

# Surface smoothing of single-crystal diamond by high-speed cluster impacts with and without reactive erosion

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The smoothness of cluster-eroded surfaces of natural and synthetic diamond (Monodite) is compared after erosion with high-speed CO<sub>2</sub> as well as Ar cluster beams. The reactive accelerated cluster erosion (RACE) of the single-crystal diamond substrates using CO<sub>2</sub> clusters leads to a fourfold larger root-mean-square roughness than the erosion with non-reactive Ar clusters. On the other hand, the erosion rate observed with the accelerated Ar clusters is lower by about the same factor than that observed with the accelerated CO<sub>2</sub> clusters. Molecular dynamics calculations reveal corresponding differences already in the simulations of single cluster impacts. Copyright © 2004 John Wiley & Sons, Ltd.

**KEYWORDS:** surface smoothing; diamond; reactive accelerated cluster erosion

## INTRODUCTION

Surfaces may be modified in the nanometre range by impacts of highly accelerated nanoparticles or clusters of atoms.<sup>1–5</sup> The impact energy (in the present contribution, 100 keV) is transferred to the substrate directly at the point of impact, leading to a high-speed collective motion of substrate as well as of cluster atoms and to a short-lived localized plasma cloud of several thousand Kelvin temperature. Ongoing bombardment with high-speed clusters erodes the surface and, by appropriate surface masking, can be used to generate two- or even three-dimensional micro- or nanostructures. Cluster erosion has been shown to yield very smooth surfaces,<sup>3</sup> with values of the root-mean-square roughness of the order of 1 nm in the case of silicon.<sup>6,7</sup> High surface finish is of paramount importance in micromechanics, because of the growing role of friction versus inertia due to the increasing surface-to-volume ratio.

Diamond is distinguished by its superior material properties, such as extreme hardness and high heat conductivity, high optical transparency and large refractive index. Low values of observed friction coefficients, as well as high wear resistance, make diamond also an ideal material for micro-

or nanomechanical applications if it can be processed with a high surface finish.

Here we compare the erosion of single-crystal diamond by high-speed impacts of CO<sub>2</sub> or Ar clusters, respectively. The two species differ by the presence, or absence, respectively, of a reactive component in the erosion process. With Ar clusters only physical erosion takes place but with CO<sub>2</sub> clusters the chemical reaction of plasma-activated CO<sub>2</sub> with the carbon substrate leads to an additional chemical erosion. Correspondingly, the process was named RACE: reactive accelerated cluster erosion.<sup>4</sup> We also investigate here the erosion of two different diamond surfaces, a natural diamond (111) surface and a polished synthetic diamond (Monodite, de Beers) (100) surface.

The first to study the cluster beam erosion of polycrystalline chemical vapour deposited (CVD) diamond films, as well as of single-crystal diamond, was our research group at Karlsruhe, Germany.<sup>8,9</sup> The treatment of the rough polycrystalline CVD films<sup>8</sup> revealed a very distinguished flattening, or polishing, by a prolonged bombardment with accelerated CO<sub>2</sub> clusters, as observed earlier with other polycrystalline specimens, e. g. quartz.<sup>4</sup> On the other hand, a comparison of the CO<sub>2</sub> cluster erosion rates of various materials, including single-crystal diamond, indicated a surprisingly large erosion rate for diamond,<sup>9</sup> which was not conceivable in the light of the model presented in Ref. 1. This model was based solely on the estimated volume of the respective cluster impact craters, which should decrease with increasing target hardness. The purely physical picture of the cluster erosion had to be extended to include a chemical erosion component, leading thus to the RACE concept.<sup>4</sup>

Later on, the polishing of polycrystalline CVD diamond was reinvestigated in Japan using non-reactive accelerated

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Ar cluster beams.<sup>10</sup> A reduction of the average roughness of thin CVD films by about an order of magnitude was observed. With O<sub>2</sub> clusters instead of CO<sub>2</sub> clusters, which according to the RACE model increases the erosion rate by about a factor of 2, an approximately tenfold higher erosion rate was quoted for O<sub>2</sub> clusters compared with Ar clusters.<sup>11</sup>

The detailed mechanism of the surface smoothing under prolonged cluster bombardment has yet to be understood, particularly when comparing reactive and non-reactive erosion. A favoured model is based on the filling of troughs by a redeposition of ejected material.<sup>10</sup> In a somewhat related study of the deposition of metal clusters on a slightly rough metal surface of the same kind, a molecular dynamics investigation of impacts on inclined parts of the surface revealed a predominant downhill motion of the involved material, viz. a filling of troughs.<sup>12</sup>

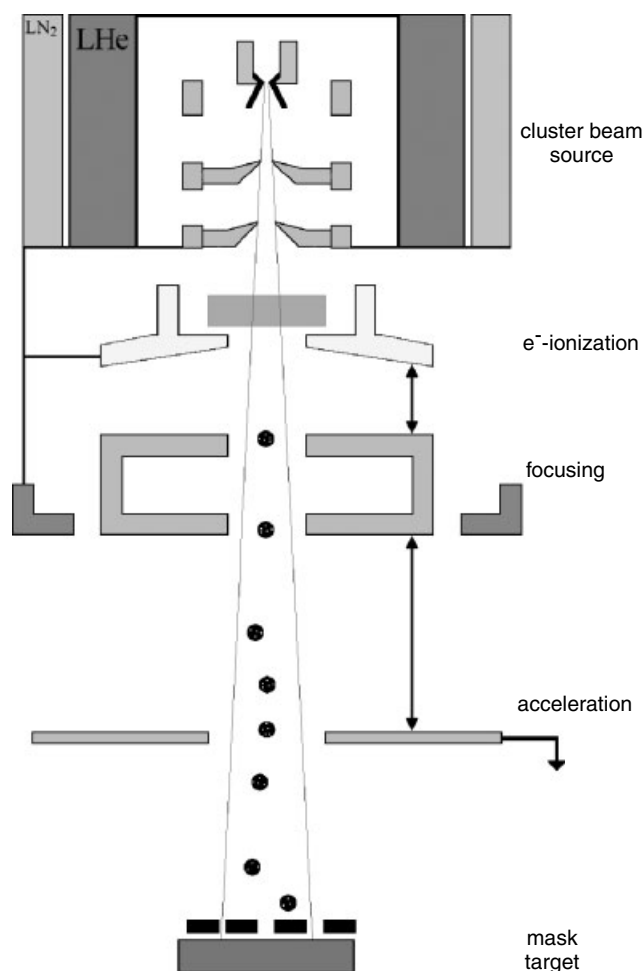
After describing the experimental facilities and procedures for cluster-eroding diamond, we shall present the results on the roughness of the surface of the various single-crystal diamond specimens as measured with atomic force microscopy (AFM). A comparison will be given with the results of molecular dynamics simulations, details of which, including references, have been reported elsewhere.<sup>13–16,18</sup>

## EXPERIMENTAL

The cluster erosion facility has been described earlier.<sup>17,18</sup> Clusters are generated by adiabatic expansion of the respective gas through a converging–diverging nozzle of 0.1 mm diameter, 10° angle of initial divergence and 28 mm length of the diverging part. The source pressures are chosen to yield mean cluster sizes of ~1000 molecules of CO<sub>2</sub>, or atoms of Ar, respectively, according to the known scaling laws.<sup>19</sup> As the molecular weights of 44 for CO<sub>2</sub> and 40 for Ar are comparable, the cluster masses should be comparable too.

The core of the partly condensed nozzle flow is transferred via two skimming orifices into a high vacuum to form the cluster beam. The beam then is partly ionized by the impact of 150 eV electrons, focused by up to 10 keV and accelerated towards the grounded target surface by a 100 keV potential difference. For that purpose, the whole cluster beam source has to be at the corresponding high positive potential. The whole set-up is housed in one common vacuum chamber whose upper part, at high potential, is electrically separated from the lower part, at ground potential, by an insulating ceramic tube acting as a part of the vacuum chamber wall.

The outer part of the nozzle flow, containing >90% of the nozzle throughput, is pumped by condensation onto bath-cooled cryopanel in the upper high-voltage part of the vacuum chamber. Thereby, the instalment of electrical power for pumping is avoided at high electrical potential. Although liquid nitrogen cooling is sufficient to condense CO<sub>2</sub>, pumping of Ar requires lower temperatures of the cryopanel. Fortunately, we could use cooling by liquid helium because the cluster beam source was originally designed for the generation of hydrogen clusters. Figure 1 shows a schematic view of the cluster erosion facility as used for argon cluster beams. The liquid helium supply in the bath cryostat allows for uninterrupted operation of ~6 h.



**Figure 1.** Experimental set-up used for the accelerated cluster erosion with an Ar cluster beam. For use with a CO<sub>2</sub> cluster beam the inner cryostat is filled with liquid nitrogen instead of liquid helium. The whole set-up is housed in a common vacuum chamber.

Initially, the nozzle is slightly pre-cooled for Ar cluster generation to ~277 K by flowing evaporated liquid nitrogen. During operation, the nozzle cools linearly down to ~250 K within 5 h, due to the radiation losses to the cryopanel. Although this may lead to a gradual increase of cluster size, this is not considered crucial because the cluster impact acts through its energy, which is determined solely by the electrical acceleration. With CO<sub>2</sub> as the beam gas, the corresponding nozzle cooling during the same time interval runs from approximately 287 K to 260 K.

Tantalum foils 100 µm thick with a lapped edge served as erosion masks, which were placed as tightly as possible onto the diamond specimen. In the case of natural diamond, optically clear type IIa diamond 'flats' were used, ~0.5 mm thick, and with two nearly parallel faces, one of which served as the erosion target. These faces are (111) single-crystal surfaces, as indicated by the typical 'trigons': Trigonal pyramid-shaped pits. The synthetic single-crystal diamond specimens were yellow de Beers Monodite bricks (diamond, type Ia) with sawed (100) surfaces.

For investigating the topology of the eroded diamond surfaces an atomic force microscope (Dimension 3100, Digital

Instruments) was used. The specimens were transferred from the vacuum chamber to the microscope under ambient atmospheric conditions without special precautions. In order to determine the respective erosion rates of synthetic diamond, a nickel stencil mask with hexagonal holes served to erode blind holes whose depths were determined with an autofocus measuring system (MicroSurf, OM Engineering).

## RESULTS

### Topography of cluster-eroded natural diamond

Using appropriate masking, eroded areas were generated on an unpolished natural diamond flat by CO<sub>2</sub> or Ar cluster beam erosion. The erosion period was 50 min in each case in order to establish a stationary situation. Figure 2 shows AFM images obtained from cluster-eroded 30  $\mu\text{m}$   $\times$  30  $\mu\text{m}$  regions and from a corresponding region of a masked pristine area. The topography was measured with AFM in the so-called tapping mode, viz. in a non-contact mode. The vertical scale in Fig. 2 is 120 nm per division. Although erosion with CO<sub>2</sub> clusters yields a roughening of the diamond surface, the Ar cluster-eroded surface turns out to be even smoother than the pristine diamond surface. (Isolated spikes in these regions may well represent dust particles.)

For Fig. 3, the inspected regions have been reduced to 5  $\mu\text{m}$   $\times$  5  $\mu\text{m}$  and the vertical scale expanded to 20 nm per division. In the Ar cluster-eroded region a surface roughness is now visible, whereas the erosion with CO<sub>2</sub> clusters still yields much larger spikes.

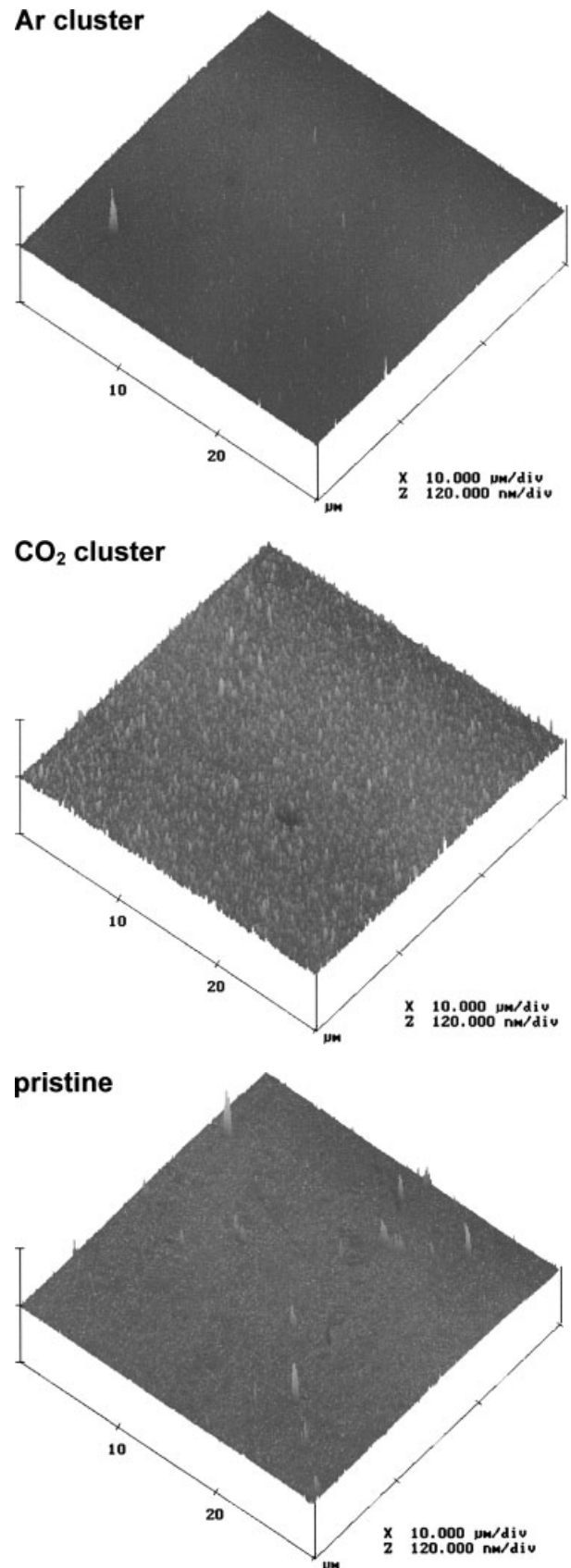
### Topography of cluster-eroded synthetic diamond (Monodite)

Figure 4 shows a comparison of the topography of 30  $\mu\text{m}$   $\times$  30  $\mu\text{m}$  regions on a synthetic diamond (Monodite) surface in either the pristine or a cluster-eroded state, with a z-scale of 100 nm per division. In this case, a sequential erosion by CO<sub>2</sub> clusters for 20 min, followed by Ar cluster erosion for 30 min, is added as a new variant. Obviously, the difference between the roughness appearance of the CO<sub>2</sub> cluster- and the Ar cluster-eroded surfaces is appreciably more pronounced than in the case of the natural diamond (111) surface. The sequentially eroded region, however, exhibits the same smoothness as the region eroded only by Ar clusters.

The results of the AFM investigation of a smaller region, 1  $\mu\text{m}$   $\times$  1  $\mu\text{m}$ , are reproduced in Fig. 5, again with 100 nm per division in the z-direction. Here, the height images were obtained in contact mode, in which loosely attached dust particles may be removed by the scanning tip. Again, the CO<sub>2</sub> cluster-eroded surface shows remarkably larger structures than the Ar cluster-eroded regions, with or without preceding CO<sub>2</sub> erosion, which are evidently smoother than even the pristine diamond surface.

### Roughness analysis

The AFM height data can be used to evaluate the respective roughness of the considered regions quantitatively. Table 1 summarizes the results for natural diamond and Table 2 for the synthetic diamond species.  $R_a$  is the arithmetic average value and  $R_q$  the root-mean-square value of the measured



**Figure 2.** Images of cluster-eroded and pristine surface areas of natural diamond of 30  $\times$  30  $\mu\text{m}^2$  with a vertical scale of 120 nm per division obtained with an atomic force microscope in tapping mode (height images). The Ar cluster erosion yields a smoother surface than the pristine surface as well as the surface eroded by CO<sub>2</sub> clusters.

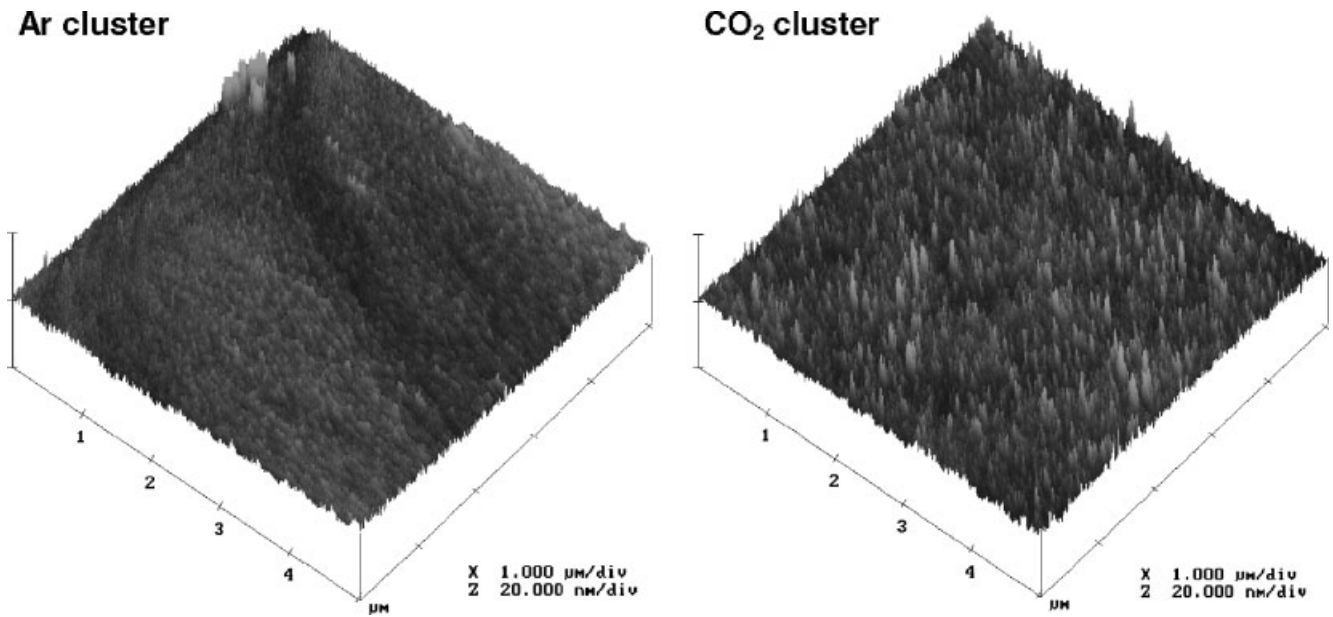


Figure 3. Images of cluster-eroded surface areas of natural diamond of  $5 \times 5 \mu\text{m}^2$  with a vertical scale of 20 nm per division (height images, tapping mode).

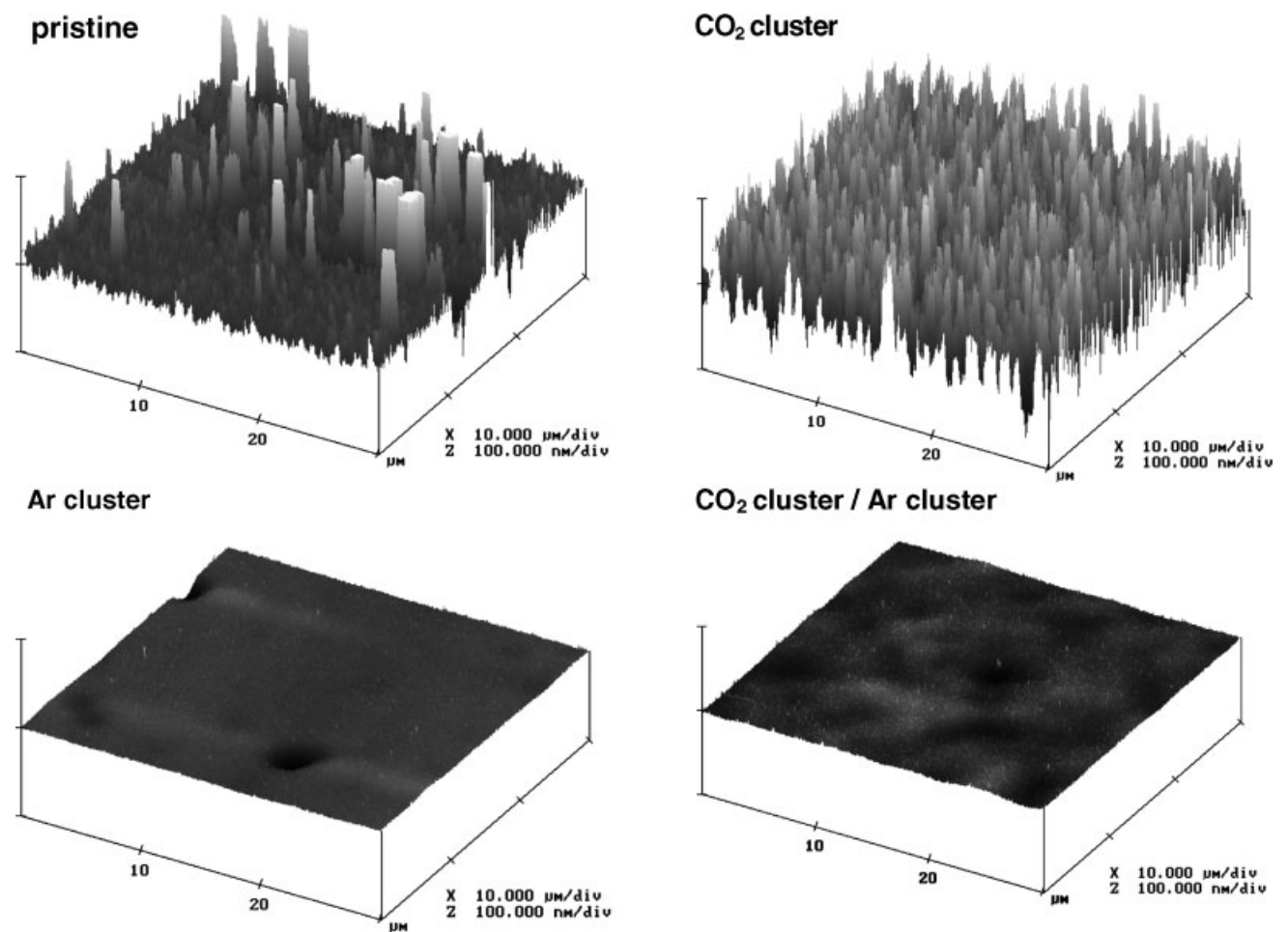
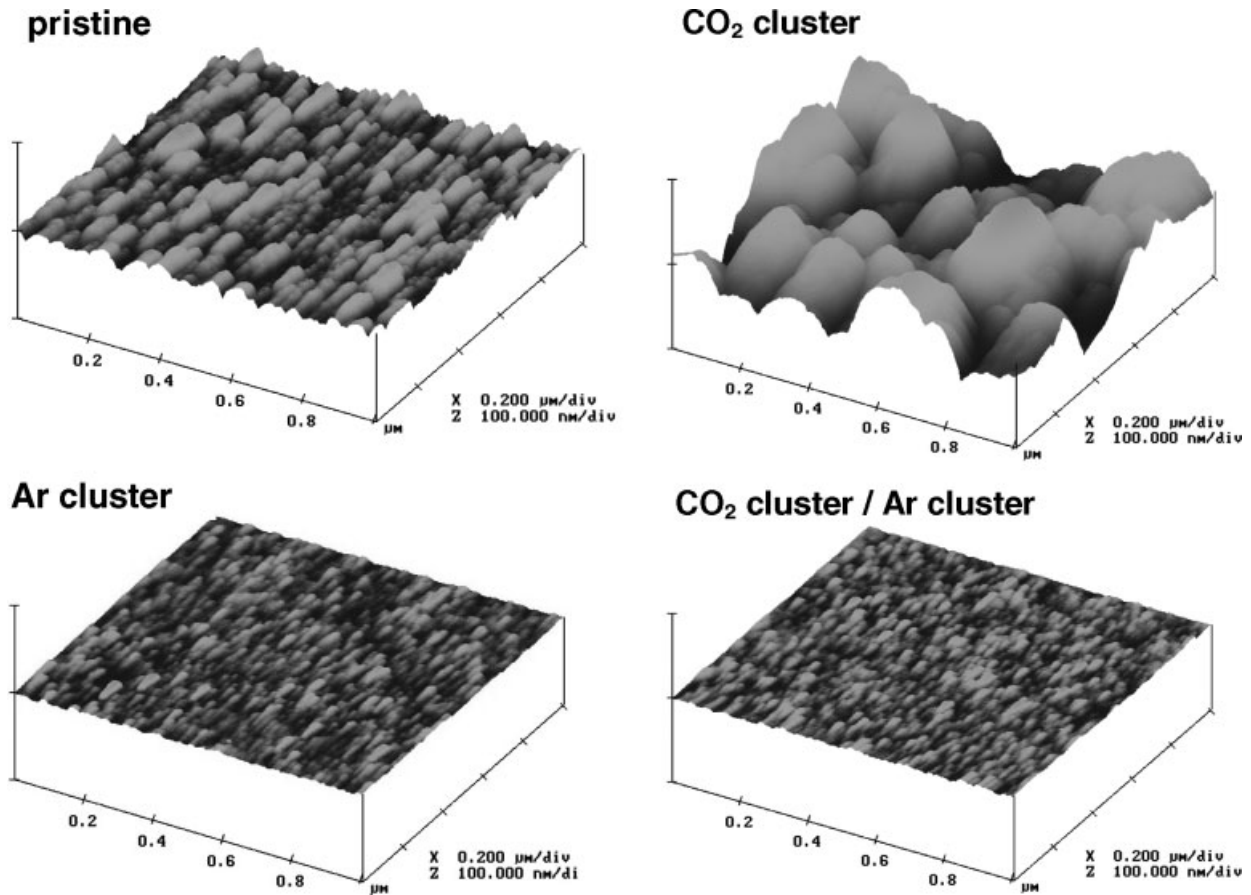


Figure 4. Images of cluster-eroded and pristine surface areas of synthetic diamond (Monodite) of  $30 \times 30 \mu\text{m}^2$  with a vertical scale of 100 nm per division (height images, tapping mode). A sequential erosion by accelerated  $\text{CO}_2$  clusters followed by accelerated Ar clusters yields a topography similar to that obtained by erosion with accelerated Ar clusters alone.



**Figure 5.** Images of cluster-eroded and pristine surface areas of synthetic diamond (Monodite) of  $1 \times 1 \mu\text{m}^2$  with a vertical scale of 100 nm per division (height images, contact mode). Both Ar cluster-eroded surfaces are smoother than the pristine surface.

height amplitude, respectively, and  $R_{\text{max}}$  is the maximum height amplitude difference in the considered region. The minimum values for the respective region are indicated in bold type and 'UB' denotes a pristine ('unbeamed') region. Statistically, the most significant parameter is  $R_q$ .

For natural diamond, the roughness  $R_q$  of the Ar cluster-eroded and the pristine  $1 \times 1 \mu\text{m}^2$  regions is practically the same (0.7 nm and 0.6 nm, respectively) and nearly a factor

**Table 1.** Roughness values of Ar cluster-eroded,  $\text{CO}_2$  cluster-eroded and pristine (UB) surface areas of a natural diamond (111) surface

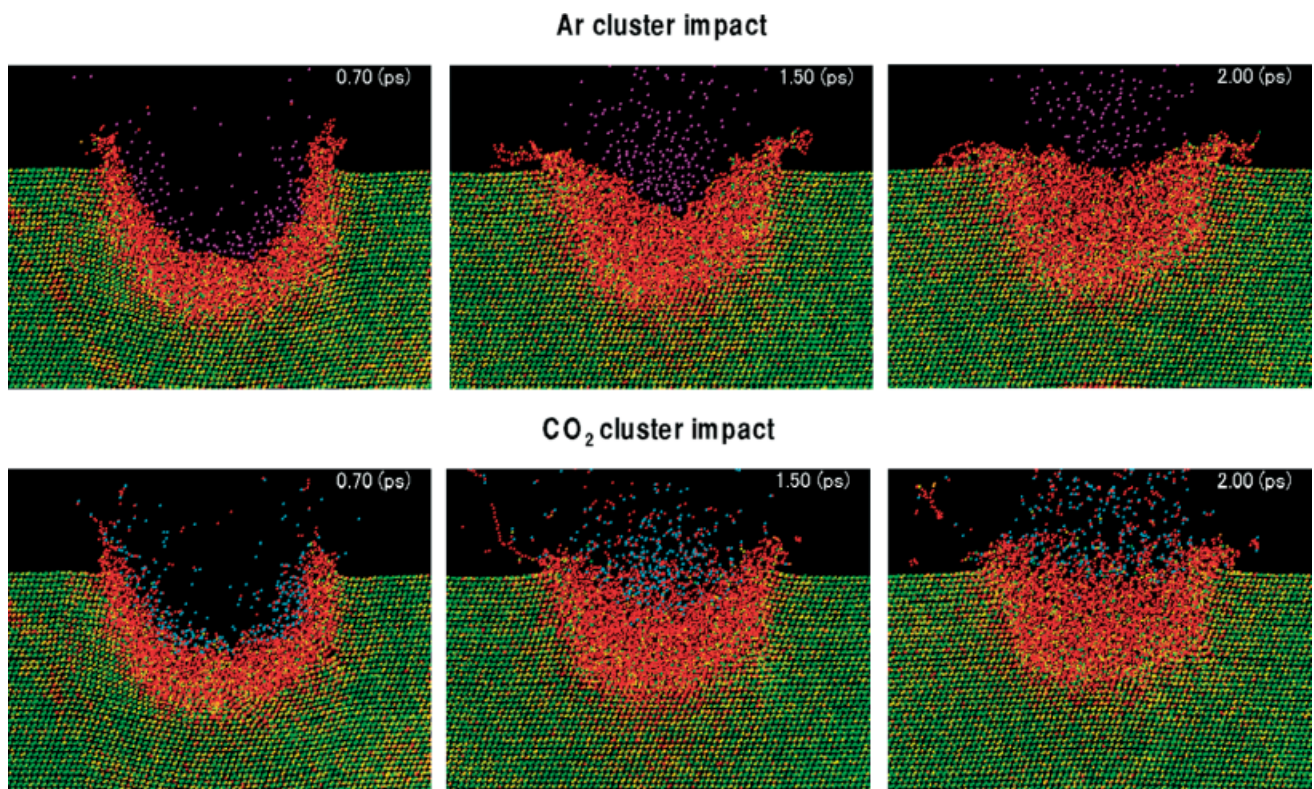
Area ( $\mu\text{m} \times \mu\text{m}$ )		$R_a$ (nm)	$R_q$ (nm)	$R_{\text{max}}$ (nm)
1 × 1	Ar	<b>0.5</b>	0.7	<b>8</b>
	$\text{CO}_2$	1.6	2.4	19
	UB	<b>0.5</b>	<b>0.6</b>	<b>8</b>
5 × 5	Ar	<b>0.9</b>	<b>1.1</b>	<b>14</b>
	$\text{CO}_2$	1.1	1.5	17
	UB	0.9	2.2	58
30 × 30	Ar	<b>1.8</b>	<b>2.6</b>	95
	$\text{CO}_2$	3.1	4.0	<b>48</b>
	UB	2.7	3.7	125

$R_a$  = arithmetic mean roughness;  $R_q$  = root-mean-square roughness;  $R_{\text{max}}$  = maximum difference of roughness height amplitudes. Bold type indicates minimum values.

**Table 2.** Roughness values of Ar cluster-eroded,  $\text{CO}_2$  cluster-eroded,  $\text{CO}_2$  cluster-eroded and subsequently Ar cluster-eroded and pristine (UB) surface areas of a synthetic diamond (100) surface

Area ( $\mu\text{m} \times \mu\text{m}$ )		$R_a$ (nm)	$R_q$ (nm)	$R_{\text{max}}$ (nm)
0.2 × 0.2	Ar	1.1	1.3	<b>6.7</b>
	$\text{CO}_2$	2.9	3.6	29.1
	$\text{CO}_2/\text{Ar}$	<b>0.8</b>	<b>1.0</b>	7.7
	UB	2.9	3.8	24.6
1 × 1	Ar	<b>0.9</b>	<b>1.2</b>	<b>9.9</b>
	$\text{CO}_2$	18.0	22.3	130.0
	$\text{CO}_2/\text{Ar}$	1.2	1.5	10.6
	UB	4.0	5.0	36.6
5 × 5	Ar	<b>0.9</b>	<b>1.1</b>	<b>11.2</b>
	$\text{CO}_2$	22.8	29.3	218.0
	$\text{CO}_2/\text{Ar}$	1.0	1.3	12.8
	UB	<b>14.3</b>	28.0	350.1
30 × 30	Ar	<b>1.6</b>	<b>3.1</b>	59.0
	$\text{CO}_2$	21.5	27.2	273.0
	$\text{CO}_2/\text{Ar}$	3.4	4.5	<b>50.0</b>
	UB	10.1	21.6	464.0

$R_a$  = arithmetic mean roughness;  $R_q$  = root-mean-square roughness;  $R_{\text{max}}$  = maximum difference of roughness height amplitudes. Bold type indicates minimum values.



**Plate 1.** Comparison of the crater profiles as obtained by molecular dynamics simulations at 0.70 ps, 1.50 ps and 2.00 ps after the impact of an accelerated Ar cluster (above) and an accelerated CO<sub>2</sub> cluster (below) onto a (111) diamond surface. A slice of the calculated specimen of 15 nm width and 1 nm thickness in the direction perpendicular to the figure plane is shown. At 2 ps after the impact the replenishment of the craters leads to a smoother and more compact surface in the case of the Ar cluster impact.

of 4 lower than that of the CO<sub>2</sub> cluster-eroded region. For the larger areas, the Ar cluster-eroded region is smoother than even the pristine UB region. In these larger areas, isolated surface features, or dust particles tend to increase the measured average roughness values.

With synthetic diamond in the smallest investigated area of 200 × 200 nm<sup>2</sup> the roughness  $R_q$  of the Ar cluster-eroded and CO<sub>2</sub> and then Ar cluster-eroded regions (1.3 nm and 1.0 nm, respectively) is again a factor of ~4 lower than that of the CO<sub>2</sub> cluster-eroded region but also lower than that of the pristine UB region, both of which practically agree in this case. For the larger investigated areas, however, the advantage in roughness reduction achieved with the Ar cluster erosion becomes very pronounced, being a factor of the order of 25 smaller than the roughness observed with the CO<sub>2</sub> cluster erosion, or even that of the pristine surface. It has to be mentioned again that the Monodite specimen had not been polished prior to the cluster beam treatment. In comparison with the natural diamond the different eroded crystal surface (100), versus (111), also might be taken into consideration.

### Maximum erosion rates

Using a nickel stencil mask with a hexagonal array of hexagonal openings, corresponding arrays of blind holes were eroded into the synthetic diamond (Monodite) (100) surface and, for comparison, into a (100) silicon wafer. The depths of these blind holes were determined with an optical autofocus system (MicroSurf), which was scanned slowly enough to ensure signal detection from the respective hole bottoms. From the maximum values of the hole depths the following maximum values of the erosion rates were determined: 4.3 μm h<sup>-1</sup> for Ar cluster erosion of diamond, 15.6 μm h<sup>-1</sup> for CO<sub>2</sub> cluster erosion of diamond and 5.6 μm h<sup>-1</sup> for CO<sub>2</sub> cluster erosion of silicon. Hence the ratio of the maximum cluster erosion rates of CO<sub>2</sub> versus Ar clusters for diamond is 3.63, or nearly 4.

## MOLECULAR DYNAMICS SIMULATIONS OF SINGLE CLUSTER IMPACTS

In order to compare with the results of the erosion measurements, some results of molecular dynamics simulations of single cluster impacts on diamond (111) are presented. Detailed reports on these simulations have been published elsewhere.<sup>13–16,18</sup>

The impacts of Ar clusters consisting of 961 atoms, or of CO<sub>2</sub> clusters consisting of 960 molecules, respectively, with an initial kinetic cluster energy of 100 keV, on a (111) surface of a hexagonal single-crystal diamond specimen of 18.2 nm vertical thickness and 34.4 nm diagonal width have been simulated. In total, this specimen contains 2 365 650 carbon atoms. At first, only an inner hexagonal cylinder containing 758 440 atoms and one-sixth of the outer region with 308 420 atoms were simulated, considering the other five-sixths of the outer region as just symmetric repetitions of the simulated one-sixth,<sup>13–15,18</sup> in order to reduce the computational effort. Later on, a simulation of the full specimen confirmed the permissibility of the prior simplification.<sup>16</sup>

The impact energy of 100 eV per atom or molecule is certainly larger than the electronic bandgap of diamond (5.5 eV) but is still in the low energy regime for ion impact phenomena where nuclear stopping phenomena dominate over electronic stopping.<sup>20</sup> Surface smoothing of metal surfaces under megaelectron-volt atomic ion irradiation has been ascribed to thermal spike-induced melting and flowing,<sup>21</sup> viz. thermal effects describable by classical collision dynamics. Here, we assume the multiple interactions between cluster and target atoms to be the most important feature of the cluster impact and use classical dynamics for the molecular dynamics simulations.

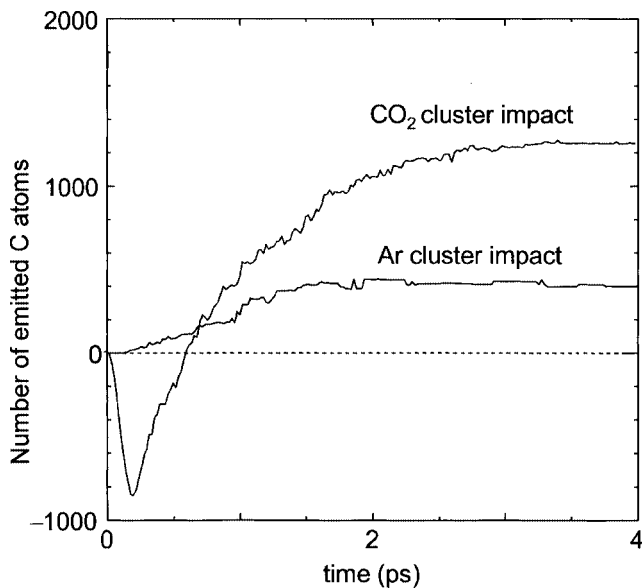
Plate 1 shows a comparison of the relaxation of the craters formed directly after the impact of an Ar cluster, or of a CO<sub>2</sub> cluster, respectively. A slice of the sample (width 15 nm and thickness only 1 nm) in the direction perpendicular to the figure plane containing the impact trajectory is presented to clarify the crater cross-section. At 0.7 ps after the impact the transient craters are nearly indistinguishable. At 2 ps, however, the crater formed by an Ar cluster impact is filled up in a rather smooth and compact way, whereas the CO<sub>2</sub> interaction leads to a more loose packing with a rather disrupted surface. Furthermore, the craters formed under CO<sub>2</sub> cluster impact were found to show, at longer times of 30 ps, so-called 'complex' crater shapes with a central peak, which is missing when the craters result from Ar cluster impact. It is assumed that the higher mass of the Ar atoms in comparison with the carbon atoms of the diamond damps the motion of the latter. In addition, oxygen atoms tend to remain on or in the sample surface, disturbing a more complete recrystallization.<sup>15</sup>

Although it is not possible to infer from these short simulations the surface roughness to be expected finally for much larger regions and much longer times, the tendency towards a rougher, less compact surface after a high-speed impact of a CO<sub>2</sub> cluster, as compared to an Ar cluster impact, seems to be illustrated.

In Fig. 6, the number of carbon atoms leaving the region of impact is depicted. After the impact of an Ar cluster, the ejection of carbon atoms, occurring mostly at the crater rim, ends already after 2 ps. After an impact of a CO<sub>2</sub> cluster, however, carbon leaves the sample until at least ~4 ps after the impact, leading to an integral loss of carbon atoms that is a factor of 3.1 larger than that caused by an Ar cluster impact. When considering the initial addition of the carbon in the cluster, which leads to the negative loss (carbon atom gain) until ~0.7 ps after impact, the integral loss ratio rises even to ~5.2. These values coincide rather well with the observed erosion ratio of nearly 4.

## DISCUSSION

The cluster erosion of single-crystal diamond has been shown to be enhanced in the interaction of the high-speed CO<sub>2</sub> clusters with the diamond surfaces, as compared to the interaction with high-speed Ar clusters. This RACE enhancement arises from the activation of CO<sub>2</sub> in the high-temperature plasma generated by the impact, yielding, for example, oxygen radicals by dissociation. Without this



**Figure 6.** Calculated number of carbon atoms emitted from the diamond surface after the respective cluster impact. The CO<sub>2</sub> cluster impact leads to a threefold higher total emission, although until ~0.7 ps the carbon atoms of the cluster add up to those of the diamond target.

chemical component of the erosion, the eroded surfaces are considerably smoother. It has also been shown here that a sequential erosion with at first CO<sub>2</sub> clusters and then Ar clusters allows a high erosion rate to be combined with a high surface finish.

At least for the smallest eroded area investigated, the smoothness ratio achieved with the two cluster species was found to be the same for a natural single-crystal diamond (111) surface and a synthetic single-crystal diamond (100) surface. The absence of a higher resistance against chemical erosion of the (111) natural diamond surface may be understood as a consequence of local destruction of the crystal order during the high-speed cluster impact. The much larger roughness observed for the larger scan areas of the reactively eroded (100) surface of the synthetic diamond remains to be explained, however.

The observed long-term ratios of the roughness values and erosion rates obtained with and without a reactive erosion component agree—in the case of the erosion rates nearly quantitatively—with corresponding data obtained in large-scale molecular dynamics simulations of isolated high-speed cluster impacts. These simulations cover up to 30 ps after the impact. The smoothing of polycrystalline CVD diamond requires an explanation of the preferential erosion of surface elevations. In the presently considered case of the cluster erosion of flat single-crystal diamond surfaces, however, the characteristic features of erosion rate and roughness show up already as short-time effects of individual accelerated cluster impacts, a succession of which then leads to the corresponding long-term properties.

## CONCLUSIONS

The smoothing of single-crystal diamond surfaces under bombardment with accelerated CO<sub>2</sub> or Ar clusters was analysed on the nanometre scale. In comparison with Ar clusters, CO<sub>2</sub> clusters yield about a fourfold higher rate of erosion of natural (111) diamond surfaces as well as of synthetic (100) diamond surfaces, due to the combination of local physical disintegration of the crystal lattice and reactive erosion by activated oxygen radicals. This RACE effect is nearly quantitatively corroborated by molecular dynamics simulations. On the other hand, the bombardment with accelerated Ar clusters leads to a root-mean-square roughness of ~1 nm, which is about a factor of 4 or more smaller than that achieved with accelerated CO<sub>2</sub> clusters. This higher smoothing effect of the Ar cluster bombardment is attributed to the more compact relaxation of the cluster impact craters, as revealed by the molecular dynamics simulations.

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