Finite-temperature effect on field evaporation under laser illumination: a time-dependent first-principles study

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Atoms of solids desorb from the surface under high dc-electric-field, which is known as field evaporation. Recently, laser-assisted atom probe tomography (La-APT) [1], which utilizes field evaporation assisted by laser illumination, has been found to be a powerful technique for three-dimensional structure and chemical-composition analysis of semiconducting and insulating materials. In a previous theoretical study on evaporation from a Si cluster, it was found that dc field and laser pulse cooperatively promote evaporation [2]. However, understanding of the microscopic mechanism of laser-assisted field-evaporation from real surfaces is not enough, so the theoretical studies that include experimental conditions in addition to the effects of dc field and laser pulse on evaporation dynamics are required.

In the present study, we investigated the field-evaporation dynamics of Si (100) surface under laser illumination by a time-dependent density functional theory (TDDFT) combined with molecular dynamics (MD), which is called TDDFT-MD method [2,3]. We implemented the Fermi-Dirac distribution function in the TDDFT-MD code to take account of finite-temperature effect [4]. The objective of the present study is to reveal the finite-temperature effect on evaporation processes.

First, we determined the electronic structures of Si (100) surface. For the clean surface, the surface dangling-bond (DB) states exist in the band gap. Electrons of the surface DB states can be excited to the conduction bands when temperature is increased. However, the surface DB states from which electrons are thermally excited to the conduction bands disappear upon H termination. To check if electronic states are modified by finite-temperature effect, we calculated the imaginary part of dielectric function that reflects the excitation properties of electronic states in the low-energy region. We found that the dielectric function shows temperature dependence for clean surface but not for H-terminated surface, which is consistent with the temperature dependence of the surface DB states of clean surface.

Second, we simulated the evaporation dynamics of clean surface under dc and laser fields. For high dc-field and low-intensity laser, which are usually used in La-APT experiments, evaporation is promoted with increasing temperature, indicating that bond weakening is assisted by thermal excitation of electrons. In contrast, the finite-temperature effect on evaporation dynamics is negligible for low dc-field and high-intensity laser because laser-induced ionization becomes a main driving-force of evaporation. We also verified that finite-temperature effect on evaporation for H-terminated surface is negligibly small owing to disappearance of the surface DB states upon H termination.

To summarize, we performed finite-temperature TDDFT-MD simulation on the field-evaporation dynamics of Si (100) surface under laser illumination for the first time. The finite-temperature effect on field evaporation is found to be more apparent for the clean surface with surface DB states than the H-terminated surface.

References:
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