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Photodetachment of H^- near a Dielectric-Covered Metal Surface

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Abstract We combine a simple model potential with closed-orbit theory to study the photodetachment of H^- near a dielectric-covered metal surface. We calculate photodetachment cross sections to show that the chemisorption of a dielectric thin layer on the metal surface can significantly affect the photodetachment of negative ions. Compared to the photodetachment of hydrogen negative ions near clean metallic surfaces, our calculated cross sections show stronger oscillations, the amplitude of the oscillation growing with the layer thickness. For fixed thickness, the amplitude depends on the dielectric constant and on the metallic surface. We expect our study to guide future experimental studies of negative-ion photodetachment from dielectric-covered metallic surfaces.

Keywords Photodetachment · Dielectric thin layer · Metal surface

1 Introduction

Studies of ion-surface interactions have become increasingly important in recent years, in view of such technological applications as the development of ion sources, control of ion-wall interactions in fusion plasma, surface chemistry and analysis, reactive ion etching, and semiconductor miniaturization via thin-film deposition [1–4]. Ion-surface interactions control many surface processes, such as low-

energy electron diffraction, field emission, photoemission, and surface-barrier tunneling. Since H^- was proposed to probe adsorbate-state lifetime and charge transfer during backscattering [5], the photodetachment of H^- near a surface has attracted special interest. In 2006, Yang et al. used an elastic surface to simulate the electron-scattering interface and relied on closed-orbit theory to compute the photodetachment cross section of H^- with and without external fields [6–9]. In addition to treating the collision between the detached electron and the surface as elastic, these studies ignored the interaction potential between the electron and the surface. The solution of the resulting integrable problem yielded cross sections similar to those found in the photodetachment of H^- in a uniform electric field [10].

Real surfaces, however, are not elastic. In a real system, the dynamics of detachment will be significantly affected by the image potential of the metal surface. To account for that potential, Yang, Zhao, and Du et al. studied the photodetachment from H^- close to a metal surface [11–16], and Huang et al. studied the photodetachment from H^- confined in a metallic micro-cavity [17], in studies of photodetachment near clean metal surfaces.

What happens if the negative ion is close to a metal surface covered with a thin dielectric film? To address this question, we have recently studied the influence of a dielectric thin film on the photodetachment from H^- near a metal surface [18]. In that work, the electrostatic imaging method yielded the potential of the detached electron and allowed computation of the trajectories of the detached electrons. Unfortunately from the computational viewpoint, the thin dielectric film introduces multiple images of the detached electron and makes the image potential unwieldy.

Here, we use a simpler model potential to study the effect of a chemisorbed dielectric film upon the photodetachment from H^- . The simplification allows us to discuss the effects of different metal surfaces, the model potential depending

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not only on the dielectric constant and thickness of the dielectric layer, but also on specific properties of the metal surface [19]. We hope that our studies will help future experimental research on the effects of chemisorption upon negative-ion photodetachment from surfaces.

The paper is organized as follows. Section 2 presents the model potential acting on the detached electron and the Hamiltonian describing H^- near a dielectric-covered metal surface. We then seek the closed orbits of the detached electron and derive the photodetachment cross section on the basis of closed-orbit theory. In Section 3, we calculate the photodetachment cross section of H^- near different dielectric-covered metal surfaces. We first choose an Ag(110) metal surface and discuss the influences of the dielectric constant and layer thickness on the photodetachment cross section. We then fix the dielectric thin layer and study how different metal surfaces affect the computed cross sections. Section 4 collects the conclusions drawn from our results. Atomic units are used throughout this work unless otherwise noted.

2 Theory and Quantitative Formula

2.1 The Model Potential and the Hamiltonian

A schematic depiction of the system appears in Fig. 1. The H^- ion sits at the origin with the active electron loosely bound by a short-range, spherically symmetric potential V_b . A z -polarized laser beam causes the photodetachment. A

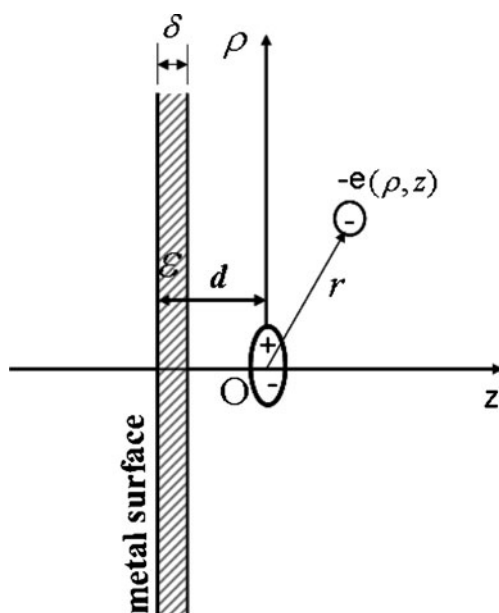


Fig. 1 Pictorial representation of the photodetachment of H^- near a dielectric-covered metal surface. The distance between the negative ion and the metal surface is d , the thickness of the dielectric layer is δ , and the dielectric constant is ε

thin dielectric layer with thickness δ and dielectric constant ε is assumed to be chemisorbed on the metal surface, which is perpendicular to the z -axis at $z=-d$. According to classical image analysis [19], the potential due to the images of the detached electron on the metal and the dielectric layer is given by the expression:

$$V_i = -\frac{1}{4|z+d-(z_0+\delta)|} \left[1 - \frac{2\delta}{(\varepsilon+1)|z+d-(z_0+\delta)|} \right]. \quad (1)$$

where z_0 is the effective image-plane location that generates the potential in low-energy electron diffraction studies, which depends on properties of the metal. The prefactor on the right-hand side of Eq. (1) is the image potential due to the metallic surface. The second term within the square brackets is a correction accounting for the thin dielectric layer.

In the cylindrical coordinate system defined in Fig. 1, our model Hamiltonian reads

$$H = \frac{1}{2} \left(P_\rho^2 + \frac{L_z^2}{\rho^2} \right) + \frac{1}{2} P_z^2 + V_i + V_0 + V_b + V_c. \quad (2)$$

where the potential $V_0 = \frac{1}{4|d-(z_0+\delta)|} \left[1 - \frac{2\delta}{(\varepsilon+1)|d-(z_0+\delta)|} \right]$, added to the right-hand side to ensure that $V(z=0)=0$, has no influence on the dynamics of photodetachment. The short-ranged potential V_c is the interaction between the detached electron and the image of the neutral hydrogen atom. The effects of the short-ranged potentials V_b and V_c of the atom and its image, respectively, can be ignored after detachment [11]. Thanks to the cylindrical symmetry, the z -component of angular momentum is conserved. For simplicity, we consider, $L_z=0$ only.

2.2 The Closed Orbit

Closed-orbit theory describes the photodetachment as follows [18, 20]. When H^- absorbs the photon energy E_{ph} , outgoing electron waves are generated, which propagate away from the hydrogen atom in all directions. With the detached electron far away from the atom, we can use a semiclassical approximation to describe the electronic motion. Since the surface potential is attractive, the waves may not propagate to infinity. Some of them turn back and return to the origin. Of all classical trajectories of the photodetached electron emanating from the origin, those bouncing back to the starting point under the image potential are called closed orbits. The returning waves traveling along the closed-orbit overlap with the outgoing source wave to generate an interference pattern in the photodetachment cross section.

It follows from the Hamiltonian equations of motion that the radial coordinate evolves freely, i.e., that

$$\rho(t) = R \sin \theta + k \sin \theta t. \quad (3)$$

Here, R is the initial spherical radius, θ is the outgoing angle of the detached electron, and $k = \sqrt{2E}$ is the momentum.

Equation (3) shows that the only closed orbit lies along the z -axis. The electron travels initially in the $+z$ -direction, but is attracted back by the image potential to the vicinity of the nucleus, to form a closed orbit. The period and action of this closed orbit can be calculated as follows. The energy of the detached electron is $E = k^2/2$, with emission angle $\theta = 0$. Its initial momentum along the z -axis is

$$p_z = \sqrt{2(E - V_i - V_0)} \quad (4)$$

From Eq. (4), we find a critical energy $E_c = V_0 = \frac{1}{4[d - (z_0 + \delta)]} \left[1 - \frac{2\delta}{(\varepsilon + 1)[d - (z_0 + \delta)]} \right]$ controlling the dynamics of the photodetached electron. If the electron energy is less than E_c , the electron reaches the highest point with $p_z = 0$. The maximum distance z_{\max} traveled by the electron along the z -axis is then given by the expression

$$z_{\max} = \frac{1 + \sqrt{1 + 16c_0(E - V_0)}}{8(V_0 - E)} - d_0$$

Here, $c_0 = \frac{2\delta}{\varepsilon + 1}$, $d_0 = d - z_0 - \delta$.

The period and the action of the closed orbit along the z -axis can be obtained from the following equalities

$$T = 2 \int_0^{z_{\max}} \frac{1}{p_z} dz \quad (0 < E \leq E_c) \quad (5)$$

and

$$S = 2 \int_0^{z_{\max}} p_z dz \quad (0 < E \leq E_c), \quad (6)$$

respectively.

2.3 Photodetachment Cross Section

According to closed-orbit theory, the photodetachment cross section of H^- near a dielectric-covered metal surface can be split into two parts:

$$\sigma(E) = \sigma_0(E) + \sigma^{\text{osc}}(E) \quad (7)$$

The first part is a smooth background that arises even in the absence of any external fields or surfaces [10]:

$$\sigma_0(E) = \frac{16\sqrt{2}\pi^2 B^2 E \sqrt{E}}{(E_b + E)^3} \quad (8)$$

The second term on the right-hand side of Eq. (7) is oscillatory and corresponds to the contribution of the returning wave. If the electron energy is less than E_c , we have to deal with a single closed orbit, along the z -direction as explained. Following the derivation of the photodetachment cross section for H^- near a metal surface [11, 18], we come to the expression

$$\sigma^{\text{osc}}(E) = \frac{8\pi^2 B^2 \sqrt{2E}}{c(E_b + E)^3 T} \cos(S) \quad (0 < E \leq E_c), \quad (9)$$

where T and S are the period and action of the orbit, given by Eqs. (5) and (6); $E_b = k_b^2/2$ is the binding energy, approximately equal to 0.754 eV; the normalization constant B is equal to 0.31552; c is the speed of light; and E is the energy of the detached electron.

If the electron energy exceeds E_c , the electron can travel to infinity. In the absence of closed orbits, the photodetachment cross section is insensitive to the surface. The total photodetachment cross section is therefore described by the expression [18]

$$\sigma(E) = \begin{cases} \sigma_0(E) + \frac{8\pi^2 B^2 \sqrt{2E}}{c(E_b + E)^3 T} \cos(S), & 0 < E \leq E_c \\ \sigma_0(E), & E > E_c \end{cases} \quad (10)$$

3 Numerical Results and Discussions

In the following calculation, we fix the distance between the negative ion and the metal surface $d = 60$ a.u. and study the effects of the chemisorbed dielectric thin layer on the photodetachment cross section. First, we choose Ag(110) as the metal surface. The distance parameter in the image potential V_i is then $z_0 = -2.35$ a.u. [19]. The resulting cross sections are shown in Fig. 2 as functions of the photon energy, the solid (dashed) line representing the photodetachment cross section for H^- near a clean Ag(110) surface [a Ag(110) surface

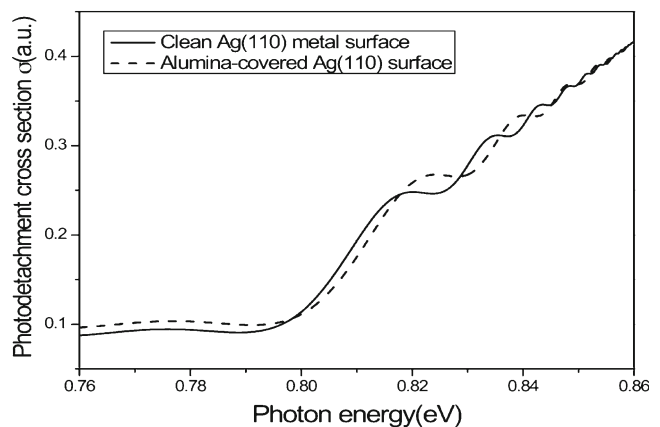


Fig. 2 Comparison of the photodetachment cross section of H^- near the clean Ag(110) metal surface with the alumina-covered Ag(110) metal surface. The thickness of alumina layer $\delta = 5.0$ a.u. and the distance between H^- and the metal surface is $d = 60$ a.u.

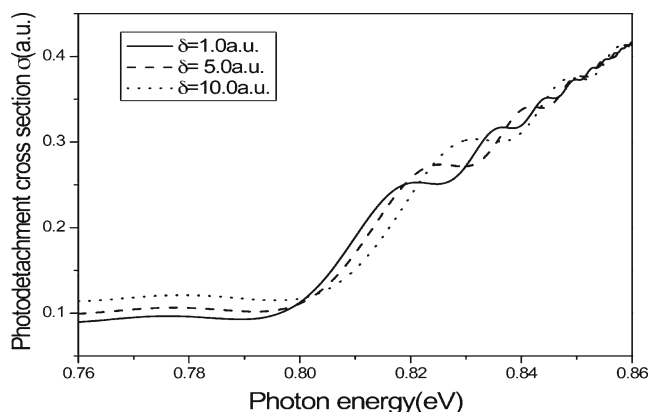


Fig. 3 Photodetachment cross section of H^- near the butyl chloride-covered Ag(110) metal surface. The distance between H^- and the metal surface is $d=60$ a.u., and the dielectric constant $\varepsilon=9.6$. The thickness of the butyl chloride layer is varied between 1.0 and 10.0 a.u.

covered by a $\delta=5.0$ a.u. layer of alumina]. The amplitude of the cross-section oscillations for the alumina-covered Ag(110)

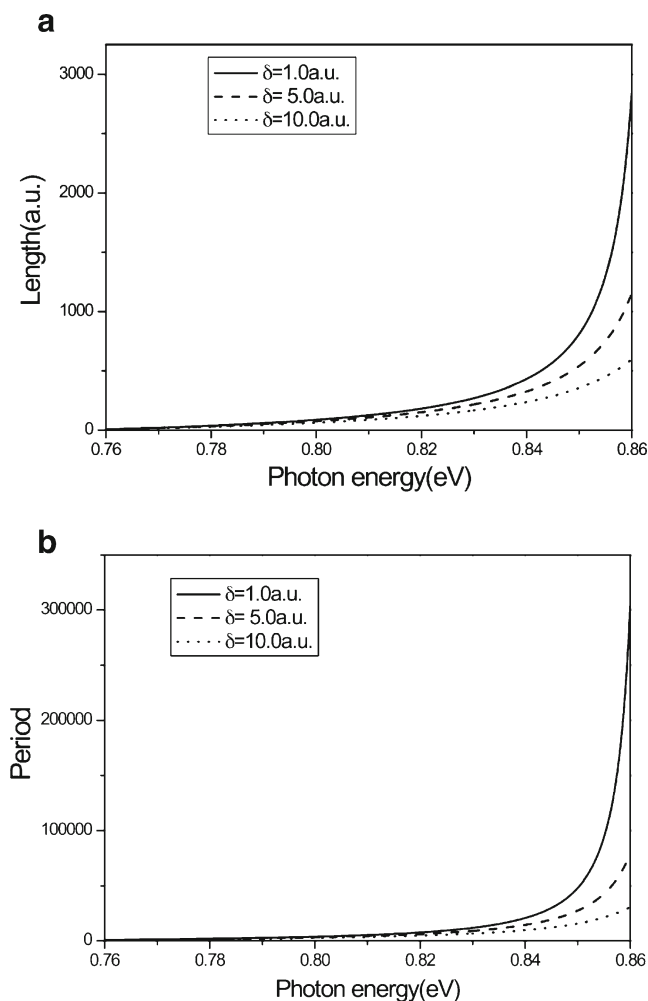


Fig. 4 Length and period of the photo-detached electron near a butyl chloride-covered Ag(110) metal surface. **a** is the length and **b** is the period. The distance between H^- and the metal surface is $d=60$ a.u., and the thickness of the butyl chloride layer is varied between 1.0 and 10.0 a.u.

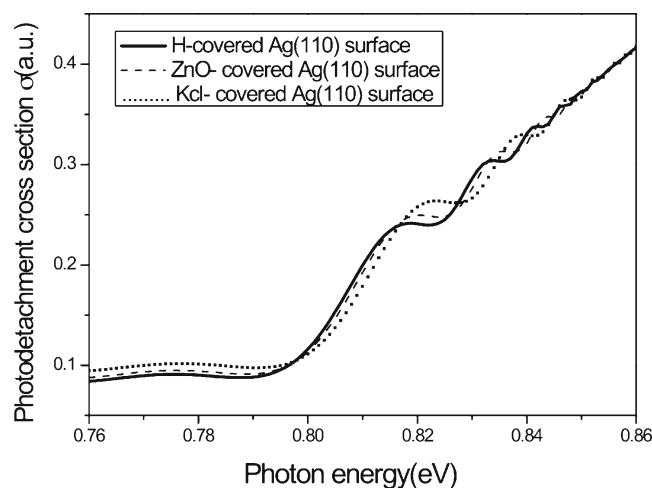


Fig. 5 Photodetachment cross section of H^- near a Ag(110) metal surface covered with a different dielectric thin layer. The distance between H^- and the metal surface is $d=60$ a.u., and the thickness of the layer is $\delta=5.0$ a.u. The dielectric constants of H, ZnO, and KCl are $\varepsilon=1.00026$, 1.6, and 4.6, respectively

surface is larger than the amplitude for the clean Ag(110) metal surface, an indication that the dielectric layer significantly affects the photodetachment.

In Fig. 3, we fix the dielectric constant and vary the thickness of the layer chemisorbed on a Ag(110) surface. The dielectric is now butyl chloride, with the dielectric constant $\varepsilon=9.6$, and the thickness varies between 1.0 and ~ 10.0 a.u. The amplitudes of the oscillations grow with the thickness of the dielectric layer, a result that was predicted in ref. [18] and is transparent to physical interpretation. As the layer becomes thicker, the attractive potential due to the images of the detached electron on the dielectric-covered metal surface becomes stronger; therefore, the length and period of the closed orbit

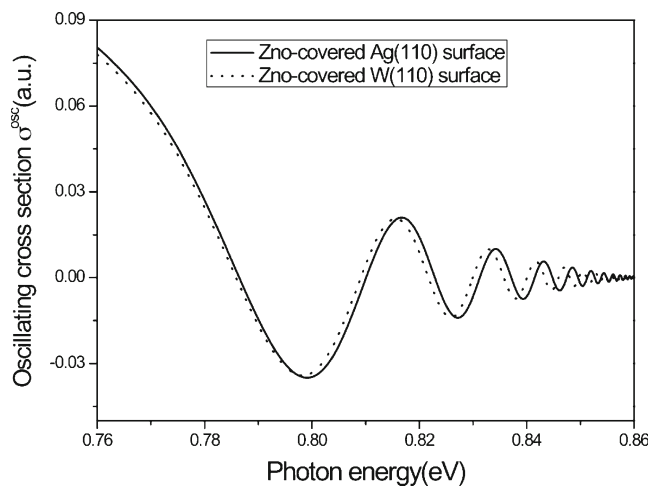


Fig. 6 Oscillating photodetachment cross section of H^- near a ZnO-covered metal surface. The *solid line* is the ZnO-covered Ag(110) surface, and the *dashed line* is the ZnO-covered W(110) surface. The distance between the H^- and the surface is $d=60$ a.u., and the thickness of the dielectric thin layer is $\delta=2.0$ a.u.

shrink. Closed-orbit theory associates the oscillation in the photodetachment cross section with the interference between the outgoing and returning wave. The smaller the period of the closed orbit, the stronger the interference between the outgoing electrons and those pulled back by the surface, and hence the greater the contribution to the oscillatory cross section. Figure 4 shows the orbital length and period as functions of the photon energy for three-layer thicknesses and confirms that the length and period decrease as the layer thickens.

Next, we fix the thickness of the dielectric layer at $\delta=5.0$ a.u. and study the influence of different dielectrics chemisorbed on Ag(100) on the photodetachment cross section. We choose layers of hydrogen, ZnO, and KCl. The results are shown in Fig. 5. The amplitude of the cross-section oscillations grows with the dielectric constant.

Finally, we study the oscillating photodetachment cross section of H^- near different metal surfaces covered with the same ZnO dielectric thin layer. We compare Ag(110) with W(110). The results show that the oscillatory part of the cross section near the ZnO-covered Ag(110) surface is a little larger than the ZnO-covered W(110) surface (Fig. 6). The difference reflects the distinct image-charge parameters: for the Ag(110) surface, the parameter in the image potential is $z_0=-2.35$ a.u., while for W(110), the parameter is $z_0=-3.28$ a.u. [19]. For fixed dielectric layer, the attractive potential of the Ag(110) surface is slightly stronger than the potential for W(110). The stronger the attractive potential, the larger the electrostatic force acting on the detached electron. Thus, the electron waves can be returned back to the origin in a short period of time, and the interference effect between the returning waves and outgoing waves becomes strong. Therefore, the oscillatory pattern in the energy dependence of the cross section becomes more obvious.

4 Conclusions

In summary, using a simple image potential and closed-orbit theory, we have studied the influence of a chemisorbed dielectric thin layer on the photodetachment of H^- near a metal surface.

The results indicate that for a given dielectric, the amplitude of the oscillations in the energy dependence of the cross section grows. If the thickness of the layer is kept fixed, the oscillatory pattern depends on the composition of the dielectric layer. Different metal surfaces covered with the same dielectric layer were shown to yield distinct energy dependences for the cross sections, but the difference is small. Our method applies equally well to other chemisorption systems and may help future experimental research on the photodetachment of negative ions in the vicinity of surfaces and micro-cavities.

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