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Photodetachment of H^- near the band-gap surfaces of the metal

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ABSTRACT

We investigate the photodetachment of H^- near the band-gap metal surface based on the semiclassical closed orbit theory. The result shows that the staircase structure of the photodetachment cross section is strongly influenced by the lifetime of the detached electron near the metal surface as the photon energy increases. When the detached electron energy is between the threshold and the image potential, the photodetachment cross section presents some more sophisticated structure in each step.

1. Introduction

H^- near the metal surface, as a typical example of the surface states in surface science, has gained much attention in recent years, which presents quite different properties from the case in free space. Our previous study has shown that the metal surface has significant influence on the photodetachment process [1]. In that case, cross section of photodetachment undergoes a transition from a staircase structure to a smooth oscillation when the incoming photon energy increases. The staircase structure is corresponding to Coulomb-like image states. This is because an electron at a distance z in the vicinity of a metal surface experiences a Coulomb-type attractive force $F(z) = -e^2/(2z)^2$ originating from the interaction between an external electron and the induced polarization charge at a distance z inside the metal [2,3], thus the electron is trapped in the potential well which is formed by the Coulomb-type attractive image potential barrier $V(z) = -e^2/4z$. In the investigation, the lifetime of the detached electron was assumed to be infinite. However, for a real system, the lifetimes of the image states are finite. Therefore, the photodetachment cross section of H^- near the metal surface is not only related to the photon energy and the distance to the surface [4], but also related to the image state lifetime of the metal surface. Fortunately, now the lifetimes of some image states of kinds of metal surfaces have been evaluated theoretically and experimentally, and found the lifetimes vary between a large range and depend on the type of metals, crystal orientation and the atom adsorption of surface [5].

With the convenience, in the present work, we investigate the photodetachment of H^- near the different band gap metal surfaces with considering the influence of the lifetimes of different image states, which is beneficial to compare with the real systems. As an extension of our previous work, the semiclassical closed orbit theory [6,7] is used to interpret the different structure of the photodetachment cross section of H^- near the gap metal surfaces. In the present study, we explore a finite lifetime system. We find that the photodetachment cross section is significantly influenced by the image state lifetime of metal surface when the photodetached electronic energy is higher than the threshold and lower than the image potential barrier.

The paper is organized as follows. In Section 2, an analytical formulation of the image-potential state lifetime of the metal surfaces is obtained. Then we systematically discuss the photodetachment cross section of H^- near the metal surface considering the influence of lifetime of the image state based on the closed orbits theory in Section 3. Finally, Section 4 gives the numerical simulation and discussions. We use atomic units in our whole work excepting designated.

2. The finite lifetime of the image states

The lifetime of the image states varies in an appropriate range and depends on the type of metal, crystal orientation and atom adsorption of metal surface [5]. Lifetime is controlled by the interaction of the excited electron with other quasiparticles, such as single-particle excitations-electrons and holes or collective

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excitations-phonons, magnons, and plasmons. These interactions change the energy and momentum of the excited electron (inelastic scattering of the electron on other quasiparticles), whereas a scattering of the electron on defects causes only a change of momentum (elastic scattering of the electron) [5]. The finite lifetime of excited states can be obtained by self-energy [8] from the knowledge of the imaginary part of the electron.

By using two-photon photoemission (2PPE) technique and two-photon photoemission in the time-resolved mode (TR-2PPE), the binding energies of the image states on metal surfaces have been extensively measured [9–12], So that the dynamical evolution of excited electrons in metals have been found [13,14], it allows one to obtain detailed information on the decay properties of surface and image-potential states in the time domain at first, then it is ready to study the lifetime experimentally. In recent years, the development of experimental techniques based on femtosecond lasers has made it more possible to study the dynamics of electronic excitations in a wide variety of systems, including clusters, electronic excitations with energies of a few electronic volts usually decay in a time scale of the order of femtoseconds. On the other hand, a theoretical methods is employed to study electron and hole dynamics in metals for calculations in which inelastic e–e scattering is a self-energy formalism of GW approximation of many-body theory [15], finally the excited electron lifetimes can be illustrated by using recent calculation results. The lifetimes in experiment agree with the theory approximation perfectly.

Based on the conclusion in experiment and theory, an order that the lifetime of the image-potential states on metal surfaces increases rapidly with n as n^3 has been obtained (n is the quantum number of image-potential states), i.e $T_{\max} = An^3$ [2], of which A is constant to a certain metal, we can use the known date to calculate it approximately [5]. The calculation result shows a regular behavior: the line width decreases with the increase of the quantum number n. Moreover, the line width of the resonance image states on all the surfaces studied obeys the n^3 scaling established theoretically [2,16,17] for the gap image states and confirmed experimentally in TR-2PPE measurements [18]. The lifetime of several metal has been shown theoretically or experimentally in Table 1 [5].

As usual the binding energy is associated with n [2], and it can be given as

$$E_{\perp} = -\frac{1}{32n^2} \tag{1}$$

Meanwhile, the lifetime of the image state is that

$$T_{\max} = An^3 \tag{2}$$

if we use $E_{\perp}$ instead of n, it can be written as following

$$T_{\max} = A\left(\frac{-1}{32E_{\perp}}\right)^{3/2} \tag{3}$$

We have to point out that the n^3 law is valid exactly for the classical image potential and a jellium model. On a real metal, the potential close to the surface deviates from $1/z$ and the energy of the different states relative to the bulk bands is not the same. For the higher n states these effects will generally have a small influence but for the $n = 1$ state they can alter the lifetime substantially. When the escaping energy of the photodetachment electron is more than $1/4z_0$ above the threshold, the cross section becomes oscillatory weakly [19], because the photodetachment cross section is not affected by the metal surface according to the closed orbit theory. Therefore when the lifetime is used in the calculate, The expressing of the binding energy is as following

$$E_{\perp} = E - \frac{1}{4z_0} \tag{4}$$

E is the excited electron energy. We only consider the situation that the detached electron energy is higher than the threshold and lower than the image potential, Above all, the lifetime of the image states can finally be written as

$$T_{\max} = A\left(\frac{1}{32(1/4z_0 - E)}\right)^{3/2} \tag{5}$$

Thus the lifetime of the image states has made in touch with the excited electron energy, we can approximately calculate the quotient A of different metals from the theory and experiment which has been obtained [5], The data is given in Table 1. The lifetime of $n = 1, 2, 3\ldots$ image potential on a certain metal can be used to calculate A approximately, then we can express the lifetime by A for a certain metal. We fit all the experimental lifetimes to Eq. (2). The estimated A is the average of all image states. For example, A of Cu(111) is 8 fs, Cu(100) is 22 fs, Ag(100) is 29 fs, and so on.

Table 1
lifetime measured and calculated for Image-Potential States on Clean Metal Suefaces: calcd = calculated; exp = experimental [17].

Metal	Ref	Lifetime (fs)				
		$n = 1$	$n = 2$	$n = 3$	$n = 4$	$n = 5$
Cu(100)	exp	40 ± 6	120 ± 15	300 ± 20	630	1200
	calcd	38	168	480		
Cu(111)	exp	18 ± 5	14 ± 3	40 ± 6		
	calcd	23				
Ag(100)	exp	55 ± 5	160 ± 10	360 ± 15		
	calcd	55	219	658		
Ag(111)	exp	32 ± 10	≤20			
	calcd	20				
Au(100)	calcd	22	93	264		
Fe(110)	exp	16 ± 2↑, 11 ± 2↓				
	calcd	31				
Ni(100)	exp	16 ± 5				
	calcd	20	101	187		
Ni(111)	exp	7 ± 3				
	calcd	15				
Co(0001)	calcd	37				

3. The photodetachment cross section

In order to investigate the dynamics of the detached electron in the photodetachment process of H^- near the metal surface, we set the system in a cylindrical coordinates (ρ,z,ϕ) as follows: a negative hydrogen ion is at the origin and a metal surface is at $-z_0$. When a z-polarized laser is shined on the ion, the valence electron absorbs the photon energy and escapes from the ionic core. The binding energy $E_b = k_b^2/2$ of the electron is approximately 0.754 eV, where k_b is associate with the initial wave function $\varphi_i = C\exp(-k_b r)/r$, C is 0.31552 which is a constant. Then, a positive charge is induced by the photodetached electron in the metal at the symmetrical position, so the photodetached electron experiences an attractive force $F(z) = -e^2/(2z)^2$ and it is trapped in the image potential $V(z) = -e^2/4z$, when the energy of the detached electron is lower than image potential. If the lifetime of the electron is infinite and the electronic energy is not dissipative, it will oscillate in the trap forever. This is the idealistic case we discussed in our previous