Optics of rough surfaces of metals

A. M. Brodskiĭ and M. I. Urbakh

A. N. Frumkin Electrochemical Institute, USSR Academy of Sciences
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The effect of surface plasmons (SP) on the optical properties of rough surfaces of metals is considered. The angular and frequency dependences of the radiation produced upon decay of SP and in diffuse scattering of light, as well as the photoemission current from a rough metal surface, are calculated. It is shown that the foregoing effects can become considerably enhanced if the SP damping lengths governed by their decay in vacuum and absorption in the metal greatly exceed the damping length connected with transitions to other SP states. The possibility of comparing the theoretical calculations with experiment are considered.

1. INTRODUCTION

The optical properties of rough metal surfaces have attracted much attention recently. The rapid pace of the study of this subject was prompted by the discovery of new phonomena connected with field enhancement at rough surfaces, such as the anomalously enhanced Raman scattering by adsorbed molecules and the enhancement of second-harmonic generation.[1,2] A similar enhancement was observed in the luminescence of molecules on the surface and in metal- semiconductor-metal tunnel junctions,[3] as well as in photoemission.[4] Advances in experimental techniques have made possible also detailed studies of the characteristics of optical surface excitation on rough surfaces, such as the dispersion law, decay, and the role in diffuse scattering of light. The basic result was obtained in Ref. 6, where it was shown that in the presence of roughnesses the spectrum of surface plasmons (SP) on silver undergoes not merely a shift, but a double splitting. It was concluded from the experimental and theoretical research that in those cases when rather long-lived surface excitations can exist, a description of the optical properties of the interface, in lowest order in the interaction between the light and the roughnesses, leads to qualitatively incorrect results. In particular, when a finite number of the perturbation-theory-series terms is retained, it is impossible to explain the form of the angular distribution of the radiation produced by the SP decay.[7] In addition, the corrections to the polarization operator that corresponds to the nth term of the perturbation-theory series are proportional to $[\varepsilon(\omega)+1]^{-n}$, where $\varepsilon(\omega)$ is the dielectric constant of the metal.[8,9] As $\varepsilon(\omega)\to -1$ this constant tends to infinity. A unique strong coupling between the field and the roughness sets in therefore, and it becomes necessary accordingly to sum some infinite sequence of the series terms.

We develop in this paper, for the description of light scattering by rough surfaces of metals, a method that makes no use of perturbation theory in the interaction between the light and the roughnesses. The field intensities near the surface and in the waves reflected from the metal we use a formalism based on representation of the Green's functions for the electromagnetic field in the form of functional (path) integrals. The problem of statistical averaging over the roughnesses is solved by the faster method of introducing functional integrals with respect to the so-called superfields, which include simultaneously commuting and anticommuting variables, in analogy with the procedure used in the similar, in many respects, problem of two-dimensional conductivity in a random medium.[10] We calculate in the framework of this method the angular distributions for the emission of SP and for diffuse scattering of light, as well as the field enhancement near the surface. It must be noted that many papers devoted to the description of the influence of roughnesses on the optical properties contain errors due to incorrect use, in the calculations, of field combinations that are discontinuous on the surface.[11,12] These difficulties are avoided here by using equations reconstructed in accordance with the scheme of Ref. 8 and containing only combination of field components and derivatives that vary slowly on the surface.

2. INITIAL EQUATIONS

The surface of a metal occupying the half-space $x_1 < 0$ is described by the function

$$x_1=\xi(\mathbf{x}_\parallel),\qquad \mathbf{x}_\parallel=(x_2,\ x_3). \tag{1}$$

We assume that this function obeys a Gaussian distribution, and the mean values designated by the angle brackets satisfy the equations

$$\begin{gathered}\langle\xi\rangle=0,\qquad \langle\xi(\mathbf{k})\xi(\mathbf{k}')\rangle=(2\pi)^2\delta(\mathbf{k}+\mathbf{k}')\overline{\xi^2}g(k),\\ g(k)=\pi a^2\exp(-a^2k^2/4),\\ \xi(\mathbf{k})=\int d^2\mathbf{x}_\parallel\xi(\mathbf{x}_\parallel)\exp(-i\mathbf{k}\mathbf{x}_\parallel),\end{gathered} \tag{2}$$

where $\mathbf{k}=(k_2,k_3)$ is a two-dimensional vector, $(\overline{\xi}^2)^{1/2}$ is the mean squared height of the roughness, and a is the correlation length. The electric field $\mathbf{E}(x,t)$ is real and satisfies the Gauss relation

$$\operatorname{div}\varepsilon(\mathbf{x},\omega)\mathbf{E}(\mathbf{x},\omega)=0. \tag{3}$$

$\varepsilon(\mathbf{x},\omega)$ in (3) denotes the dielectric function of the investigated system at the frequency ω. We confine ourselves for simplicity to a local dielectric function $\varepsilon(\mathbf{x},\omega)$ in the form

$$\varepsilon(\mathbf{x},\omega)\equiv\varepsilon(x_1-\xi(\mathbf{x}_\parallel),\omega)\to\begin{cases}1 & \text{at}\quad x_1-\xi(\mathbf{x}_\parallel)>\delta\\ \varepsilon(\omega) & \text{at}\quad x_1-\xi(\mathbf{x}_\parallel)<-\delta\end{cases}, \tag{4}$$

which corresponds to a smooth transition, over atomic distances σ, from the dielectric constant of the metal to the dielectric constant of the outer medium. We can neglect in

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this case the spatial dispersion far from the bulk-plasmon frequency interval.[8,3] To shorten the intermediate calculations and to make them more lucid, we assume for the time being that there is no absorption in the system under consideration, i.e., $\operatorname{Im}\varepsilon(\mathbf{x},\omega)=0$. The final expressions for the observed quantities will be written for arbitrary $\operatorname{Im}\varepsilon(\mathbf{x},\omega)$. In the calculations that follow we consider only electromagnetic-field components corresponding to p-polarization, since they make the main contribution to the field intensity near the surface, and hence to all the observable quantities considered in this paper. Additional allowance for the field components corresponding to s-polarization does not lead to significant complications. Under the assumptions made, the vector field $E(x_1,\mathbf{k},\omega)$ can be expressed in terms of a scalar function $a(x_1,\mathbf{k},\omega)$ as follows[8]:

$$\mathbf{E}(x_1,\mathbf{k},\omega)=\int\frac{d^2\mathbf{k}'}{(2\pi)^2}\,\varepsilon^{-1}(x_1,\mathbf{k}-\mathbf{k}',\omega)\,\mathbf{e}_p(k_1,\mathbf{k}')\,a(x_1,\mathbf{k}',\omega),\tag{5}$$

where

$$e_p^{\,i}(k_1,\mathbf{k})=\frac{1}{k}\,(k_1k_i-\delta_{i1}(k_1^2+k^2)),\qquad i=1,2,3;$$
$$\varepsilon^{-1}(x_1,\mathbf{k},\omega)=\int d^2\mathbf{x}_{\parallel}\varepsilon^{-1}(\mathbf{x},\omega)\exp(-i\mathbf{k}\mathbf{x}_{\parallel}),\qquad k_1=-i\frac{\partial}{\partial x_1}.\tag{6}$$

We introduce, as proposed in Ref. 14, a complete set of eigenfunctions $a^0(x_1,\omega,\mathbf{k},\tilde\omega)$ that satisfy the following one-dimensional equation

$$\left[\frac{d}{dx_1}\varepsilon_0^{-1}(x_1,\omega)\frac{d}{dx_1}-k^2\varepsilon_0^{-1}(x_1,\omega)\right]a^0(x_1,\omega,\mathbf{k},\tilde\omega)=-\frac{\tilde\omega^2}{c^2}a^0(x_1,\omega,\mathbf{k},\tilde\omega),\tag{7}$$

which has the structure of the equation for a p-polarized field in the absence of roughnesses. We have introduced in (7) the parameter $\tilde\omega^2$ that has the meaning of an eigenvalue. By $\varepsilon_0(x_1,\omega)$ we denote the function $\varepsilon(\mathbf{x},\omega)$ defined by Eq. (4) at $\xi\equiv0$. It is convenient to choose solutions $a^0(x_1,\omega,k,\tilde\omega)$ that are real and are normalized as follows:

$$\int dx_1a_r^{\,0}(x_1,\omega,k,\tilde\omega)\,a_r^{\,0}(x_1,\omega,k,\tilde\omega')=\delta(\tilde\omega^2-\tilde\omega'^2),$$
$$\int dx_1a_{sp}^{0}(x_1,\omega,k,\tilde\omega)\,a_{sp}^{0}(x_1,\omega,k,\tilde\omega')=\delta_{\tilde\omega^2,\tilde\omega'^2},\tag{8}$$
$$\int dx_1a_{sp}^{\,0}(x_1,\omega,k,\tilde\omega)\,a_r^{\,0}(x_1,\omega,k,\tilde\omega')=0.$$

The subscripts sp and r denote here solutions corresponding respectively to the discrete and continuous spectrum. We assume here that the inequality[1)] $\varepsilon(\omega)<0$ is satisfied, so that a continuous spectrum exists at values of $\tilde\omega^2$ located in a region $\mathfrak{M}$ that consists of two intervals: $\tilde\omega^2>k^2c^2$ and $\tilde\omega^2<k^2c^2/\varepsilon(\omega)$. We assume for simplicity that the discrete spectrum has only one branch:

$$\tilde\omega_{sp}^2=\tilde\omega_{sp}^2(\omega,k).\tag{9}$$

In particular, in the case of an abrupt boundary,

$$\tilde\omega_{sp}^2=k^2c^2\gamma(\omega),\qquad\gamma(\omega)=\frac{\varepsilon(\omega)+1}{\varepsilon(\omega)}.\tag{10}$$

From the asymptotic form of the solutions $a_{sp}{}^0(x_1,\omega,\mathbf{k},\tilde\omega)$ as $x_1\to\pm\infty$ it follows that the condition for the existence of a discrete spectrum in (7) is the inequality $\varepsilon(\omega)<0$, i.e., the frequency ω must be lower than the frequency ω_p of the bulk plasmons.

We expand the function $a(\mathbf{x},\omega)$ of (5) in terms of the solutions of Eq. (7):

$$a(\mathbf{x},\omega)=\sum_{\alpha=sp,r}\int d\Omega_\alpha b_\alpha(\Omega_\alpha)\,a_\alpha{}^0(x_1,\omega,\Omega_\alpha)\exp(i\mathbf{k}\mathbf{x}_{\parallel}).\tag{11}$$

We have introduced here the notation

$$\Omega_{sp}=\{\mathbf{k}\},\qquad\Omega_r=\{\mathbf{k},\tilde\omega\},$$
$$\int d\Omega_{sp}=\int\frac{d^2\mathbf{k}}{(2\pi)^2},\qquad\int d\Omega_r=\int\frac{d^2\mathbf{k}}{(2\pi)^2}\int_{\mathfrak{M}}d\tilde\omega^2.\tag{12}$$

Substituting (5) and (11) in the equations for the field $\mathbf{E}(\dot{\mathbf{x}},\omega)$ we arrive at a system of equations for the quantities $b_\alpha(\Omega_\alpha)$:

$$b_\alpha(\Omega_\alpha)(\omega^2-\tilde\omega_\alpha^2)+\sum_{\beta=sp,r}\int d\Omega_\beta\xi(\mathbf{k}-\mathbf{k}')f_{\alpha\beta}(\Omega_\alpha,\Omega_\beta)\,b_\beta(\Omega_\beta)=0,\tag{13}$$
$$\tilde\omega_r^2=\tilde\omega^2.$$

Here

$$f_{\alpha\beta}=c^2\int dx_1\left\{\frac{(\mathbf{k}\mathbf{k}')}{kk'}\frac{\delta\varepsilon(x_1,\mathbf{k}-\mathbf{k}')}{\xi(\mathbf{k}-\mathbf{k}')}\times\left[\varepsilon_0^{-1}(x_1)\frac{d}{dx_1}a_\alpha{}^0(x_1,\omega,\Omega_\alpha\right]\times\left[\varepsilon_0^{-1}(x_1)\frac{d}{dx_1}a_\beta{}^0(x_1,\omega,\Omega_\beta)\right]-kk'\frac{\delta\varepsilon^{-1}(x_1,\mathbf{k}-\mathbf{k}')}{\xi(\mathbf{k}-\mathbf{k}')}a_\alpha{}^0(x_1,\omega,\Omega_\alpha)\,a_\beta{}^0(x_1,\omega,\Omega_\beta)\right\}.\tag{14}$$

We have introduced in (14) the quantities $\delta\varepsilon(x_1,\mathbf{k})$ and $\delta\varepsilon^{-1}(x_1,\mathbf{k})$, which differ from zero only at atomic distances (along the coordinate x_1) from the surface and are given by

$$\delta\varepsilon(x_1,\mathbf{k})=\varepsilon(x_1,\mathbf{k})-(2\pi)^2\delta(\mathbf{k})\,\varepsilon_0(x_1)\approx-\xi(\mathbf{k})\frac{d}{dx_1}\varepsilon_0(x_1),$$
$$\delta\varepsilon^{-1}(x_1,\mathbf{k})=\varepsilon^{-1}(x_1,\mathbf{k})-(2\pi)^2\delta(\mathbf{k})\,\varepsilon_0^{-1}(x_1)\approx-\xi(\mathbf{k})\frac{d}{dx_1}\varepsilon_0^{-1}(x_1).\tag{15}$$

The reason for retaining in (15) only terms of first order in $\xi(\mathbf{k})$ is that addition to $\delta\varepsilon$ and $\delta\varepsilon^{-1}$ of terms of second order in $\xi(\mathbf{k})$ can be reduced (in the first nonvanishing approximation in the interaction with the roughnesses) to a change of the real components of the quantities $\tilde\omega^2$ and $\tilde\omega_{sp}^2$ in Eqs. (13). The resultant additional effects reduce to an insignificant shift of the surface-plasmon spectrum and to a change of the coefficient of specular reflection of the light.

By virtue of the properties (7), the functions $a_\alpha{}^0(x_1,\omega,\Omega_\alpha$ and $\varepsilon_0^{-1}(x_1)(d/dx_1)a_\alpha^0(x_1\omega,\Omega_\alpha)$ vary slowly with x_1. We expand these functions in power of x_1 at the point $x_1=0$ under the integral sign in (14), where they are multiplied by the quantities $\delta\varepsilon(x_1, \mathbf{k})$ and $\delta\varepsilon^{-1}(x_1, \mathbf{k})$. Retaining in (15) only the principal terms of the expansion in the parameter $\delta\omega/c$, we obtain the following expressions for the values of (14) that will be needed later on:

$$f_{sp,sp}(\mathbf{k}, \mathbf{k}') = 2c^2(-\varepsilon(\omega))^{-1/2}(kk')^{1/2}[-(\mathbf{k}\mathbf{k}')+kk'],$$

$$f_{sp,r}(\mathbf{k}, \mathbf{k}', \tilde\omega) = \frac{\sqrt{2}\, ck^{1/2}}{\pi^{1/2}[k'^2(\varepsilon+1)-\tilde\omega^2\varepsilon/c^2]^{1/2}}\left(\frac{\tilde\omega^2}{c^2}-k'^2\right)^{1/4}\left[-kk'(-\varepsilon)^{1/4} + \frac{(\mathbf{k}\mathbf{k}')}{k'}\left(k'^2-\frac{\tilde\omega^2}{c^2}\varepsilon\right)^{1/2}(-\varepsilon)^{-1/4}\right], \quad \tilde\omega^2>c^2k'^2;$$

$$f_{r,r}(\mathbf{k}, \tilde\omega, \mathbf{k}', \tilde\omega') = \pi^{-1}\left[k^2(\varepsilon+1)-\frac{\tilde\omega^2}{c^2}\varepsilon\right]^{-1/2}\left[k'^2(\varepsilon+1)-\frac{\tilde\omega'^2}{c^2}\varepsilon\right]^{-1/2} \times\left(\frac{\tilde\omega^2}{c^2}-k^2\right)^{1/4}\left(\frac{\tilde\omega'^2}{c^2}-k'^2\right)^{1/4} \times\left[-\varepsilon kk' - \frac{(\mathbf{k}\mathbf{k}')}{kk'}\left(k^2-\frac{\tilde\omega^2}{c^2}\varepsilon\right)^{1/2}\left(k'^2-\frac{\tilde\omega'^2}{c^2}\varepsilon\right)^{1/2}\right]$$

$$\tilde\omega^2>c^2k^2, \quad \tilde\omega'^2>c^2k'^2. \tag{16}$$

Here $a^0_\alpha(x_1, \omega, \Omega_\alpha)$ and $\varepsilon_0^{-1}(x_1)(d/dx_1)a^0_\alpha(x_1, \omega, \Omega_\alpha)$ replace, at the accuracy considered, the quantities obtained by solving the abrupt-boundary problem. Use of the reasoning that led to (16) raises no problems connected with the series expansions of the functions $\delta\varepsilon(x_1, \mathbf{k})$ and $\delta\varepsilon^{-1}(x_1, \mathbf{k})$ at singular points or with the appearance of products of singular functions in the integrals. Such problems are encountered, e.g., in Refs. 11 and 12.

The formal solution of (13) can be represented in the form

$$b_\alpha(\Omega_\alpha) = b_\alpha{}^0(\Omega_\alpha) - \sum_{\beta,\gamma=sp,r}\int d\Omega_\beta\, d\Omega_\gamma \times\{D_{\alpha\beta}(\Omega_\alpha, \Omega_\beta)\,\xi(\mathbf{k}'-\mathbf{k}'')\,f_{\beta\gamma}(\Omega_\beta, \Omega_\gamma)\,b_\gamma{}^0(\Omega_\gamma)\}. \tag{17}$$

The quantities

$$b_{sp}{}^0(\mathbf{k}) = (2\pi)^2\delta(\mathbf{k}-\mathbf{k}_0)\,b_{sp}{}^0$$

$$\text{and } b_r{}^0(\mathbf{k}, \tilde\omega) = (2\pi)^2\delta(\mathbf{k}-\mathbf{k}_0)\,\delta(\tilde\omega^2-\omega^2)\,b_r{}^0$$

in (17) denote the coefficients of the expansion of $a(\mathbf{x}, \omega)$ in (11) at $\xi = 0$, while $D_{\alpha\beta}$ denote the Green's functions for Eqs. (13). The equations for $D_{\alpha\beta}$ are

$$D_{\alpha\beta}(\Omega_\alpha, \Omega_\beta) = D_\alpha{}^0(\Omega_\alpha)\,\delta_{\alpha\beta} - \sum_{\gamma=r,sp}\int d\Omega_\gamma \times\{D_{\alpha\gamma}(\Omega_\alpha, \Omega_\gamma)\,\xi(\mathbf{k}'-\mathbf{k}'')\,f_{\gamma\beta}(\Omega_\gamma, \Omega_\beta)\,D_\beta{}^0\}, \tag{18}$$

where

$$D_{sp}{}^0 = \frac{1}{\omega^2-\tilde\omega_{sp}{}^2-i\eta}, \quad D_r{}^0 = \frac{1}{\omega^2-\tilde\omega^2-i\eta}, \quad \eta\to+0 \tag{19}$$

is the Green's function for a smooth metal surface. The fields $b_\alpha(\Omega_\alpha)$ can be expressed with the aid of (17) and (18) in the form

$$b_{sp}(\mathbf{k}) = D_{sp,sp}(\mathbf{k}, \mathbf{k}_0)\,D_{sp}^{0\,-1}(\mathbf{k}_0)\,b_{sp}{}^0 + D_{sp,r}(\mathbf{k}, \mathbf{k}_0, \omega)\,D_r^{0\,-1}(\mathbf{k}_0, \omega)\,b_r{}^0,$$

$$b_r(\mathbf{k}, \tilde\omega) = D_{r,sp}(\mathbf{k}, \tilde\omega, \mathbf{k}_0)\,D_{sp}^{0\,-1}(\mathbf{k}_0)\,b_{sp}{}^0 + D_{r,r}(\mathbf{k}, \tilde\omega, \mathbf{k}_0, \omega)\,D_r^{0\,-1}(\mathbf{k}_0, \omega)\,b_r{}^0. \tag{20}$$

Equations (20) are analogous to the corresponding scattering-theory equations.[15] They are valid in the absence of eigensolutions of (13) that are localized along the surface. The possible existence and the properties of localized solutions not considered in the present paper is at present under discussion.[16] They certainly do not arise at sufficiently low values of $\bar\xi^2$.

3. EQUATIONS FOR MEASURABLE QUANTITIES

In the optics of rough metal surface, it is of interest to detect observable quantities such as the electromagnetic-field intensity near the surface, or the angular distributions of the radiation produced in diffuse scattering of light and in SP decay. In the parameter interval of interest to us, which corresponds to creation of SP, the surface-wave intensity near the interface exceeds considerably the intensity of the bulk r-waves. We therefore consider hereafter the interaction of the r-waves with one another and with the sp-waves in the first nonvanishing order of perturbation theory. Principal attention will be paid to the description of the interaction between sp-waves.

Using (4), (11), and (20), we can represent the mean value of the electromagnetic field intensity near the surface at $x_1 = \xi(\mathbf{x}_\parallel) + x_1^0$ in the form

$$J_{ii}(x_1{}^0) \equiv \langle E_i(x_1{}^0, \mathbf{x}_\parallel)\,E_i{}^*(x_1{}^0, \mathbf{x}_\parallel)\rangle = |\varepsilon_0{}^{-1}(x_1{}^0)\,e_p{}^i(k_1, \mathbf{k}_0)\,a_r{}^0(x_1{}^0, \omega, \mathbf{k}_0, \omega)\,b_r{}^0|^2 + \frac{1}{S}\int\frac{d^2\mathbf{k}}{(2\pi)^2}|\varepsilon_0{}^{-1}(x_1{}^0)\,e_p{}^i(k_1, \mathbf{k})\,a_{sp}{}^0(x_1{}^0, \omega, k)\,b_r{}^0|^2 \times\langle D_{sp,r}(\mathbf{k}, \mathbf{k}_0, \omega)$$

$$\times D^*_{sp,r}(\mathbf{k}, \mathbf{k}_0, \omega)\rangle \approx |\varepsilon_0{}^{-1}(x_1{}^0)\,e_p{}^i(k_1, \mathbf{k}_0)\,a_r{}^0(x_1{}^0, \omega, \mathbf{k}_0, \omega)\,b_r{}^0|^2 + \frac{1}{S}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{d^2\mathbf{k}'}{(2\pi)^2}|\varepsilon_0{}^{-1}(x_1{}^0)\,e_p{}^i(k_1, \mathbf{k})\,a_{sp}{}^0(x_1{}^0, \omega, k)|^2 \times\langle D_{sp,sp}(\mathbf{k}, \mathbf{k}')$$

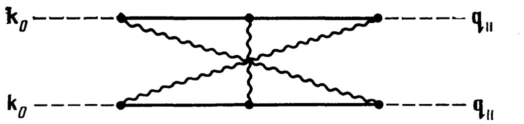
$$\times D^*_{sp,sp}(\mathbf{k}, \mathbf{k}')\rangle\overline{\xi^2}g(|\mathbf{k}'-\mathbf{k}_0|)\,|f_{sp,r}(\mathbf{k}', \mathbf{k}_0, \omega)\,b_r{}^0|^2, \tag{21}$$

where S is the surface area extended to infinity. We obtain similarly an expression for the angular distribution of diffusely scattered light. The time- and surface-averaged energy flux in the $\mathbf{q}/q$ direction, where $\mathbf{q} = \{(\omega^2/c^2 - q_\parallel^2)^{1/2}, \mathbf{q}_\parallel\}$, is equal to

$$P_{diff}(q) \approx \frac{c^2}{8\pi\omega}\frac{\mathbf{q}}{q}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{d^2\mathbf{k}'}{(2\pi)^2}[e_p(k_1, \mathbf{q}_\parallel)[\mathbf{q}e_p(k_1', \mathbf{q}_\parallel)]] \times\int_{m} d\tilde\omega^2\, d\tilde\omega'^2 a_r{}^0(x_1, \omega, q_\parallel, \tilde\omega)\,a_r{}^0(x_1', \omega, q_\parallel, \tilde\omega') \times D_r{}^0(q_\parallel, \tilde\omega)\,D_r{}^{0*}(q_\parallel, \tilde\omega')\,|b_r{}^0|^2 \times\overline{\xi^2}g(\mathbf{q}_\parallel-\mathbf{k})\Big\{f_{rr}(\mathbf{q}_\parallel, \tilde\omega, \mathbf{k}_0, \omega)\,f_{rr}{}^*(\mathbf{q}_\parallel, \tilde\omega', \mathbf{k}_0, \omega)$$

$$\times(2\pi)^4\delta(\mathbf{k}-\mathbf{k}_0)\,\delta(\mathbf{k}'-\mathbf{k}_0)$$
$$+f_{r,sp}(\mathbf{q}_\|,\tilde{\omega},\mathbf{k})\,f_{r,sp}(\mathbf{q}_\|,\tilde{\omega}',\mathbf{k})\,\frac{1}{S}$$
$$\times\langle D_{sp,sp}(\mathbf{k},\mathbf{k}')D_{sp,sp}(\mathbf{k},\mathbf{k}')\rangle\overline{\xi}^2 g(\mathbf{k}'$$
$$-\mathbf{k}_0)\,|f_{sp,r}(k',k_0,\omega)|^2\Big\}_{x_1=x_1'\to\infty}. \tag{22}$$

Here $k_1=-i\partial/\partial x_1$ and $k_1'=-i\partial/\partial x_1'$. In the expression for $\langle D_{rr}D^*_{rr}\rangle$ in (22) we have retained only the terms corresponding to the ladder approximation.[17] It must be noted that when $P_{\text{diff}}(\mathbf{q})$ is calculated in a narrow angle interval in the backward direction at $|k_0+q_\||l_{sp}<1$, where l_{sp} is the SP damping length, account must be taken also of terms corresponding to the so-called fan diagrams for $\langle D_{rr}D^*_{rr}\rangle$, which can be represented in the following manner[18]:



The dashed lines correspond here to the Green's functions D^0_r and D^{0*}_r the solid lines to the Green's functions $\langle D_{sp,sp}\rangle$ and $\langle D^{*p}_{sp,sp}\rangle$, and the wavy lines to the interactions with the roughnesses. At $\mathbf{q}_\|=-\mathbf{k}_0$ the contribution of the fan diagrams to $P_{\text{diff}}(\mathbf{q})$ is equal to the contribution of the ladder diagrams, and allowance for them doubles the second term in (22). Away from the backward direction, the terms corresponding to the fan diagrams decrease rapidly[18] and contain at $|\mathbf{k}_0+\mathbf{q}_\||l_{sp}>1$ an additional factor $(|\mathbf{k}_0+\mathbf{q}_\||l_{sp})^{-2}$ that is small compared with the corresponding terms in (22). The contribution of the fan diagrams is therefore significant only in a very narrow angle interval in the backward direction; this angle does not exceed 1′ at reasonable values of the parameters ($\overline{\xi}^2a^2\approx10^6$ Å^4, $2\pi c/\omega\approx5\cdot10^3$ Å and $\varepsilon\approx5$). It is as yet impossible to measure the intensity of diffusely scattered light at such small deviations from the backward direction. All other terms not included in (22) contain, in the entire range of scattering angles, an additional small factor of order $(k_{sp}l_{sp})^{-2}$ (Refs. 17 and 18), where k_{sp} is the SP wave vector.

The first term of (22), which describes a direct transformation of the incident light into diffusely scattered light, was calculated earlier, for example in Ref. 11, in which the interaction of electromagnetic waves of *sp* and *r* type with roughnesses was taken into account only in the lowest nonvanishing orders of perturbation theory. The first term in (22) describes the creation of surface waves, the transitions between them, and finally their decay into radiation. It will be shown below that in the case of highly reflecting metals the second term, which was not considered in earlier papers, can exceed the first. Finally, the angular distribution of the radiation produced upon decay of SP is expressed as follows:

$$P_{sp}(\mathbf{q})\approx\frac{c^2}{8\pi\omega}\frac{\mathbf{q}}{q}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\Big\{[e_p(k_1,\mathbf{q}_\|)[\mathbf{q}e_p(k_1',\mathbf{q}_\|)]]$$
$$\times\int_m d\tilde{\omega}^2\,d\tilde{\omega}'^2 a_r^0(x_1,\omega,q_\|,\tilde{\omega})\,a_r^0(x_1,\omega,q_\|,\tilde{\omega}')\,D_r^0(\mathbf{q}_\|,\tilde{\omega}')$$
$$\times D_r^{0*}(\mathbf{q}_\|,\tilde{\omega})\,\overline{\xi}^2 g(\mathbf{q}_\|-\mathbf{k})\,f_{r,sp}(\mathbf{q}_\|,\tilde{\omega},\mathbf{k})$$
$$\times f_{r,sp}(\mathbf{q}_\|,\tilde{\omega}',\mathbf{k})\,f^*_{r,sp}(\mathbf{q}_\|,\tilde{\omega}',\mathbf{k})$$
$$\times\frac{1}{S}\langle D_{sp,sp}(\mathbf{k},\mathbf{k}_0)D^*_{sp,sp}(\mathbf{k},\mathbf{k}_0)\rangle\,|D^{0\,-1}_{sp}(\mathbf{k}_0)\,b_{sp}^0|^2\Big\}_{x_1=x_1'\to\infty}. \tag{23}$$

Representing the Green's functions $D_{sp,sp}$ and $D^*_{sp,sp}$ that enter in (21)–(23) as ratios of functional integrals,[19] we can express the quantities $J_{ii}(x_1^0)$, P_{diff}, P_{sp} in terms of the following integral:

$$J=\frac{1}{S}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{d^2\mathbf{k}'}{(2\pi)^2}h_1(\mathbf{k})h_2(\mathbf{k}')$$
$$\times\langle D_{sp,sp}(\mathbf{k},\mathbf{k}')D^*_{sp,sp}(\mathbf{k},\mathbf{k}')\rangle$$
$$=\frac{1}{S}\int D\xi D\hat{b}_{sp}D\hat{b}_r\Big\{\exp\Big[-\int\frac{d^2\mathbf{p}}{(2\pi)^2}\xi(\mathbf{p})\xi^*(\mathbf{p})\frac{1}{\overline{\xi}^2g(p)}\Big]$$
$$\times\int\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{d^2\mathbf{k}'}{(2\pi)^2}h_1(\mathbf{k})h_2(\mathbf{k}')\,b_{sp}^{(1)}(\mathbf{k})\,b_{sp}^{(2)*}(\mathbf{k})\,b_{sp}^{(1)*}$$
$$\times(\mathbf{k}')\,b_{sp}^{(2)}(\mathbf{k}')$$
$$\times\exp[-S(\hat{b}_{sp},\hat{b}_r,\xi)]$$
$$\times Q^{-1}\Big[\int D\hat{b}_{sp}D\hat{b}_r\exp(-S(\hat{b}_{sp},\hat{b}_r,\xi))\Big]^{-1}\Big\}. \tag{24}$$

The actual form of the functions h_1 and h_2 in (24) follows from a comparison of (24) with Eqs. (21)–(34); the functional $S(\hat{b}_{sp},\hat{b}_r,\xi)$ is the effective action, variation of which yields Eqs. (13):

$$S(\hat{b}_{sp},\hat{b}_r,\xi)$$
$$=i\sum_{n=1}^{2}\Big\{\int\frac{d^2\mathbf{k}}{(2\pi)^2}b_{sp}^{(n)*}(\mathbf{k})\,[(\omega^2-\tilde{\omega}_{sp}^2(k,\omega))(-1)^{n+1}-i\eta]$$
$$\times b_{sp}^{(n)}(\mathbf{k})+\int\frac{d^2\mathbf{k}}{(2\pi)^2}\int_m d\tilde{\omega}^2 b_r^{(n)*}(\mathbf{k},\tilde{\omega})$$
$$\times[(\omega^2-\tilde{\omega}^2)(-1)^{n+1}-i\eta]\,b_r^{(n)}(\mathbf{k},\tilde{\omega})$$
$$+(-1)^{n+1}\sum_{\alpha,\beta=sp,r}\Delta S_{\alpha,\beta}(b_{sp}^{(n)},b_r^{(n)},\xi)\Big\},\qquad\eta\to+0,$$

$$\Delta S_{\alpha,\beta}=\int d\Omega_\alpha\,d\Omega_\beta\,b_\alpha^{(n)*}(\Omega_\alpha)\,b_\beta^{(n)}(\Omega_\beta)\,\xi(-\mathbf{k}-\mathbf{k}')\,f_{\alpha\beta}(\Omega_\alpha,\Omega_\beta),$$
$$\hat{b}=\begin{pmatrix}b^{(1)}\\ b^{(2)}\end{pmatrix},\qquad b^{(n)*}(\mathbf{k})=\int d^2\mathbf{x}_\|\,b^{(n)*}(\mathbf{x}_\|)\exp(-i\mathbf{k}\mathbf{x}_\|), \tag{25}$$
$$Q=\int D\xi\exp\Big[-\int\frac{d^2\mathbf{k}}{(2\pi)^2}\xi(\mathbf{k})\xi^*(\mathbf{k})\frac{1}{\overline{\xi}^2g(k)}\Big].$$

The indices 1 and 2 in (25) pertain respectively to the Green's functions $D_{sp,sp}$ and $D^*_{sp,sp}$. The functional integration with respect to Db in (24) is defined in accordance with Ref. 19, and it can be verified that (24) is the inverted perturbation-theory series. The functional integrals (24) are calculated in the Appendix by introducing the superfields. As a result, if the inequalities

$$l_{sp}k_{sp}\gg1,\quad k_{sp}a<1\quad\text{и}\quad\overline{\xi}^2/a^2<\gamma^2(\omega) \tag{26}$$

are satisfied, we obtain

$$J=[4\pi c^2\gamma(\omega)]^{-2}|k_{sp}(\omega)|^{-1}$$

$$\times\sum_{l=-2}^{2}\frac{1}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(sp)})^{-1}(\varkappa_0-\varkappa_l)/\varkappa_0}$$

$$\times\int_0^{2\pi}d\varphi\,d\varphi'\,h_1(k_{sp}\hat{\mathbf{k}})h_2(k_{sp}\hat{\mathbf{k}}')\exp[il(\varphi-\varphi')];\qquad(27)$$

$$\hat{\mathbf{k}}=(\cos\varphi,\sin\varphi),\quad\hat{\mathbf{k}}'=(\cos\varphi',\sin\varphi'),\quad k_{sp}^2=\frac{\omega^2}{c^2}\gamma^{-1}(\omega).$$

Here $\varkappa_0=3/2$, $\varkappa_{\pm1}=-2$, $\varkappa_{\pm2}=1/2$, while the quantities $l_{sp}^{(r)}$ and $l_{sp}^{(sp)}$ have the meaning of the SP damping lengths due respectively to radiation into the vacuum and to transitions into other SP states. If (26) is satisfied, these quantities can be expressed in the form[8,12]

$$(l_{sp}^{(sp)})^{-1}\approx\frac{3}{2}\pi\overline{\xi}^2a^2\frac{\omega^5}{c^5}\frac{|\varepsilon|^{7/2}}{|\varepsilon+1|^{9/2}},$$
$$(l_{sp}^{(r)})^{-1}\approx\frac{1}{2}\overline{\xi}^2a^2\frac{\omega^5}{c^5}\frac{|\varepsilon|^{3/2}}{|\varepsilon+1|^3}\left[-\frac{2}{3}|\varepsilon|-\frac{1}{3}+\frac{3|\varepsilon|^2}{|\varepsilon+1|}(1-|\varepsilon+1|^{-1/2}\operatorname{arctg}|\varepsilon+1|^{1/2})\right].\qquad(28)$$

We emphasize that the cancellation of $l_{sp}^{(sp)}$ in the denominator of the term (27) with $l=0$ is due to the following exact relation (sum rule):

$$(l_{sp}^{(sp)})^{-1}=\frac{1}{c^2\gamma|k_{sp}|}\int\frac{d^2\mathbf{k}'}{(2\pi)^2}\frac{d^2\mathbf{k}''}{(2\pi)^2}U(k_{sp}\hat{\mathbf{k}},\mathbf{k}')\times\operatorname{Im}\langle D_{sp,sp}(\mathbf{k}',\mathbf{k}'')\rangle\times(1+O(l_{sp}^{(sp)}/l_{sp}^{(r)})).\qquad(29)$$

$U(\mathbf{k},\mathbf{k}')$ denotes here the kernel of an equation of the Bethe-Salpeter type[8,17] for $\langle D_{sp,sp}(\mathbf{k},\mathbf{k}')D^*_{sp,sp}(\mathbf{k},\mathbf{k}')\rangle$. The validity of (29) can be verified by comparing the sequence of irreducible diagrams for $l_{sp}^{(sp)}$ and $U(\mathbf{k},\mathbf{k}')$, as is done when considering the conductivity of random media.[20] We note that expression (27) for the integral J can also be obtained by directly summing the ladder sequence of diagrams in the crossing technique.[17]

4. COMPARISON WITH EXPERIMENT

The results of the preceding sections allow us to write down formulas for various measured quantities. Thus, taking into account the explicit form of the functions $a_\alpha^0(x_1,\omega,\Omega_\alpha)$, we obtain from (22) and (27) the following expression for the partial radiation intensity $P_{\text{diff}}(\theta_f,\varphi_f)$, normalized to the incident flux, into a solid angle $d\Omega_f=\sin\theta_f d\theta_f\,d\varphi_f$ for diffuse scattering of light:

$$P_{diff}(\theta_f,\varphi_f)=\frac{|\varepsilon-1|^2}{16\pi\cos\theta}\frac{\omega^4}{c^4}\overline{\xi}^2a^2\exp\left[-\frac{a^2}{4}(\mathbf{q}_\parallel-\mathbf{k}_0)^2\right]|(1+r_p(q_\parallel))\times(1+r_p(k_0))\cos\theta\cos\theta_f\cos\varphi_f-\varepsilon^{-1}\times(1-r_p(q_\parallel))(1-r_p(k_0))\sin\theta\sin\theta_f|^2$$
$$+\frac{|\varepsilon-1|^2}{24\pi\cos\theta}\frac{\omega^4}{c^4}\overline{\xi}^2a^2\left\{\left[\left[|\varepsilon|^{-1}|1-r_p(q_\parallel)|^2\sin^2\theta_f+\frac{1}{2}|1+r_p(q_\parallel)|^2\times\cos^2\theta_f\right]\right]\left[|\varepsilon|^{-1}|1-r_p(k_0)|^2\sin^2\theta+\frac{1}{2}|1+r_p(k_0)|\cos^2\theta\right]\frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}}\right.$$
$$+2|\varepsilon|^{-1}\operatorname{Im}r_p(q_\parallel)\operatorname{Im}r_p(k_0)\sin2\theta\sin2\theta_f\cos\varphi_f\times\frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}+{}^7/_3(l_{sp}^{(sp)})^{-1}}$$
$$\left.+\frac{1}{8}|1+r_p(q_\parallel)|^2|1+r_p(k_0)|^2\cos^2\theta\times\cos^2\theta_f\cos2\varphi_f\frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}+{}^2/_3(l_{sp}^{(sp)})^{-1}}\right\}$$
$$\times\exp\left[-\frac{a^2}{4}(k_0^2+q_\parallel^2+2k_{sp}^2)\right]\times I_0\left(\frac{1}{2}a^2k_0k_{sp}\right)I_0\left(\frac{1}{2}a^2q_\parallel k_{sp}\right).\qquad(30)$$

Here

$$\mathbf{k}_0=\frac{\omega}{c}(0,\sin\theta,0)$$

and $\mathbf{q}_\parallel=\frac{\omega}{c}(0,\sin\theta_f\cos\varphi_f,\sin\theta_f\sin\varphi_f)$

are the projections of the wave vectors in the wave incident on the metal and in the scattered wave on the plane of the surface, $I_0(z)$ is a modified Bessel function, $r_p(k)$ is the amplitude of the coefficient of reflection of p-polarized light from a smooth metal surface, calculated from Fresnel's formulas, θ_f is an angle in the incidence plane and measured from the normal to the surface, and the quantity

$$l_{sp}^{(m)}=\left[2\frac{\omega}{c}\operatorname{Im}\left(\frac{\varepsilon}{\varepsilon+1}\right)^{1/2}\right]^{-1}$$

has the meaning of the SP damping length due to absorption in the metal.

An expression for the intensity $P_{sp}(\theta_f,\varphi_f)$ of the radiation due to the SP decay and propagating along the surface with a wave vector k_{sp} and an amplitude $\mathscr{E}_{sp}$, is similar in form:

$$P_{sp}(\theta_f,\varphi_f)=\frac{|\varepsilon-1|^2}{4^3\pi^2|\varepsilon+1|}\frac{\omega^4}{c^3}\overline{\xi}^2a^2\times\exp\left[-\frac{a^2}{4}(q_\parallel-k_{sp})^2\right]|\mathscr{E}_{sp}|^2|i(1+r_p(q_\parallel))\times\sin\theta_f\cos\varphi_f+(-\varepsilon)^{-1/2}(1-r_p(q_\parallel))\cos\theta_f|^2$$
$$+\frac{|\varepsilon-1|^2}{4^3\pi^2|\varepsilon+1|}\frac{\omega^4}{c^3}\overline{\xi}^2a^2|\mathscr{E}_{sp}|^2\times\exp\left[-\frac{a^2}{4}(q_\parallel^2+k_{sp}^2)\right]I_0\left(\frac{1}{2}a^2q_\parallel k_{sp}\right)\times\left\{[|\varepsilon|^{-1}|1-r_p(q_\parallel)|^2\cos^2\theta_f+\frac{1}{2}|1+r_p(q_\parallel)|^2\sin^2\theta_f]\frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}}-\frac{8}{3}|\varepsilon|^{-1/2}\right.$$

$$\times \operatorname{Im} r_p(q_\parallel) \sin 2\theta_f$$

$$\times \cos\varphi_f \frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}+{}^7/_3(l_{sp}^{(sp)})^{-1}}$$

$$+\frac{1}{6}|1+r_p(q_\parallel)|^2 \sin\theta_f \cos 2\varphi_f$$

$$\times \left.\frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}+{}^2/_3(l_{sp}^{(sp)})^{-1}}\right\} . \qquad (31)$$

In (31), θ_f is an angle in the SP incidence plane and measured from the SP propagation direction. The first terms in (30) and (31) describe a direct transformation of the light incident on the metal and of the SP into scattered light. The second terms of (30) and (31) take into account, respectively, the possibility of transformation of the incident light into SP, transitions between various SP states, and their subsequent decay into vacuum. These terms do not arise if only the first terms of the perturbation-theory series are retained. Therefore the formulas used up to now in the literature[7,8,11,12] to calculate P_{diff} and P_{sp} comprised the first two terms of (30) and (31). In the case $(l_{sp}^{(sp)})^{-1} > (l_{sp}^{(r)})^{-1} + (l_{sp}^{(m)})^{-1}$, however (these inequalities are valid, e.g., for silver in the interval 2.5 eV $< \hbar\omega <$ 3.5 eV), the second terms of (30) and (31) are larger than the first. The frequency and angular dependences of the first and second terms of (30) and (31) are different, so that the contributions of the different processes to the scattered-light intensity can be separated. It follows from (30), in particular, that allowance for only effects connected with creation and rescattering of SP can lead to the abrupt decrease, observed in Ref. 21, of the scattered-light intensity when the limiting SP frequency (more accurately, the limiting frequencies at which $k_{sp}a > 1$) is approached from below. The first term of (31) as a function of θ_f has, at fixed φ_f, one maximum as θ_f varies from 0 to 180°. The second term of (31) has two maxima symmetric about the normal. This picture is due to the fact that SP with different wave-vector directions exist on the surface on a par if $(l_{sp}^{(sp)})^{-1} > (l_{sp}^{(2)})^{-1} + (l_{sp}^{(m)})^{-1}$. Plots of $P_{sp}(\theta_f)$ with two maxima were observed in a study[7] of the decay of SP on silver, and found heretofore no natural explanation.

Attention must also be called to one more difference between our results and those of Refs. 11 and 12 and used to reduce the experimental data. The calculations carried out in Refs. 11 and 12, where only the first terms of the perturbation-theory series are retained, lead to the conclusion that even as $\operatorname{Im}\varepsilon(\omega) \to 0$ the SP are absorbed in the surface layer of the metal, and that at $|\varepsilon(\omega)| \sim 1$ this effect makes the principal contribution to the change, due to roughnesses, of the specular-reflection coefficient. Our results show that, as expected in the case $\operatorname{Im}\varepsilon(\omega) \to 0$, the SP cannot be absorbed in a metal, but allowance for the transitions between the various SP states leads to a corresponding change of the specular-reflection coefficient.

Equation (27) yields also an expression for the field intensity Jiinear the surface, defined in (21). For the normal field components, for example, it takes the form

$$J_{11}(x_1^0) = |\varepsilon_0(x_1^0)|^{-2} |D_1(0,k_0)|^2$$

$$+\frac{1}{3}|\varepsilon|\,|\varepsilon_0(x_1^0)|^{-2} \frac{(l_{sp}^{(sp)})^{-1}}{(l_{sp}^{(r)})^{-1}+(l_{sp}^{(m)})^{-1}}$$

$$\times [|E_\parallel(0,k_0)|^2 - 2|\varepsilon|^{-1}|D_1(0,k_0)|^2], \qquad k_{sp}a<1. \qquad (32)$$

Here $E_\parallel(0, k_0) = -[1 + r_p(k_0)]\sin\theta\mathscr{E}_p$ and $D_1(0, k_0) = [1 - r_p(k_0)]\cos\theta\mathscr{E}_p$ are the intensities of the tangential field component and the normal induction component at the surfaces, calculated for the case of a smooth metal surface, and $\mathscr{E}_p$ is the field amplitude in the wave incident on the metal. Expression (32) is close to that obtained earlier in Refs. 22. It follows from (32) that in the presence of roughnesses, when $(l_{sp}^{(sp)})^{-1} > (l_{sp}^{(r)})^{-1} + (l_{sp}^{(m)})^{-1}$, the field intensity near the surface can increase substantially (by ~ 10 times in the case of silver at $\hbar\omega = 3$ eV) compared with the field intensity on a smooth surface. This increase of the field leads to a number of experimentally observable effects, e.g., to an increase of the photoemission current from a rough surface. Recognizing that the photoemission current is proportional to the field intensity near the surface,[8] we obtain the following expression for the measured ratio δY_p of the quantum yields of photoemission from a rough and a smooth surface in p-polarized light:

$$\delta Y_p \approx \frac{J_{11}t+J_{22}+J_{33}}{|D_1(0,k_0)|^2 t+|E_\parallel(0,k_0)|^2}. \qquad (33)$$

The factor t introduced in (33) takes into account the possible difference between the photoemission matrix elements corresponding to transitions induced by the normal and tangential field Figure 1 shows the results of a comparison of calculations in accordance with Eqs. (32) and (33) with the experimental data obtained in Ref. 4 in a study of photoemission from silver in a 0.5 mol solution of K_2SO_4. It can be seen from the figure that at parameter values $\bar{\xi}^2 a^2 \approx 4 \cdot 10^7$ Å^4 it is possible to achieve quantitative agreement between theory and experiment. We note one other important consequence of (32) and (33). It can be seen from these equations that when the mean squared dimension $\bar{\xi}^2$ of the roughnesses is increased the photoemission current should first increase, and then become constant at values $\bar{\xi}^2 > \bar{\xi}_0^2$, for which the

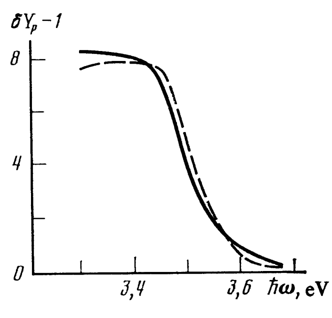


FIG. 1. Comparison of the theoretically calculated frequency dependence of $\delta Y_p - 1$ with experiment. The solid and dashed curves are respectively plots of the experimental data of Ref. 4 and of the values calculated from Eqs. (32) and (33) at $t = 1/|\varepsilon|^2$. The optical constants of silver, used in the calculations, were taken from Ref. 25.

inequality $(l_{sp}^{(r)})^{-1} > (l_{sp}^{(m)})^{-1}$ is satisfied, but $\overline{\xi}^2$ is still less than a^2. Such a dependence of the photoemission current on was observed in experiment.[5]

APPENDIX

We rewrite (24) in the form of a single functional integral over superfields ψ that contain as their components both commuting variables b and anticommuting (Grassmann) ones χ. Using notation analogous to that of Ref. 10, we introduce the eight-component supervectors

$$\psi^{(n)}=\begin{pmatrix} u^{(n)} \\ v^{(n)} \end{pmatrix},\quad u^{(n)}=\frac{1}{\sqrt{2}}\begin{pmatrix} \chi^{(n)*} \\ \chi^{(n)} \end{pmatrix},\quad v^{(n)}=\begin{pmatrix} b^{(n)*} \\ b^{(n)} \end{pmatrix},$$
$$\overline{\psi}=(\hat{C}\psi)^{Tr},\ n=1,\ 2;$$
$$\hat{C}_{mn}=\hat{\Lambda}_{mn}\begin{pmatrix} \hat{c}_1 & 0 \\ 0 & \hat{c}_2 \end{pmatrix},\quad \hat{\Lambda}_{mn}=\begin{pmatrix} \hat{1} & 0 \\ 0 & -\hat{1} \end{pmatrix},$$
$$\hat{c}_1=\begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix},\quad \hat{c}_2=\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{A.1}$$

Introduction of integrals over the superfields ψ makes possible averaging over ξ in general form, up to direct calculation of the Green's function. After integration with respect to ξ and χ_r the evaluation of the integral (24) reduces to calculation of a generating functional $Z(\alpha_i, \beta_i)$ $i = 1,2$ with effective action S_{eff}:

$$J=\frac{1}{4S}\left(\frac{1}{i}\frac{\partial}{\partial\alpha_1}+\frac{\partial}{\partial\beta_1}\right)\left(\frac{1}{i}\frac{\partial}{\partial\alpha_2}-\frac{\partial}{\partial\beta_2}\right)$$
$$Z(\alpha_i,\beta_i)|_{\alpha_i,\beta_i=0};$$
$$Z=\int D\psi_{sp}\exp[-S_{eff}(\psi_{sp})];$$
$$S_{eff}=S_0(\psi_{sp})+S(\psi_{sp},h_1,h_2)+\frac{1}{4}\overline{\xi^2}\int\frac{d^2\mathbf{k}^{(1)}}{(2\pi)^2}\frac{d^2\mathbf{k}^{(2)}}{(2\pi)^2}\frac{d^2\mathbf{k}^{(3)}}{(2\pi)^2}$$
$$\times\frac{d^2\mathbf{k}^{(4)}}{(2\pi)^2}\{(2\pi)^2\delta(\mathbf{k}^{(1)}+\mathbf{k}^{(2)}+\mathbf{k}^{(3)}+\mathbf{k}^{(4)})g(|\mathbf{k}^{(1)}+\mathbf{k}^{(2)}|)$$
$$\times(\overline{\psi}_{sp}(\mathbf{k}^{(1)})\psi_{sp}(\mathbf{k}^{(2)}))$$
$$\times(\overline{\psi}_{sp}(\mathbf{k}^{(3)})\psi_{sp}(\mathbf{k}^{(4)}))f_{sp,sp}(-\mathbf{k}^{(1)},\mathbf{k}^{(2)})f_{sp,sp}(-\mathbf{k}^{(3)},\mathbf{k}^{(4)})\}. \tag{A.2}$$

Here

$$S_0(\psi_{sp})=i\int\frac{d^2\mathbf{k}}{(2\pi)^2}\overline{\psi}_{sp}(\mathbf{k})$$
$$\times[\omega^2-\tilde{\omega}_{sp}{}^2(k)-i\Gamma_r(k)\hat{\Lambda}-\Delta_r(k)]\psi_{sp}(\mathbf{k}),$$
$$S(\psi_{sp},h_1,h_2)=i\int\frac{d^2\mathbf{k}}{(2\pi)^2}\overline{\psi}_{sp}(\mathbf{k})\{\hat{\Sigma}_1(\alpha_1h_1(\mathbf{k})+\alpha_2h_2(\mathbf{k})]$$
$$-i\hat{\Sigma}_2[\beta_1h_1(\mathbf{k})+\beta_2h_2(\mathbf{k})]\}\psi_{sp}(\mathbf{k});$$
$$(\hat{\Sigma}_1)_{3,7}=(\hat{\Sigma}_1)_{7,3}=(\hat{\Sigma}_2)_{3,7}=-(\hat{\Sigma}_2)_{7,3}=1,$$
$$(\hat{\Sigma}_{1(2)})_{ij}=0\ \text{при}\ ij\neq21,$$
$$\Gamma_r(k)=\pi\overline{\xi^2}\int\frac{d^2\mathbf{k}'}{(2\pi)^2}g(|\mathbf{k}+\mathbf{k}'|)f_{sp,r}^2(\mathbf{k},\mathbf{k}',\omega). \tag{A.3}$$

The quantity Γ_r characterizes the SP damping due to their decay into r-waves emitted from the metal. The SP-spectrum shift $\Delta\Gamma$ resulting from the interaction of the sp- and r-fields reduces to an insignificant modification of the function $\tilde{\omega}_{sp}^2$ and will be disregarded. At $\overline{\xi}^2/a^2 < \gamma^2$ the last term in the expression for S_{eff} turns out to be significant only at values $k^{(1)}$, $k^{(2)}$, $k^{(3)}$ and $k^{(4)}$ that are close to the root k_{sp} of the equation $\omega^2 - \tilde{\omega}_{sp}^2(k) = 0$. Accordingly, when the inequalities (26) are satisfied, the calculation results do not depend on the details of the law that governs the decrease of $g(k)$ at large k. Taking the foregoing into account, we can simplify greatly the calculations, if (26) is satisfied, by using the substitution

$$g(|\mathbf{k}^{(1)}+\mathbf{k}^{(2)}|)\approx\pi a^2\exp\left[-\frac{a^2}{4}((k^{(1)})^2+(k^{(2)})^2)\right]. \tag{A.4}$$

Direct calculations show that allowance for the additional terms of order $(a^2k^{(1)}k^{(2)})^n$ in the expression for $g(|\mathbf{k}^{(1)}+\mathbf{k}^{(2)}|)$ leads, if (26) is satisfied, to small corrections of order $(a^2k_{sp}{}^2)^n$ in the final expressions.

It follows from (A.2) that the determination of the generating functional reduces to solution of a nonlinear problem for $\psi_{sp}(\mathbf{k})$. It is convenient[10,23] to seek an approximate solution by using the Hubbard-Stratonovich transformation,[24] which permits the action to be linearized by introducing additional fields. Introducing the tensor fields $\hat{Q}$, $\hat{P}$, and $\hat{R}_{ij}$, we get

$$Z=\int D\psi_{sp}D\hat{Q}D\hat{P}D\hat{R}\exp\left\{-(\overline{\xi^2}a^2)^{-1}\mathrm{S\,Sp}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\right.$$
$$\times\left[\hat{Q}(\mathbf{q})\hat{Q}(-\mathbf{q})\right.$$
$$\left.+2\sum_{i=2,3}\hat{P}_i(\mathbf{q})\hat{P}_i(-\mathbf{q})+\sum_{i,j=2,3}\hat{R}_{ij}(\mathbf{q})\hat{R}_{ij}(-\mathbf{q})\right]$$
$$\left.-S(\psi_{sp},\hat{Q},\hat{P},\hat{R})-S(\psi_{sp},h_1,h_2)\right\};$$
$$S(\psi_{sp},\hat{Q},\hat{P},\hat{R})=S_0(\psi_{sp})$$
$$+2\pi^{1/2}c^2(-\varepsilon)^{-1/2}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\frac{d^2\mathbf{q}}{(2\pi)^2}\overline{\psi}_{sp}(\mathbf{k})$$
$$\times\left\{\hat{Q}(\mathbf{q})k|\mathbf{k}+\mathbf{q}|+2\sum_{i=2,3}\hat{P}_i(\mathbf{q})k_i|\mathbf{k}+\mathbf{q}|\right.$$
$$\left.+\sum_{i,j=2,3}\hat{R}_{ij}(\mathbf{q})k_i(\mathbf{k}+\mathbf{q})_j\right\}$$
$$\times\psi_{sp}(-\mathbf{k}-\mathbf{q})(k|\mathbf{k}+\mathbf{q}|)^{1/2}\exp\left[-\frac{a^2}{8}(k^2+(\mathbf{k}+\mathbf{q})^2)\right]. \tag{A.5}$$

Each of the matrices $\hat{Q}$, $\hat{P}_i$, and $\hat{R}_{ij}$ in (A.5) is made up of four (4×4) supermatrices, for example

$$\hat{Q}=\begin{pmatrix}\hat{Q}^{11} & \hat{Q}^{12}\\ \hat{Q}^{21} & \hat{Q}^{22}\end{pmatrix},\quad \hat{Q}^{11}=\begin{pmatrix}\hat{a}^{11} & \hat{\sigma}^{11}\\ \hat{\sigma}^{11+} & i\hat{b}^{11}\end{pmatrix},$$
$$\hat{Q}^{12}=i\begin{pmatrix}\hat{a}^{12} & i\sigma^{12}\\ \hat{\sigma}^{21+} & ib^{12}\end{pmatrix},\quad \hat{Q}^{21}=-i\begin{pmatrix}\hat{a}^{12+} & \hat{\sigma}^{21}\\ i\hat{\sigma}^{12+} & ib^{12+}\end{pmatrix},$$
$$\hat{Q}^{22}=\begin{pmatrix}\hat{a}^{22} & i\hat{\sigma}^{22}\\ i\hat{\sigma}^{22+} & i\hat{b}^{22}\end{pmatrix},$$

$$\hat{a}^{mn*}=\hat{a}^{mn}=\hat{c}_1 a^{mnT_r}\hat{c}_1,$$
$$\hat{b}^{mn*}=\hat{b}^{mn}\hat{c}_2\hat{b}^{mnT_r}\hat{c}_2^{T_r},$$
$$\hat{\sigma}^{mn*}=\hat{c}_2\hat{\sigma}^{mnT_r}\hat{c}_1^{T_r}. \tag{A.6}$$

The elements of matrices $\hat{a}_{mn}$ and $\hat{b}_{mn}$ are commuting variables, while those of matrices $\hat{\sigma}_{mn}$ are anticommuting. The symbol S Sp denotes a supertrace over the matrix variables

$$\mathrm{S\,Sp}\, Q=\mathrm{S\,Sp}\,\hat{Q}^{11}+\mathrm{S\,Sp}\,\hat{Q}^{22}=\mathrm{Sp}(\hat{a}^{11}+\hat{a}^{22})-i\mathrm{Sp}(\hat{b}^{11}+\hat{b}^{22}). \tag{A.7}$$

Comparison of (24) with Eqs. (21)–(23) shows that the functions $h_1(\mathbf{k})$ and $h_2(\mathbf{k})$ in the expression for $S(\psi_{sp}, h_1, h_2)$ can be represented in the form

$$h_n(\mathbf{k})=2\pi^{1/2}c^2(-\varepsilon)^{-1/2}k\Big[d_0^{(n)}k^2+2\sum_{i=2,3}d_i^{(n)}kk_i+\sum_{i,j=2,3}d_{ij}^{(n)}k_ik_j\Big],\quad n=1,2, \tag{A.8}$$

where d_0, d_i, and d_{ij} are coefficients independent of the wave vector $\mathbf{k}$. Taking (A.8) into account, we make in (A.5) the linear substitution

$$\hat{Q}(\mathbf{q})\to\hat{Q}(\mathbf{q})-(2\pi)^2\delta(q)\times\{i\hat{\Sigma}_1[\alpha_1 d_0^{(1)}+\alpha_2 d_0^{(2)}]+\hat{\Sigma}_2[\beta_1 d_0^{(1)}+\beta_2 d_0^{(2)}]\},$$
$$\hat{P}_i(\mathbf{q})\to\hat{P}_i(\mathbf{q})-(2\pi)^2\delta(\mathbf{q})\times\{i\hat{\Sigma}_1[\alpha_1 d_i^{(1)}+\alpha_2 d_i^{(2)}]+\hat{\Sigma}_2[\beta_1 d_i^{(1)}+\beta_2 d_i^{(2)}]\}, \tag{A.9}$$
$$\hat{R}_{ij}(\mathbf{q})\to\hat{R}_{ij}(\mathbf{q})-(2\pi)^2\delta(\mathbf{q})\times\{i\hat{\Sigma}_1[\alpha_1 d_{ij}^{(1)}+\alpha_2 d_{ij}^{(2)}]+\hat{\Sigma}_2[\beta_1 d_{ij}^{(1)}+\beta^2 d_{ij}^{(2)}]\}.$$

The substitution (A.9) cancels out the contribution of $S(\psi_{sp}, h_1, h_2)$ to the functional (A.5). After calculating the derivatives with respect to α_1, α_2, β_1, and β_2 we obtain the following expression for the integral J:

$$J=\frac{2}{S}(\overline{\xi^2 a^2})^{-2}\int D\hat{Q}\,D\hat{P}\,D\hat{R}\,\Big\{Q_{33}^{12}(0)Q_{33}^{21}(0)d_0^{(1)}d_0^{(2)}+2\sum_{i,j=2,3}[\hat{P}_i(0)]_{33}^{12}[\hat{P}_j(0)]_{33}^{21}d_i^{(1)}d_j^{(2)}+\sum_{i,j,m=2,3}[R_{ij}(0)]_{33}^{12}[R_{lm}](0)]_{33}^{21}d_{ij}^{(1)}d_{lm}^{(2)}\Big\}$$
$$\times\exp[-F(\hat{Q},\hat{P},\hat{R})]-2(\overline{\xi^2 a^2})^{-1}\times\Big[d_0^{(1)}d_0^{(2)}+\sum_{i=2,3}d_i^{(1)}d_i^{(2)}+\sum_{i,j=2,3}d_{ij}^{(1)}d_{ij}^{(2)}\Big]. \tag{A.10}$$

The functional $F(\hat{Q},\hat{P},\hat{R})$ in (A.10) is given by

$$F(\hat{Q},\hat{P},\hat{R})=(\overline{\xi^2 a^2})^{-1}\mathrm{S\,Sp}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\Big[\hat{Q}(\mathbf{q})\hat{Q}(-\mathbf{q})+2\sum_{i=2,3}\hat{P}_i(\mathbf{q})\hat{P}_i(-\mathbf{q})+\sum_{i,j=2,3}\hat{R}_{ij}(\mathbf{q})\hat{R}_{ij}(-\mathbf{q})\Big]$$
$$-\ln\int D\psi_{sp}\exp[-S(\psi_{sp},\hat{Q},\hat{P},\hat{R})]. \tag{A.11}$$

Since the factors preceding the exponentials in the integrals of (A.10) contain variables only at $q=0$, it suffices to obtain the explicit form of the functional F for the fields $\hat{Q}(\mathbf{q})$, $\hat{P}_i(\mathbf{q})$ and $\hat{R}_{ij}(\mathbf{q})$ with $\mathbf{q}$ close to zero. In analogy with the procedure in Refs. 10 and 23, we represent the functional F as an expansion in the deviations $\delta\hat{Q}$, $\delta\hat{P}_i$ and $\delta\hat{R}_{ij}$ from the minimum $\hat{Q}=\hat{Q}^0$, $\hat{P}_i=\hat{P}_i^0$ and $\hat{R}_{ij}=\hat{R}_{ij}^0$, which is obtained by equating to zero the variations of (A.11) with respect to the variables $\hat{Q}$, $\hat{P}_i$, and $\hat{R}_{ij}$. Analysis of the equations $\delta F/\delta Q(\mathbf{q})=0$, $\delta F/\delta P_i(\mathbf{q})=0$ and $\delta F/\delta R_{ij}(\mathbf{q})=0$ by perturbation theory shows that the minimum of (A.11) is reached on a class of matrices $\hat{Q}^0$, $\hat{P}_i^0$, $\hat{R}_{ij}^0$ which are constant in coordinate space. Taking the foregoing into account, we obtain from (A.5) and (A.11) the following equations for $\hat{Q}^0$, $\hat{P}_i^0$, $\hat{R}_{ij}^0$.

$$\hat{Q}^0=-i\overline{\xi^2}a^2c^2\pi^{1/2}(-\varepsilon)^{-1/2}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\hat{D}_{sp,sp}(\mathbf{k})k^3\exp\Big(-\frac{a^2}{4}k^2\Big),$$
$$\hat{P}_i^0=-i\overline{\xi^2}a^2c^2\pi^{1/2}(-\varepsilon)^{-1/2}\int\frac{d^2\mathbf{k}}{(2\pi)^2}\hat{D}_{sp,sp}(\mathbf{k})k^2k_i\exp\Big(-\frac{a^2}{4}k^2\Big),$$
$$\hat{R}_{ij}^0=-i\overline{\xi^2}a^2c^2\pi^{1/2}(-\varepsilon)^{-1/2}\times\int\frac{d^2\mathbf{k}}{(2\pi)^2}\hat{D}_{sp,sp}(\mathbf{k})kk_ik_j\exp\Big(-\frac{a^2}{4}k^2\Big), \tag{A.12}$$

where $\hat{D}_{sp}$, sp is the Green's function corresponding to the action $S(\psi_{sp},\hat{Q}^0,\hat{P}^0,\hat{R}^0)$:

$$\hat{D}_{sp,sp}(k)=\Big\{\omega^2-\tilde{\omega}_{sp}^2(k)-i\Gamma_r(k)\hat{\Lambda}-2\pi^{1/2}c^2i(-\varepsilon)^{-1/2}k \tag{A.13}$$
$$\times\Big[\hat{Q}^0k^2+2\sum_{l=2,3}\hat{P}_l^0kk_l+\sum_{l,m=2,3}\hat{R}_{lm}^0k_lk_m\Big]\exp\Big(-\frac{a^2}{4}k^2\Big)\Big\}.$$

Following the calculation method described in Ref. 17, we obtain[2)] from (A.12)

$$\hat{Q}^0=\frac{\pi^{1/2}}{3}\frac{\overline{\xi^2}a^2}{c^2(-\varepsilon)^{1/2}\gamma}k_{sp}^3\hat{\Lambda},$$
$$\hat{R}_{33}^0=\hat{R}_{22}^0={}^1/_2\hat{Q}^0,\quad \hat{P}_i^0=0,\quad \hat{R}_{ij}^0=0\quad\text{at}\quad i\neq j. \tag{A.14}$$

From the structure of the matrices $\hat{Q}^0$, $\hat{P}_i^0$, and $\hat{R}_{ij}^0$ it follows that the functional F is zero at the minimum. Expanding F about the minimum and confining ourselves to small deviations, we obtain at $(l_{sp}^{(sp)})^{-1}>(l_{sp}^{(r)})^{-1}$

$$F\approx-(\overline{\xi^2}a^2)^{-1}\Big[1+O\Big(\frac{l_{sp}}{l_{sp}^{(r)}}\Big)\Big]\times\int\frac{d^2\mathbf{q}}{(2\pi)^2}\Big\{\frac{2}{3}\Big[\frac{l_{sp}}{l_{sp}^{(r)}}+2(l_{sp}^{(sp)})^2q^2\Big]$$
$$\times\mathrm{S\,Sp}\Big[\delta\hat{Q}^{12}(\mathbf{q})+\frac{1}{2}(\delta\hat{R}_{33}^{12}(\mathbf{q})+\delta\hat{R}_{22}^{12}(\mathbf{q}))\Big]\Big[\delta\hat{Q}^{21}(-\mathbf{q})+\frac{1}{2}(\delta\hat{R}_{33}^{21}(-\mathbf{q})+\delta\hat{R}_{22}^{21}(-\mathbf{q}))\Big]$$
$$+\mathrm{S\,Sp}\Big[\frac{1}{3}(\delta\hat{Q}^{12}(\mathbf{q})-\delta\hat{R}_{33}^{12}(\mathbf{q})$$

$$-\delta\hat{R}_{22}^{12}(\mathbf{q}))(\delta\hat{Q}^{21}(-\mathbf{q})-\delta\hat{R}_{33}^{21}(-\mathbf{q})-\delta\hat{R}_{22}^{21}(-\mathbf{q}))$$

$$+\frac{5}{12}(\delta R_{33}^{12}(\mathbf{q})-\delta R_{22}^{12}(\mathbf{q}))(\delta R_{33}^{21}(-\mathbf{q})-\delta R_{22}^{21}(-\mathbf{q}))$$

$$+\frac{2}{3}\sum_{i=2,3}\delta\hat{P}_i^{12}(\mathbf{q})\delta\hat{P}_i^{21}(-\mathbf{q})$$

$$+\frac{1}{2}(\delta\hat{R}_{23}^{12}(\mathbf{q})-\delta\hat{R}_{32}^{12}(\mathbf{q}))(\delta\hat{R}_{23}^{21}(-\mathbf{q})$$

$$-\delta R_{32}^{21}(-\mathbf{q}))+\frac{5}{12}(\delta R_{23}^{12}(\mathbf{q})+\delta R_{32}^{12}(\mathbf{q}))$$

$$\times(\delta R_{23}^{21}(-\mathbf{q})+\delta R_{32}^{21}(-\mathbf{q}))\Big]\Big\}. \qquad (A.15)$$

Substituting (A.15) in (A.10) and evaluating the Gaussian integrals we arrive at expression (27) of the text. Allowance for terms of order $(\delta Q)^4$, $(\delta P_i)^4$ and $(\delta R_{ij})^4$ in the expansion of F leads to corrections[3)] of relative order $(k_{sp}l_{sp})^{-2}\ln(l_{sp}^{(r)}/l_{sp}^{(sp)})$ to the quantity J.

[1)]Since surface plasmons can exist only at $\varepsilon(\omega)<0$, it is just this frequency region which is of further interest.

[2)]Note that the system of nonlinear equations (A.12) has several solutions at large enough $\bar{\xi}^2$. We have retained, following Ref. 10, only that solution which yields the correct expression for the SP damping and goes over continuously, in the limit as $\bar{\xi}^2\to 0$, into the results of perturbation theory. A separate analysis is necessary to determine the meaning of the remaining solutions.

[3]The influence of terms of higher order in δQ, δP_i and δR_{ij} in F can be analyzed, using the renormalization-group equations, by the same procedure as in the analysis of surface conductivity.[10,23]

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