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Citation: The Journal of Chemical Physics **109**, 5175 (1998); doi: 10.1063/1.477133
View online: http://dx.doi.org/10.1063/1.477133
View Table of Contents: http://scitation.aip.org/content/aip/journal/jcp/109/13?ver=pdfcov
Published by the AIP Publishing

# Transport coefficients of liquid transition metals: A computer simulation study using the embedded atom model

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(Received 11 June 1998; accepted 27 July 1998)

Using the Voter and Chen version of the embedded atom model, we carried out molecular dynamics simulations to compute the diffusion constants and shear viscosities of liquid Pd, Pt, Cu, Ag, and Au as representative of single-particle and collective dynamic properties, respectively. The generally good agreement between the calculated values and available experimental data is evidence that the Voter and Chen embedded atom model allows a reliable description of the dynamic properties of liquid transition metals in spite of being derived from solid-state data and the properties of the diatomic molecule. The discrepancy between the reported experimental diffusion constant of liquid Cu and the values calculated in this work, together with the consistency of these latter, suggests that the reported experimental value may be in error. © *1998 American Institute of Physics.* [S0021-9606(98)51937-X]

During the last decade, considerable experimental and theoretical effort has been devoted to gaining a better understanding of the dynamic properties of liquid alkali metals (see, e.g., Refs. 1–8). Molecular dynamics (MD) simulations based on realistic interionic potentials, such as those derived from the empty core model pseudopotential[9] or, like the neutral pseudoatom pseudopotential,[10,11] from the first principles, have proved to be very useful in this respect, since they can afford information on properties that would be very difficult or impossible to determine experimentally, or simply have not yet been measured.

We recently used the Voter and Chen (VC) version[12] of the embedded atom model (EAM) in MD simulations to compute both single-particle and collective time-dependent properties of the face-centered-cubic (fcc) transition metal Ni in the liquid state, and thereby obtain its diffusion constant $D$ and shear viscosity $\eta$.[13] The calculated values of the dynamic and self-dynamic structure factors were in good general agreement with the results of inelastic neutron scattering experiments.[14] Moreover, although there are no experimental values of $D$ for liquid Ni with which to compare calculated values, the values obtained from our MD simulations by several independent routes were mutually consistent. Our simulations were also supported by the values of $\eta$ obtained by several different methods, which were very similar to available experimental data.[15]

The agreement between our VC EAM MD predictions for liquid Ni and available experimental data suggests that this model is capable of describing the dynamical properties of liquid transition metals. Unfortunately, extensive comparison between simulated and experimental results for the dynamic structure factors of liquid transition metals is not possible at present because very few experimental results are available for any transition metal except Ni.[14] There are, however, some experimental data for the diffusion constant and the shear viscosity, transport coefficients that are related to single-particle and collective dynamic properties, respectively. We have therefore explored the VC EAM model further by performing MD simulations to compute $D$ and $\eta$ for the fcc metals Pd, Pt, Cu, Ag, and Au in the liquid phase, and comparing these values with experimental data where possible.[15]

The VC EAM version used in this work has been described in detail elsewhere,[12,13] and will not be explained here. It differs from other EAM versions (in particular, from that of Foiles, Baskes, and Daw,[16] which has been used extensively to calculate a wide variety of bulk and surface properties of metals and alloys; see, e.g., Ref. 17) in two main ways: (a) its core–core pair interaction has a medium range attractive contribution, rather than being entirely repulsive; and (b) properties of the diatomic molecule (bond length and bond energy) were used as well as bulk properties (the elastic constants and vacancy formation energy) in fitting the embedding function and pair interaction. The inclusion of properties of the diatomic molecule among the data to which the VC EAM potential is fitted has proved to be very useful for describing the characteristic features of small metal clusters.[18–20] However, it is not obvious *a priori* that such a potential (derived from solid-state data and the properties of the diatomic molecule) can correctly describe the dynamic properties of the bulk liquid phase.

We studied Pd, Pt, Cu, Ag, and Au in liquid states that are near their corresponding melting points[21] and are characterized by the temperatures $T$ and atomic number densities $\rho$ listed in Table I. The parameters of the VC model for these metals were optimized by a least squares procedure.[19] MD simulations were performed for a system of 500 atoms following the computational procedure described in detail in Ref. 13. The diffusion constant $D$ was calculated from the mean square displacement using Einstein's formula and from the velocity autocorrelation function using the Green–Kubo relation.[22,23] Similarly, the shear viscosity was computed using the Green–Kubo and generalized Einstein formulas.[6,22]

Table I lists the computed values of $D$ and $\eta$ together

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TABLE I. Values of the diffusion constant $D$ and the shear viscosity $\eta$ for the liquid metals considered, as computed using the Green–Kubo (GK) and Einstein (E) relations at the temperatures and atomic densities specified, and available experimental data (Ref. 15).

| | | | $D$ (Å$^2$ ps$^{-1}$) | | | $\eta$ (eV ps Å$^{-3}$) | | |
|---|---|---|---|---|---|---|---|---|
| | $T$ (K) | $\rho$ (Å$^{-3}$) | GK | E | Expt. | GK | E | Expt. |
| Pd | 1853 | 0.0594 | 0.380±0.004 | 0.383±0.004 | ⋯ | 0.025±0.003 | 0.025±0.004 | ⋯ |
| Pt | 2053 | 0.0577 | 0.284±0.003 | 0.288±0.002 | ⋯ | 0.037±0.003 | 0.037±0.004 | ⋯ |
| Cu | 1423 | 0.0755 | 0.303±0.004 | 0.306±0.003 | 0.471 | 0.025±0.002 | 0.026±0.003 | 0.0248 |
| Ag | 1273 | 0.0517 | 0.260±0.004 | 0.265±0.003 | 0.281 | 0.023±0.003 | 0.024±0.003 | 0.0230 |
| Au | 1423 | 0.0525 | 0.256±0.002 | 0.259±0.002 | ⋯ | 0.026±0.003 | 0.027±0.003 | 0.0271 |

with the available experimental data.[15] The values of $D$ computed using the Green–Kubo and Einstein relations are mutually consistent, as are the values of $\eta$ obtained using the Green–Kubo and generalized Einstein formulas. For Cu, Ag, and Au the computed values of $\eta$ agree very well with the experimental data;[15] for the other metals, there are no experimental values of this transport coefficient. The only metals studied here for which an experimental value of the diffusion constant $D$ is available are Cu and Ag.[15] For Ag, our MD results agree quite well with the experimental value reported in Ref. 15, which was obtained from two different sources.[24,25] However, our values for liquid Cu differ considerably from the experimental value reported in Ref. 15, which was obtained from a single source.[26] In principle, this discrepancy could be attributed to inaccuracy of the VC EAM potential for liquid Cu. However, the VC EAM result was corroborated by our obtaining a similar value, $D=0.27$ Å$^2$ ps$^{-1}$, in MD simulations based on an $N$-body potential constructed using the second moment approximation to the tight-binding method.[27] This accord, the good general agreement between the computed values of $D$ and $\eta$ and such experimental values as are available, and the consistency of the description of dynamic properties afforded by the VC EAM for liquid Ni,[13] together suggest that the reported experimental value for the diffusion constant of liquid Cu may not be accurate, and that this property of liquid Cu should be measured again. It is perhaps also worth pointing out that, if the diffusion constants of the noble metals Cu and Ag are roughly calculated from the experimental shear viscosities using the Stokes–Einstein relation[23] with particle radii deduced from the atomic densities, the value obtained for Cu, 0.268 Å$^2$ ps$^{-1}$, differs much more widely from the reported experimental value than does the value obtained for Ag, 0.228 Å$^2$ ps$^{-1}$.

To sum up, in this work we checked the reliability of the VC EAM for describing the dynamical properties of liquid transition metals by computing the diffusion constant and the shear viscosity of Pd, Pt, Cu, Ag, and Au and comparing the results with available experimental data. The good general agreement between calculated and experimental values confirms that the VC EAM is capable of describing the dynamic properties of liquid transition metals, in spite of the liquid phase interatomic separation distributions being quite different from those of the solids, whose properties (together with the properties of the diatomic molecules) were used to fit the VC EAM potentials. Pending experimental values of $D$ or $\eta$ for the other liquid metals studied in this communication, the VC EAM MD results listed in Table I may prove useful to researchers who need these data.

This work was supported by the DGICYT, Spain (Project No. PB95-0720-C02-02) and the Xunta de Galicia (Project No. XUGA20606B96).

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