Molecular Dynamics study on size dependencies of melting dynamics in Gold thin film

R Fahdiran*, I Sugihartono, E Handoko, E Budi, A B Susila, T B Prayitno and S Sunaryo

Department of Physics, State University of Jakarta, Jalan Rawamangun Muka, Jakarta Timur, 13220, Indonesia

*riser-fahdiran@unj.ac.id

Abstract. We explore the size dependencies of melting dynamics of Gold in the form of thin film. The sizes are 4.896 nm, 7.344 nm and 9.792 nm which is comparable between nanoparticle diameter and thin film thickness. The systems are treated by increasing temperature from 300 K to 1400 K. Molecular Dynamics (MD) scheme is employed to follow the trajectories of the systems up to 20 ps. Structure factor analysis indicated that the melting is suppressed by the increasing size.

1. Introduction
Gold as a functional material could melt below melting point in the form of nanoparticle. This was reported due to size dependencies of the system [1,2]. For bulk form of Gold, the melting point is at $T_m = 1338$ K [3]. The size of the system exhibits conditional influence on the melting mechanism due to pressure oscillation.

The case for Gold nanoparticle melting due to size dependencies has been done with significant results [1,2,4]. For the case of thin film, the structural change due to melting was done for Aluminum [5]. Structural change could be used to analyze the phase transformation due to heat treatment.

This article studies the influence of thin film thickness of Gold due to heat treatment. The system is heated from room temperature to 1400 K where it is already above melting point. Structural change of the system is performed in order to visualize the transformation of each system.

2. Simulation methods
Thin film of Gold was constructed with different thicknesses employing Large-scale atomic/molecular massively parallel simulator (LAMMPS) [6]. The initial thicknesses ($d_0$) are 4.896 nm, 7.344 nm and 9.792 nm correspond to 6912 atoms, 23328 atoms and 55296 atoms, respectively. The thin film is heated in the from 300 K to 1400 K within 20 ps with same temperature increment. The interatomic potential is based on Foiles, et.al. using cut-off radius 5.55 Å [7]. All system is relaxed at temperature of 300 K and 0.0 GPa for 20 ps. Visualization of the system was done using OVITO software [8]. Virial theorem is included in the analysis of thermodynamics properties in MD scheme [9,10]. Debyer package is employed to calculate structure factor of the system [11], with method based on crystal structure theory [12-14].
3. Results and discussion

The thickness evolution as a function of increasing temperature is shown in Figure 1 (left). The system thickness is increasing as the interatomic distances between atoms is expanding due to heating. The increasing of temperature breaks the atomic bond within the system. Since the heating is gradually increases, the velocity of the atoms is also increase. Hence the atoms are coordinating with surrounding and phase transformation begin. It can be clearly seen that for smaller diameter, the extension of the thickness is the smallest. As the system transform from crystalline state to liquid state, the thickness is extended. The extension of the thickness is correlated with the pressure of the system.

The pressure profile in Figure 1 (right) indicated oscillation of the suppressed system due to thermal induced melting process. The oscillation brings the system back and forth in the surface area in thickness direction and change the thickness size. The compressive pressure (positive value) is responsible for the suppression of the system, while the tensile pressure (negative value) is capable on extending the system. The pressure oscillation is in the range of -28 to 20 MPa for our case of thickness. For higher tensile pressure, in the order of GPa, the system could form a void and at higher tensile pressure materials could be spallated as for the case of laser ablation [15,16].

![Figure 1](image1.png)

**Figure 1.** Thickness (left) and Pressure (right) evolution of Gold thin film with thickness 4.896 nm, 7.344 nm and 9.792 nm.

The oscillation pattern on pressure indicated the thermal induced melting profile. The case at higher temperature indicated that the system evolves as a function of thickness.

![Figure 2](image2.png)

**Figure 2.** Local atomic temperature at the end of simulation. Thickness: (a) 4896 nm, (b) 7.344 nm and (c) 9.792 nm. Color scale are given in K. Size are not to scale.
The thermodynamics analysis on local atomic temperature (Figure 2) at the end of simulation indicated that the system is completely melted. More than 95% of the system in each thickness has temperature around 1400 K which already above melting point of Gold. It is also seen from the combination of pressure and temperature analysis, the smaller thickness suffered for early melting which happened below melting point as can also be seen from structure factor. This was coherent with the results of nanoparticle [4].

The structure factor at the initial of melting stage is shown in Figure 3. For all thickness, at larger scattering vector ($6 \leq Q \leq 10$ Å$^{-1}$) the value is approaching unity. This indicate that the system is going for transformation to liquid state. The inset of the figure shows the broadening of structure factor peak at lower scattering vector. The broadening comes from the merging of neighboring peak which blended due to temperature increase. For lower thickness, the initial stage of melting occurred at 1152 K which is earlier than the larger thicknesses where it occurred at 1210 K and 1296 K. This confirmed the local atomic temperature point of view, where the system is completely melted at the end of simulation.

**Figure 3.** Structure Factor of thickness: (a) 4.896 nm, (b) 7.344 nm and (c) 9.792 nm at initial stage of melting.

4. **Conclusion**
Gold thin film melting are depending of the thickness of the system. The thickness influences the system to melted earlier when the size is smaller. The structure factor and local atomic temperature confirmed that at the end of simulation, the system is already in liquid state.

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**References**
[1] Qiao Z, Feng H and Zhou J 2014 Molecular dynamics simulations on the melting of gold nanoparticles *Phase Transitions* **87** 59–70
[2] Font F and Myers T G 2013 Spherically symmetric nanoparticle melting with a variable phase change temperature *J. Nanoparticle Res.* **15** 2086
[3] Kittel C 1996 *Introduction to Solid State Physics* (New York: John Wiley & Sons, Inc.)
[4] Fahdiran R, Handoko E and Sugihartono I 2019 Size dependencies on melting of Gold
nanoparticle: A Molecular Dynamics study. *Journal of Physics: Conference Series* 1402 (IOP Publishing) p 66002

[5] Fahdiran R, Handoko E, Sugihartono I, Susila A B, Budi E and Budi S 2018 Structural change of aluminum thin film in the temperature range from 300 K to 1000 K *MATEC Web of Conferences* vol 197 (EDP Sciences) p 2016

[6] Plimpton S 1995 Fast Parallel Algorithms for Short – Range Molecular Dynamics *J. Comput. Phys.* 117 1–19

[7] Foiles S M, Baskes M I and Daw M S 1986 Embedded-atom-method functions for the fcc metals Cu, Ag, Au, Ni, Pd, Pt, and their alloys *Phys. Rev. B* 33 7983

[8] Stukowski A 2009 Visualization and analysis of atomistic simulation data with OVITO–the Open Visualization Tool *Model. Simul. Mater. Sci. Eng.* 18 15012

[9] Colla T J and Urbassek H M 1997 Visualization of ke V-ion- induced spikes in metals *Radiat. Eff. Defects Solids* 142 439–47

[10] Upadhyay A K and Urbassek H M 2005 Melting and fragmentation of ultra-thin metal films due to ultrafast laser irradiation: a molecular-dynamics study *J. Phys. D. Appl. Phys.* 38 2933

[11] Wojdyr M 2016 Debyer https://github.com/wojdyr/debyer

[12] Takeshi E and Billinge S J L 2003 *Underneath The Bragg Peaks: Structural Analysis of Complex Materials* (Oxford: Pergamon Materials Series)

[13] Howell R C, Proffen T and Conradson S D 2006 Pair distribution function and structure factor of spherical particles *Phys. Rev. B* 73 94107

[14] Lin Z and Zhigilei L V 2006 Time-resolved diffraction profiles and atomic dynamics in short-pulse laser-induced structural transformations: Molecular dynamics study *Phys. Rev. B* 73 184113

[15] Fahdiran R and Urbassek H M 2015 Ultrafast laser irradiation of spherical nanoparticles: Molecular-dynamics results on fragmentation and small-angle scattering *Eur. Phys. J. D* 69 35

[16] Fahdiran R, Handoko E, Sugihartono I and Urbassek H M 2018 Laser induced ablation of aluminum nanoparticle: a molecular dynamics study *MATEC Web Conf.* 197 04004