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Composition dependence of $Si_{1-x}Ge_x$ sputter yield

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Sputtering yields have been measured for unstrained $Si_{1-x}Ge_x$ ($x=0-1$) alloys when bombarded with Ar^+ ions within the linear cascade regime. Nonlinear S-shape dependence of the sputter yield as a function of the alloy composition has been revealed. The dependence is analyzed within the frameworks of the cascade theory conventionally accepted to be the most systematic to date theoretical approach in sputtering. In view of a linear composition dependence predicted for the sputter yield by the cascade theory adapted for polyatomic substrates, the nonlinearity observed in our experiments is shown to be related to the alloying effect on the surface binding energies of the alloy components. Based on this analysis, an interpretation is proposed for the experimentally observed nonlinear composition dependence of $Si_{1-x}Ge_x$ sputter yield. The yield is expressed by an equation derived from the cascade theory with additional terms of the composition parameter x. The form of the equation implies that for a polyatomic substrate the surface binding energy of an individual atom is determined not only by its own chemical identity but to a considerable degree by the identities of its neighbors.

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Today, SiGe becomes a commercial technology offering new products on a market and challenging those niches where traditionally different kinds of compound semiconductors dominated.[1] The market sectors where SiGe is believed to allow next generation cost-effective solutions already in the nearest future are those related to high-performance radio frequency integrated circuits and systems for wireless communication that span from sub-GHz to 10^2 GHz. What SiGe can offer for optoelectronics, MEMs and nanotechnology is being extensively studied nowadays. Application of the compound semiconductor in the context of CMOS technology enables solutions based on the tradeoff between performance and cost. As SiGe is just one step from Si, the mature silicon technology with highly developed arsenal of tools and well established processing is at hand being fully compatible with new schemes and design platforms in which the compound semiconductor and silicon co-exist. By changing the alloy composition the electrical and optical properties can be modified at will via so-called band gap engineering to meet the requirements of a particular application. Introducing additional strain and enhancing the charge carrier mobility via Si/SiGe heterostructure architecture enables those applications in which traditionally III-V's dominate to migrate nowadays to SiGe and low-power CMOS technologies delivering appropriate performance at much lower cost. In the arsenal of silicon technology there is a number of plasma based and/or assisted processing techniques that form a set of standard tools in nanofabrication and microfabrication (e.g., ultrashallow ion implantation, reactive ion etching, sputter etching, deposition and coating, etc.) and surface analytics and monitoring (wide variety of mass-spectrometry techniques). These are based on use of ionized energetic atomic particles interacting with the surface of a semiconductor. An important phenomenon occurring on the surface when bombarded with ions is sputtering, i.e., emission of the surface atoms as a result of an atomic collision cascade induced in the subsurface layers by the incident particle. Though a relatively large amount of information has been collected about various aspects of sputtering, it is yet impossible to make a generalization for compound semiconductors within available theoretical models due to the lack of data for different groups of semiconductors (for review see Ref. 2 and references therein). Information about SiGe sputtering available in the literature is quite scarce. This is explained, at least partly, by the fact that high quality $Si_{1-x}Ge_x$ alloys with low density of defects are only available recently due to advances in epitaxial growth. To date the most systematic theoretical approach in sputtering is Sigmund cascade theory[3] initially developed for a monatomic structureless medium. Theory of sputtering for polyatomic substrates is lacking. In the present paper we report on experimental study of $Si_{1-x}Ge_x$ sputtering, viz. the alloying effect on sputtering, covering the whole composition range from elemental Si to Ge. On the basis of the experimental data the applicability of the cascade theory to the compound semiconductor is evaluated.

Unstrained and uniform crystalline $Si_{1-x}Ge_x$ ($x=0-1$) layers approximately 1 μm thick were grown by chemical vapor deposition on top of compositionally graded $Si_{1-y}Ge_y$ buffers with y ranging from 0 to x. The buffer layers resided on p-type Si (100) substrates with resistivity of less than 0.02 Ω cm. Quality of the grown layers was ensured by transmission electron microscopy (TEM) and ion channeling with Rutherford backscattering spectrometry (RBS). Most of the extended defects revealed by TEM were those of threading dislocations and stacking faults that were found to concentrate within the buffers and thread to the silicon substrates rather than through the top $Si_{1-x}Ge_x$ layers. Surface dislocation density measured by TEM was only $\sim 10^5$ cm^{-2}. Ion channeling analysis was performed with 500 keV He^{2+} RBS that measured the minimum backscattering yield $\chi_{\min}$ values around 4% for $\langle 100\rangle$ axial channeling assuring high quality of the grown $Si_{1-x}Ge_x$ layers.

The samples were bombarded with a fine collimated beam of Ar^+ ions in an UHV chamber with a base pressure 10^{-8} mbar. Energy of ions was 3 keV and the beam current density was $0.05\ \mathrm{mA/cm^2}$ to ensure a dynamic sputtering of surface. To measure a total sputter yield, i.e., a number of the alloy atoms eroded away per each bombarding ion, the sputter crater that developed on the surface of bombarded samples was analyzed with a profiler Tencor P15 in two regimes, (a) line scan across the crater measuring a step height between the sputtered and virgin parts of the sample, and (b) three-dimensional mapping that provides a 3D picture of the crater with sequential integrating of the sputtered volume. Both methods turned out to give very close values for the sputter yields within an accuracy of the experiments. The samples were bombarded to a fluence of the order of $10^{18}\ \mathrm{cm}^{-2}$. As-sputtered samples were analyzed with optical and scanning electron microscopies to ensure that no significant surface topography was developed after the high-fluence irradiation that could bring about an ambiguity in interpretation of the measured sputter yields.

In order to reduce ambiguity of the experiments and avoid artifacts when comparing measured sputter yields with those predicted by the cascade theory, the experimental conditions and the choice of compound should be adequate to the physical model described by the theory.

First of all, the cascade theory was developed for a structureless medium. Due to a lattice mismatch of approximately 4% between Si and Ge, unstrained high quality $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ alloys with very low density of defects are available only in the form of crystalline layers epitaxially grown on compositionally graded buffers. It is well known that sputter yields measured on monocrystals are dependent on a particular crystallographic orientation and typically lower than for both polycrystalline and amorphouslike substrates due to the channeling effect.[4] Fortunately, most of the semiconductors are readily amorphized when bombarded with high fluence of ions at room temperature. Although mechanisms of radiation damage accumulation are different for crystalline Si and Ge, bombardment with medium mass ions of keV energies to a critical fluence of $10^{14}-10^{15}\ \mathrm{cm}^{-2}$ is known to lead to complete surface amorphization. In $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ amorphous layers are produced by ion irradiation even more easily than in pure Si.[5] As the experimental fluences were three to four orders of magnitude larger than the critical fluence, $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ samples in our experiments may be considered as structureless being preamorphized at the very early stage of irradiation. Therefore, we believe that in our experiments a possibility for experimental artifacts in sputter yield measurements related to the crystalline nature of the $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ samples was minimal.

Second, to apply cascade theory to a binary compound requires that it be a homogeneous random solid solution. Si and Ge are chemically similar elements and they form ideal liquid and solid solutions. Phase diagram for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ system shows that Ge and Si are completely miscible in both liquid and solid phases.[6] Stable solid phase is a cubic diamond type substitutional solid solution with the lattice parameter showing almost linear dependence on the Ge content with just minor deviations from the Vegard's law for medium compositions.[7] No other intermediate phases are known that could give rise to a second phase nucleation and stoichiometry dependent heterogeneous decomposition. Unfortunately, very little is known about ordering in $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ alloys. No detailed atomistic picture is available and information reported in the literature is rather controversial.[8] According to the phase diagram $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ solid solution with a positive enthalpy of mixing does not have any ordered phases at room temperature. Observations of long-range ordering of Si and Ge atoms reported in some experimental works was attributed to a particular growth condition rather than being an equilibrium property. Even if some uncontrolled ordering existed in our samples the collision cascade induced by the bombarding ions would mix up the subsurface atomic layers bringing about additional randomization.

Third, as sputtering is a surface phenomenon in the sense that most of the sputtered atoms emerge from the topmost monolayer, surface composition is of concern as it may differ from the bulk stoichiometry after high fluence ion bombardment due to preferential sputtering, and radiation induced segregation and/or diffusion. In our study surface composition of as-sputtered samples was not analyzed. As $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ forms ideal solid solution for the whole range of composition, preferential surface segregation of one of the constituents is very unlikely due to the chemical similarity between Si and Ge. To the best of our knowledge, neither preferential sputtering nor radiation induced segregation and/or diffusion have ever been reported for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$. On the contrary, no surface composition changes have been confirmed in some related experimental studies where $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ alloys were bombarded by different ions with energies in the range from sub-keV to tens of keV.[9–12] In the context of the cascade theory, preferential sputtering for a binary substrate can be characterized by so-called sputter preferentiality δ related to the difference in partial sputtering yields of the constituents due to the mass difference (mass effect) and the surface binding energy difference (bonding effect). For $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ preferentiality δ is given by[13]

$$\delta = \frac{Y_{\mathrm{Ge}} \cdot C_{\mathrm{Si}}^{\mathrm{S}}}{Y_{\mathrm{Si}} \cdot C_{\mathrm{Ge}}^{\mathrm{S}}} - 1 = \left(\frac{M_{\mathrm{Si}}}{M_{\mathrm{Ge}}}\right)^{2m} \left(\frac{U_{\mathrm{Si}}}{U_{\mathrm{Ge}}}\right)^{1-2m} - 1, \qquad (1)$$

where Y_i, C_i^S, M_i, U_i $(i=\mathrm{Si},\mathrm{Ge})$ are partial sputtering yield, equilibrium surface concentration, atomic mass, and surface binding energy for Si and Ge, respectively; m is a parameter in the cascade theory related to the interatomic interaction potential. The exact value for m is uncertain as it depends on the energy of interaction but for the energies related to sputtering the relevant values are assumed to be in the region $0 \leqslant m \leqslant 0.2$.[14,15] Preferentiality δ for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ is shown for the whole range of m in Fig. 1. According to Eq. (1) the mass effect will cause preferential sputtering of the lighter component Si ($M_{\mathrm{Si}}=28$) and, hence, surface enrichment with the heavier element Ge ($M_{\mathrm{Ge}}=72$). Bonding effect, on the other hand, will lead to preferential removal from the surface of the component that has lower surface binding energy. Being a serious difficulty in the cascade theory, exact values of the surface binding energy are not known even for monatomic substrates and, therefore, the heat of formation of gaseous atoms or cohesive energy is usually used instead. However,

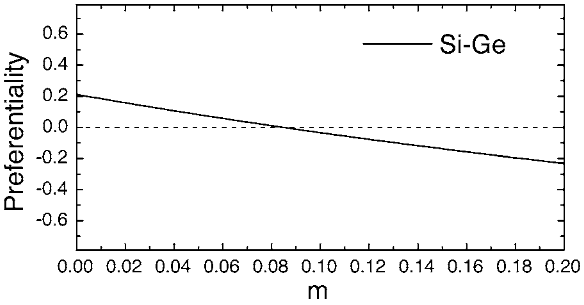

FIG. 1. Preferentiality δ in sputtering of Si-Ge system calculated from Eq. (1) as a function of parameter m.

this does not necessarily need to be the case. For multicomponent substrates like alloys and compounds the surface binding is even less defined as the alloying effect should inevitably make it stoichiometry dependent. Though exact values of U_{Si} and U_{Ge} are not known for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$, for estimations in Eq. (1) it is generally accepted to use instead the ratio of cohesive energies for elemental Si ($E_{c\mathrm{Si}}=4.7$ eV) and Ge ($E_{c\mathrm{Ge}}=3.88$ eV). Having lower cohesive energy and, hence, presumably lower surface binding energy, Ge has a greater ejection probability that will cause an enrichment of Si on the surface. Consequently, the bonding effect opposing and compensating the mass difference effect will result in only minor surface composition changes in $\mathrm{Si}_{1-x}\mathrm{Ge}_x$, if any, due to a negligible preferential sputtering (see Fig. 1).

All in all, $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ forms a model system possessing a variety of technologically relevant properties while being accessible to theoretical analysis within the frameworks of the cascade theory of sputtering.

Experimentally measured composition dependence of $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ sputtering is presented in Fig. 2. The sputter yield is shown as a function of Ge concentration in the alloy. The experimental yields are compared with predictions of the cascade theory applied to a binary $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ target. In the elemental sputter theory[3] the yield (Y) is derived from a linearized Boltzman transport equation solved for the atomic collision cascades and has the form

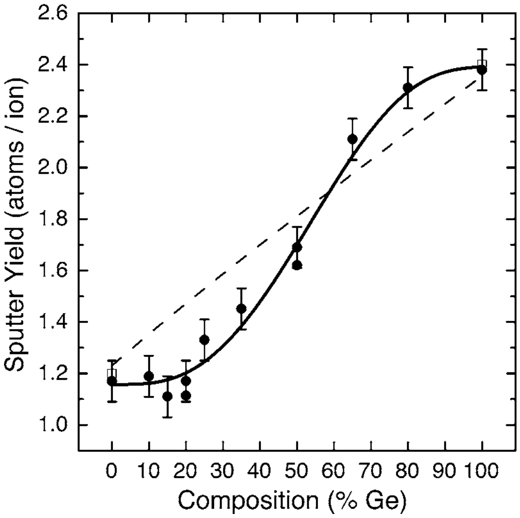

FIG. 2. The composition dependence of the sputter yield for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ bombarded with 3 keV Ar^+ ions. Experimentally measured yields shown by circles (this work) and squares (data from Refs. 18 and 19) are compared with theoretical values calculated from Eq. (2) with U_0 in the form of Eq. (4) (dashed curve) and Eq. (13) (solid curve).

$$Y = \Lambda \cdot F_D(E_p), \tag{2}$$

where Λ is a material constant related to the substrate properties and having no parameters of the projectile, i.e., the bombarding ion,

$$\Lambda = \frac{0.042}{N \cdot U_0}, \tag{3}$$

with atomic density of the target N, and the surface binding energy of the target atoms U_0 that is represented for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ by a weighted average of the constituents:

$$U_0 = U_0(x) = (1-x) \cdot U_{\mathrm{Si}} + x \cdot U_{\mathrm{Ge}}. \tag{4}$$

The factor F_D in Eq. (2) is the density of energy deposited at the surface by the projectile with primary energy E_p and given by the following equation:

$$F_D(E_p) = \alpha(M_t/M_p;\theta) \cdot \left(\frac{dE}{dz}\right)_n \tag{5}$$

where α is a dimensionless factor representing the fraction of the energy deposited available for sputtering and is seen in Eq. (5) to be a function of the target to projectile mass ratio M_t/M_p and the incidence angle θ the primary ions impinge on the surface. The energy deposited by the projectile is introduced by the nuclear energy loss $(dE/dz)_n$, i.e., energy transfer via screened Coulomb interactions with the target atoms. Sputtering is governed by this mechanism of energy loss dominating at the experimental ion energy under discussion,

$$\left(\frac{dE}{dz}\right)_n = 4 \cdot \pi \cdot a(x) \cdot N(x) \cdot Z_p \cdot Z_t(x) \cdot e^2 \times \frac{M_p}{M_p + M_t(x)} \cdot S_n(x,\varepsilon), \tag{6}$$

where the screening distance a is given by

$$a(x) = \frac{0.8853 \cdot a_0}{[Z_p^{1/2} + Z_t(x)^{1/2}]^{2/3}}, \quad a_0 = 0.529 \text{ Å (the Bohr radius)}, \tag{7}$$

with Z_p and Z_t being atomic numbers of projectile and target, respectively. For application of the elemental cascade theory Eq. (2) to sputtering of compounds both atomic number and mass number of the target are considered to be weighted averages of the constituents, i.e., for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$,

$$Z_t(x) = (1-x) \cdot Z_{\mathrm{Si}} + x \cdot Z_{\mathrm{Ge}} \tag{8}$$

and

$$M_t(x) = (1-x) \cdot M_{\mathrm{Si}} + x \cdot M_{\mathrm{Ge}}. \tag{9}$$

There is no one universal form of the function $\alpha(M_t/M_p;\theta)$ for both mass and angular dependence. However, for the low energies of sputtering as in our experiments and for angles θ close to normal incidence $(\theta < 10°)$ the following form of $\alpha(M_t/M_p;\theta)$ gives an accurate approximation:[14,16]

$$\alpha(M_t/M_p;\theta) = 0.15 \cdot [1 + M_t(x)/M_p]^{0.85} \cdot \cos^{-1}\theta. \quad (10)$$

The nuclear stopping cross-section $S_n(x,\varepsilon)$ in Eq. (6) depends on the particular form of the screened Coulomb interatomic potential. In our calculations we use the form derived by Wilson *et al.* that was proven to be an accurate estimate of the cross section due to proper taking into account the interaction potential screening,[17]

$$S_n(x,\varepsilon) = \frac{0.5\ln(1+\varepsilon)}{\varepsilon + 0.14 \cdot \varepsilon^{0.42}} \quad (11)$$

in terms of the dimensionless reduced energy

$$\varepsilon = \varepsilon(x) = \frac{a(x) \cdot M_t(x)}{Z_p \cdot Z_t(x) \cdot e^2 \cdot [M_p + M_t(x)]} \cdot E_p. \quad (12)$$

For calculation of the surface binding energy $U_0(x)$ in Eq. (4) the values U_{Si} and U_{Ge} for Si and Ge are needed. As mentioned above, even for pure elements the exact values are not known. Instead, the corresponding cohesive energies per atom are conventionally used although there is no ample justification that such an identification is valid for semiconductors. Use of cohesive energy as a substitute for the surface binding energy is based upon an assumption that the number of bonds for an atom on the surface is equivalent to half of that for a bulk atom. Under ion bombardment semiconductors are easily amorphized and, therefore, atoms on the surface are on average more coordinated than just half of the bulk bonds. On the basis of experiments and computer simulations many authors pointed out that actual surface binding energy should be larger than the corresponding cohesive energy.[2,13,16,20–23] For Si and Ge the values $U_{\mathrm{Si}}=6.5$ eV and $U_{\mathrm{Ge}}=5.37$ eV are consistent with approaches of different authors for calculation of the effective surface binding energy and were found to lead to a considerably better agreement between theory and experiments. Therefore, we have used these values in our calculations.

Theoretical sputter yield for $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ calculated from Eq. (2) as a function of Ge concentration is shown in Fig. 2 by the dashed curve. While the calculated yields for elemental Si and Ge fit the experimental values very well, it is obvious that Eq. (2) fails to predict the experimental composition dependence of the sputter yield. Theory predicts almost linear dependence that is, however, not a surprise as all the terms of fraction x enter Eq. (2) as weighted averages of the alloy constituents.

In discussion of the nonlinear alloying effect on sputtering of $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ we assume that a total sputter yield for a polyatomic substrate may differ from the weighted average of the individual yields for the pure constituents due to surface binding differences of the atomic species. As was pointed out above, in sputtering of alloys and compounds mass and surface binding energy differences may cause the preferential emission of one of the components.[24] Since the momentum and energy transfer in a linear collision cascade is different for different components, the mass effect may lead to preferential sputtering of the lighter component. However, due to a high degree of randomization in the atomic cascade with respect to both energy and momentum transfer it is very unlikely that the mass effect can cause a deviation from a linear composition dependence for the total sputter yield unless, of course, the mass difference is extremely big. On the other hand, since sputtering is a surface phenomenon, any nonlinearity related to surface binding may have a dramatic impact on the sputtering process. Using in Eq. (3) surface binding energy U_0 in the form of the weighted average of $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ constituents implies that binding of the emitted atom is determined only by its own chemical identity and does not depend on the identities of the surrounding neighbors. Intuitively, it is clear that emission of, say, a Si atom surrounded by only Si neighbors is governed by the binding energy similar to that of pure Si whereas the binding should be different if the surroundings are populated with Ge atoms. Consequently, the probability of emission should be dependent on the local composition and, in general, be a function of the alloy stoichiometry. For first order approximation we can make an assumption that binding of a particular atom is determined by the nearest neighbors. The average number of nearest neighbors in a diamond structure is four for both crystal and amorphous phases. Therefore, we can assume that in $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ alloy only those Si atoms which are surrounded in the bulk by four Si neighbors are sputtered with the same probability as in pure Si and, hence, is characterized by the elemental surface binding energy U_{Si}. Such Si atoms and the corresponding nearest neighbors (nn) configuration can be considered as Si-like. Non-Si-like configurations are those that include at least one neighboring Ge. A Si atom sputtered from such a configuration makes a non-Si-like contribution to sputtering associated with a surface binding energy different as compared with U_{Si}. Similar considerations can be applied to Ge so that Ge-like and non-Ge-like nn configurations can be introduced in the sense of their contribution to sputtering yield. Since surface binding energy in the cascade theory is a parameter, i.e., it is not derived from the theory, in order to take into account contributions to sputtering from different nn configurations, U_0 in Eq. (3) is replaced with the following expression:

$$\frac{1}{U_0} = \sum_{\mathrm{nn}} \frac{a_{\mathrm{nn}}^{\mathrm{Si}} \cdot f_{\mathrm{nn}}^{\mathrm{Si}}(x)}{U_{\mathrm{nn}}^{\mathrm{Si}}} + \frac{a_{\mathrm{nn}}^{\mathrm{Ge}} \cdot f_{\mathrm{nn}}^{\mathrm{Ge}}(x)}{U_{\mathrm{nn}}^{\mathrm{Ge}}}, \quad (13)$$

where f_{nn}^{i} and U_{nn}^{i} ($i=$Si and Ge) are atomic fractions of Si and Ge atoms associated with different nearest neighbors configurations and corresponding effective surface binding energies, respectively. In a $\mathrm{Si}_{1-x}\mathrm{Ge}_x$ alloy, $(1-x)$ gives the fraction of all Si atoms. Then, $(1-x)^5$ is the fraction of Si atoms that have all four Si nearest neighbors and, hence, the configuration nn is Si-4Si (nearest neighbors). Following this nomenclature, Table I shows a summary of all the nn configurations with corresponding fractions and effective surface binding energies. The latter were calculated on the basis of available data on diatomic molecule bond energies.[25] Constants a_{nn}^{i} ($i=$Si, Ge) in Eq. (13) are fitting parameters as, first, we consider the constraint that binding energy is deter-

TABLE I. Summary of nearest neighbors configurations with corresponding fractions and effective surface binding energies.

Configuration	Fraction	Surface binding energy (eV)
Si-4Si	$(1-x)^5$	6.5
Si-3Si, 1Ge	$(1-x)^4x$	6.36
Si-2Si, 2Ge	$(1-x)^3x^2$	6.21
Si-1Si, 3Ge	$(1-x)^2x^3$	6.07
Si-4Ge	$(1-x)x^4$	5.92
Ge-4Si	$x(1-x)^4$	6.04
Ge-1Ge, 3Si	$x^2(1-x)^3$	5.87
Ge-2Ge, 2Si	$x^3(1-x)^2$	5.70
Ge-3Ge, 1Si	$x^4(1-x)$	5.54
Ge-4Ge	x^5	5.37

mined only by the nearest neighbors, and, second, some uncontrolled ordering of Si and Ge atoms in our $Si_{1-x}Ge_x$ samples could favor certain types of configurations at the expense of others, thereby changing eventually their fractions.

Sputter yield of $Si_{1-x}Ge_x$ calculated from Eq. (2) using Eq. (13) for U_0 is overlaid on the experimental values of the composition dependence in Fig. 2. The agreement between experimental and theoretical values suggests that the nonlinear composition dependence of the alloy sputter yield can be reliably predicted by the cascade theory over the entire composition range provided that the surface binding energy term has the form of Eq. (13).

In summary, the total sputter yield of $Si_{1-x}Ge_x$ bombarded with 3 keV Ar^+ ions has been measured for alloy composition from $x=0$ to 1. Sputtering in the linear cascade regime was found to result in a nonlinear S-shape composition dependence for the sputter yield. It has been shown that the composition dependence can be calculated from the linear cascade theory using a simple expression for the surface binding energy taking into account the alloying effect on binding of atoms in the binary compound. The form of the expression implies that in a polyatomic substrate binding of atoms is determined to a great extent by the identities of the surrounding atoms and not only by its own chemical identity. Accurate predictions of the sputter yield for the whole range of $Si_{1-x}Ge_x$ alloy composition are indicative supporting the interpretation of the alloying effect on the surface binding.

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