

Dynamical simulation of brittle fracture using order-N electronic-structure method

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We have developed a variational and/or perturbative order-N algorithm for large-scale electronic-structure calculations, based on the localized-state theory [1]. This algorithm was tested as molecular-dynamics simulations of upto about 34000 atoms, using a single-CPU workstation with 1GB RAMs. Such localized states are formally equivalent to a unitary transformation of eigen states and satisfy the Kohn-Sham equation with off-diagonal matrix elements ($H_{\text{eff}}\psi_j = \sum_i^{\text{occ.}} \varepsilon_{ij}\psi_i$).

As an application of our algorithm, we perform dynamical fracture simulations of silicon with external loads, where we use a sp tight-binding Hamiltonian and isolated clusters with orientation-fixed sp³ states at their surfaces. In smaller systems, we observe a satisfactory agreement of the stress values and the crack-propagating velocity among the order-N method, the exact diagonalization method, and experimental results. We also observe that the fracture occurs, when the band-gap goes to be zero. This is microscopically interpreted as bond-breaking processes or as vanishing the gap between local bonding- and antibonding- levels. Such microscopic pictures can be directly monitored in the present order-N simulation.

For investigation of the microscopic mechanism, we monitor the occupation ratio of s-orbital for each localized state: $f_s^{(j)} \equiv |\langle s|\psi_j\rangle|^2$. This quantity describes the extent of the sp hybridization and, for instance, is 1/4 for an ideal sp³ state. In general, the extent of the sp hybridization is a quantum-mechanical freedom and plays a crucial role for atomic structures. For detailed investigation of the electronic states, we also monitor the projected density of states of the localized states in the bond-breaking processes. We observe that after bond-breaking processes, the localized states on the broken bonds are stabilized with a high value of $f_s^{(j)}$ ($0.5 \leq f_s^{(j)} \leq 0.8$). This means that such states are similar to an atomic s-state, rather than a sp³ dangling-bond state. These ‘s-rich’ states seem to be transitional and tend to re-bond somewhere later in the simulation.

These observations show a general theory of the brittle fracture. The atomic s- and p- levels are $\varepsilon_s = -5.45\text{eV}$ and $\varepsilon_p = 1.2\text{eV}$, respectively. Since the atomic configuration is s²p², the averaged occupation ratio of s-orbital is the half ($f_s = 1/2$) and the electronic energy is given by $\varepsilon_{\text{atom}} \equiv (\varepsilon_s + \varepsilon_p)/2 = -2.13\text{eV}$ per electron. In perfect crystals, all the localized states form the sp³ bonds and the corresponding energy level is $\varepsilon_{\text{bulk}} = -5.2\text{eV}$, located at the weighted band center of the valence band [1]. So the bulk system is stabilized with the cohesive electronic-energy of $\Delta\varepsilon \equiv \varepsilon_{\text{atom}} - \varepsilon_{\text{bulk}} (> 0)$ per electron. Here it is important that the atomic s-level is *lower* than the bulk level ($\varepsilon_s < \varepsilon_{\text{bulk}}$). Therefore the occupation ratio (f_s) is crucial to the cohesion of the perfect crystal. In crack tips, on the other hand, the sp³-bonded network is *locally* broken and the existence of the low s level stabilizes the transitional ‘s-rich’ states. Therefore the bonds near at crack tips tend to be broken much easier than those at the bulk regions. This can be interpreted as the microscopic origin of the brittle fracture.

References

- [1] Takeo Hoshi and Takeo Fujiwara, J. Phys. Soc. Jpn, vol. 69, No.12, pp3773-3776 (2000).