Electron-phonon interaction at the Be(0001) surface.

A. Eiguren\textsuperscript{1}, S. de Gironcoli\textsuperscript{2}, E. V. Chulkov\textsuperscript{1,3}, P. M. Echenique\textsuperscript{1,3}, and E. Tosatti\textsuperscript{2,4}

1. Departamento de Física de Materiales and Centro Mixto CSIC-UPV/EHU, Facultad de Ciencias Químicas, Universidad del País Vasco/Euskal Herriko Unibertsitatea, Apto. 1072, 20018 San Sebastián/Donostia, Basque Country, Spain

2. International School for Advanced Studies (SISSA) and Istituto Nazionale di Fisica della Materia (INFM/DEMOCRITOS), via Beirut 2-4, I-34014, Trieste, Italy

3. Donostia International Physics Center (DIPC), Paseo de Manuel Lardizabal, 4, 20018 San Sebastián/Donostia, Spain and

4. International Centre for Theoretical Physics (ICTP), P. O. Box 586, I-34014, Trieste, Italy

(Dated: November 20, 2018)

We present a first principle study of the electron-phonon (e-p) interaction at the Be(0001) surface. The real and imaginary part of the e-p self energy ($\Sigma$) are calculated for the $\bar{\Gamma}$ surface state in the binding energy range from the $\bar{\Gamma}$ point to the Fermi level. Our calculation shows an overall good agreement with several photoemission data measured at high and low temperatures. Additionally, we show that the energy derivative of Re$\Sigma$ presents a strong temperature and energy variation close to $E_F$, making it difficult to measure its value just at $E_F$.

PACS numbers: 71.10.-w, 68.35.Ja, 73.20.-r, 74.78.-w

For many years now a considerable effort has been directed toward the study of electron-phonon (e-p) effects and quasiparticle dynamics in low dimensional systems. Important examples include semiconductor heterostructures, superconducting materials with layered structures and electrons in two-dimensional surface states. Experimentally, e-p effects are becoming increasingly accessible on surfaces, through powerful techniques like high resolution angular-resolved photoemission spectroscopy (PES), scanning tunnelling spectroscopy (STS), and time-resolved two-photon photoemission spectroscopy (TR-2PPE).

Be(0001) offers in this respect a textbook study case. Here the surface states form a fully metallic, high density two-dimensional electron gas, rather well decoupled from the bulk Be substrate, itself only of semi-metal character\textsuperscript{1,2}. The surface states increase the density of electron states (DOS) at the Fermi energy ($E_F$) by a factor of 4-5 in the surface layer\textsuperscript{2}. This radical change of character between surface and bulk goes along with some other very special properties of the Be(0001) surface\textsuperscript{3}. Following the DOS arguments, the surface $\lambda$ parameter, representative of the e-p interaction, is believed to be about a factor of 4-5 larger than in bulk Be\textsuperscript{4,5,6,7}. Values of $\lambda$ ranging from 0.65 to 1.2 were obtained from the high temperature surface state broadening\textsuperscript{4}, or by fitting the real part of the low temperatures surface state self-energy to simple models\textsuperscript{5,6,7}. The scatter between these values...
A norm conserving pseudopotential was used to have used the density-functional theory in the local density approximation (LDA). The e-p matrix elements and phonon frequencies were calculated using density functional perturbation theory \[^{10}\] through the PWSCF program \[^{11}\]. A norm conserving pseudopotential was used with a cutoff of 22\(R_y\) for the expansion of the electronic wave function in plane waves. For the Brillouin zone electronic structure integrations an order one Hermite-Gauss smearing technique was used, with smearing parameter \(\sigma = 0.05\) \(\text{Ry}\) and as many as 102 special \(\mathbf{k}\) points in the irreducible wedge of the 2D Brillouin zone (IWBZ). The linear response calculation for phonon modes \[^{10}\] have been performed with 30 special \(\mathbf{q}\) points in the IWBZ, yielding surface phonons in excellent agreement with experiments \[^{8}\] and previous calculations \[^{12}\].

The basic quantity in the study of the e-p interaction is the so called Eliashberg function (ELF) \[^{13}\]. This function measures the phonon density of states weighted by the e-p interaction. It represents the probability of emitting a phonon of energy \(\omega\) at \(T = 0\)

\[
\alpha^2 F_i(\omega, E) = \int d\mathbf{q}_G \frac{\delta(E - E_{i,\mathbf{k}})}{\Omega_{\mathbf{BZ}}} \times \\
\times \int \frac{d^2 \mathbf{q}}{\Omega_{\mathbf{BZ}}} \sum_{\nu} |g^{i,f,\nu}_{\mathbf{k},\mathbf{k}+\mathbf{q}}|^2 \delta(E - E_{f,\mathbf{k}+\mathbf{q}}) \delta(\omega - \omega^\nu_{\mathbf{q}}). \tag{1}
\]

The ELF contains all the information needed for the calculation of the real and imaginary part of the self-energy. In Eq. \(\ref{eq:elf}\) \(g^{i,f,\nu}_{\mathbf{k},\mathbf{k}+\mathbf{q}} = \langle \Psi_{i,\mathbf{k}} | \Delta_{\text{CSR}} V^\nu_{\mathbf{q}} | \Psi_{f,\mathbf{k}+\mathbf{q}} \rangle\) is the e-p matrix element where \(\Psi\) are the electronic state wave functions and \(\Delta_{\text{CSR}}V^\nu_{\mathbf{q}}\) denotes the change of the self-consistent electron potential induced by a phonon mode with momentum \(\mathbf{q}\) and mode index \(\nu\); \(i\) refers to the electronic state (surface state) index, \(f\) to the final state, \(E\) denotes the binding energy, \(N_i(E)\) the \(i\) state partial density of states at energy \(E\) and \(\Omega_{\mathbf{BZ}}\) is the area of the 2D Brillouin zone.

The imaginary part of the self-energy related to the lifetime of the excited electronic state through \(\tau^{-1} = 2\) \(Im\Sigma\) can be written in terms of the ELF,

\[
Im\Sigma_i^0(E; T) = \pi \int_0^\infty d\omega \alpha^2 F_i(\omega, E) \frac{1 - f(E - \omega) + f(E + \omega) + 2n(\omega)}{\omega}, \tag{2}
\]

where \(f\) and \(n\) are the Fermi and Bose distribution functions, respectively. The real part reflects renormalization of the electronic band due to the interaction with phonons and can also be calculated from the ELF \[^{13}\],

\[
Re\Sigma_i(E; T) = \int_{-\infty}^{\infty} d\nu \int_0^\infty d\omega \alpha^2 F_i(\omega, E) \frac{2\omega}{\nu^2 - \omega^2} f(\nu + E). \tag{3}
\]

For the calculation of the ELF, the \(\mathbf{k}\) integration was carried out with 102 special points in the IWBZ and with 30 points for the \(\mathbf{q}\) integration. The two Dirac deltas containing the electron band energies were again replaced by order one Hermite-Gauss smearing functions. In order to gain confidence in the convergence of the calculation, we performed a set of calculations with different smearing parameters in the range 0.0025 — 0.1\(R_y\), which gives an estimated error of \(\approx 5\%\) for the surface state broadening at all energies except inside a window of 0.4 eV around \(E_F\) where we estimate an error of \(\approx 15\%\).

Fig. \(\ref{fig:elf}\) shows the electron and phonon band structures. In the upper panel of Fig. \(\ref{fig:elf}\) the \(\Gamma\) point surface state is disturbing. Also, a value as large as 1.2 may raise questions concerning the stability of the normal metallic state of this surface. On the other hand, it is well known that simple model calculations, such as Debye models, are surely too crude, neglecting band structure effects and the important contribution of the surface phonon modes \[^{8}\]. Electron energy-loss spectroscopy (EELS) measurements \[^{7}\], showed that the surface phonon dispersions at Be(0001) do qualitatively differ from those obtained by using truncated bulk models \[^{2}\].

In this letter we present the first parameter free \textit{ab initio} calculation of the e-p interaction at a real metal surface, Be(0001), that contains all the ingredients entering the e-p interaction, namely, surface electron and phonon states, as well as electronic screening. Our detailed analysis results in excellent agreement with all experimental data. We also motivate more future experimental work.

A fully relaxed 12 layer slab has been considered. We have used the density-functional theory in the local density approximation (LDA). The e-p matrix elements and phonon frequencies were calculated using density functional perturbation theory \[^{10}\], through the PWSCF program \[^{11}\]. A norm conserving pseudopotential was used with a cutoff of 22\(R_y\) for the expansion of the electronic wave function in plane waves. For the Brillouin zone electronic structure integrations an order one Hermite-Gauss smearing technique was used, with smearing parameter \(\sigma = 0.05\) \(\text{Ry}\) and as many as 102 special \(\mathbf{k}\) points in the irreducible wedge of the 2D Brillouin zone (IWBZ). The
The first reciprocal moment of the ELF \[\lambda_0(E)\] indicates an enhanced role of the ELF at low energies. Fig. 2 shows that the weight of the surface phonon modes is concentrated around \(\omega_D/2\) (\(\omega_D \approx 80\) meV), thus a fitting procedure of \(\lambda_0\) parameter through a Debye model \((\alpha^2F = \lambda_0(\omega/\omega_D)^2)\) will give rise in this case to an excessive low temperature broadening compared to a more realistic calculation using the same Debye energy \(\omega_D\) and \(\lambda_0\). The top panel of Fig. 3 shows the real (orange) and imaginary part (black) of the self-energy calculated for \(T=0\) and as function of the binding energy. The imaginary part is resolved into contributions coming from intraband scattering (green) and the contribution of the Rayleigh mode (red). The imaginary part varies by a factor of two going from \(E_F\) to the \(\bar{\Gamma}\) point. The bottom panel of Fig. 3 shows the variation of the \(\lambda_0\) parameter defined through Eq. 4 as function of the binding energy. From this figure we deduce that the temperature derivative of the high-\(T\) surface state broadening is twice for the hole at \(E_F\) compared to that at the \(\bar{\Gamma}\) point.

Recently, the identity between the partial derivative of \(\Re\Sigma\) at Fermi level and \(\lambda_0(E_F)\), that is \(\lambda \equiv \lambda_0(E_F) = \Lambda(E_F, T = 0)\),

\[
\Lambda(E, T) \equiv -\frac{\partial \Re \Sigma(E, T)}{\partial E}
\]

(5)

has been used to determine the e-p coupling parameter \(\lambda\) \[\ref{5} \ref{6} \ref{7}\]. However, it is of crucial importance here to distinguish \(\Lambda\) and \(\lambda_0\). The results of Fig. 3 do not take into account the renormalization of electron bands due to the real part of the self-energy. We denote by \(E_k^0\) the pure LDA unrenormalised bands and by \(E_k\) the renormalised ones including \(\Re\Sigma\), obtained by solving numerically the equation \(E_k = E_k^0 + \Re\Sigma(E_k)\) \[\ref{8}\] (see Fig. 4). It is well known that close to \(E_F\) the electron velocity, e-p coupling form factor and the lifetime itself are renormalised \[\ref{8}\].

Provided that the system is isotropic and \(|\Lambda(E, T)| \ll 1\), an estimation of the renormalised imaginary part \(\Re\Sigma\) can be obtained from

\[
\Im \Sigma(E_k) \approx \frac{\Im \Sigma^0(E_k)}{1 + \Lambda(E_k)}
\]

(6)

where \(\Im \Sigma\) denotes the renormalised quantity. While the breakdown of the approximation expressed by Eq. 6 is clear for values of \(\Lambda(E, T)\) close to -1, this form generally helps us to understand how the deformation of the electronic band caused by the real self-energy affects itself the lifetime of the surface state.

In the top panel of Fig. 4 we show the unrenormalised imaginary part of the self-energy (\(\Im \Sigma^0\), dotted line) calculated from Eq. 2 and the renormalised ones at \(T = 40\) (orange) and at \(T = 12K\) (violet). The result for 12K is displayed only for reasonable values of \(1/(1 + \Lambda)\). These two temperatures, 12K and 40K, are the same as those considered experimentally, Refs. \[\ref{8} \ref{9}\] respectively. In the bottom panel, the real part of the self-energy at \(T = 12K\) (violet) and at \(T = 40K\) (orange) are depicted.
The inset of the bottom panel shows the derivatives of the real part of the self-energy at these two temperatures.

In Ref. 3 a detailed measurement of \( \text{Im} \Sigma \) and \( \text{Re} \Sigma \) is reported for \( T = 40K \). An 'unusual' peak of \( \text{Im} \Sigma \) for the hole at binding energies around the maximum phonon energy (\( \sim \omega_D \)) was observed. This behaviour is well described by our calculation at the same temperature (orange). The calculated real part of the self-energy also shows excellent agreement with the measurement, and our \( \Lambda(E_F, T = 40K) \approx 0.7 \), is in excellent agreement with their measured 0.7 ± 0.1 (Inset of the bottom panel of Fig. 4). In Ref. 2, the derivative of the real self-energy part was presented for \( \omega = 3/2 \omega_D \) binding energies. Black lines represent the renormalised broadening according to Eq. 4. Note the minimum at temperatures \( k_B T \sim 13 \) meV. Black dotted line for \( \omega_D \) indicates region where Eq. 5 is not reliable. The inset shows the \( \text{Im} \Sigma \) dependence on binding energy (\( E_b \)).

FIG. 5: Temperature dependence of the broadening at \( \omega_D/2 \), at \( \omega_D \) and at \( 3/2 \omega_D \) binding energies. Black lines represent the renormalised broadening according to Eq. 4. Note the minimum at temperatures \( k_B T \sim 13 \) meV. Black dotted line for \( \omega_D \) indicates region where Eq. 5 is not reliable. The inset shows the \( \text{Im} \Sigma \) dependence on binding energy (\( E_b \)).

Analysing the temperature dependence of the broadening in more detail, we note that for larger \( T \), the maximum value of the T-derivative of \( \text{Re} \Sigma \) decreases, and so does the effect of the renormalising denominator in Eq. 6 (Fig. 4). By contrast, increasing temperature favours phonon occupation: thus we have two competing tendencies and a minimum can be expected in the temperature dependence of \( \text{Im} \Sigma \). Detailed calculation of \( \text{Im} \Sigma \) predicts a minimum of \( \text{Im} \Sigma \) around \( T \sim 150K \) for states close to \( \omega_D \) binding energy (see Fig. 5). It would be interesting to verify this minimum in the temperature dependence of the broadening. This effect should be even more pronounced for binding energies close to \( \sim \omega_D \) where the effect of the renormalisation is stronger.

Here in fact the group velocity selectively rises above its bare value, and so does the low temperature hole decay rate, giving rise to a local maximum as a function of hole energy. Removal of this effect by temperature causes the local minimum predicted in Fig 5.

In summary we presented the first parameter-free \( ab - initio \) calculation of the e-p interaction in a surface state on a real metal surface, Be(0001). The surface state renormalisation and its broadening were calculated, and the latter was found to agree well with existing experimental data. The energy derivative of \( \text{Re} \Sigma \) presents a strong energy and temperature dependence near \( E_F \), so that an accurate measurement of \( \lambda \equiv \Lambda(E_F) = \lambda_0(E_F) \) requires temperatures \( T \lesssim 12K \) and energies very close to \( E_F \). Measurements done under different conditions can produce very different \( \lambda \) values which explains the variety of values in literature. Additionally, the effect of the renormalisation of the surface state band on its own broadening is pointed out and new experiments are suggested to confirm a new minimum predicted in the temperature dependence. Other conceivable consequences of a medium size surface state e-p coupling, including the possible stabilisation of a 2D BCS state at \( T=0 \) and/or the factors hindering it, remain to be explored.

We acknowledge financial support from the Basque Government, the Max Planck Research Award Funds, the Spanish Ministerio de Educación y Ciencia (MEC), and the Basque Country University. Work in SISSA was supported through MIUR COFIN and FIRB projects, by INFN/F and by the 'Iniziativa Trasversale Calcolo Parallelo' of INFM.

\[ 1 \] R. A. Bartynski et al., Phys. Rev. B 32, 1921 (1985).
\[ 2 \] E. V. Chulkov et al., Surface Sci. 188, 287 (1987).
\[ 3 \] E. W. Plummer and J. B. Hannon, Prog. Surf. Sci. 46, 149 (1994); L. I. Johansson, et al., Phys. Rev. Lett. 71, 2453 (1993); P. T. Sprunger et al., Science 275, 1764 (1997).
\[ 4 \] T. Balasubramanian et al., Phys. Rev. B 57, R6866 (1998).
\[ 5 \] S. LaShell et al., Phys. Rev. B 61, 2371 (2000).
\[ 6 \] M. Hengsberger et al., Phys. Rev. Lett. 83 592 (1999).
\[ 7 \] M. Hengsberger et al., Phys. Rev. B 60, 10796 (1999).
\[ 8 \] A. Eiguren et al., Phys. Rev. Lett. 88, 066805 (2002).
\[ 9 \] J. B. Hannon et al., Phys. Rev. B 53, 2090 (1996).
\[ 10 \] S. Baroni et al., Rev. Mod. Phys. 73, 515 (2001).
\[ 11 \] S. Baroni et al., \url{http://www.pwscf.org}.
\[ 12 \] M. Lazzeri et al., Surf. Sci. 402 715 (1998).
\[ 13 \] G. Grimvall, in The Electron-Phonon Interaction in Metals, Selected Topics in Solid State Physics, edited by E. Wohlfarth (North-Holland, New York, 1981).
\[ 14 \] E. Tosatti, in Electronic surface and interface states on metallic systems, eds, E. Bertel and M. Donath (World Scientific, Singapore, 1995), p.67.