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Molecular dynamics study of the surface scattering and capture of nanoparticles at high velocities

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ABSTRACT

Classical molecular dynamics has been used to simulate the elastic scattering of single solid nano-particles from a flat rigid surface. The results show that with the inclusion of a weighted dissipative force the method is capable of reproducing experimental measurements of the critical velocities required for sodium chloride and silver nano-particles to experience elastic scattering from a surface. The velocity at which impacting particles undergo a transition away from elastic scattering and towards plastic deformation is also reproduced.

1. Introduction

The outcome of a collision between a nanoparticle and a surface has previously been thought to fall broadly into two categories; either the incident kinetic energy is high and the particle fragments (Pettersson & Markovic, 1993) and/or becomes embedded (Kenny et al., 2002), or the kinetic energy is sufficiently low as to not cause any significant deformation of the particle and it adheres intact to the surface (Cheng & Landman, 1995). Obviously, for the latter to happen the attraction between the particle and the surface has to be sufficient to overcome the particle's residual kinetic energy as it lifts from the surface. However, recent experiments have identified a third or intermediate pattern of behaviour whereby particles have been observed to bounce and rebound from surfaces (Partridge et al., 2004; Ayesh et al., 2010; Kuuluvainene et al., 2013; Rennecke & Weber, 2013a,b; Schöner et al., 2014); behaviour that has been found to depend on particle size, density, and velocity. In a series of recent experiments, Rennecke & Weber (2013a,b) recorded the critical velocities required for sodium chloride and silver nanoparticles (10 – 100 nm in diameter) to rebound from a surface. At the smaller end of the size range, rebound was observed once particle velocities were of the order of $\sim 20\ \mathrm{m\ s^{-1}}$. The implications associated with a better understanding of bouncing by nano-particles, range from modelling filtration processes for the removal of particles through to the development of particle deposition methods for the assembly of wires and layers (Partridge et al., 2004; Ayesh et al., 2010; Kuuluvainene et al., 2013; Krinke et al., 2002; Zarutskaya & Shapiro, 2000). With respect to the latter, a better understanding of how nanoparticles transfer from the gas phase to a surface could contribute to improvements in the methods used to fabricate nanostructured materials (Krinke et al., 2002).

In the calculations presented here, a molecular dynamics method in the form of Dissipative Particle Dynamics (DPD) has been used to model the elastic scattering of sodium chloride and silver nanoparticles from a flat rigid surface. To describe the dissipative mechanism the calculations have been performed with a single set of parameters, which although not necessarily unique, are linked in such a way as to exhibit systematic variations from which patterns of behaviour using alternative

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variables could be predicted. Many calculations on the impact of nanoparticles with surfaces have treated the former and sometimes the latter as collections of Lennard–Jones atoms (see for example, Fangli et al., 2005; Sato, & Pui, 2007; Awasthi et al., 2007, 2010; Ayesh et al., 2010; Jung et al., 2012; Kuninaka & Hayakawa, 2012; Takato & Sen, 2014). The intention here is to see if, by treating the nanoparticle as a single solid entity, it is possible to increase significantly the size and time scale over which simulations are performed, whilst still retaining an ability to reproduce experimental data.

2. Molecular dynamic method

Details of how calculations are performed to simulate the collision of a rotating particle with a surface have been given elsewhere (Stace, 2014). To model the capture of a particle by a surface there needs to be a dissipative element to the calculations, and this was also introduced previously in the form of the following equation (Stace, 2014):

$$\boldsymbol{F}_{\boldsymbol{N}}^{\boldsymbol{D}} = \left[|\boldsymbol{F}_{\boldsymbol{N}}| - \gamma M \boldsymbol{v}.\hat{\boldsymbol{z}}/2\right]\hat{\boldsymbol{z}} \tag{1}$$

where $\boldsymbol{F}_{\boldsymbol{N}}$ is the force between a particle and the surface, which in the calculations reported here is derived from a Lennard–Jones (9,3) potential (Stace, 2014), M is the mass of the particle, $\boldsymbol{v}$ is the component of the particle's velocity that is normal to the surface, γ is a damping coefficient, $\hat{\boldsymbol{z}} = \boldsymbol{z}/z$, and z is the distance between the centre of the particle and the surface. Equation (1) has been adopted from Dissipative Particle Dynamics (DPD); a technique that has been used very widely to simulate inelastic collisions in particulate and granular matter (Thompson & Grest, 1991; Groot & Warren, 1997; Kondic, 1999; Lantermann & Hänel, 2007; Kondic et al., 2012; Bai et al., 2009).

The nanoparticles being studied here are those of sodium chloride and silver and their masses have been determined from the density of the bulk material (sodium chloride at 2.16 g cm^{-3} and silver at 10.49 g cm^{-3}). Using equations derived from a Lennard–Jones (9,3) potential (Stace, 2014), γ can be expressed in the form:

$$\gamma = 2(1-\alpha)/t_{coll} \tag{2}$$

where t_{coll} is the estimated duration of a collision, and is given by $\pi\sqrt{M/(2k_{LJ})}$ (Thompson & Grest, 1991; Kondic, 1999; Kondic et al., 2012), where k_{LJ} is a force constant derived from a Lennard–Jones (9,3) potential (Stace, 2014). α is a coefficient of restitution, which provides a measure of kinetic energy loss by a particle to the surface during a collision. The parameter α is expected to be linked to the properties of the materials being studied (Kondic, 1999; Kondic et al., 2012). $\boldsymbol{F}_{\boldsymbol{N}}^{\boldsymbol{D}}$ has the form of a damped oscillator, which provides a mechanical connection between a particle and a surface, and where the damping force, γM, is proportional to the mass and velocity of the particle. In addition to systems of particles, stochastic trajectory methods with a dissipative (damped oscillator) component have also featured prominently in theoretical studies of the scattering of atoms and molecules from solid surfaces (Adelman & Garrison, 1976; Shugard & Tully, 1977).

For particles with high mass and/or velocity, the term γM in Eq. (1) can introduce a problem that is illustrated in Fig. 1, where curve 1 shows an example of how the velocity of a 40 nm diameter sodium chloride particle starting 1500 nm above a solid surface evolves as a function of time for a trajectory where the force has been calculated using Eqs. (1) and (2). The combination of high velocity and large particle mass mean that the dissipative force takes effect immediately after the trajectory has started. Therefore, even before the particle has struck the surface, its velocity has dropped by almost 0.3 ms^{-1}. Once past the collision stage, the dissipative force continues to influence the particle's velocity such that by the end of the

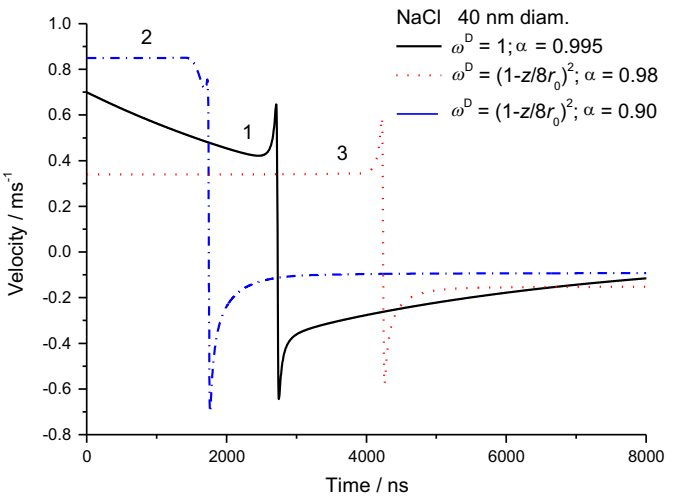

Fig. 1. Plots of velocity against time for particle trajectories involving inelastic collisions with a surface. Trajectories were run with and without the inclusion of a dissipative component and with different values assigned to α, the coefficient of restitution. The particles start at 1500 nm above the surface and their velocities change sign following a collision.