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Molecular dynamics simulation of amorphization in forsterite by cosmic rays

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Abstract

We have examined cosmic ray interactions with silicate dust grains by simulating a thermal spike in a 1.25 million atom forsterite (Mg_2SiO_4) crystal with periodic boundaries. Spikes were generated by giving a kinetic energy of 1 or 2 eV to every atom within a cylinder of radius 1.73 nm along the [0 0 1] direction. An amorphous track of radius ~3 nm was produced for the 2 eV/atom case, but practically no amorphization was produced for 1 eV/atom because of effective dynamic annealing. Chemical segregation was not observed in the track. These results agree with recent experimental studies of ion irradiation effects in silicates, and indicate that cosmic rays can cause the amorphization of interstellar dust.

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Keywords: Cosmic ray interactions; Interstellar dust; Amorphization; Silicate; Forsterite; Molecular dynamics simulation

1. Introduction

Interstellar dust grains play a crucial role in the dynamics of star formation and influence the appearance of galaxies due to their role in the reradiation of starlight [1]. Crystalline silicates, such as forsterite (Mg_2SiO_4), are important components of interstellar dust. Up to 15% of the silicates in stellar ejecta are estimated to be crystalline; in sharp contrast, the fraction of crystalline silicates in the interstellar medium (ISM) is only about 0.2% [2]. Bombardment by the heavy-ion component of cosmic rays over a time scale of the order of several million years is considered to be a possible cause of the amorphization of silicates in the ISM [2]. However, the processes of amorphization and dynamic annealing during energetic processing of silicate dust grains are not well understood. These phenomena need to be critically examined, because interstellar dust influences the process of star formation [1].

The effect of cosmic ray heavy-ion bombardment on the crystallinity of interstellar dust can be examined in laboratory experiments using an ion beam with a large electronic stopping ($\frac{dE_e}{dx}$) component. Recently, Bringa et al. [3] irradiated single crystals of Mg_2SiO_4 with 10 MeV Xe^{3+} ($\frac{dE_e}{dx} = 4.5$ keV/nm) at room temperature and observed the formation of amorphous tracks. The authors [3] estimated the amorphous track radius to be $\sim 2.8 \pm 0.6$ nm based on Rutherford backscattering/channeling and a defect overlap model. In order to interpret the results of such laboratory experiments and extend them to conditions relevant to the ISM, atomistic simulations of damage accumulation are required.

Atomistic modeling of the interaction of highly ionizing radiation with matter is not straightforward, because classical molecular dynamics (MD) simulations commonly used to examine beam–solid interactions (typically at keV energies) do not account for electronic excitations. These

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effects associated with a high $\frac{dE_e}{dx}$, typical of cosmic rays, can be indirectly incorporated into the simulation by assuming that a fraction of the electronic energy deposited in the material, typically 0.4 for a 10 MeV Xe^{3+} ion [4] and represented by f, is subsequently transferred to the atoms. This transfer can be modeled as a thermal spike [5] by giving a velocity in a random direction to atoms within a cylinder. The radius of the cylinder (also known as a track) and the kinetic energy given to each atom within the cylinder are key parameters of the simulation. By varying the kinetic energy imparted, one can simulate different $\frac{dE_e}{dx}$ conditions. Such simulations have been limited in the past to elemental materials using simple interatomic potentials [6], because it is computationally intensive to simulate complex materials, such as silicates, with realistic potentials that include long-range Coulombic interactions.

In the present work, we have performed classical molecular dynamics simulation of thermal spikes in Mg_2SiO_4 using system sizes of more than a million atoms for times of the order of 0.1 ns. By imparting different kinetic energies to the atoms, we show that ionizing radiation can amorphize Mg_2SiO_4, and the amorphous track radius depends on the value of $\frac{dE_e}{dx}$. In addition, our observation of dynamic annealing in irradiated Mg_2SiO_4 at 300 K complements experimental studies by providing information about defect evolution that cannot be obtained directly by experiment. Some preliminary results have been presented in a recent paper [7].

2. Simulation details

We used the DL_POLY code [8] to perform molecular dynamics simulations using a simulation cell containing 1249920 atoms ($62 \times 30 \times 24$ unit cells measuring $30 \times 30.5 \times 14.4$ nm). The initial temperature was 300 K, as in the experiment of Bringa et al. [3]. We performed the simulations in the micro canonical ensemble with periodic boundaries. The interactions, based on the Teter model [9], consisted of a long-range Coulombic term and a short-range Buckingham-type term. The Coulombic term is given by

$$V_{ij}^{C} = \frac{q_i q_j e^2}{4\pi\varepsilon_0 r_{ij}}, \tag{1}$$

where q_i and q_j are the charges of the ions (Mg: 1.2; Si: 2.4; O: −1.2) interacting at a distance r_{ij}, e is the electron charge, and ε_0 is the vacuum permittivity. We evaluated the Coulombic interaction using the smooth particle mesh Ewald summation method [8]. The Buckingham-type term, used at distances less than 1 nm for Mg–O, Si–O and O–O interactions, is given by

$$V_{ij}^{B} = A_{ij} \exp\left(-\frac{r_{ij}}{\rho_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6}. \tag{2}$$

The parameters A_{ij}, ρ_{ij} and C_{ij} for Si–O and O–O interactions and the details of fitting the potential have been reported previously [10]. For the Mg–O interaction, these parameters are 7060.0 eV, 0.0211 nm and 1.92×10^{-5} eV nm^6, respectively [9]. The parameters were obtained by fitting to the structural and mechanical properties of several silicates [9].

We joined the above interaction potential to the repulsive Ziegler–Biersack–Littmark potential [11] at distances less than 0.1 nm using an exponential function in order to realistically model the close-pair repulsion. Table 1 lists the structural data and elastic constants obtained from the Teter model and experiments [12, and references therein]. It can be seen that the Teter model [9] provides a good fit to the experimentally observed structural parameters and elastic constants of forsterite. The exceptions are c_{44} and c_{55}, which are considerably smaller than the corresponding experimental values. While this reflects on the limitations of empirical potentials for modeling complex oxides, it is not expected to adversely impact the conclusions of the present study.

We simulated two cases of thermal spikes by giving random velocities corresponding to kinetic energies of 1 and 2 eV/atom, respectively, to atoms within a cylinder of radius 1.73 nm (13 040 excited atoms). The cylinder was along the [0 0 1] direction (Z axis of length 14.4 nm) and located at the center of the simulation cell ($X = 0$, $Y = 0$). If we assume $f = 0.4$, the two thermal spikes correspond to $\frac{dE_e}{dx}$ of 2.25 and 4.5 keV/nm, respectively. The final temperature was 338 K after 0.074 ns for the case of 1 eV/atom, and 374 K after 0.116 ns for 2 eV/atom. Energy conservation was excellent and fluctuations in internal energy, $\frac{\Delta E}{E}$, were less than 10^{-6}. We integrated the equations of motion with the velocity Verlet algorithm using a variable time step with a minimum step size of 0.01 fs and a maximum of 2 fs. We have not performed detailed statistical studies by simulating multiple thermal spikes for each energy nor have we examined spikes along different crystallographic directions due to computational cost.

The identification of amorphous regions in simulations of radiation damage in complex ceramics is a challenging

Table 1
Structural data and elastic constants of forsterite

	Teter model	Experiment [12]
Lattice constants (nm)		
a	0.475	0.475
b	1.019	1.019
c	0.598	0.598
Cell volume (nm^3)	0.290	0.290
Elastic constants (GPa)		
c_{11}	312	328–331
c_{22}	176	191–200
c_{33}	220	230–236
c_{44}	29	66–67
c_{55}	56	81–83
c_{66}	67	81–82
c_{12}	69	64–66
c_{23}	77	72–74
c_{13}	75	68–69
Bulk modulus	122	140