

# Electron–phonon coupling and lifetimes of excited surface states

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## Abstract

Many important chemical and physical phenomena are influenced by inherent dissipative processes, which involve energy transfer between the electrons (electron–electron scattering) and between the electrons and the ionic motion (electron–phonon scattering). The non-adiabatic interaction between the valence electrons and the ion motion in a solid reveals the break down of the Born–Oppenheimer approximation. To pin down the influence of the electron and phonon structure on these scattering processes the two-dimensional surface states are ideal both from an experimental and theoretical point of view. Several experimental techniques presently in use are able to give information about the lifetime of an excited electron or hole in the surface state band. With help from advanced theoretical calculations it is possible to sort out the relative importance of the electron–electron and electron–phonon scattering processes responsible for the quenching of the excitation and to point out the key parameters of the electron and phonon structure.

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## 1. Introduction

In the spirit of the Born–Oppenheimer approximation we consider that the electrons respond

instantaneously to the motion of the ions in a metal. The argument is that the electrons are light in comparison and move rapidly. However, the electrons do not move infinitely fast and thus if an ion is displaced the electron system will for a short but finite time be in an excited state. Is this something we have to be worried about? Well, it depends on what we are concerned with. To obtain

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information about crystal structure and in general also the electron structure—as long as we are interested in ground state properties such as electron densities and density of states—the Born–Oppenheimer picture works fine.

In surface science, adsorption, desorption and reaction often proceeds via an intermediate electronically excited state. This workshop is concerned with desorption induced by electronic transitions, DIET. From a theoretical point of view DIET is connected to two fundamental tasks, to calculate from *first principles* (1) Potential energy surfaces (PES) of excited states and (2) energy dissipation due to non-adiabatic coupling between nuclear motion and the electron density. Progress in the field of *ab initio* studies of excited state PES will for example, be presented by Roling et al. at this meeting. Our contribution is an attempt to obtain further understanding of the non-adiabatic coupling and to point out relevant calculation schemes and experiments for this purpose.

Having in mind a typical DIET process, one should ask the question if the lifetime of the excited state is compatible with the time it takes for the adsorbate to leave the surface, or at least long enough that a significant fraction will. For this reason it is important to find the parameters that determine the lifetime of the excitation. The way we experimentally can get a grip of the non-adiabatic coupling between the ionic motion and the conduction electrons is by performing a well defined excitation of the system, either by a vibrational excitation or an electronic excitation and then monitoring, by some means, the decay time to the ground state. In the first case, considering for instance a molecule adsorbed on a metal surface, the observed vibrational line width,  $\Gamma$ , in an electron energy loss spectroscopy (EELS) or infrared spectroscopy (IRS) measurement give information about the vibrational lifetime,  $\tau = \hbar/\Gamma$ . Considering an electronic excitation, angular resolved photoemission spectroscopy (ARPES) and lately also scanning tunneling spectroscopy (STM) are able to give information about the lifetime of an excited one-electron state.

In this paper we will discuss the decay of a hole state in a surface state band. The reason why there

has been such a focus on surface states in this context is rather obvious as from an experimental and theoretical point of view these states are excellent for benchmark studies. The influence of the solid on the photoelectron is minimized and the surface states, two-dimensional in character, are for many systems well isolated from bulk bands in a large fraction of the surface Brillouin zone. For example, the surface state wave function for Cu(111) decays exponentially in the direction toward the bulk within about 4 atomic layers [1].

Today, several ways to experimentally investigate lifetimes of surface localized electron states have been presented, ARPES [2–8], Two-photon photoemission (2PPE) techniques [9,10], Time resolved two-photon photoemission technique for direct determination of excitation lifetimes [11–22] and different methods based on STM [23–29].

## 2. Inherent scattering processes

The decay channel of the hole in the surface state band via excitations of secondary electrons, the so-called electron–electron (e–e) interaction has been investigated theoretically in detail for many systems [1,30,31]. In Fig. 1 an example of

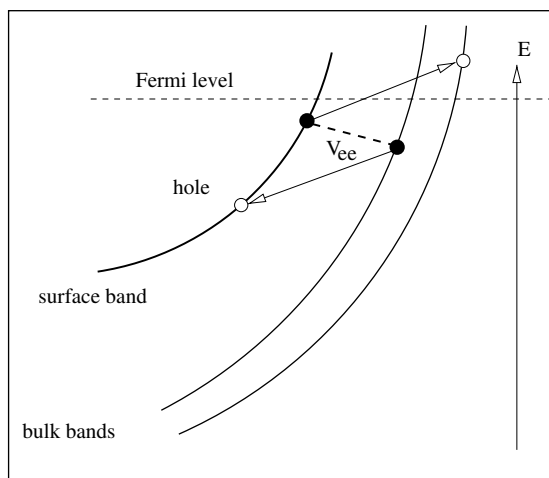


Fig. 1. Schematic illustration of the energy and parallel momentum conservation in an electron–electron scattering event, which leads to filling of the photohole in the surface state band.  $V_{ee}$  denotes the screened electron–electron interaction.

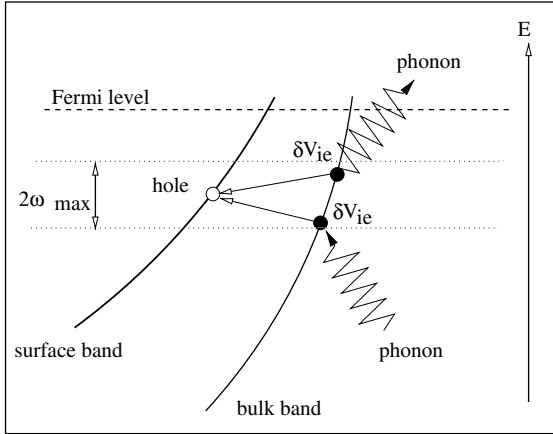


Fig. 2. Schematic illustration of the energy conservation in electron–phonon scattering events, phonon absorption and emission, which leads to filling of the photohole in the surface state band.  $\delta V_{ie}$  denotes the change of the screened ion–electron interaction  $V_{ie}$  due to the vibrational motion.

an e–e scattering event is shown schematically. The figure illustrate an event when a bulk electron, due to the screened interaction  $V_{ee}$  with another electron, is scattered into the hole of the surface state band and simultaneously an electron is scattered from the occupied part of the surface state band into the unoccupied part of a bulk band. All types of scattering events, which conserve energy and momentum have to be considered. The non-adiabatic ion–electron interaction, or as more often phrased, the electron–phonon coupling e–p, is illustrated in Fig. 2. In this case, what drives the scattering of an electron into the hole is the field, generated by the vibrating ions giving rise to a change in the screened ion–electron interaction,  $\delta V_{ie}$ . In this case we have to sum up all events including phonon absorption and emission, which fulfill energy and momentum conservation.

### 3. Qualitative discussion of non-adiabatic ion–electron coupling

Before giving the detailed expressions for the phonon induced lifetime broadening  $\Gamma_{ep}$  we can discuss qualitatively the expected binding energy and temperature dependence.

#### 3.1. Binding energy dependence

For the e–p scattering the energy window of scattering events is twice the maximum phonon energy,  $\omega_{max}$ , typically 40–60 meV (indicated in Fig. 2). Thus we should expect that the phonon induced lifetime broadening  $\Gamma_{ep}$  versus hole binding energy—energy below Fermi level—essentially will saturate for binding energies exceeding the  $\omega_{max}$ .

#### 3.2. Temperature dependence

For a qualitative discussion we can start off with a simple one-dimensional model. Consider a single ion in an electron gas. If the ion is displaced a typical vibrational amplitude from the equilibrium position ( $R = 0$ ) we can expand the screened ion–electron interaction potential  $V_{ie}$  to first order and the Hamiltonian is written

$$H = H_0 + \delta V_{ie} = H_0 + \frac{\partial V_{ie}(x)}{\partial R} R, \quad (1)$$

where  $x$  is the electron coordinate and the derivative of the ion–electron potential is evaluated at the equilibrium position ( $R = 0$ ). The solutions to  $H_0$  are the adiabatic states,  $|I, n\rangle = \Psi_I(R)\phi_n(R, x)$ , where  $\Psi_I(R)$  is the wave function of the ion and  $\phi_n(R, x)$  the electron wave function, which depends parametrically on the ion position. Consider now the initial state where an electron is photoemitted, leaving behind a hole. Applying the “golden-rule”, first order time dependent perturbation theory, we can calculate the electron scattering rate into the hole state  $n = n_i$ , assuming the ion is initially in the vibrational state  $I$ .

$$\begin{aligned} \Gamma_{I, n_i} &= 2\pi \sum_{J, n} \left| \left\langle J, n \left| \frac{\partial V_{ie}(x)}{\partial R} R \right| I, n_i \right\rangle \right|^2 \\ &\quad \times \delta(\hbar\omega_{IJ} - \hbar\omega_{nn_i}) \\ &= \sum_J |\langle I|R|J\rangle|^2 \sum_n \left| \left\langle n \left| \frac{\partial V_{ie}(x)}{\partial R} \right| n_i \right\rangle \right|^2 \\ &\quad \times \delta(\hbar\omega_{IJ} - \hbar\omega_{nn_i}), \end{aligned} \quad (2)$$

where  $\hbar\omega_{IJ} = E_I - E_J$  and  $\hbar\omega_{nm} = \epsilon_n - \epsilon_m$ . We assume the vibrating ion to be a harmonic oscillator with a single frequency  $\Omega_0$ . If we now consider for

example, processes involving absorption of one single vibrational quanta  $\hbar\Omega_0$ , we have  $J = I + 1$ . Having the oscillating ion in thermal equilibrium with a temperature  $T$  we take the expectation value of the vibrational quantum number  $I$ ,  $\langle I \rangle = n_B(T)$  where  $n_B(T) = [\exp(\hbar\Omega_0/k_B) - 1]^{-1}$ . This will give us the essential part of the temperature dependence. Later on we will also include the temperature dependent Fermi factors of the electrons, but unless temperature are very high or we are concerned with states very close to the Fermi level these are of no importance. So, assuming that the binding energy of the hole state exceeds  $\hbar\Omega_0$  and summing up electron scattering events, which involves absorption of a vibration quanta we have

$$\Gamma_{n_i} = 2\pi \frac{\hbar}{2M\Omega_0} [1 + n_B(T)] \times \sum_n \left\langle \left| n \left| \frac{\partial V_{ic}(x)}{\partial R} \right| n_i \right\rangle \right|^2 \delta(\hbar\Omega_0 - \hbar\omega_{ni}), \quad (3)$$

where  $M$  is the ion mass. In the case of emission of a vibrational quanta the temperature factor  $1 + n_B(T)$  is replaced by  $n_B(T)$ . Thus we can deduce the qualitative temperature dependence. In the limit  $k_B T \ll \hbar\Omega_0$  we have  $\Gamma_{n_i} \sim 1$  and for high temperatures  $k_B T \gg \hbar\Omega_0$  we obtain  $\Gamma_{n_i} \sim T$ . Thus for low temperatures  $\Gamma_{n_i}$  takes a finite value while at high temperatures it becomes linear with temperature. This qualitative behavior can be seen in Fig. 4.

Now, we proceed and consider a solid terminated by a surface. We will apply the slab model, which means we consider a finite number of atomic layers and periodic boundary conditions in the lateral directions. From the point of view of vibrational properties we now have coupled vibrating ions, which yields collective vibrational modes, phonons. Applying a simple single force constant model, we consider springs attached between neighboring ions and then we calculate the eigenvectors (phonon polarization vectors) and eigenvalues (phonon dispersion relation) of the dynamical matrix. The phonons will be characterized by a mode index  $\nu$  and a momentum,  $\vec{q}$  parallel to the atomic layers of the slab. In a metal slab model we have one-electron wave functions and energies

$$\psi_{n,\vec{k}_{\parallel}}(z, \vec{x}) = \frac{1}{\sqrt{A}} \phi_n(z) e^{i\vec{k}_{\parallel} \cdot \vec{x}},$$

$$\epsilon_n(\vec{k}_{\parallel}) = \epsilon_n^0 + \hbar^2 k_{\parallel}^2 / 2m_n, \quad (4)$$

where  $n$  is the band index,  $\vec{k}_{\parallel}$  the momentum parallel to the surface and  $A$  the surface area. In the following we will suppress the  $\parallel$  index. The  $z$  coordinate is along the surface normal and  $\vec{x}$  in the surface plane.

#### 4. Phonon induced lifetime broadening

Within our slab model we now outline the equations for calculating the phonon induced lifetime broadening of a hole in a surface state band. Summing up the contributions from phonon emission, corresponding to Eq. (3) in the case of an isolated vibrating ion, and phonon absorption and also taking into account the temperature dependence of the occupancy of the electron states,  $f$ , we have

$$\Gamma_{\text{ep}}(\omega, \vec{k}_i) = 2\pi \sum_{\nu, \vec{q}, f} |g_{i,f}^{\nu}(\vec{q})|^2 \{ [1 + n_B(\hbar\omega_{\nu}) - f(\epsilon_{f,\vec{k}_i - \vec{q}})] \delta(\hbar\omega - \epsilon_{f,\vec{k}_i - \vec{q}} - \hbar\omega_{\nu}) + [n_B(\hbar\omega_{\nu}) + f(\epsilon_{f,\vec{k}_i - \vec{q}})] \delta(\hbar\omega - \epsilon_{f,\vec{k}_i - \vec{q}} + \hbar\omega_{\nu}) \}. \quad (5)$$

Eq. (5) is obtained from a Master type of equation where the time dependent filling of the initially empty hole state is determined by the scattering rate into the hole minus the scattering rate out of the hole [38]. The so-called electron–phonon coupling function squared is given by

$$|g_{i,f}^{\nu}(\vec{q})|^2 = \frac{\hbar}{2MN\omega_{\nu}(\vec{q})} \times \left\langle \left| f \left| \sum_{\mu} \vec{\epsilon}_{\vec{q}\nu}(\vec{R}_{\mu}) \cdot \vec{\nabla}_{\vec{R}_{\mu}} \tilde{V}_q^{\mu} \right| i \right\rangle \right|^2. \quad (6)$$

Comparing Eqs. (5) and (6) and Eq. (3) we note the similar structure. We consider the static screening of the electron–ion potential, since the phonon frequencies are in general small in comparison with the energies of the scattered electrons. The coupling function in Eq. (6) is the result of the standard first order expansion of the screened

electron–ion potential,  $\tilde{V}_{q,\mu}^\mu$ , with respect to the vibrational coordinate  $\vec{R}_\mu$ .  $N$  is the number of ions in each layer,  $M$  is the ion mass,  $\mu$  is the layer index and  $\vec{\epsilon}_{\vec{q},\nu}(\vec{R}_\mu)$  are the phonon polarization vectors.

To relate to a conceptually more simple picture of the phonons we introduce the Eliashberg spectral function  $\alpha^2 F(\omega)$  [32], which is the phonon density of states weighted by the e–p coupling function  $g$

$$\alpha^2 F_{\vec{k}_i}(\omega) = \sum_{\nu,\vec{q},f} |g_{i,f}^\nu(\vec{q})|^2 \delta(\hbar\omega - \hbar\omega_\nu(\vec{q})) \delta(\epsilon_f - \epsilon_i), \quad (7)$$

where the last delta function indicates that we consider the quasi-elastic approximation, neglecting the change of the energy of the scattered electron due to absorption or emission of a phonon. Thus we can write  $\Gamma_{\text{ep}}$  as an integral over phonon energies. If we consider an initial hole state  $(\omega, \vec{k}_i)$  and take into account phonon absorption and emission processes we obtain

$$\Gamma_{\text{ep}}(\omega, \vec{k}_i) = 2\pi \int_0^{\hbar\omega_{\text{max}}} \alpha^2 F_{\vec{k}_i}(\epsilon) [1 + 2n_B(\epsilon) + f(\hbar\omega + \epsilon) - f(\hbar\omega - \epsilon)] d\epsilon, \quad (8)$$

where  $\omega_{\text{max}}$  is the maximum phonon frequency. We then obtain the  $T=0$  result ( $\Rightarrow n_B=0$ ) for  $\Gamma_{\text{ep}}$  as a function of hole binding energy  $|\omega|$ .

$$\Gamma_{\text{ep}}(\omega, \vec{k}_i) = 2\pi \int_0^{|\omega|} \alpha^2 F_{\vec{k}_i}(\epsilon) d\epsilon. \quad (9)$$

The mass enhancement parameter  $\lambda$  is in terms of the Eliashberg function just its first reciprocal moment [33]

$$\lambda(\vec{k}_i) = 2 \int_0^{\omega_m} \frac{\alpha^2 F_{\vec{k}_i}(\omega)}{\omega} d\omega. \quad (10)$$

If the high  $T$ -limit ( $k_B T \gg \hbar\omega_{\text{max}}$ ) of Eq. (8) is considered, Grimvall [34] has pointed out a very useful result, which enables an experimental determination of the mass enhancement parameter

$$\Gamma_{\text{ep}}(\omega, \vec{k}_i) = 2\pi\lambda(\vec{k}_i)k_B T. \quad (11)$$

We thus conclude that the Eliashberg function  $\alpha^2 F$  is a basic function to calculate. Given this function most of the interesting quantities can be calculated, such as the temperature and also bind-

ing energy dependence of the lifetime broadening and the mass enhancement parameter. However, this is no simple task, as all the physics connected to the e–p interaction is buried in  $\alpha^2 F$ , the phonon dispersion relation, phonon polarization vectors, one-electron wave functions and last but not least, the gradient of the screened electron–ion potential—the deformation potential.

## 5. Calculations

We here present results from a recent calculation of the lifetime broadening of the intrinsic surface states of the noble metal surfaces Cu(111) and Ag(111) compared to high resolution ARPES data [36].

Considering the e–p interaction the aim was to take into account in some details the phonons, both bulk and surface modes. To achieve the phonon dispersion and the phonon polarization vectors we performed a slab calculation applying a single force constant model. The force constant is adjusted to fit to the maximum phonon frequency [38]. We adopted the Ashcroft pseudo potentials as bare electron–ion potentials, parametrized according to Ashcroft and Langreth [37]. We have investigated the screening of the bare potentials by applying the dielectric function according to both Thomas–Fermi and RPA (constructed by the eigenwave functions and energy eigenvalues from a 31-layer slab calculation). The two different types of screening gave a difference of about 1% for the mass enhancement factor ( $\lambda$ ) and the phonon induced lifetime broadening ( $\Gamma_{\text{ep}}$ ) [35,38].

Calculating  $\Gamma_{\text{ep}}$  we have to take into account intra band and inter band scattering of electrons and also possibly Umklapp processes. For the surface states of the studied noble metals the Umklapp processes can be neglected as the Fermi momentum of the surface states are small ( $<0.12$  a.u.) in comparison with half the minimum reciprocal vector ( $|\vec{G}|/2 < 0.75$  a.u.) [35]. Furthermore the intra band scattering has been shown by us to be of minor importance [38].

We now turn to the results of the calculations concerning the hole binding energy and temperature dependence of the studied surface states. In

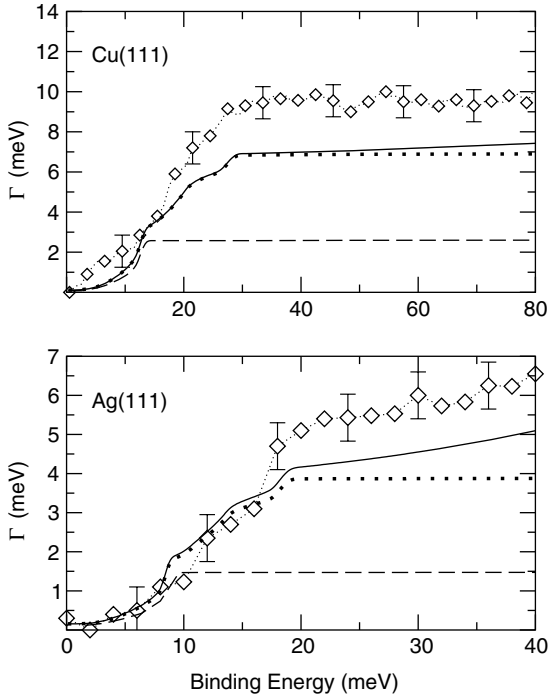


Fig. 3. Lifetime broadening of the Cu(111) and Ag(111) surface hole state as a function of binding energy,  $\Gamma_{ee} + \Gamma_{ep}$  (solid line),  $\Gamma_{ep}$  (dotted line) and the Rayleigh mode contribution to  $\Gamma_{ep}$  (dashed line).

Fig. 3 we present the calculated hole binding energy dependence of  $\Gamma_{ep}$ , at  $T = 30$  K, for the surface states of Cu(111) and Ag(111). The calculated structure of  $\Gamma_{ep}$  in the small binding energy region reflects the structure of the Eliashberg function which in turn depends on the phonon density of states of the system. The high resolution ARPES data show some of these structures, indicating the possibility to experimentally obtain the Eliashberg function at low temperatures. We also note, as mentioned in the introduction that the saturation of  $\Gamma_{ep}$  at  $\omega = \omega_{\max}$  ( $\approx 30$  meV for Cu(111) and 20 meV for Ag(111)) is also seen in the experimental data.

Adding the contribution from the e–e interaction, values close to the experiment are obtained [36]. We note from Fig. 3, that the contribution from the Rayleigh surface mode gives an important contribution in particular for very small binding energies.

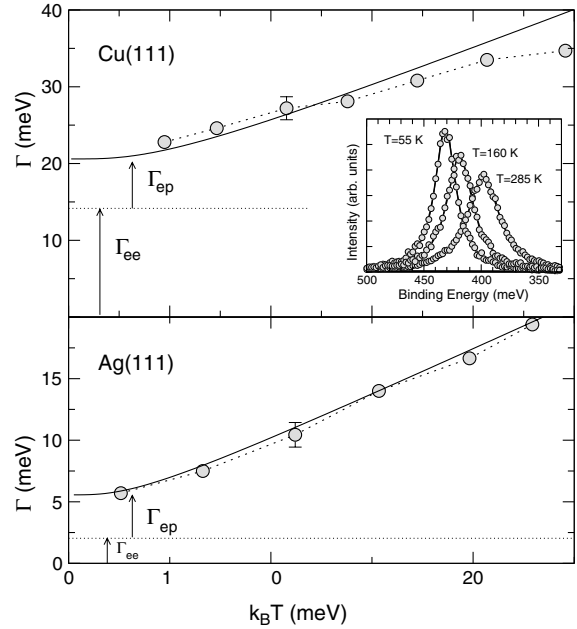


Fig. 4. Lifetime broadening of the Cu(111) and Ag(111) surface hole states as a function of temperature (solid line),  $\Gamma_{ee}$  (dotted line).

The main signature of the e–p contribution to the lifetime broadening is the temperature dependence. In Section 3.2 we argued that if the binding energy of the hole state exceeds the vibrational energies  $\Gamma_{ep}$  takes a finite value while at high temperatures it becomes linear with temperature. This qualitative behavior can be seen in our calculated temperature dependence of  $\Gamma_{ep}$  presented in Fig. 4 considering a hole state in the  $\bar{\Gamma}$  point for Cu(111) and Ag(111). The calculated full lifetime broadenings for both Cu(111) and in particular for Ag(111), are in excellent agreement with the experimental data [36].

## 6. Concluding remarks

In this paper we give a presentation of the electron–phonon coupling starting off by describing the coupling between a single vibrating ion and the surrounding electron gas. We are then focused on evaluating the phonon induced lifetime broadening of an electronic surface band state. We have



demonstrated that it is possible to reasonably well understand experimental data concerning the e–p induced lifetime broadening of surface states. Of major importance is to take into account bulk and surface electron and phonon states.

There are presently many interesting theoretical investigations of the e–p interaction on metal surfaces to be done. For example, to explain the observed strong e–p coupling for metallic overlayer structures, e.g. Na/Cu(111) [39] and Ag/Fe(100) [40].

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