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Nonlinear effects in the sputtering of gallium arsenide and silicon by bismuth cluster ions

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An experimental study on the influence of the energy and the number of atoms in the bombarding ions Bi_n^+ ($n = 1-4$) on the sputter yield of GaAs was carried out. It was shown that the specific sputter yield Y_{sp} non-additively increases with increasing n and specific kinetic energy E_{sp} per an atom in the bombarding ion, and the efficiency of energy transfer from bombarding ions to target atoms also increases with increasing n. A comparison was made with the previously obtained results for Si targets.

Keywords: ion sputtering, non-additivity factor, cluster ions, bismuth.

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Focused beams of atomic and cluster heavy–metal ions, foremost, Sn, Au, Pb and Bi obtained by using electro-hydrodynamic (liquid-metal) ion sources [1] are nowadays used to solve an extensive range of fundamental and applied problems of micro— and nano–technologies, aerospace engineering and microprobe analysis. The use of cluster ions in ion–beam lithography and nano-structuring allows increasing the sputter yield. A great number of studies (see, e. g., [2–8]) are devoted to computer simulation of sputtering processes and to the sputter yield calculation. Along with this, the number of publications on experimental measurements of yields of sputtering by cluster ions is significantly lower, and even lower is the number of papers presenting data on comparative efficiency of sputtering by atomic and cluster ions under similar experimental conditions [9,10]. In our previous work [11], the influence of the energy and number of atoms (nuclearity) in bombarding ions Bi_n^+ ($n = 1-4$) on the Si sputter yield was studied. It was found out that specific sputter yield Y_{sp}, namely, the number of target atoms sputtered per an atom contained in the cluster ion, increases non-additively with increasing n and kinetic energy E_{sp} per an atom of the bombarding ion. It was proposed to use as the non-additivity k factor the slope ratio of the straight line approximating the $Y_{sp}(n)$ dependence at the same E_{sp} value. In the case of Si irradiated with 10 keV ions Bi_n^+ ($n = 1-4$) , this factor appeared to be 0.4 ± 0.1. In this study we have extended the range of test materials by adding to them the two-component compound GaAs that is a very important semiconductor material, the third after silicon and germanium with respect to the scale of industrial application.

The experiments were performed at the commercial ion-beam lithographer VELION (Raith Nanofabrication, Germany [12]); its performance characteristics may be found in [1]. Mass–separated beams of atomic ions Bi^+ ($n = 1$) and cluster ions Bi_n^+ ($n = 2-4$) [13] were directed normally to the target surface. The accelerating voltage was varied in the 10–40 kV range with the step of 10 kV; diameter of the beam spot on the target did not exceed 100 nm (FWHM) for all the ion types, while the ion current ranged from 0.02 to 1 nA depending on the energy and nuclearity of the cluster ion. Long-term stability of the ion current was better than 1% per 7 h of continuous operation; the ion current at the target was automatically measured each 10 s.

The total sputter yield Y was determined based on the sputtered substance volume V defined as a product of the crater area and depth, and total bombarding ion dose F_{ion}:

$$Y = \frac{VN}{F_{ion}}. \quad (1)$$

Here N is the atomic density of the substance to be sputtered (target),

$$F_{ion} = \frac{It}{e}, \quad (2)$$

where I is the ion current at the target, t is the sputtering time, e is the elementary charge.

In addition to the total sputter yield Y, we have determined specific yield Y_{sp} equal to Y/n; this means that the total ion dose in relation (1) was substituted with the total bombarding atom dose $F_{at} = nF_{ion}$. Notice that in this method, referred to as Volume Loss Method (VLM) [14], serious requirements were imposed on the primary ion

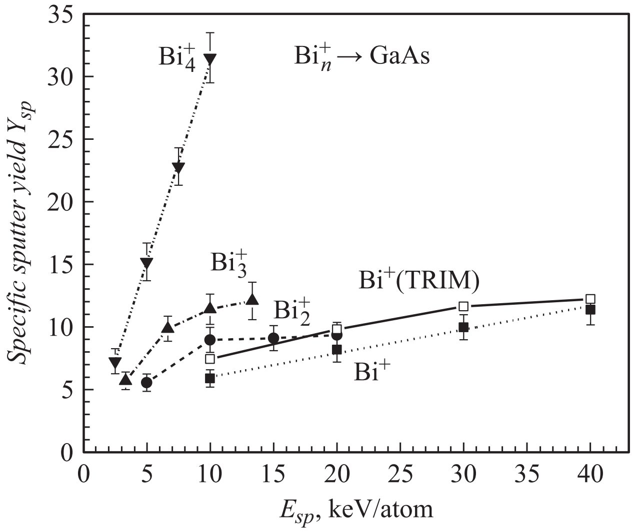

Figure 1. Dependences of specific sputter yields on the energy per a bombarding ion atom for the case of GaAs sputtering with ions Bi_n^+ ($n = 1-4$); the figure also presents linear approximations of these dependences for Bi^+ and Bi_4^+. For comparison, the calculated sputter yield dependence obtained with TRIM for ions Bi^+ is given (a solid line with open squares).

beam column (ion gun) that has to generate sharply focused and mass-separated beams of atomic and cluster ions with equal energies E_{sp}. This requirement should be met in order to ensure reliable distinguishing of contributions to the sputter yield of such parameters as energy and nuclearity of the bombarding ion. Along with this, the ion current density and dose of the bombarding particles should be sufficient for formation of a crater whose linear size and depth can be precisely measured with a profilometer. In our experiments, a contact profilometer Veeco DEKTAK 150 with vertical resolution of 0.1 nm was used. The measurements were taken at three points arbitrary chosen at the bottom of each crater; the measurement error did not exceed ± 0.5 nm.

The total number of craters prepared by irradiating GaAs with ions Bi_n^+ ($n = 1-4$) 10, 20, 30 and 40 keV in energy was 16 for each type of ions. The crater depths and areas were 65$-$1425 nm and 400$-$700 μm^2 , respectively. For each ion type and energy, linear sizes of the craters significantly exceeded 100 diameters of the ion beam; this fact, jointly with fast scanning of this beam directed normally to the target surface, ensured formation of craters with vertical walls and even bottoms. In addition, yields of sputtering with atomic bismuth ions were estimated with the TRIM code [15]; the sample composition was assumed to be equal to the nominal one, i. e., the approximation of a target not excited with ion irradiation was used.

Fig. 1 presents specific sputter yields $Y_{sp}(n)$ for the case of GaAs sputtering with ions Bi_n^+ ($n = 1-4$) versus energy E_{sp}. Slope ratios of linear approximations of these dependences reflect the efficiency of ion sputtering. For GaAs, the slope ratios were 0.19 (Bi^+) and 3.2 (Bi_4^+. For comparison: these values for silicon were 0.09 (Bi^+), 0.17 (Bi_2^+) and 0.25 (Bi_3^+) [11]. In our experiments, the total sputter yield Y for 40 keV Bi_4^+ ions was 125 (GaAs) and 15 (Si) [11].

Fig. 2 presents dependences $Y_{sp}(n)$ for GaAs and Si irradiated with ions Bi_n^+ 10 keV/atom in energy. Slope ratios of the straight lines approximating these dependences are 2.8 ± 0.2 and 20 (GaAs) and 0.4 ± 0.1 (Si) [11]. We used these slope ratios as non-additivity factors k, i. e., as a quantitative characteristic of contribution of nonlinear processes to the sputtering mechanism(s). Notice that in this case calculation of the k factor does not need additional computer$-$aided calculations as, e. g., in [9].

In such a brief report it is impossible to discuss in details all the mechanisms responsible for the Y_{sp} increase with increasing nuclearity n in the bombarding ion Bi_n^+ ($n = 1-4$) and specific energy E_{sp}. Most probably, the greatest contribution comes from thermal spikes (see [16,17] with references) in the volume of which the nonlinear sputtering mode accompanied by local melting and evaporation of the substance is realized; the efficiency of these processes increases with increasing n and E_{sp} (Fig. 1). It was also established (Fig. 2) that at the same E_{sp} the increase in number of atoms in the bombarding ion results in non-additive increase in Y_{sp}. Non-additivity factors k appeared to be significantly different for the materials studied in this work and in [11]. This may be associated with less thermal spike sizes in GaAs compared to those in Si because of shorter free paths of primary ions and recoil atoms in this material. It is also impossible to exclude the influence of thermal conductance which is more than three times lower in GaAs than in Si [18], which can promote localization of thermal spikes in smaller volumes, lead to a higher

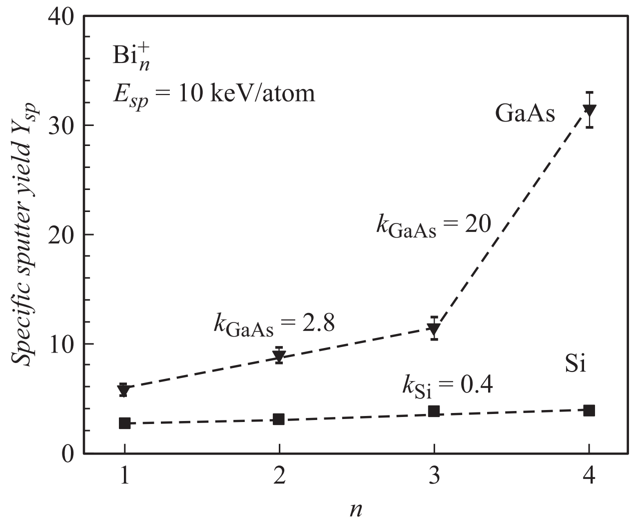

Figure 2. Dependences of specific sputter yields on number n of atoms contained in a cluster ion for the case of sputtering GaAs and Si [11] with Bi_n^+ ions 10 keV/atom in energy. The values of non-additivity k factors that are slope ratios of the straight lines approximating experimental dependences $Y_{sp}(n)$ are presented in the figure.

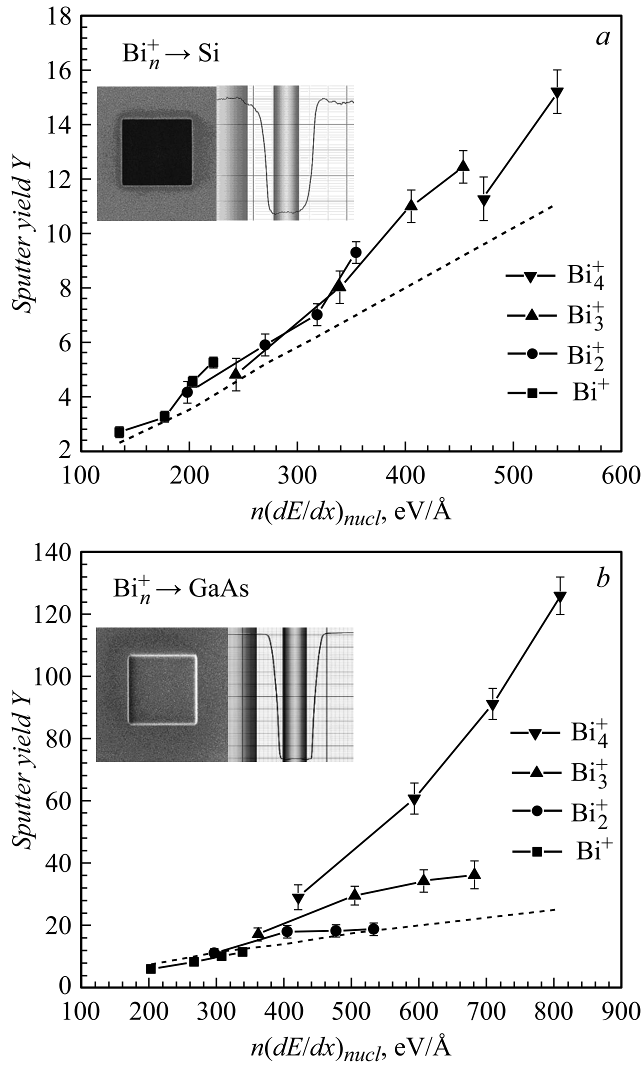

Figure 3. Total sputter yields of Si (as per [11]) (*a*) and of GaAs (*b*) under irradiation with ions Bi_n^+ versus nuclear energy loss of the bismuth cluster. The dashed line represents the TRIM approximation of calculations. The insets present the sputter crater images obtained with a lithium ion microscope and also profiles of these craters measured with profilometer Veeco DEKTAK 150.

temperature and, hence, considerably affect the target atoms sputtering in the nonlinear mode.

GaAs is known to melt and then evaporate incongruently (with decomposition), namely, with formation of liquid gallium and gaseous molecular arsenic [19]. Most probably, variation in the GaAs composition in the thermal spike volume affects the $Y_{sp}(E_{sp})$ dependences, and a transient mode (Fig. 1) is observed for bombarding ions Bi_2^+ and Bi_3^+ as compared with Bi^+ and Bi_4^+; this mode is accompanied by nonlinear variation in dependence $Y_{sp}(E_{sp})$. A sharp increase in the non-additivity factor in the case of turning to ions Bi_4^+ (Fig. 2) may also result from the enhancement of the incongruence effect on the sputtering efficiency. Fig. 3 demonstrates the dependences of total sputter yields of Si (as per [11]) (*a*) and GaAs (*b*) on the bismuth ion nuclear energy loss for clusters with different *n*. In calculating the cluster energy losses, contributions of individual atoms were assumed to be independent [20,21], i.e.

$$\left(\frac{dE}{dx}(n, E)\right)_{nucl} = n\left(\frac{dE}{dx}\left(1, \frac{E}{n}\right)\right)_{nucl}. \quad (3)$$

In the same figure, the dashed line represents the dependence obtained by TRIM simulation at the energies below the elastic loss peak, and also extrapolation of this dependence to higher energies. This line approximately represents the contribution of the linear sputtering component. The difference between the experimental data and this line is much greater for GaAs than for Si.

Thus, this study has established that the sputtering non-additivity factor is higher for GaAs than for Si. It has been also shown that, while for silicon the dependences of sputter yields on elastic energy loss for clusters with different *n* almost fully coincide with each other, those for gallium arsenide exhibit significant differences. This feature is possibly associated with the effect of incongruence of GaAs melting and evaporation.

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Conflict of interests

The authors declare that they have no conflict of interests.

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