# Localization of the Surface Plasmon Polariton Caused by Random Roughness and its Role in Surface-Enhanced Optical Phenomena

K. Arya, Z. B. Su,[a] and Joseph L. Birman
*Department of Physics, The City College of the City University of New York, New York, New York 10031*
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We show explicitly that in a certain frequency range a surface plasmon polariton (SPP) is localized parallel to the metal surface by the presence of random roughness. By use of a diagrammatic method developed earlier, a self-consistent equation is derived for the renormalized diffusion coefficient of the SPP. We interpret surface-enhanced phenomena in terms of localized SPP and obtain an expression for the enhanced intensity near the metal surface. We find important differences in the enhancement results due to localization; these are consistent with experiments.

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In recent years, there has been considerable interest in the study of surface-enhanced optical properties from rough metal surfaces.[1-3] In particular, surface-enhanced Raman scattering (SERS) has attracted maximum attention because the enhancement by as much as six orders of magnitude in the cross section has made it possible to detect signals even from monolayers of admolecules.[1,2]

The enhancement on a rough surface is believed to be due to the resonant excitation of the surface plasmon polariton (SPP) by the incident photon which has large electromagnetic (em) field amplitude near the metal surface.[1-4] The presence of surface roughness is necessary because on a smooth surface, the dispersion curve for the SPP always lies below the photon line; thus SPP cannot be excited by a photon because energy and momentum cannot be conserved simultaneously.[5]

We consider here the case of random roughness which is commonly present in most surface-enhanced experiments. The SPP excited by the photon is an extended mode parallel to the smooth metal surface. On a rough surface, it undergoes elastic scattering as a result of the spatial fluctuation in the dielectric function near the (smooth) metal-vacuum interface. This multiple scattering of the SPP is considered by Arya and Zeyher[6] in the diffusion approximation similar to that in the conventional transport theory. However, in the latter case, it is well known that one should include effects due to interference between scattering waves, which can become very important. For example, in the case of an electron in a random potential, such an interference leads to the Anderson localization; i.e., the renormalized diffusion coefficient for the electron goes to zero.[7,8] Recently, there have been similar discussions about the localization of the elastic wave in a disordered medium,[9] and also the existence of a mobility edge has been suggested for the em wave in a disordered dielectric.[10]

In this Letter, we report the results of investigations of the localization of the SPP due to random roughness. For this we have used the diagrammatic method for the polariton Green's functions developed earlier by Arya and Zeyher,[6] and following Ref. 8 we derive a self-consistent equation for the renormalized diffusion coefficient $D$. Using a plasmon-pole approximation for the metal dielectric function, and for typical values of the roughness parameters obtained from surface-enhanced experiments, we show that $D$ goes to zero, which means that the SPP is localized parallel to the surface in the presence of random roughness. The localization of SPP is found to have important effects on the surface-enhanced phenomena and we discuss how these effects can be observed in surface-enhanced experiments.

We consider a semi-infinite metal with a local dielectric function

$$\epsilon(\mathbf{r},\Omega)=\epsilon^{(0)}(z,\Omega)+[\epsilon(\Omega)-1][\theta(\zeta(\mathbf{r}_{\parallel})-z)-\theta(-z)], \tag{1}$$

where $\epsilon^{(0)}(z,\Omega)=\theta(z)+\epsilon(\Omega)\theta(-z)$ is the dielectric function for a plane dielectric-vacuum interface. $\zeta(\mathbf{r}_{\parallel})$ gives the height of roughness with respect to the plane surface $z=0$, and $\mathbf{r}_{\parallel}=(x,y)$. Let $\hat{d}_{ij}(\mathbf{k}_{\parallel},\mathbf{k}'_{\parallel},\Omega|z,z')$ denote the polariton Green's function which satisfies the Maxwell equation for the metal-vacuum system defined by Eq. (1); we have performed a Fourier transformation with respect to $\mathbf{r}_{\parallel}$ and $\mathbf{r}'_{\parallel}$. For a randomly rough surface, the Green's function has to be averaged over the distribution of the profile function $\zeta$. We assume this distribution to be Gaussian and define averages by $\langle\zeta(\mathbf{r}_{\parallel})\rangle=0$ and $\langle\zeta(\mathbf{r}_{\parallel})\zeta(\mathbf{r}'_{\parallel})\rangle=\delta^2\exp(-|\mathbf{r}_{\parallel}-\mathbf{r}'_{\parallel}|^2/a^2)$, where $\langle\ \rangle$ denotes the ensemble average and $\delta$ and $a$ are the root-mean-square roughness amplitude and transverse correlation length, respectively.

To calculate average Green's functions, a diagrammatic expansion has been developed in Ref. 6 and by Arya, Zeyher, and Maradudin[11] by using the second term (describing random roughness) in Eq. (1) as a perturbation to the unperturbed part $\epsilon^{(0)}(z,\Omega)$ (corre-

sponding to a smooth surface). For small roughness, it is shown that these diagrams and the rules to calculate them are similar to those of the case of an electron in a random potential. Also, in $\hat{d}_{ij}$ only the arguments $z=0-$ and $z'=0+$ are needed, where $0\pm$ denote a small positive and a small negative quantity, respectively. Furthermore, the roughness coupling between the $p$-polarized SPP and the $s$ photon, which is small in the frequency range under consideration, can also be neglected for simplification. Within this approximation, the average $p$-polariton Green's function

$$\langle \hat{d}_{ij}(\mathbf{k}_\parallel, \mathbf{k}'_\parallel, \Omega | 0-, 0+) \rangle = (2\pi)^2 \delta(\mathbf{k}_\parallel - \mathbf{k}'_\parallel) d_{ij}(\mathbf{k}_\parallel, \Omega)$$

can be written as[12,6]

$$d_{ij}(\mathbf{k}_\parallel, \Omega) = \frac{e_i^-(\mathbf{k}_\parallel, \Omega) e_j^+(\mathbf{k}_\parallel, \Omega) \alpha(\mathbf{k}_\parallel, \Omega)}{\Omega - \Omega^{(0)}(k_\parallel) - i\gamma}, \tag{2}$$

$$\alpha(k_\parallel, \Omega) = \frac{4\pi c^4 k_\parallel^2 (\nu - \epsilon\nu_0)}{(1-\epsilon)[\Omega^2 - \Omega^{(+)2}(k_\parallel)][\Omega + \Omega^{(-)}(k_\parallel)]}, \tag{3}$$

where $\mathbf{e}^-(\mathbf{k}_\parallel, \Omega) = \hat{\mathbf{z}} + i\hat{\mathbf{k}}_\parallel \nu(k_\parallel, \Omega)/k_\parallel$ and $\mathbf{e}^+(\mathbf{k}_\parallel, \Omega) = \hat{\mathbf{z}} + i\hat{\mathbf{k}}_\parallel \nu_0(k_\parallel, \Omega)/k_\parallel$ are polarization vectors, $\nu^2(k_\parallel, \Omega) = k_\parallel^2 - \epsilon(\Omega)\Omega^2/c^2$, $\nu_0^2 = k_\parallel^2 - \Omega^2/c^2$, and $\mathrm{Im}\nu_0$ is positive. $\Omega^{(\pm)}(k_\parallel)$ corresponds to upper and lower branches of the SPP[5] and are given by the dispersion relation $k_\parallel^2 = k_s^2(\Omega) = (\Omega/c)^2\epsilon(\Omega)/[\epsilon(\Omega)+1]$. Since the upper branch is too high we will assume that only the lower branch is excited by the photon. In Eq. (2), only the imaginary part of the self-energy $S(\mathbf{k}_\parallel, \Omega)$ is retained, i.e., $\gamma = \mathrm{Im}S(\mathbf{k}_\parallel, \Omega)$. For small roughness, we approximate this by the first term in the series expansion of $S$. For $k_\parallel = k_s$, we thus have

$$\gamma \approx \pi a^2 \delta^2 \Lambda^2 \alpha(k_s, \Omega) \mathrm{Im} \int \frac{d^2k_\parallel}{(2\pi)^2} \exp[-\tfrac{1}{4}(\mathbf{k}_s - \mathbf{k}_\parallel)^2 a^2] \mathbf{e}^+(\mathbf{k}_s, \Omega) \cdot \mathbf{d}(\mathbf{k}_\parallel, \Omega) \cdot \mathbf{e}^-(\mathbf{k}_s, \Omega) = \gamma_{sc} + \gamma_{rad}, \tag{4}$$

where $\gamma_{sc}$ and $\gamma_{rad}$ correspond to the contributions from the region $k_\parallel > \Omega/c$ and $k_\parallel < \Omega/c$, respectively, in the integration and $\Lambda = [\epsilon(\Omega)-1]\Omega^2/4\pi c^2$. Note that for the SPP, $k_\parallel = k_s(\Omega) > \Omega/c$ and $\nu_0(k_\parallel, \Omega)$ and $\alpha(k_\parallel, \Omega)$ are real. Thus $\gamma_{sc}$ corresponds to the elastic scattering of the SPP into other SPP states. For $k_\parallel < \Omega/c$, $\nu_0(k_\parallel, \Omega)$ and $\alpha(k_\parallel, \Omega)$ have a cut in the complex $k_\parallel$ plane and $d_{ij}(\mathbf{k}_\parallel, \Omega)$ represents the $p$-photon mode. Therefore, $\gamma_{rad}$ denotes scattering of SPP into radiative modes. In a frequency range of interest to us, where the SPP branch is far away from the photon line, $\gamma_{rad}$ is very small. Therefore, in the following discussion, we neglect contributions from the region $k_\parallel < \Omega/c$ and assume Eq. (2) to define the SPP Green's function for all $k_\parallel$ values. We will, however, include the radiative losses ($\gamma_{rad}$) phenomenologically at the end along with other losses, e.g., these due to interband scattering in the metal.

As is well known, the essential physics of localization is contained in the calculation of the average two-particle Green's function,[8] which for the SPP can be written as[6]

$$\langle \hat{d}_{ij}(\mathbf{k}_\parallel +, \mathbf{k}'_\parallel +, \Omega + | 0-, 0) \hat{d}^*_{kl}(\mathbf{k}_\parallel -, \mathbf{k}''_\parallel -, \Omega | 0-, 0+) \rangle$$
$$= (2\pi)^2 \delta(\mathbf{k}'_\parallel - \mathbf{k}''_\parallel) L(\mathbf{k}_\parallel, \mathbf{k}'_\parallel, \Omega, \mathbf{q}, \omega) e_i^-(\mathbf{k}_\parallel +, \Omega +) e_j^+(\mathbf{k}'_\parallel +, \Omega +) e_k^{-*}(\mathbf{k}_\parallel -, \Omega) e_l^{+*}(\mathbf{k}'_\parallel -, \Omega), \tag{5}$$

where $\mathbf{k}_\parallel \pm = \mathbf{k}_\parallel \pm \mathbf{q}/2$ and $\Omega + = \Omega + \omega$. It is easy to verify that

$$L(\Omega, \mathbf{q}, \omega) = \int \frac{d^2k_\parallel}{(2\pi)^2} \int \frac{d^2k'_\parallel}{(2\pi)^2} L(\mathbf{k}_\parallel, \mathbf{k}'_\parallel, \Omega, \mathbf{q}, \omega) \tag{6}$$

is related to the (em) energy density-density correlation function on the surface. In the localization theory and in surface-enhancement results, $L(\Omega, \mathbf{q}, \omega)$ is the main quantity which needs to be calculated.

As discussed in Ref. 6, $L$ involves similar diagrams as in the case of an electron in a random potential. Therefore, we follow the diagrammatic treatment of Ref. 8 to calculate $L$. Furthermore, the SPP Green's function is very similar to that of the electron except with a different dispersion relation. Thus in a frequency range where the coupling of the SPP and the photon is small, it can be easily shown that for small roughness all the approximations of Ref. 8 are also applicable to our case. For example, we obtain the continuity equation ($\omega \to 0$, $q \to 0$)

$$L(\Omega, \mathbf{q}, \omega) - \mathbf{q} \cdot \int \frac{d^2k_\parallel}{(2\pi)^2} \int \frac{d^2k'_\parallel}{(2\pi)^2} \mathbf{v}(\mathbf{k}_\parallel) L(\mathbf{k}_\parallel, \mathbf{k}'_\parallel, \Omega, \mathbf{q}, \omega) = 2\pi i N(k_s), \tag{7}$$

where the second term on the left-hand side is the SPP current density, $N(k_s) = (\Omega/2\pi c^2)[\epsilon^2(\Omega)+1]/[\epsilon(\Omega)+1]^2$ is the SPP density of states near $k_s$, and $\mathbf{v}(\mathbf{k}_\parallel) = \partial\Omega^{(-)}(k_\parallel)/\partial\mathbf{k}_\parallel$ is the velocity of the SPP. Using Eq. (7) and a similar expression for the SPP current density which can be derived (for weak roughness) by follow-

ing Ref. 8, we obtain

$$L(\Omega,\mathbf{q},\omega)=2\pi iN(k_s)/[\omega+iD(\mathbf{q},\omega)q^2]. \tag{8}$$

In Eq. (8), $D(\mathbf{q},\omega)=2i\gamma_{sc}D_0/M(\mathbf{q},\omega)$ is the renormalized diffusion coefficient, where $D_0=v^2(k_s)/4\gamma_{sc}$ is the bare diffusion coefficient and

$$M(\mathbf{q},\omega)=2i\gamma_{sc}-\frac{1}{N(k_s)}\int\frac{d^2k_{\parallel}}{(2\pi)^2}\int\frac{d^2k'_{\parallel}}{(2\pi)^2}\Delta D(\mathbf{k}_{\parallel})U(\mathbf{k}_{\parallel},\mathbf{k}'_{\parallel},\Omega,\mathbf{q},\omega)\Delta D(\mathbf{k}'_{\parallel}). \tag{9}$$

In Eq. (9), $\Delta D(\mathbf{k}_{\parallel})=\hat{\mathbf{q}}\cdot\mathbf{k}_{\parallel}[d(\mathbf{k}_{\parallel},+,\Omega,+)-d^*(\mathbf{k}_{\parallel},-,\Omega)]$, where $d(\mathbf{k}_{\parallel},\Omega)$ is given by Eq. (2) without polarization vectors $e_i^-$ and $e_j^+$. $U(\mathbf{k}_{\parallel},\mathbf{k}'_{\parallel},\Omega,\mathbf{q},\omega)$ is the sum of all irreducible diagrams in the four-point vertex and in the lowest-order approximation (ladder summation) $M(\mathbf{q},\omega)$ is just given by the first term in Eq. (9) and thus $D(q,\omega)=D_0$.

Equation (9) can be solved for $\mathbf{M}(\mathbf{q},\omega)$ following Ref. 8. For $\gamma_{sc}\ll\Omega$, the main contribution comes from the maximal cross diagrams and the contribution of all other diagrams goes to zero in the limit $\omega\to 0$, $q\to 0$. The sum of maximal cross diagrams is calculated from the ladder summation by use of the fact that the SPP, being a Bose field, has time-reversal symmetry, and we thus have

$$U(\mathbf{k}_{\parallel},\mathbf{k}'_{\parallel},\Omega,\mathbf{q},\omega)=U[\tfrac{1}{2}(\mathbf{k}_{\parallel}-\mathbf{k}'_{\parallel}+\mathbf{q}),\tfrac{1}{2}(\mathbf{k}'_{\parallel}-\mathbf{k}_{\parallel}+\mathbf{q}),\Omega,\mathbf{k}_{\parallel}+\mathbf{k}'_{\parallel},\omega]\approx\frac{2\gamma_{sc}U_0}{-i\omega+D(q,\omega)(\mathbf{k}_{\parallel}+\mathbf{k}'_{\parallel})^2}, \tag{10}$$

where $U_0=\frac{3}{2}\pi a^2\delta^2\Lambda^2\exp(-k_s^2a^2/2)$ and we have used the renormalized diffusion coefficient $D$ instead of the bare $D_0$.[8] Inserting Eq. (10) into (9) and after some simplification, one finds a simple expression for $M(\mathbf{q},\omega)$, which when used in the defining relation of $D$ gives the required equation for the renormalized diffusion coefficient as

$$D(q,\omega)=D_0-\frac{1}{\pi N(k_s)}\int_0^{k_c}\frac{dk_{\parallel}}{2\pi}k_{\parallel}\left[k_{\parallel}^2-\frac{i\omega}{D(q,\omega)}\right]^{-1}. \tag{11}$$

In Eq. (11), an upper cutoff $k_c=1/l$ has been used in the integration, where $l=2D_0/v(k_s)$ is the diffusion length. In the limit $\omega\to 0$, it is easy to show from Eq. (11) that $D(q,\omega)\propto -i\omega$, which is the condition for localization. One can also define the localization length $\xi=[iD(q,\omega)/\omega]^{1/2}_{\omega\to 0}$, which from Eq. (11) is given by

$$1=-i(\omega+i\gamma_l)\xi^2/D_0+(k_c/\pi k_s)\ln(1+\xi^2k_c^2). \tag{12}$$

In Eq. (12) we have included the damping term $\gamma_l$ for the SPP due to losses by radiation and other interband scattering in the metal. The localization, however, is meaningful only if $\gamma_l\ll\gamma_{sc}\ll\Omega$. Furthermore, since a sufficient amount of elastic scattering of the SPP is necessary for building up localization, we also have the condition that $\xi>l>1/k_s$. From Table I, where calculated values of these parameters are given for $\epsilon(\Omega)=1-\omega_p^2/\Omega^2$, we find that these conditions for localization are satisfied in a reasonable frequency range close to $\omega_s=\omega_p/\sqrt{2}$ where radiative losses are very small. In real metals we have additional losses due to interband scattering [$\gamma_{int}=\Omega\epsilon_2/2(1+\epsilon_1)^2$, where $\epsilon_1$ and $\epsilon_2$ are real and imaginary parts of $\epsilon(\Omega)$] which are also small at least for noble metals like silver.

We now discuss how localization effects can be observed in surface-enhanced experiments where enhancement is due to the resonant excitation of the SPP. One can view surface-enhanced phenomena as a response of the SPP to the incident photon in the presence of roughness. This is analogous to the response of electrons in a random potential to the external electric field where the well known effect of localization is that in two dimensions, conductivity $\sigma(\omega)\to 0$ as $\omega\to 0$. There is, however, an important difference; in the latter case the system is in thermal equilibrium and only electrons near the Fermi level are excited by the external field. For the SPP, there is no equivalent to the Fermi energy because of its Bose statistics. In surface-enhanced experiments, SPP's are in a nonequilibrium state obtained by excitation with an almost monochromatic beam of photons. Therefore, any process used to observe localization effects in this case should be faster than the SPP's thermalization time. This condition is satisfied in almost all experiments.

In Ref. 6, it is shown that in the presence of an external photon, the average field intensity

TABLE I. Typical parameters calculated as a function of the SPP frequency. $\epsilon(\Omega)=1-2\omega_s^2/\Omega^2$, and roughness parameters $a=1.5\omega_s/c$, $\delta=0.5\omega_s/c$.

| $\Omega/\omega_s$ | $ck_s/\Omega$ | $\gamma_{sc}/\Omega$ | $\gamma_{rad}/\Omega$ | $\xi\Omega/c$ | $l\Omega/c$ |
|---|---|---|---|---|---|
| 0.50 | 1.08 | 0.0133 | 0.0024 | 68.2 | 26.9 |
| 0.55 | 1.10 | 0.0249 | 0.0033 | 40.4 | 14.1 |
| 0.60 | 1.13 | 0.0450 | 0.0042 | 24.7 | 7.6 |
| 0.65 | 1.17 | 0.0787 | 0.0046 | 15.4 | 4.1 |
| 0.70 | 1.22 | 0.1340 | 0.0042 | 9.5 | 2.2 |
| 0.75 | 1.28 | 0.2231 | 0.0028 | 2.3 | 1.2 |

$\langle|\mathbf{E}(\mathbf{r},\Omega|^2\rangle$ near the metal surface is related to $\langle\hat{d}_{ij}(\mathbf{r},\mathbf{r}',\Omega)\hat{d}^*_{kl}(\mathbf{r},\mathbf{r}'',\Omega)\rangle$. For example, the $z$ component of the intensity just outside the surface can be written as[6]

$$\langle|E_z(0,\Omega|0+)|^2\rangle = |E_z^{(0)}(\mathbf{Q}_\parallel,\Omega)|^2\int_0^{q_c}\frac{dq}{2\pi}[1+U_0L(\Omega,q,0)], \tag{13}$$

where we have assumed a $p$-polarized narrow incident beam [of wave vector $\mathbf{Q}=(\mathbf{Q}_\parallel,Q_z\hat{\mathbf{z}})$ and $\Omega=cQ$] with a spread of $q_c$ in $Q_\parallel$. In Eq. (13), the first term on the right-hand side is the contribution from a flat surface and the second term is due to the excitation of the SPP via roughness. Comparing the second with the first term and using Eq. (8) for $L(\Omega,q,0)$, we find an enhancement $\rho(\Omega)$ in the intensity,

$$\rho(\Omega)=1+\frac{2\gamma_{sc}}{\gamma_I}\frac{1}{\xi^2q_c^2}\ln(1+\xi^2q_c^2). \tag{14}$$

A similar equation for $\rho(\Omega)$ can also be derived in the case of extended SPP by using the bare $D_0$ instead of $D$ in Eq. (8):

$$\rho^{\text{ext}}(\Omega)=1+\frac{2\gamma_{sc}}{D_0q_c^2}\ln(1+D_0q_c^2). \tag{15}$$

Comparing Eq. (14) with (15), we find at least two differences due to localization: (i) $\rho(\Omega)$ obtained from Eq. (14) is always larger than that given by (15) because, in the case of localization, electric field is concentrated near the incident point rather than being uniformly distributed on the surface as in the case of extended SPP. This is shown in Fig. 1 for $\epsilon(\Omega)=1-2\omega_s^2/\Omega^2$ and for Ag where the complex $\epsilon(\Omega)$ is taken from Johnson and Christy.[13] In both cases, localization gives larger $\rho(\Omega)$ and the effects become more pronounced near $\omega_s$ because of small radiative losses ($\gamma_{\text{rad}}/\gamma_{\text{sc}}\to 0$, see Table I). $\rho(\Omega)$ also increases with $\Omega$ because of the increase in roughness coupling due to the large SPP density of states. However, one should not extrapolate these results for $\Omega$ too close to $\omega_s$ because we have approximated $\gamma$ in Eq. (4) only by the first term of the self-energy. The values of the roughness parameters $(a,\delta)$ used in these calculations are typically those obtained from enhancement experiments and we find the magnitude of $\rho(\Omega)$ to be consistent also. (ii) Another important effect of localization is that in a frequency range where $\gamma_{\text{rad}}\ll\gamma_{\text{int}}$ [$\gamma_{\text{int}}\propto\epsilon_2(\Omega)$], we have from Eq. (14) $\rho(\Omega)\propto 1/\epsilon_2(\Omega)$, which is consistent with experiments.

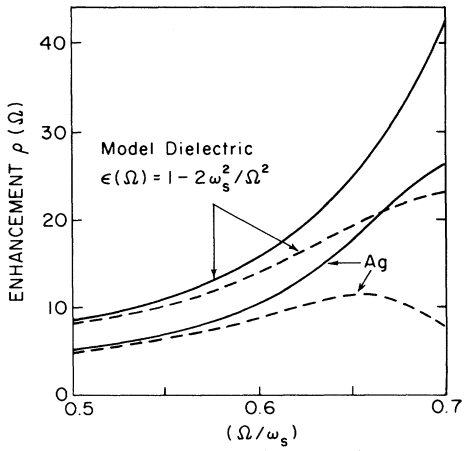


FIG. 1. Enhancement $\rho(\Omega)$ with (solid curves) and without (dashed curves) localization effects calculated from Eqs. (14) and (15), respectively. Roughness parameters used for the model dielectric, $c\delta/\omega_s=0.5$, $ca/\omega_s=1.5$; for Ag, $\delta=250$ Å, $a=750$Å. Also $q_c\sim 1/\xi$. In the case of Ag, to obtain the $\Omega$ scale in electronvolts use $\omega_s=5$ eV.

In conclusion, on a semi-infinite metal, the SPP is localized parallel to the surface in the presence of random roughness. The localization effects may be appreciable only in a certain frequency range where radiative losses are small. However, in other geometries, e.g., where a thin dielectric layer sandwiched between two thick metal films with rough metal-dielectric interfaces, radiation losses can be reduced considerably.

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[a]Permanent address: Institute of Theoretical Physics, Academia Sinica, Beijing, China.

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