^{-12}$ s [24]. The characteristic time for lattice atoms cooling will be $\tau_i \cong r_0^2/\chi_i < 10^{-14}$ s. The condition $\tau_i << \tau_h$ means that lattice atoms will not heat and swift heavy ions will not create hot tracks in gold. Nevertheless at the volume with high local crystal lattice disturbance the condition $\tau_i << \tau_h$ may be breaks. First of all, time τ_h decreases up to 10^{-13} s [16, 24], secondly, the lattice thermal conductivity decreases and consequently the temperature conductivity significantly decreases too consequently.

So, one can conclude that the sputtering-evaporation coefficient strongly depends on the defect concentration such as point defects, defect clusters and dislocations as drawing defects. That conclusion is in agreement with the results obtained for irradiated metals, alloys and HOPG [23-27].

CONCLUSION

The presence of radiation defects in metals essentially increases the influence of inelastic energy loss of fast heavy ions on the sputtering yield. Thus, the sputtering yield for metal having a small defect number in its crystalline structure is in the range 1-10 atom/ion [3-6]. Experiments show that the sputtering yield for coarse-grained metals under swift heavy ion irradiation increases very quickly with the increasing irradiation fluences and with accumulation of radiation defects in the target crystalline structure. The experimentally observed high sputtering yield for gold can be explained by atom evaporation from the surface with the tracks which has been heated up to the temperature higher than the boiling temperature (T_b) (i.e. a thermal spike model works here). In this way we experimentally proved that inelastic energy loss $(dE/dx)_{inel}$ of fast heavy ion strongly affects on the sputtering of metals with damaged crystal structure. The hot electron plasma model [29] does not contradict the experimental results presented in given work.

The scanning electron microscopy studies show strong inhomogeneity of the metal surface sputtering with fast heavy ions. The sputtering yield near the grain boundaries is greater than that from the grain bodies.

The explanation of experimental results is given. On the base of data obtained for Ni, W and HOPG [23-28] we have concluded that the track formation and consequently the high evaporation (sputtering) coefficients take place in case of high disordered systems or in materials with relatively low concentration of free electrons (like dielectrics and some semiconductors). It has been shown that for conductive materials, when the condition $\tau_L \ll \tau_h$ takes place at low defect zone, the lattice atoms around the swift heavy ion trajectory are cold and do not create a hot track. The condition $\tau_L << \tau_h$ is disturbed on the boundaries between grains and also in the area with high defect concentration, as one can see for Au previously cold deformed. As a result, when the swift heavy ion pass through such areas, the hot track is created and the target atom evaporation takes place.

This conclusion is proved by the result of the studies about track formation in single crystal InP semiconductor [30, 31] irradiated previously with 5 MeV electrons (fluence 5×10^{17} electron/cm^2) and then with 250 and 340 MeV ^{129}Xe and 245 and 210 MeV ^{86}Kr ions. Heavy ion tracks were also found in amorphous Si and Ge [32]. The processes of damage concentration saturation in some materials (like Ge, C, W etc.) versus swift heavy ion fluence were observed in a cycle of publications (see, e.g. [33]). A model of disordering and following crystal recrystallization under heavy ion irradiation was developed in ref.[34].

The importance of the sputtering problem for accelerator engineering and for the high-energy heavy ion implantation into novel materials makes it necessary to continue the experimental and theoretical researches.

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Received on November 29, 2002.

Дидык А. Ю., Хофман А., Семина В. К. E14-2002-270
Распыление деформированного золота
при облучении быстрыми тяжелыми ионами криптона

Представлены результаты по уровню распыления золота, облученного ионами ^{86}Kr, в режиме высоких неупругих потерь энергии флюенсом вплоть до 10^{14} ион/см^2. Показано, что уровень распыления (испарения) сильно зависит от исходной концентрации дефектов в золоте. Коэффициент распыления начинает очень быстро расти с увеличением числа дефектов, созданных за счет упругих и неупругих потерь энергии тяжелыми ионами. Согласно расчетам с помощью различных выражений и моделей температура на поверхности в области траектории иона криптона намного выше температуры плавления и испарения золота.

Работа выполнена в Лаборатории ядерных реакций им. Г. Н. Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2002

Didyk A. Yu., Hofman A., Semina V. K. E14-2002-270
The Sputtering of the Deformed Gold
under Irradiation with Krypton Swift Heavy Ions

The results about sputtering yield of gold irradiated by ^{86}Kr ions with high inelastic energy losses up to a fluence of 10^{14} ion/cm^2 are presented. It was shown that the sputtering (evaporation) yield strongly depends on the initial defect concentration in gold. The sputtering yield begins to grow very strongly with the increasing of damage created by heavy ion elastic and inelastic energy losses. The temperature on the surface in the area around krypton ion trajectory is much higher than the melting and evaporation temperatures for gold as follows from calculations with the various expressions and models.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2002

Макет *Т. Е. Попеко*

Подписано в печать 24.12.2002.
Формат 60 × 90/16. Бумага офсетная. Печать офсетная.
Усл. печ. л. 1,0. Уч.-изд. л. 1,10. Тираж 280 экз. Заказ № 53682.

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