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Long-range scattering of electrons by phonons at metal surfaces

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We examine the inelastic scattering of electrons by phonons at metal surfaces. A dielectric-function formulation for calculating the loss probability is first summarized. Then, using the Bohm-Staver description of a metal's response, we derive formulas for the spectra of single-phonon losses near the specular electron beam. Both bulk and surface phonons contribute. Finally, we compare the thermal diffuse scattering predicted in our model to that due to impact scattering. The former can dominate the latter at low incident-electron energies.

I. INTRODUCTION

There is at present considerable interest in the inelastic scattering of low-energy electrons from surfaces.[1,2] The spectra allow one to study a wide range of surface excitations, extending from vibrations of adsorbed species to collective motions of the bare substrate. As has been emphasized by Mills,[3] one may understand the excitation of all such losses as due (at least in part) to a common mechanism: the long-ranged Coulomb coupling between the charged probe and the electric field fluctuations associated with the surface modes. There is, of course, additional coupling from short-range scattering as the probe bounces off the surface, but its detailed calculation is quite difficult[4] and we shall generally neglect it here. By limiting ourselves to the effects of the long-range interaction and by further treating it as a weak perturbation, we can study a wide variety of systems in a common fashion, allowing us to compare the strength and spectral structure of various loss processes. In particular, we can theoretically examine modes that are not easily observed and compare their characteristics with modes that have often been seen.

Our specific interest in this paper is with phonons on clean metal surfaces. We use a very simple model for their description and focus on its implications for the long-range scattering of electrons outside the metal. In Sec. II we derive for completeness our basic approach, which is based on a linear-response treatment of the loss process. This dielectric-function approach has been used many times before and is useful in providing a clear and common physical picture for a host of model systems. We apply the formalism in Sec. III to a Bohm-Staver model of phonons at metal surfaces, deriving the single-excitation loss spectra of low-energy electrons to both bulk and surface phonons. Since such losses involve energies near the limit of present-day resolution, we also examine in Sec. IV the thermal diffuse scattering implied by these excitations, both when excited by the long-range mechanism of interest here and when excited by short-range impact scattering.

II. DIELECTRIC-FUNCTION FORMULATION

Now we briefly describe the spirit of our approach. As noted above the basic method is not new—see Ref. 1—nor does it directly yield a general result. Its advantages are that it is easy to apply, that it provides a common perspective, and that with only a little extra interpretation[5] it can be extended to treat some of its omissions.

The basic scheme is the following. One imagines that the external charge that is to be (weakly) inelastically scattered follows in lowest order its classical specular trajectory. From the point of view of the metal, this prescribed trajectory of the external charge represents a dynamic perturbation to which the metal responds. We treat this response to linear order, calculating the induced charge density in the metal and the induced force $\vec{F}$ that it exerts back on the external charge. The net effect is that the external charge actually loses energy at the rate

$$\frac{dW}{dt} = -\vec{v}(t)\cdot\vec{F}(t) \ , \tag{1}$$

where t is time and $\vec{v}$ is the velocity of the external charge. As t runs from $-\infty$ to $+\infty$, the external charge completes its specular trajectory and its total

energy loss is

$$W = -\int_{-\infty}^{\infty} dt\, \vec{v}(t)\cdot\vec{F}(t) \ . \tag{2}$$

We will show below that solving the linear-response problem using Fourier analysis allows us to reexpress (2) as

$$W = \int_0^{\infty} d\omega \int d^2Q\, \hbar\omega P(\vec{Q},\omega) \ , \tag{3}$$

where $\vec{Q}$ is a two-dimensional vector in the plane of the (smooth) metal surface and ω is a frequency. With W written as in (3), we may interpret $P(\vec{Q},\omega)d^2Q\, d\omega$ as the probability that the external charge is inelastically scattered into the range of energy losses between $\hbar\omega$ and $\hbar(\omega + d\omega)$ and into the range of momentum losses parallel to the surface between $\hbar\vec{Q}$ and $\hbar(\vec{Q} + d\vec{Q})$. Specifying ω and $\vec{Q}$ completely determines the actual kinematics of an electron at the detector.

As is clear from this description the scattering analysis is not self-consistent. Yet if the losses are small, this lack of self-consistency should only be a higher-order correction to P. Hence the theory we are constructing only applies to small energy losses and near-specular scattering. From the point of view of excitations in the metal it only describes the excitation of single quanta in a system initially in its ground state. One may construct more general theories[2,3,6–8] that allow multiple losses or gains and, in addition, treat the external charge quantum mechanically. However these theories in the limit of weak scattering reduce essentially to what we use here,[2] so we prefer to continue with our simple but limited approach.

To derive an explicit formula for the loss probability function, $P(\vec{Q},\omega)$, we need the results of linear response. Imagine the metal is located in $x < 0$ and describe the external charge's trajectory by

$$\vec{x}(t) = (v\,|\,t\,|,\vec{V}t) \ , \tag{4}$$

where v sgn$t$ and $\vec{V}$ are the components of velocity perpendicular and parallel, respectively, to the surface. Here we ignore any refraction of the trajectory due to an image potential and also assume specular scattering occurs just outside the metal. This last assumption is the extent of our treatment of short-range impact scattering. Since specular scattering is in general not the only possible scattering process at the surface, we should view our derived P as a relative probability.

The perturbing potential the metal sees is

$$\phi_{\text{ext}}(\vec{x},t) = \frac{e}{|\vec{x} - \vec{x}(t)|}$$

or

$$\phi_{\text{ext}}(\vec{Q},x;\omega) = \frac{2\pi e}{Q} e^{Qx}\left[\frac{2Qv}{Q^2v^2 + (\vec{Q}\cdot\vec{V} - \omega)^2}\right] \ , \tag{5}$$

where $e < 0$ is an electron's charge and x lies in the metal, i.e., $x < 0$. In the second line we have carried out a Fourier transform in order to simplify the response calculation. Presuming a smooth metal surface, we find that the metal's response leads to an induced potential outside the metal of

$$\phi_{\text{ind}}(\vec{Q},x;\omega) = \frac{2\pi e}{Q} e^{-Qx}\frac{\bar{\epsilon}(\vec{Q},\omega) - 1}{\bar{\epsilon}(\vec{Q},\omega) + 1} \times \left[\frac{2Qv}{Q^2v^2 + (\vec{Q}\cdot\vec{V} - \omega)^2}\right] \ , \tag{6}$$

where $x > 0$ and

$$\frac{1}{\bar{\epsilon}(\vec{Q},\omega)} = \frac{2}{\pi}\int_0^{\infty} dq \frac{Q}{Q^2 + q^2} \Big/ \epsilon(\vec{q},\omega) \ , \tag{7}$$

with $\vec{q} = (q,\vec{Q})$. In deriving this result we have retained only the linear response and, furthermore, have used a common approximation for it wherein the surface response of the semi-infinite metal is expressed in terms of the bulk dielectric function $\epsilon(\vec{q},\omega)$ via (7). Since we ignore retardation effects, ϵ is the metal's longitudinal dielectric function. The approximation that leads to (7) has been labeled quasiclassical or step density. It arose originally in surface-plasmon theory,[9] but has had a widespread use in surface response problems since then. Although it has several interpretations, we prefer to view it as an approximation to the susceptibility that appears in the random-phase integral equation,[10] roughly replacing a surface-sensitive kernel by its bulk limit. Note that if one can ignore the $\vec{q}$ dependence of ϵ, then $\bar{\epsilon}$ has no $\vec{Q}$ dependence and $\bar{\epsilon}(\omega) = \epsilon(\omega)$. The derivation of (6) from (5) in this case is trivial.

To complete our formal development is now straightforward. We calculate the induced force acting back on the external electron,

$$\vec{F}(t) = -e\,\vec{\nabla}\phi_{\text{ind}}\,|_{\vec{x}=\vec{x}(t)} \tag{8}$$

and then integrate (2). The result implies (Im denotes "imaginary part")

$$P(\vec{Q},\omega)=\frac{e^2}{\pi^2\hbar Q}\left[\frac{2Qv}{Q^2v^2+(\vec{Q}\cdot\vec{V}-\omega)^2}\right]^2 \times \mathrm{Im}\left[\frac{-1}{1+\bar{\epsilon}(\vec{Q},\omega)}\right] . \quad (9)$$

The squared factor strongly favors losses in the forward direction while the dielectric-function term determines the relative strength of various possible contributing modes. We stress that an evaluation of (9) does not first require a derivation of surface-mode eigenfunctions and eigenvalues. The relevant information is contained in $\bar{\epsilon}$, which is often easily calculated from ϵ via (7).

Before treating phonons at metal surfaces, for which the $\vec{q}$ dependence of ϵ is crucial, we close this section with several examples of the application of our approach which only use an $\epsilon(\omega)$. First consider surface-plasmon losses. The appropriate ϵ for this possibility is

$$\epsilon(\omega)=1-\omega_p^2/\omega^2 , \quad (10)$$

where ω_p is the bulk plasma frequency. Then for $\omega>0$,

$$P(\vec{Q},\omega)=\frac{e^2\omega_s}{4\pi\hbar Q}\left[\frac{2Qv}{Q^2v^2+(\vec{Q}\cdot\vec{V}-\omega)^2}\right]^2\delta(\omega-\omega_s) , \quad (11)$$

where $\omega_s=\omega_p/\sqrt{2}$, which is the standard result.[6] One can just as easily treat losses to Fuchs-Kliewer modes on ionic crystals.[11]

Next consider a metal of dielectric constant $\epsilon_b(\omega)$ on top of which is a surface layer of thickness d of a material characterized by $\epsilon_s(\omega)$. Although (7) is not relevant, one can still solve in straightforward fashion for ϕ_{ind}, given ϕ_{ext}. The result has the same form as (6) but with

$$\bar{\epsilon}(\vec{Q},\omega)=\epsilon_s(1-\Delta e^{-2Qd})/(1+\Delta e^{-2Qd}) , \quad (12)$$

where

$$\Delta=(\epsilon_s-\epsilon_b)/(\epsilon_s+\epsilon_b) . \quad (13)$$

These formulas were first obtained by Mills[3] who subsequently used them to describe losses to surface electronic excitations on semiconductors.[12] Here we use them to describe losses to vibrational modes of adsorbed species on metals. To this end we set $\epsilon_b=\infty$ so $\Delta=-1$. Then treating the adsorbed species as independent we write

$$\epsilon_s(\omega)=1+4\pi n_0\alpha(\omega) , \quad (14)$$

and expand as $n_0, d\to 0$,

$$\bar{\epsilon}(\vec{Q},\omega)\approx(1+4\pi n_0\alpha)/Qd \quad (15)$$

and

$$\mathrm{Im}\left[\frac{-1}{1+\bar{\epsilon}}\right]\approx(Qd)(4\pi n_0\,\mathrm{Im}\alpha) . \quad (16)$$

Identifying the surface density of the adsorbate as $n_s=n_0 d$, we find

$$P(\vec{Q},\omega)=\frac{4e^2n_s}{\pi\hbar}\left[\frac{2Qv}{Q^2v^2+(\vec{Q}\cdot\vec{V}-\omega)^2}\right]^2\mathrm{Im}\alpha(\omega) , \quad (17)$$

which is the standard result.[2] Notice the extra factor of Q between (17) and (11).

III. PHONONS AT METAL SURFACES

In this section we apply a crude description of coupled electron and ion motion to the loss formulas just derived. There has been considerable work on phonons at surfaces,[13] but we could find none that for metals retains the fluctuating fields outside the surface. Our model is essentially a Bohm-Staver theory applied at a surface.[14] Following a textbook description,[15] we write the total bulk dielectric constant as

$$\epsilon=\epsilon^{\mathrm{el}}+\epsilon^{\mathrm{ion}}_{\mathrm{bare}}-1 , \quad (18)$$

where we set

$$\epsilon^{\mathrm{el}}=1+k_s^2/|\vec{q}|^2 , \quad (19)$$

$$\epsilon^{\mathrm{ion}}_{\mathrm{bare}}=1-\Omega_p^2/\omega^2 . \quad (20)$$

We are treating the electronic response as if it were static; the k_s^2 in (19) is the square of the Thomas-Fermi screening wave vector. On the other hand, the ion response is dynamic but is taken to be local in space; the Ω_p^2 in (20) is the square of the (bare) ion plasma frequency.

Equations (18)–(20) are sufficient to specify our model. For the bulk metal we have a Bohm-Staver theory as can be seen by setting $\epsilon(\vec{q},\omega)=0$ to find the bulk dispersion law

$$\frac{\omega^2}{\Omega_p^2}=\frac{|\vec{q}|^2}{k_s^2+|\vec{q}|^2} . \quad (21)$$

Just as easily we can calculate $\bar{\epsilon}(\vec{Q},\omega)$ from (7)

$$\frac{1}{\bar{\epsilon}(\vec{Q},\omega)}=\frac{a\omega^2}{\omega^2-\Omega_p^2}\frac{Q}{Q_0} , \quad (22)$$

where

$$Q_0 = \left| Q^2 + \frac{k_s^2 \omega^2}{\omega^2 - \Omega_p^2} \right|^{1/2} \geq 0 \ . \tag{23}$$

The parameter a depends on where one is in the $\omega - Q$ plane, as shown in Fig. 1. In region I, $\omega > \Omega_p$, one has $a = 1$ and $\bar{\epsilon}$ is real and positive. In region II, $0 < \omega < \Omega_p[Q^2/(k_s^2 + Q^2)]$, again $a = 1$ but now $\bar{\epsilon}$ is real and negative. Lastly, in region III $a = i$, so $\bar{\epsilon}$ is positive imaginary. With regard to the loss formula (9) we will get contributions only from regions II and III. Those from the latter region are due to bulk phonons which still produce fields outside the metal. In region II there are contributions only along the dashed line in Fig. 1 which is the solution of $\bar{\epsilon}(\vec{Q},\omega) = -1$:

$$\frac{\bar{\omega}^2(Q)}{\Omega_p^2} = \frac{Q^2}{k_s^2} + \frac{1}{2} - \left[\frac{Q^4}{k_s^4} + \frac{1}{4} \right]^{1/2} \ . \tag{24}$$

The dispersion of this surface phonon begins linearly with Q and saturates for large Q at $\Omega_p/\sqrt{2}$.

Combining (9) and (22) one can readily calculate the loss spectrum. Note that the surface phonon loss at any fixed angle of observation (i.e., fixed $\vec{Q}$) is a δ function lying below the bulk phonon losses:

$$\mathrm{Im}\left[\frac{-1}{1+\bar{\epsilon}} \right] \Bigg|_{\omega \simeq \bar{\omega}(Q)} = \lambda \delta(\omega - \bar{\omega}(Q)) \ , \tag{25}$$

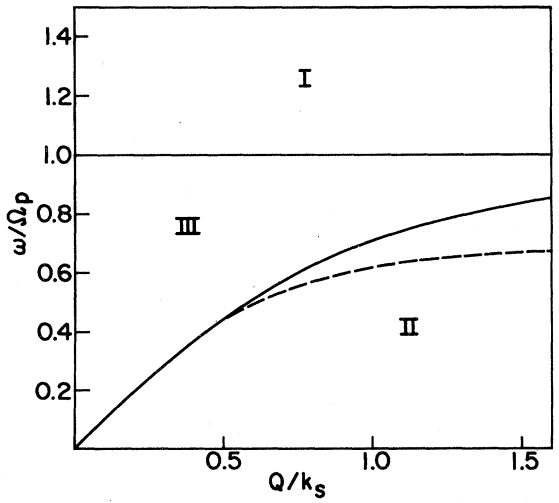

FIG. 1. Phonon dispersion at a metal surface versus wave vector Q parallel to the surface. The bulk phonons lie in region III, the surface phonon (dashed line) in region II. The parameter a of equation (22) is 1 in regions I and II and is i in region III.

where

$$\lambda = \pi \frac{\omega^3}{\Omega_p^2} \frac{Q^2}{k_s^2} \left[1 + \frac{2\omega^2 Q^2/k_s^2}{\Omega_p^2 - \omega^2} \right]^{-1} \ . \tag{26}$$

The rapid decay of λ at low frequencies is remarkable.

We shall not present model calculations of the loss spectra at various angles for several reasons. First, one should realize that these modes are at rather low energies; e.g., for silver surface we estimate (see Sec. IV) $\hbar\Omega_p \sim 20$ meV. There have, in fact, been few reported observations of substrate phonons on metals, and even these have used either stepped surfaces[16] or overlayers[17,18] to enhance the coupling and to see phonons at the zone edge or above the bulk bands. Second, our model of the phonon dispersion is quite crude. A better treatment in connection with loss spectra was done some time ago by Roundy and Mills,[19,20] using force-constant models. Yet their calculation was rather sensitive to parameter choices and more importantly did not allow for the long-range coupling used exclusively here. The utility of our results is, in fact, not the shape of the spectra but rather the estimate of its strength.

An attempt to bring out this point is made in the next section, but before presenting it we wish to discuss work on a related surface model by Wanser.[21] His paper deals with the influence of surface distortion on the static image potential. He uses classical elasticity theory to describe atomic distortions near a metal surface and finds a nonlinear correction to the image potential law, tending to strengthen the attraction. Our model is quite different from his but can be applied to the same problem. In linear response the image force is for a charge e at R above the metal

$$-\frac{\partial}{\partial R}\Delta V = -e^2 \int_0^\infty Q \, dQ \, e^{-2QR} \frac{\bar{\epsilon} - 1}{\bar{\epsilon} + 1} \ , \tag{27}$$

where $\bar{\epsilon}$ is to be evaluated in the static limit, $\omega = 0$. For our choice (18) of ϵ, one has $\epsilon(\vec{Q},\omega = 0) = -\infty$, and hence an image force of $e^2/4R^2$ directed towards the metal. This diverges as $R \to 0$ because we have assumed a spatially local ion response, but compared to its value when only the electron response (19) is included,[22] we, too, have found a strengthening of the image interaction. This qualitative feature is reasonable since the ions, if allowed to move, will certainly act to lower the system's energy.

IV. THERMAL DIFFUSE SCATTERING

A further implication of our electron-loss formulas is a new mechanism of thermal diffuse scattering. One need only at each $\vec{Q}$ to integrate over the possible loss energies to derive a prediction for the strength of thermal diffuse scattering due to long-range scattering. This contribution will be an addition (if we neglect interference) to that which arises from impact scattering. The latter mechanism has been studied extensively before, both theoretically and experimentally,[23] and we will try to compare the two.

This comparison is not completely straightforward because of differing theoretical approaches. The loss function $P(\vec{Q},\omega)$ that we use represents a probability relative to that for the specular beam and we have not discussed how one should estimate the strength of the elastic specular beam. On the other hand, theories that use only short-range scattering can calculate the elastic and thermal diffuse spectra simultaneously, but there is a simple relation between them only in a crude kinematic model which allows only one scattering event on the surface and limits it to the top layer of atoms. Yet since our interest is merely to estimate roughly the relative strength of the two mechanisms, these different approaches can be forced to a common basis. The quantity we will examine in each case is the ratio α of the thermal diffuse strength to the elastic specular scattering strength.

For our long-range-scattering model

$$k_s^{-2}\alpha = \int_0^\infty d\omega\, P(\vec{Q},\omega)\coth\left[\frac{\beta\hbar\omega}{2}\right], \qquad (28)$$

where $\beta = 1/kT$ with T the surface temperature. The coth factor has been inserted in (28) to account for the fact that whereas our derivation in Sec. II only included the spontaneous creation of surface excitations due to the external charge, one expects stimulated losses and gains to also occur. Hence (28) estimates the total single-phonon-excitation strength at fixed $\vec{Q}$ and T, relative to the specular beam.

To develop an analogous expression from a short-range-scattering model, we simplify further the kinematic model discussed above. Specifically, we assume that incoming electrons scatter in first Born approximation from the short-range potentials of a single layer of atoms whose vibrations may be described as if they were in bulk.[15] Thus we ignore any change in phonon amplitudes at the surface. We also neglect polarization effects and assume that the single-atom-scattering amplitude and electron-momentum change depend only weakly on angle. With this last assumption the scattering amplitude drops out of the ratio α, and we obtain

$$k_s^{-2}\int d^2Q\,\alpha = (2|\vec{k}_i|\cos\theta_i)^2\sum_{\vec{q}}\frac{\hbar}{2MN\omega}\coth\left[\frac{\beta\hbar\omega}{2}\right], \qquad (29)$$

where $\vec{k}_i$ is the incident electron wave vector and θ_i its angle of incidence with respect to the normal so $2|\vec{k}_i|\cos\theta_i$ is the change in the electron wave vector after specular scattering. The other parameters in (29) are M, the mass of an atom, and N, the number of atoms in the crystal. The frequency $\omega = \omega(\vec{q})$ will be computed from the bulk dispersion relation (21) and the sum on $\vec{q}$ done by the replacement

$$\frac{1}{N}\sum_{\vec{q}}\cdots = \int d^2Q\left[\int\frac{dq}{(2\pi)^3 n_B}\cdots\right], \qquad (30)$$

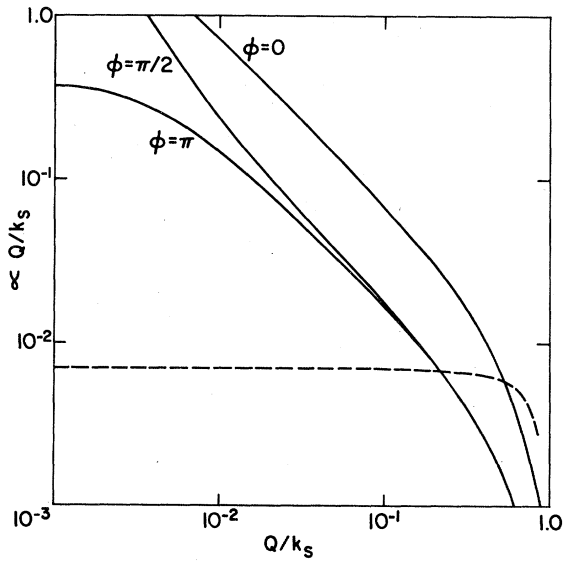

FIG. 2. Thermal diffuse scattering by various mechanisms. We plot the function α times Q/k_s versus Q/k_s. The dashed curve describes impact scattering, while the solid curves come from long-range scattering. The angle ϕ is that between the surface projection of the electron velocity before and after scattering. See text for other parameter values.

where n_B is the bulk density of atoms.

Since most of the experimental work was done on silver,[24,25] we chose our model parameters to be those appropriate to silver,[15] taking $\hbar\Omega_p = 20.1$ meV and $k_s = 1.71$ Å^{-1}. To make the q integral in (30) well defined we use a Debye cutoff. In Fig. 2 are plotted some typical results for the one-phonon thermal diffuse scattering using an incident energy of $E_i = 5$ eV and an incident angle of $\theta_i = \pi/4$. The surface temperature is 300 K. With several assumptions—high temperature, linear dependence of ω on $|\vec{q}|$, and no cutoff—one can show that α for impact scattering should scale as Q^{-1}.[26,27] We have consequently plotted out results as $\alpha(Q/k_s)$. The impact-scattering case is indeed nearly constant, falling off only when Q approaches the Debye cutoff, which here has the value $Q_D/k_s = 0.885$. Such large Q values are not properly treated by our models, which are reasonable only near specular scattering, i.e., for $Q/k_s << 1$.

For the smaller values of Q the interesting feature of Fig. 2 is that the thermal diffuse scattering for the long-range coupling mechanism α_D lies well above that for impact scattering α_I. The growth of α_D is roughly as Q^{-2} as Q decreases, at least until the squared factor in (9) forces a saturation of αQ, which here occurs near $Q/k_s \sim 10^{-3}$ for all orientations of $\vec{Q}$ in the plane. This change in dependence is probably at too small an angle with respect to the specular direction to be presently resolved. Note, too, that the relative strength of α_D and α_I depends sensitively on the incident energy. If we use instead an $E_i = 100$ eV, which was a typical value in past experiments, then the ratio of α_I/α_D would be roughly enhanced at intermediate values of Q by a factor of $(100/5)^2 = 400$ and α_I would dominate, as has in fact been observed.[24,25] The primary distinguishing dependences of α_I and α_D are $E_i Q^{-1}$ and $E_i^{-1}Q^{-2}$, respectively, for $0.01 \lesssim Q/k_s \lesssim 0.1$. Hence evidence for the presence of α_D would require the observation of a stronger dependence of α on Q as E_i is lowered. We hope that such experiments might be done soon. They would establish that the long-range-scattering mechanism is operative and allow one to check the estimates of its strength that have been given here.

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