Interaction of fast charges with a rough metal surface

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ABSTRACT

We use the Green function formulation of a dielectric response formalism to study the dynamic polarization of a rough metal surface by a single charged particle and by a pair of charged particles that move parallel to the surface. While the surface roughness is treated nonperturbatively, the plasmon excitation of the metal electron gas is described locally. We find that the magnitudes of both the image potential and the stopping power of a single particle are increased by the increasing roughness and decreasing correlation length of the surface. On the other hand, both the long-range wake potential of a single charged particle and the interaction potential between two particles are weakly affected by the surface roughness. However, the strongest effects of the surface roughness are seen in the correlated stopping power of two charged particles, giving rise to oscillations in the dependence of the stopping ratio on their distance, both when the interparticle axis is perpendicular to their direction of motion and when the wake-related oscillations are damped by adiabatic suppression of plasmon excitations at low particle speeds.

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1. Introduction

Interaction of fast moving charged particles with metal surfaces has been studied in the context of ion-surface grazing scattering [1] and reflection electron energy loss spectroscopy (REELS) [2]. Those experimental techniques provide important information about excitation of the collective electron modes, or plasmons, of interest in the ongoing research in nano-plasmonics [3] and nano-optics [4]. In particular, this problem was recently revived by the novel work of Nelayah et al. who have developed a method of selectively exciting plasmon eigenmodes of nanometer-sized metal particles with aloof electron beams [5].

The effects of the roughness of metal surfaces were thoroughly studied in the context of nano-optics of surface plasmon–polaritons [4], as well as in the plasmon spectroscopy via REELS, both theoretically [6–8] and experimentally [9,10]. It was shown that the roughness of sputtered metal surfaces may be characterized by a standard deviation of $\delta \sim 1$ nm and the lateral correlation length $a \sim 10$ nm [11], whereas the surfaces of metal films grown on various substrates via molecular beam epitaxy may exhibit much larger roughness with $\delta \sim 10$ nm and $a \lesssim \mu$m [10]. In both classes of targets, the REELS exhibited clear signatures of surface roughness in the measured plasmon spectra [9,10]. However, much less is known about the effects of surface roughness in the case of fast ions moving under glancing angle of incidence upon metal surfaces [1,12,13]. Since dynamic polarization of a metal surface provides

the dominant mechanisms of energy loss and the image attraction when the incident charged particles move at large distances (almost) parallel to the surface, it is clearly desirable to estimate the effects of the atomic scale roughness on the process of surface plasmon excitation.

Moreover, motivated by observations made in the scattering of molecules on solid surfaces [14–16], it is worthwhile exploring the phenomenology that may arise in grazing scattering of the molecular projectiles in the presence of surface roughness. Such processes may include dissociation of the incident molecule [17,18], Coulomb explosion of its ionic fragments [19,20], vicinage effect in the energy loss of these fragments [21], and secondary electron emission from the surface [16]. Therefore, it is interesting to explore, e.g., the effects of roughness of nano-structured metal surfaces [11] on the dynamics of Coulomb explosion, as well as on the vicinage effects in the energy loss of a pair of spatially correlated ions moving parallel to the surface.

In this work we use the nonperturbative results for the electrostatic Green's function for a rough metal surface described by a local dielectric function, which was developed by Rahman and Maradudin [7,8], and invoke the formalism that was recently used to evaluate the dynamic polarization of a supported graphene by correlated external charges [22]. In particular, we calculate the wake potential due to single point charge [23], its energy loss rate and the dynamic image potential, as well as the dynamically screened inter-particle interaction energy and the vicinage energy loss ratio for two co-moving point charges above a rough metal surface [24]. In particular, we investigate particles moving in the range of speeds on the order of several units of the Bohr velocity v_B (corresponding to a proton kinetic energy of 25 keV), for which the rate of plasmon excitation on metal surfaces reaches peak values [25].

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After outlining the theoretical model, we present and discuss our numerical results, and conclude this work with a short summary. A summary of the expressions used for the Green's function is given in the Appendix A. Note that we use Gaussian electrostatic units.

2. Theory

We use a Cartesian coordinate system with coordinates r = {**R**, z}, where **R** = {x, y} is a position in the $z = 0$ plane. We assume that metal occupies region V_1 defined by $z \leq s(\mathbf{R})$ and its electrostatic response is described by a frequency-dependent relative dielectric constant $\epsilon(\omega)$, whereas the region V_2 defined by $z > s(\mathbf{R})$ is vacuum or air. Here $s(\mathbf{R})$ is a random function that describes the profile of the metal surface, which is assumed to have a zero mean,

$$\langle s(\mathbf{R}) \rangle = 0, \tag{1}$$

where $\langle \cdots \rangle$ stands for an ensemble average over all realizations of the metal surface roughness. If we further assume that the surface presents a translationally invariant and isotropic landscape, then the auto-correlation function of the process $s(\mathbf{R})$ may be written as

$$\left\langle s(\mathbf{R}) s\left(\mathbf{R}'\right) \right\rangle = \delta^2 \ C\left(\left\| \mathbf{R} - \mathbf{R}' \right\|\right), \tag{2}$$

where $\delta^2 \equiv \langle s^2(\mathbf{R}) \rangle$ is the variance in the single-point height fluctuations, and $C(R)$ is the reduced auto-correlation function, such that $C(0) = 1$ and $C(R) \to 0$ when $R \to \infty$ with a characteristic length scale $R \sim a$ that describes the longitudinal correlation length of the surface roughness.

Rahman and Maradudin have shown that the Green's function (GF) for the Poisson equation, $G(\mathbf{r}, \mathbf{r}'; t - t')$, in the presence of such surface may be evaluated in a nonperturbative manner assuming that $s(\mathbf{R})$ is a Gaussian process with small magnitude [7,8]. Hence, the ensemble average of the GF retains the translational invariance of the surface, $G(\mathbf{r}, \mathbf{r}'; t - t') = G(\mathbf{R} - \mathbf{R}', z, z'; t - t')$, allowing one to define a Fourier transform (FT) of the GF (FTGF) with respect to position **R** and time t, $\widetilde{G}(\mathbf{Q}, z, z'; \omega)$, via

$$G\left(\mathbf{R} - \mathbf{R}', z, z'; t - t'\right) = \int \frac{d^2 \mathbf{Q}}{(2\pi)^2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} e^{i\mathbf{Q} \cdot (\mathbf{R} - \mathbf{R}') - i\omega(t - t')} \widetilde{G}\left(\mathbf{Q}; z, z'; \omega\right), \tag{3}$$

where $\mathbf{Q} = \{Q_x, Q_y\}$ is a two-dimensional (2D) wavevector. When the source point z' is in the region V_2, we may write the FTGF as $\widetilde{G}\left(\mathbf{Q}, z, z'; \omega\right) = \widetilde{G}C\left(Q, z, z'\right) + \widetilde{G}_{\text{ind}}\left(\mathbf{Q}, z, z'; \omega\right)$, where $\widetilde{G}C\left(Q, z, z'\right) = (2\pi/Q)e^{-Q|z - z'|}$ corresponds to the bare Coulomb interaction, whereas the screened interaction by the induced charge in the metal surface may be written as [7,8]

$$\widetilde{G}_{\text{ind}}\left(\mathbf{Q}; z, z', \omega\right) = \frac{2\pi}{Q} e^{-Qz} \begin{cases} A(Q, \omega) e^{-Qz} & \text{for } z \in V_2, \\ B(Q, \omega) e^{Qz} & \text{for } z \in V_1, \end{cases} \tag{4}$$

where $Q = \sqrt{Q_x^2 + Q_y^2}$, and the coefficients $A(Q, \omega)$ and $B(Q, \omega)$ are listed in the Appendix A. We note that the statements $z \in V_{1,2}$ strictly mean $z < \min\{s(\mathbf{R})\}$ and $z > \max\{s(\mathbf{R})\}$, respectively, but we shall assume that the point z may be considered "sufficiently" far outside the rough region of the surface if $|z| \gtrsim 2 \, \delta$. In the limit of a flat surface with $\delta = 0$, the coefficients A and B are simply reduced to

$$A_0(\omega) = \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \tag{5}$$

$$B_0(\omega) = \frac{2}{1 + \epsilon(\omega)}. \tag{6}$$

We further assume that N point-charge particles with charges $Z_j e$ (where $e > 0$ is the proton charge and $j = 1, 2, ..., N$) move with the same velocity **v** parallel to the $z = 0$ plane. Thus, their distribution may be described by a time-dependent density $\rho(\mathbf{r}, t) = \rho_0(\mathbf{R} - \mathbf{v}t, z)$, where $\rho_0(\mathbf{R}, z)$ is the distribution in their own moving frame of reference. This picture corresponds to a distribution of ionic fragments resulting from Coulomb explosion of a cluster grazingly scattered from a metal surface, where the relative motion of the fragments with respect to each other may be treated as adiabatic within the moving frame of reference [16]. Denoting the position of the jth particle in that frame by $\mathbf{r}_j = \{\mathbf{R}_j, z_j\}$ with $z_j \in V_2$, we may write

$$\rho_0(\mathbf{R}, z) = e \sum_{j=1}^{N} Z_j \delta\left(\mathbf{r} - \mathbf{r}_j\right), \tag{7}$$

where $\delta(\mathbf{r} - \mathbf{r}_j) = \delta(\mathbf{R} - \mathbf{R}_j) \delta(z - z_j)$ is a 3D delta function. Therefore, with the induced potential given by

$$\Phi_{\text{ind}}(\mathbf{r}, t) = \int d^3 \mathbf{r}' \int_{-\infty}^{\infty} dt' \ G_{\text{ind}}\left(\mathbf{r}, \mathbf{r}'; t - t'\right) \rho\left(\mathbf{r}', t'\right), \tag{8}$$

we may express the total induced electrostatic (self-) energy of the assembly of N particles as [16]

$$\begin{aligned} U_{\text{ind}} &= \frac{1}{2} \int d^3 \mathbf{r} \ \rho(\mathbf{r}, t) \Phi_{\text{ind}}(\mathbf{r}, t) \\ &= \frac{1}{2} \int dz \int dz' \int \frac{d^2 \mathbf{Q}}{(2\pi)^2} \widetilde{G}_{\text{ind}}\left(\mathbf{Q}, z, z'; \mathbf{Q} \cdot \mathbf{v}\right) \widetilde{\rho}_0^*(\mathbf{Q}, z) \widetilde{\rho}_0\left(\mathbf{Q}, z'\right) \\ &= \frac{1}{2} \int \frac{d^2 \mathbf{Q}}{(2\pi)^2} \frac{2\pi}{Q} \mathcal{F}(\mathbf{Q}) \Re[A(Q, \mathbf{Q} \cdot \mathbf{v})], \end{aligned} \tag{9}$$

where $\widetilde{\rho}_0(\mathbf{Q}, z)$ is a 2D spatial FT of the charge density in Eq. (7), while the charge form factor of the assembly is defined by

$$\begin{aligned} \mathcal{F}(\mathbf{Q}) &= e^2 \left| \sum_{j=1}^{N} Z_j \ e^{-i\mathbf{Q} \cdot \mathbf{R}_j - Qz_j} \right|^2 \\ &= e^2 \sum_{j=1}^{N} Z_j^2 \ e^{-2Qz_j} + e^2 \sum_{j=1}^{N} \sum_{j \neq \ell = 1}^{N} Z_j Z_\ell \ e^{i\mathbf{Q} \cdot (\mathbf{R}_\ell - \mathbf{R}_j)} e^{-q(z_{j + z\ell})}. \end{aligned} \tag{10}$$

In a similar manner, one may also define the rate of energy loss of the assembly of N particles by [26]

$$\begin{aligned} \frac{d\mathcal{E}}{dt} &= -\int d^3 \mathbf{r} \ \rho(\mathbf{r}, t) \frac{\partial}{\partial t} \Phi_{\text{ind}}(\mathbf{R}, z, t) \\ &= -\int \frac{d^2 \mathbf{Q}}{(2\pi)^2} \frac{2\pi}{Q} \mathcal{F}(\mathbf{Q})(\mathbf{Q} \cdot \mathbf{v}) \Im[A(Q, \mathbf{Q} \cdot \mathbf{v})]. \end{aligned} \tag{11}$$

In the final expressions of Eqs. (9) and (11) we have used the symmetry of the coefficient, $A^*(Q, \omega) = A(Q, -\omega)$.

Using Eq. (10) in Eq. (9), we may express the induced self-energy as

$$U_{\text{ind}} = \sum_{j=1}^{N} Z_j^2 V_{\text{im}}\left(z_j\right) + U_{\text{vic}}, \tag{12}$$

where the first term represents independent contributions due to the self-energies of individual particles, which may be expressed in terms