

Research Article

Geometry Optimization Calculations for the Elasticity of Gold at High Pressure

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Received 2 May 2013; Revised 18 September 2013; Accepted 23 September 2013

Academic Editor: Augusto Deus

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We present embedded atom method based geometry optimization aspects of pressure effect on some elastic and mechanical properties of gold. During study, we determined the pressure dependency of equilibrium volume, typical cubic elastic constants, bulk modulus, shear modulus, young modulus, and elastic wave velocities of the considered metal with varying pressure between 0 GPa and 1000 GPa. Finally, we carried out a benchmark between our results and other available theoretical calculations and experimental data. The results of the study mutually agree with the previous findings and provide a deeper outlook for high pressure behavior of the studied metal.

1. Introduction

The effect of pressure on the propagation of elastic waves in materials is essential for predicting and understanding some physical properties comprising the interatomic forces, mechanical stability, phase transition mechanisms, dynamic fracture, earthquakes, and the internal structures of Earth. Unfortunately, there is not much experimental information about the elasticity of solids at high pressure, since measurement of the elastic constants is challenging under high pressure.

The two traditional experimental methods for moderate pressures are ultrasonic technique and Brillouin spectroscopy measurements. From these methods, ultrasonic methods utilize a few gigapascals (GPa) where Brillouin spectroscopy measurements are limited to 25 GPa [1]. Furthermore, gold (Au) is often used as an internal pressure calibrant for high pressure studies, because of its stable nature over wide temperature and pressure ranges. Au is also chemically inert, compressible, and displays a simple X-ray diffraction pattern. However, as reported by Matsui there has been much debate especially on the pressure-volume-temperature (P - V - T) equation of state (EOS) of Au, even at room temperature. Also, estimated pressure differences between proposed EOSs often reach more than 10% with respect to experiments

[2]. Therefore, to overcome the experimental limitations, computational researches on the high pressure characteristics of Au are particularly critical for ongoing academic studies and still current research topics [3–6].

Over the past few decades, computer simulation techniques have become an important tool in science as an aid to the interpretation of experimental data for yielding an atomic level model [7]. Notably, two popular calculation methods, namely, classical physics based molecular dynamics (MD) and quantum-mechanics based density functional theory (DFT) methods, play a major role on the materials science and condensed matter scene to get reliable results on the physical properties of metals being dependent to accurate interatomic potentials [8]. For instance, embedded atom method (EAM) and its potentials are well-known schemes for the atomistic simulations of metallic materials [9–11]. In the early 1980s, Daw and Baskes introduced EAM for calculating the ground state properties of realistic metal systems to remove the insufficiency of pairwise potentials [12], and, afterwards, researchers applied this semiempirical method extensively with various types of many improved body potentials for distinct physical problems in metallic systems. Exemplarily, Finnis-Sinclair type EAM potentials adequately explain the elastic and defect properties of base centered cubic (bcc) metals [13, 14], whereas Sutton-Chen

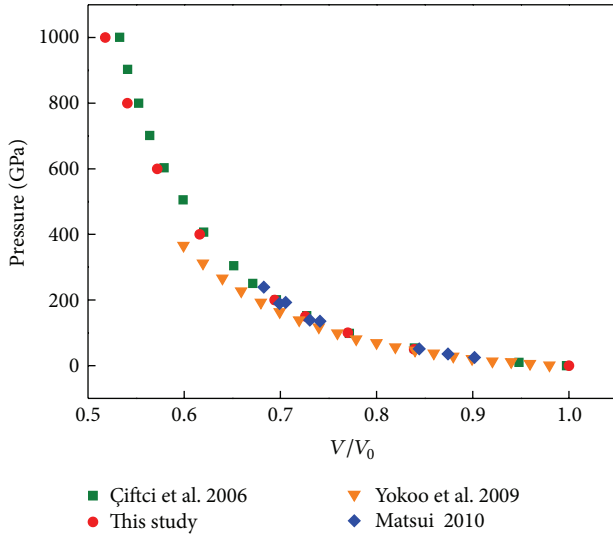


FIGURE 1: A comparison for the P - V equation of state for Au between 0 GPa and 1000 GPa.

type EAM is a proper alternative for face centered cubic (fcc) metals for atomistic calculations [15]. Noteworthy to mention here, the use of EAM with geometry optimization calculations with General Utility Lattice Program (GULP) code reveals veracious research results on some versatile features of metals such as bulk modulus, elastic constants, and elastic moduli [16, 17].

In the present study, we focus on clarifying the individual high pressure elastic constants of Au by EAM based geometry optimization calculations. The main purpose of this study is not only to provide results that can be used to evaluate quantitatively the high pressure elasticity of Au from 0 GPa up to 1000 GPa, but also to contribute to enlightening the abovementioned doubts on Au.

2. Computational Procedure

During this computational study, we performed all calculations using General Utility Lattice Program (GULP) code 4.0. This simulation code allows performing wide-range property calculations for 3D periodic solids, 2D surfaces and gas phase clusters, and so forth. Besides, this code also allows the use of two-body, three-body, four-body, six-body, and many body (EAM) potentials depending on demands of research. Most calculations in GULP code consist of the optimization of a trial structure to the local energy minimum, under given conditions of pressure and temperature. Moreover, several types of standard minimization techniques are available in GULP. The two common techniques are to optimize the related structures at constant pressure, in which all internal and cell variables are included or at constant volume, where the unit cell remains frozen [16, 17]. So, we applied a constant pressure optimization for the Au. The geometry of the studied cells was optimized by the Newton-Raphson method [21] based on the Hessian matrix calculated from the second derivatives. The Hessian matrix is recursively updated during

optimization using the Broyden-Fletcher-Goldfarb-Shanno algorithm [22–25]. We employed Sutton-Chen type EAM potential to study fcc Au metal, under periodic boundary conditions with a temperature configuration at $T = 0$ Kelvin. Then, we increased the pressure from 0 GPa to 1000 GPa in the steps of 200 GPa with a lattice constant $a_0 = 4.08$ Å for Au. Following the EAM based optimization results for Au, we particularly concentrated on some elastic parameters such as typical cubic elastic constants (C_{11} , C_{12} , and C_{44}), bulk modulus (B), shear modulus (G), young modulus (E), longitudinal sound velocity (V_L), and shear velocity (V_T) of this metal for the studied pressure range where all these physical parameters are circumstantiated in very recent study of authors [26]. At the end of our calculations we presented a benchmark between our results and earlier experimental and DFT findings of fcc Au.

3. Results and Discussion

Since it is not possible get data on the EOS of Au up to 1000 GPa experimentally, we deliberately focused on the former theoretical results from the literature. Figure 1 shows P - V diagram of Au. In Figure 1 the circle symbols represent our present calculation data where squares show the molecular dynamics results of Çiftci et al. [5]. In addition, diamonds symbols stand for Matsui's data [2], and triangles belong to Yokoo et al. calculations [27]. As it is obvious from Figure 1, our present results on the P - V diagram of Au are very close to molecular dynamics data in [5] and cover both the Matsui's findings and Yokoo's data in which Yokoo and his coworkers calculations also subsumed the electronic-free energy contribution. As a result, our data strongly supports previous findings on the EOS of Au and show significant similarities on the P - V curve.

Figure 2 represents the typical elastic constants (C_{11} , C_{12} , and C_{44}) of Au at the pressure interval between 0 GPa and 150 GPa where the authors could be able to make a comparison between current results and former experimental data and DFT values. The closed circles in Figure 2 indicate the present calculations while open-crossed circles denote experimental data of Duffy et al. [28] and squares show Greeff and Graf DFT data [29]. It is quite clear in Figure 2 that our elastic constant data, appears in the range of both experimental and DFT data except the little disagreement of C_{44} . Additionally, Figure 3 outlines the pressure dependency of the elastic constants in the 0 GPa–1000 GPa range obtained during this research. As seen in Figure 3, the calculated values of C_{11} , C_{12} , and C_{44} elastic constants are positive and exhibit a smooth increment as a function of the increasing pressure. Besides, the increment of the elastic constant C_{11} is higher than both elastic constants C_{12} and C_{44} . Physically, C_{11} describes the longitudinal elastic behavior, whereas C_{12} and C_{44} portray the off-diagonal and shear elastic characteristic of cubic crystals related with shearing, respectively. So, a longitudinal strain produces a change in volume without a change in shape. This volume change is highly related to pressure and thus reflects a large change in C_{11} . On the other hand, a transverse strain or shearing causes a change in shape

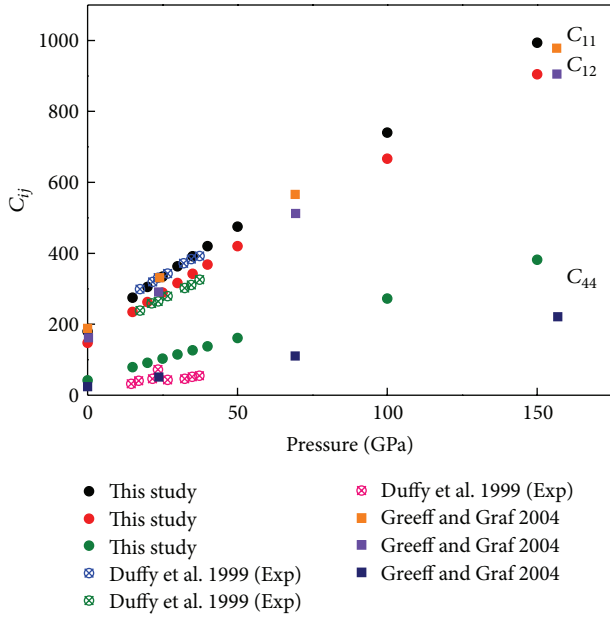


FIGURE 2: Comparing typical cubic elastic constants of Au as a function of pressure.

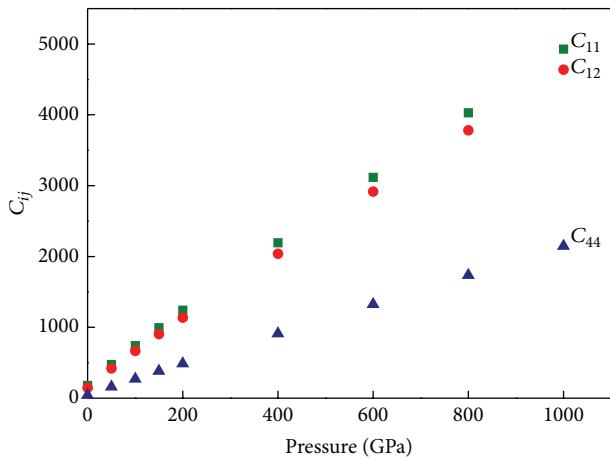


FIGURE 3: Typical cubic elastic constants of Au from 0 GPa to 1000 GPa.

without a change in volume. Therefore, C_{12} and C_{44} are less sensitive to pressure when compared with C_{11} .

Figure 4 displays the behavior of three elastic moduli (B , G , and E) of Au for the applied pressure range. From the prevalent physical definition of bulk modulus $B = \Delta P / \Delta V$, an increment for B is expected due to its direct proportionality to applied pressure. Thus, it is visible in Figure 4 that bulk modulus of Au represents a straight increment as expected. In this regard, the other elastic moduli G and E depending strictly on bulk modulus also expose similar behavior in Figure 4. The behavior of these curves is also consistent within the former results of moderating pressures in metals and alloys.

Pressure effect on the longitudinal sound velocity (V_L) and shear velocity (V_T) of Au up to 1000 GPa can be seen in

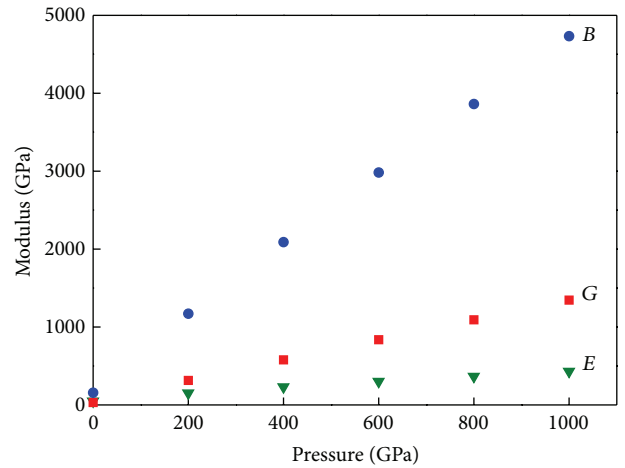


FIGURE 4: Bulk modulus, shear modulus, and young modulus of Au as a function of studied pressure range.

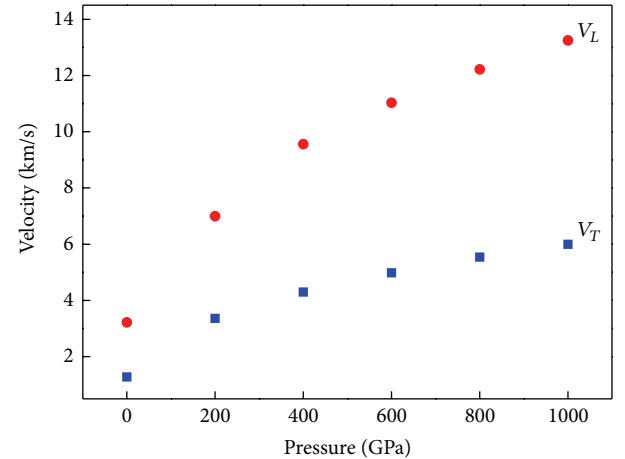


FIGURE 5: V_L and V_T behavior of Au under pressure up to 1000 GPa.

Figure 5. Under high pressure both of the sound velocities display an increment trend with the increasing of pressure; this characteristic confirms the former findings of pressure effect on both V_L and V_T [30]. When compared with each other V_L has a more uniform increasing fashion than V_T .

An additional comparison is given in Tables 1 and 2 for the present typical cubic elastic constants C_{11} , C_{12} , and C_{44} and other calculated parameters, respectively. As seen in Table 1, our obtained values for C_{11} , C_{12} , and C_{44} are reasonable when compared with the experimental measurements. On the other hand, Table 2 summarizes the present and former experimental results (originally extrapolated from 4 Kelvin measurements) of B , G , E , V_L , and V_T constants for $T = 0$ Kelvin and $P = 0$ GPa. From a more physical perspective, from Tables 1 and 2, it is easy to see the existence of traditional Born mechanical (dynamical) stability conditions ($P = 0$ GPa); $C_{11} > 0$, $C_{44} > 0$, $C_{11} - C_{12} > 0$, $C_{11} + C_{12} > 0$, and $C_{12} < B < C_{11}$ [18] for Au over the entire pressure range.

TABLE 1: Comparing previous experimental results and present calculation data for C_{11} , C_{12} , and C_{44} constants of Au.

Elastic constants	Reference [18] (Exp)	Reference [19] (Exp)	This study
C_{11} (GPa)	186.0	190.0	179.8
C_{12} (GPa)	157.0	161.0	147.7
C_{44} (GPa)	42.0	42.3	42.1

TABLE 2: Comparing earlier experimental data of B , E , G , V_L , and V_T values for Au with our results.

Elastic parameters	Reference [20] (Exp)	This study
B (GPa)	180.32	158.4
G (GPa)	29.23	31.6
E (GPa)	83.19	46.5
V_L (km/s)	3.35	3.22
V_T (km/s)	1.22	1.28

Apparently our presently calculated data shows a fair consistency again for all considered parameters and their related characteristics under high pressures.

4. Conclusion

In summary, we surveyed the effect of pressure on several mechanical and elastic properties of Au metal with embedded atom model by geometry optimization calculations. As an outcome, the obtained findings put forth reasonable results for the high pressure behavior of Au when compared with the other experimental, molecular dynamics, and density functional results. We found that the calculated P - V EOS of Au (Figure 1) up to 1000 GPa is fully consistent with the former data of available experiments and theoretical results. From the high pressure point of view, with the increasing of pressure, all the elastic constants (Figures 2 and 3) and other elastic parameters (Figures 4 and 5) increased almost linearly. This finding is also compatible with the foregoing studies (Tables 1 and 2) of the considered metal. Finally, as it is obvious from all figures and tables, EAM based geometry optimization provides satisfactory results for the concerned study parameters and a convenient alternative for atomistic modeling.

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