Multiple-scattering effect of surface-plasmon polaritons in light emission from tunnel junctions

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We have investigated the multiple-scattering effect of surface-plasmon polaritons in the light-emission process from metal-insulator-metal tunnel junctions. The theory developed by Arya and Zeyher [Phys. Rev. B **24**, 4048 (1983)] is extended to take into account the roughness at all interfaces of the junction. The light intensity due to multiple scattering from roughness at the metal-oxide interfaces first increases steeply, takes a maximum value, and then decreases slowly with the increase in the root-mean-squared amplitude of roughness δ. This behavior is very different from that due to roughness at the top surface of the junction investigated by Arya and Zeyher. In their case the predicted intensity due to multiple scattering increases monotonically with increasing δ. The origin of this difference is discussed.

I. INTRODUCTION

Visible light emission from the metal-insulator-metal (M-I-M) tunnel junction was first observed by Lambe and McCarthy in 1975. They conjectured that the light is radiated by surface-plasmon polaritons (SPP's) that are excited by tunneling electrons.[1] Since SPP's are nonradiative at a perfectly flat surface, they assumed that the surface roughness of the junction plays the role of scattering SPP's into external radiation. After their original discovery, light-emitting tunnel junctions (LETJ's) were investigated by many researchers, and the conjecture of Lambe and McCarthy has been confirmed experimentally.[2–6]

A theoretical study of LETJ's was started by Davis,[7] and a theory that quantitatively takes into account the effect of surface roughness was first presented by Laks and Mills.[8] They formulated light emission as a dipole radiation process from a current source embedded in a multilayered structure. The surface roughness was treated as a perturbative term in the electromagnetic wave equation. They placed roughness only at the top surface of the tunnel junction, and solved the electromagnetic wave equation to the lowest-order term (first-order perturbation theory).

Takeuchi *et al.* extended the theory of Laks and Mills to include the effect of roughness at all interfaces in the tunnel junction.[9] They found that the roughness at the metal-oxide interfaces is more effective in inducing light emission than the roughness at the top surface (the case treated by Laks and Mills).

Physically, the first-order perturbation theories mentioned above take into account only the single scattering process of SPP's by roughness. Then the theories predict an emission intensity proportional to δ^2, where δ is the root-mean-squared amplitude of the roughness. When the amplitude δ is over some critical value, it is clear that multiple scattering of SPP's from roughness becomes significant. To study the light-emission properties of junctions with roughness beyond the critical level, we need to develop a theory that includes not only the lowest-order term but also higher-order terms.

Arya and Zeyher developed a theory that takes into account the higher-order terms, and discussed the multiple-scattering effect due to roughness at the surface of the top metal layer. They found that the emission intensity due to roughness increases faster than δ^2 with increasing δ.[10] Since the roughness at the metal-oxide interfaces are now known to be more important than the roughness at the top surface as a result of the work by Takeuchi *et al.*,[9] it is desirable to extend the multiple-scattering theory of Arya and Zeyher[10] to include the effect of roughness at other interfaces than the top surface of the junction.

The purpose of the present paper is to investigate the multiple-scattering effect due to individual interface roughness in the junction. The content of this paper is as follows. A brief description of the theoretical framework is presented in Sec. II. The results of numerical calculations are given in Sec. III. The multiple-scattering effect on light emission is discussed in Sec. IV, and Sec. V is the conclusion.

II. THEORY

A LETJ is modeled as a multilayered structure, and light emission is formulated as a dipole radiation process from a fluctuating current source embedded in the structure. We calculate the radiation intensity from a current source embedded in an n-layered structure with $n-1$ interfaces, as illustrated in Fig. 1. Roughness is placed only at the mth interface ($m=1\sim n-1$). m is always used to denote the interface with roughness throughout the present paper. In Fig. 1, z_i ($i=1\sim n-1$) is the z position of the ith (averaged z position for $i=m$) interface plane and $\zeta_m(\mathbf{x}_\parallel)$ is the profile function of roughness at the mth interface. Then $\langle \zeta_m(\mathbf{x}_\parallel)\rangle=0$, where $\langle\ \rangle$ means a statistical average over the interface plane. The directions of the electric field for p- and s-polarized light are defined as shown in Fig. 1. The present theory gives the radiation intensity emitted into the nth layer. Thus the $(n-1)$th interface is the top surface of the junction.

When there is a current source $J_\nu(\mathbf{x}';\omega)$ at $\mathbf{x}'$ with frequency ω, the electric field E_μ at position $\mathbf{x}$ is given by

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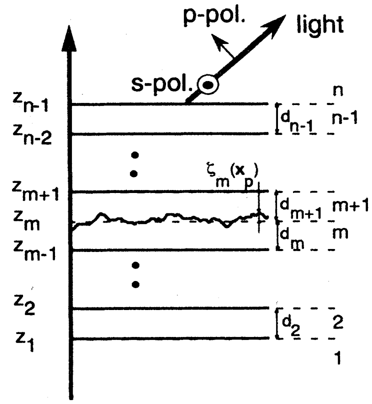

FIG. 1. Model structure for the theoretical calculation of LETJ's. Light emission is formulated as a dipole radiation process from a current source embedded in the n-layered structure. Roughness is placed only at the mth interface ($m=1\sim n-1$).

$$E_\mu(\mathbf{x},\omega)=-i\frac{\omega}{c^2}\sum_\nu \int d^3x' D_{\mu\nu}(\mathbf{x},\mathbf{x}';\omega)J_\nu(\mathbf{x}';\omega) \ . \tag{1}$$

Here c is the speed of light in vacuum, and $D_{\mu\nu}(\mathbf{x},\mathbf{x}';\omega)$ is the electromagnetic Green's function that is a solution of the wave equation:

$$\sum_\mu \left[\frac{\omega^2}{c^2}\varepsilon(\mathbf{x},\omega)\delta_{\lambda\mu}-\frac{\partial^2}{\partial x_\lambda \partial x_\mu}+\delta_{\lambda\mu}\nabla^2\right]D_{\mu\nu}(\mathbf{x},\mathbf{x}';\omega) =4\pi\delta_{\lambda\nu}\delta(\mathbf{x}-\mathbf{x}') \ . \tag{2}$$

$\varepsilon(\mathbf{x},\omega)$ in Eq. (2) is a frequency- and position-dependent dielectric function defined by

$$\varepsilon(\mathbf{x},\omega)=\varepsilon^{(0)}(z,\omega)+\Delta\varepsilon(\mathbf{x}_\parallel,z,\omega) \ ,$$

where

$$\varepsilon^{(0)}(z,\omega)=\varepsilon_i \quad \text{for } z_{i-1}<z<z_i \ ,$$

and

$$\Delta\varepsilon(\mathbf{x}_\parallel,z,\omega)=\begin{cases}\varepsilon_m-\varepsilon_{m+1} & \text{for } z_m<z<z_m+\zeta_m(\mathbf{x}_\parallel)\\ 0 & \text{otherwise} \ .\end{cases}$$

The far-field radiation intensity per unit solid angle per unit frequency range at distance R from the source is

$$P(\mathbf{k}_\parallel^{(0)},\omega)\equiv\frac{d^3W}{d\Omega\, d\omega\, dt} =|R|^2\frac{\sqrt{\varepsilon_n}c}{8\pi}\sum_\mu\langle E_\mu(\mathbf{x},\omega)^*E_\mu(\mathbf{x},\omega)\rangle \ . \tag{3}$$

with

$$|\mathbf{k}_\parallel^{(0)}|=\sqrt{\varepsilon_n}\frac{\omega}{c}\sin(\theta_0) \ .$$

Here $\sqrt{\varepsilon_n}$ is the refractive index of the nth layer and θ_0 is the radiation angle measured from the surface normal. $\langle\ \rangle$ represents an ensemble average over all different profile functions.[8] By substituting Eq. (1) into Eq. (3), one obtains

$$P(\mathbf{k}_\parallel^{(0)},\omega)=\frac{\varepsilon_n^{3/2}\omega^4\cos^2\theta_0}{32\pi^3c^5}\sum_\mu\sum_{\lambda,\lambda'}\int d^2\mathbf{Q}_\parallel dz'dz''\langle D_{\mu\lambda}(\mathbf{k}_\parallel^{(0)},\mathbf{Q}_\parallel,\omega|z,z')^*D_{\mu\lambda'}(\mathbf{k}_\parallel^{(0)},\mathbf{Q}_\parallel,\omega|z,z'')\rangle J_{\lambda\lambda'}(\mathbf{Q}_\parallel,\omega|z',z'') \ , \tag{4}$$

where $D_{\mu\nu}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega|z,z')$ and $J_{\lambda\lambda'}(\mathbf{Q}_\parallel,\omega|z',z'')$ are the two-dimensional Fourier transforms of $D_{\mu\nu}(\mathbf{x},\mathbf{x}';\omega)$ and $\langle J_\lambda(\mathbf{x}',\omega)^*J_{\lambda'}(\mathbf{x}'',\omega)\rangle$, respectively.

A semiphenomenological form of $J_{\lambda\lambda'}(\mathbf{Q}_\parallel,\omega|z',z'')$ for M-I-M tunnel junctions proposed by Laks and Mills[8] is

$$J_{\lambda\lambda'}(\mathbf{Q}_\parallel,\omega|z,z')=\begin{cases}\dfrac{eI_0(1-\hbar\omega/eV_0)}{2\pi^2A}\dfrac{\Delta(z,z')}{(1+\mathbf{Q}_\parallel^2\xi^2)^{3/2}}\delta_{z\lambda}\delta_{z\lambda'}, & eV_0>\hbar\omega\\ 0, & eV_0<\hbar\omega \ ,\end{cases} \tag{5}$$

where e is the electronic charge, I_0 is the total current in the junction of area A, V_0 is the bias voltage across the junction, and ξ is a phenomenological electron coherence length. Following the argument by Laks and Mills,[8] $\Delta(z,z')$ is taken to be unity in the tunneling barrier and zero outside. Thus the remaining task in calculating Eq. (4) is to obtain the two-photon Green's function

$$\langle D_{\mu\lambda}(\mathbf{k}_\parallel^{(0)},\mathbf{Q}_\parallel,\omega|z,z')^*D_{\mu\lambda'}(\mathbf{k}_\parallel^{(0)},\mathbf{Q}_\parallel,\omega|z,z'')\rangle \ .$$

The Fourier-transformed electromagnetic Green's function satisfies the following integral equation which is a natural generalization of Eq. (18) of Ref. 10:

$$D_{\mu\nu}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega|z,z')=(2\pi)^2\delta(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)d_{\mu\nu}^{(0)}(\mathbf{k}_\parallel,\omega|z,z') +\Lambda_m(\omega)\sum_\lambda d_{\mu\lambda}^{(0)}(\mathbf{k}_\parallel,\omega|z,z_{m+})\int\frac{d^2k''_\parallel}{(2\pi)^2}\zeta_m(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)D_{\lambda\nu}(\mathbf{k}''_\parallel,\mathbf{k}'_\parallel,\omega|z_{m-},z') \ , \tag{6}$$

where

$$\Lambda_m(\omega)=-\frac{\omega^2}{4\pi c^2}(\varepsilon_m-\varepsilon_{m+1}) .$$

In this equation $\zeta_m(\mathbf{k}_\parallel)$ is the two-dimensional Fourier transform of the roughness profile function $\zeta_m(\mathbf{x}_\parallel)$. $d_{\mu\nu}^{(0)}(\mathbf{k}_\parallel,\omega|z,z')$ is the electromagnetic-Green's function for the n-layered structure with flat interfaces $[\zeta_m(\mathbf{k}_\parallel)=0]$.[12]

Following Arya and Zeyher,[10] we introduce $L_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega|z_1,z'_1,z_2,z'_2)_m$ defined by

$$\langle D_{ij}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega|z_1,z_2)^*D_{kl}(\mathbf{k}_\parallel,\mathbf{k}''_\parallel,\omega|z'_1,z'_2)\rangle=(2\pi)^2\delta(\mathbf{k}'_\parallel-\mathbf{k}''_\parallel)L_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega|z_1,z'_1,z_2,z'_2\rangle_m .$$

Then we obtain the following expression that corresponds to Eq. (30) of Ref. 10 for $m=n-1$:

$$\begin{aligned}
&L_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega|z_1,z'_1,z_2,z'_2)_m\\
&\quad=(2\pi)^2\delta(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)d_{ij}^{(0)}(\mathbf{k}_\parallel,\omega|z_1,z_2)^*d_{kl}^{(0)}(\mathbf{k}_\parallel,\omega|z'_1,z'_2)\\
&\qquad+\delta^2|\Lambda_m(\omega)|^2\sum_{\alpha\alpha'}d_{i\alpha}^{(0)}(\mathbf{k}_\parallel,\omega|z_1,z_{m+})^*d_{k\alpha'}^{(0)}(\mathbf{k}_\parallel,\omega|z'_1,z_{m+})g_{mm}(|\mathbf{k}_\parallel-\mathbf{k}'_\parallel|)d_{\alpha j}^{(0)}(\mathbf{k}'_\parallel,\omega|z_{m-},z_2)^*d_{\alpha' 1}^{(0)}(\mathbf{k}'_\parallel,\omega|z'_{m-},z'_2)\\
&\qquad+\{\delta^2|\Lambda_m(\omega)|^2\}^2\sum_{\alpha\alpha'\beta\beta'}d_{i\alpha}^{(0)}(\mathbf{k}_\parallel,\omega|z_1,z_{m+})^*d_{k\alpha'}^{(0)}(\mathbf{k}_\parallel,\omega|z'_1,z_{m+})\\
&\qquad\qquad\times\int\frac{d^2k_\parallel^{(1)}}{(2\pi)^2}g_{mm}(|\mathbf{k}_\parallel-\mathbf{k}_\parallel^{(1)}|)\int\frac{d^2k_\parallel^{(2)}}{(2\pi)^2}L_{\alpha\beta\alpha'\beta'}(\mathbf{k}_\parallel^{(1)},\mathbf{k}_\parallel^{(2)},\omega)_m\\
&\qquad\qquad\times g_{mm}(|\mathbf{k}_\parallel^{(2)}-\mathbf{k}'_\parallel|)d_{\beta j}^{(0)}(\mathbf{k}'_\parallel,\omega|z_{m-},z_2)^*d_{\beta' l}^{(0)}(\mathbf{k}'_\parallel,\omega|z_{m-},z'_2) ,
\end{aligned}\tag{7}$$

where $g_{mm}(|\mathbf{k}_\parallel|)$ is defined by $\langle\zeta_m(\mathbf{k}_\parallel)\zeta_m(\mathbf{k}'_\parallel)\rangle=(2\pi)^2\delta(\mathbf{k}_\parallel+\mathbf{k}'_\parallel)\delta^2g_{mm}(|\mathbf{k}_\parallel|)$. $L_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega)_m$ in Eq. (7) satisfies the Bethe-Salpeter equation[10]

$$\begin{aligned}
L_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel,\omega)_m=&\sum_{\lambda\lambda'}d_{i\lambda}^{(0)}(\mathbf{k}_\parallel,\omega|z_{m-},z_{m+})^*d_{k\lambda'}^{(0)}(\mathbf{k}_\parallel,\omega|z_{m-},z_{m+})\\
&\times\left[(2\pi)^2\delta(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)\delta_{j\lambda}\delta_{l\lambda'}+\delta_m^2|\Lambda_m(\omega)|^2\int\frac{d^2k_\parallel^{(2)}}{(2\pi)^2}g_{mm}(|\mathbf{k}_\parallel-\mathbf{k}''_\parallel|)L_{\lambda j\lambda' l}(\mathbf{k}''_\parallel,\mathbf{k}'_\parallel,\omega)_m\right] .
\end{aligned}\tag{8}$$

The Bethe-Salpeter equation can be solved in the same way that is described in Ref. 10 under the assumption that the roughness has a Gaussian profile:

$$g_{mm}(|\mathbf{k}_\parallel|)=\pi a^2\exp(-\tfrac{1}{4}a^2|\mathbf{k}_\parallel|^2) .\tag{9}$$

Here δ and a are the root-mean-squared amplitude and the autocorrelation distance of the roughness, respectively.

By substituting the solution of Eq. (8) into Eq. (7), we obtain the two-photon Green's function, and then obtain the radiation intensity from Eq. (4). The final form of the radiated power from Eq. (4) is

$$\begin{aligned}
P(\mathbf{k}_\parallel^{(0)},\omega)=&\frac{A\varepsilon_0^{3/2}\omega^4\cos^2\theta_0}{16\pi^2c^5}\left|\frac{4\pi}{W(k_\parallel^{(0)},\omega)}\right|^2\\
&\times\sum_\mu|e_\mu^{>}(\mathbf{k}_\parallel^{(0)},\omega|z)|^2\\
&\times\int dQ_\parallel Q_\parallel dz'dz''J_{zz}(Q_\parallel,\omega|z',z'')\\
&\times\left[\frac{2\pi}{k_\parallel^{(0)}}\delta(k_\parallel-Q_\parallel)E_z^{<}(Q_\parallel,\omega|z')^*E_z^{<}(Q_\parallel,\omega|z'')+\pi a^2\delta^2|\Lambda_m(\omega)|^2\left|\frac{4\pi}{W_\parallel(Q_\parallel,\omega)}\right|^2\right.\\
&\times\sum_{nn'}(a_n^0a_{n'}^0)^{1/2}f_{nm}^0(k_\parallel|2)\{[1-P^0(\omega)]^{-1}\}_{nn'}\\
&\times\{g_{n'm}^0(Q_\parallel,\omega|4)E_z^{<}(Q_\parallel,\omega|z')^*E_z^{<}(Q_\parallel,\omega|z'')\theta(z_{m-}-z')\theta(z_{m-}-z'')\\
&+g_{n'm}^0(Q_\parallel,\omega|3)E_z^{<}(Q_\parallel,\omega|z')^*E_z^{>}(Q_\parallel,\omega|z'')\theta(z_{m-}-z')\theta(z''-z_{m-})\\
&+g_{n'm}^0(Q_\parallel,\omega|2)E_z^{>}(Q_\parallel,\omega|z')^*E_z^{<}(Q_\parallel,\omega|z'')\theta(z'-z_{m-})\theta(z_{m-}-z'')\\
&\left.+g_{n'm}^0(Q_\parallel,\omega|1)E_z^{>}(Q_\parallel,\omega|z')^*E_z^{>}(Q_\parallel,\omega|z'')\theta(z'-z_{m-})\theta(z''-z_{m-})\}\right] ,
\end{aligned}\tag{10}$$

where a_n^0 is defined in the same way as in Ref. 10, and $P_{nn'}^0(\omega)$ is

$$P_{nn'}^0(\omega)=\pi a^2\delta^2|\Lambda_m(\omega)|^2\sqrt{a_n^0 a_{n'}^0}\int\frac{dk_\parallel}{2\pi}k_\parallel g_{nm}^0(k_\parallel,\omega|1)|\tilde{d}(k_\parallel,\omega)|^2 f_{n'm}^0(k_\parallel,\omega|1)\ . \tag{11}$$

$\tilde{d}(k_\parallel,\omega)$ is given by

$$\tilde{d}(k_\parallel,\omega)=\frac{4\pi}{W_\parallel(k_\parallel,\omega)}\left[-1+\delta^2|\Lambda_m(\omega)|^2\int\frac{d^2k'_\parallel}{(2\pi)^2}\frac{\mathbf{e}^>(\mathbf{k}_\parallel,\omega|z_{m+})}{\left[\frac{W_\parallel(k_\parallel,\omega)}{4\pi}\right]^{1/2}}\cdot\frac{\mathbf{e}^<(\mathbf{k}'_\parallel,\omega|z_{m-})}{\left[\frac{W_\parallel(k'_\parallel,\omega)}{4\pi}\right]^{1/2}}\times\frac{\mathbf{e}^>(\mathbf{k}'_\parallel,\omega|z_{m+})}{\left[\frac{W_\parallel(k'_\parallel,\omega)}{4\pi}\right]^{1/2}}\cdot\frac{\mathbf{e}^<(\mathbf{k}_\parallel,\omega|z_{m-})}{\left[\frac{W_\parallel(k_\parallel,\omega)}{4\pi}\right]^{1/2}}g_{mm}(|\mathbf{k}_\parallel-\mathbf{k}'_\parallel|)\right]^{-1}\ . \tag{12}$$

$W(k_\parallel,\omega)$, $\mathbf{e}_\mu^>(\mathbf{k}_\parallel,\omega|z)$, $\mathbf{e}_\mu^<(\mathbf{k}_\parallel,\omega|z)$, $f_{\rm nm}^0$, and $g_{\rm nm}^0$ in Eqs. (10)–(12) have different forms for *p*- and *s*-polarized light. Their explicit forms are shown with the definition of $E_z^<(\mathbf{Q}_\parallel,\omega|z)$ and $E_z^>(\mathbf{Q}_\parallel,\omega|z)$ in the Appendix.

The first term of Eq. (10) corresponds to direct emission[8] that is independent of the interface roughness, and its properties are not discussed here. The remaining terms of Eq. (10) correspond to light intensity radiated due to roughness. We express the roughness induced intensity as $I_{n\rm th}$, where the subscript *n*th indicates that the intensity includes the higher-order terms. If we substitute $P_{nn'}^0(\omega)=0$ into $I_{n\rm th}$, we obtain $I_{\rm 1st}$ that coincides with the first-order perturbation result. The radiation intensity due to the higher-order terms are given by $I_{n\rm th}-I_{\rm 1st}$.

III. RESULTS

To understand the properties of the multiple-scattering effect that arises from the roughness at the metal-oxide interfaces not included in the work by Arya and Zeyher, we calculate the light-emission properties of the Al-Ox-Ag junction with identical geometrical parameters to those used by Arya and Zeyher.[10] The junction is modeled by a four-layered structure: an Al layer with semi-infinite thickness extending downward, a 3-nm-thick, oxide layer, a 20-nm thick Ag layer, and a vacuum layer that extends upward as illustrated in Fig. 1. The dielectric functions of Al and Ag are obtained from Ordal *et al.*[14] and Johnson and Christy,[15] respectively. The dielectric function of the oxide was taken to be constant at 3.1 in the relevant photon energy range (from 1.5 to 2.5 eV). The electron coherence length ξ is assumed to be 10 nm.[8] The autocorrelation distance of roughness a is fixed at 30 nm. All numerical results presented here were obtained for $V_0=4$ V and $I_0=55$ mA.[16]

Figures 2(a) and 2(b) show the *p*-polarized spectra emitted at 60° from the surface normal for $\delta=1.0$ and 7.0 nm, respectively. m in Fig. 2 represents the interface with roughness ($m=1$, 2, and 3 correspond to the interfaces of Al-oxide, oxide-Ag, and Ag-vacuum, respectively). The spectral shapes are similar for all three cases.

An interesting property is seen in the relative strengths for $m=1$, 2, and 3. For $\delta=1$ nm the emission intensity due to the roughness at the Ag-vacuum interface ($m=3$) is appreciably smaller than that due to the roughness at other two interfaces. In contrast, the emissions due to the roughness at the interfaces have similar intensities when $\delta=7$ nm. This difference is due to the higher-order terms that become significant for large δ. The emission intensities due to the first-order perturbation term are proportional to δ^2. Thus the change of the relative strengths with increasing δ from 1 to 7 nm, as seen in Fig. 2, does not arise for the first-order perturbation term.

In order to evaluate the fraction of emission intensity due to the higher-order terms, we calculate the ratio R between the *p*-polarized emission intensity due to the

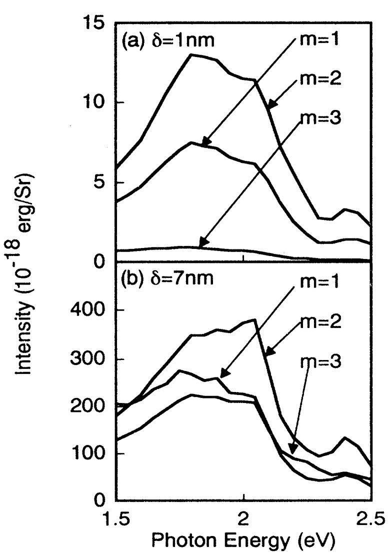

FIG. 2. *p*-polarized light-emission spectra from the Al-Ox-Ag junction. The bias voltage is 4 V and the tunneling current is 55 mA. The autocorrelation distance a is 30 nm and the root-mean-squared amplitude δ is 1 nm for (a) and 7 nm for (b). m represents the interface where the roughness is placed.

higher-order terms and the intensity due to the first-order perturbation terms:

$$R \equiv \frac{I_{n\mathrm{th}} - I_{1\mathrm{st}}}{I_{1\mathrm{st}}} . \tag{13}$$

Figures 3(a), 3(b), and 3(c) show the δ dependence of R at the photon energy of 2 eV for the three different locations of interface roughness ($m=1$, 2, and 3), respectively. For $m=1$ and 2, where the roughness is placed at the metal-oxide interfaces, R first increases steeply, takes a maximum value of ~ 0.5, and then decreases slowly with increasing δ. That is to say, the emission intensities due to the higher-order terms are at most half of the first-order perturbation contribution for the roughness at the metal-oxide interfaces. However, for $m=3$ it increases monotonically with the increase in δ, in agreement with the result reported by Arya and Zeyher.[10] The change in the relative strengths for $m=1$, 2, and 3 between Figs. 2(a) and 2(b) arises from the difference in the δ dependence for $m=1$ and 2, and $m=3$.

Figures 4(a), 4(b), and 4(c) show the δ dependence of the intensity ratios of the p-polarized light to the s-polarized light (p/s ratio) at the photon energy of 2 eV for $m=1$, 2, and 3, respectively. The emission angle is 60° from the surface normal. The dashed lines represent the p/s ratios calculated by the first-order perturbation theory. Since the emission intensities due to the first-order perturbation term are proportional to δ^2 for both p- and s-polarized light, the p/s ratio is independent of δ.

For $m=1$ and 2 in Fig. 4 the p/s ratio increases first, takes a maximum value, and then decreases to approach the asymptotic value determined by the first-order perturbation term. For $m=3$ it increases monotonically with increasing δ. By comparing Fig. 4 with Fig. 3 we see a strong correlation between the δ dependence of the p/s ratio and that of the p-polarized emission intensity due to the higher-order terms.

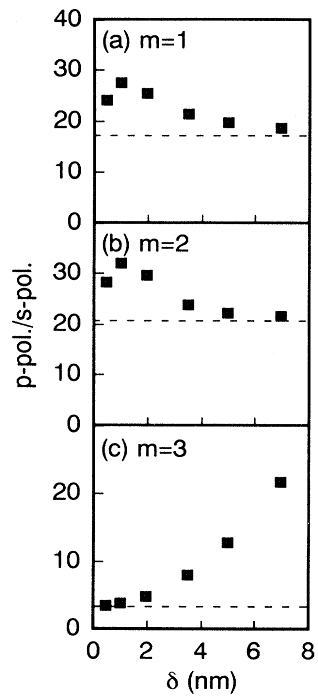

FIG. 4. δ dependence of the ratio of the p-polarized light intensity to the s-polarized intensity at the photon energy of 2 eV. m denotes the interface where roughness is placed.

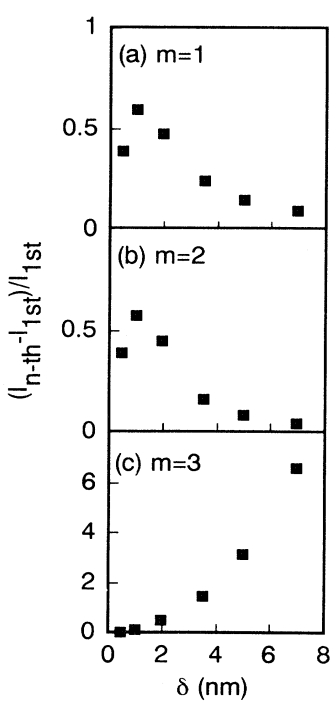

FIG. 3. δ dependence of the emission intensities due to the higher-order terms at the photon energy of 2 eV. m represents the interface where the roughness is placed.

IV. DISCUSSION

As we have remarked earlier, first-order perturbation theory[9] shows that the light-emission intensity due to the roughness at the metal-oxide interfaces ($m=1$ and 2) is stronger than that due to the roughness at the top surface of the junction ($m=3$); Arya and Zeyher[10] found that the light-emission intensity due to higher-order terms for $m=3$ increases faster with increasing δ than the first-order result which is proportional to δ^2. Thus one might expect that the light-emission intensity due to the higher-order terms for $m=1$ and 2 could increase faster than that for $m=3$ with increasing δ. However, the result we have seen above contradicts this expectation. In what follows we will see why we obtained the unexpected result.

Let us consider the δ dependence of the light-emission intensity $I_{n\mathrm{th}}$. As seen from Eq. (10), it has the following form:

$$I_{n\mathrm{th}} = (a\delta)^2 \sum_{nn'} F_n [1 - P^0(\omega)]^{-1}_{nn'} G_{n'} , \tag{14}$$

where F_n and $G_{n'}$ are independent of δ. Equation (14) can be expanded into a series of the matrix $P^0(\omega)$:

$$I_{n\text{th}}=(a\delta)^2\sum_{nn'}F_n[1+P^0(\omega)+P^0(\omega)^2+P^0(\omega)^3+\cdots]_{nn'}G_n \ . \qquad (15)$$

The first term (unity) and the remaining terms in the square bracket correspond to the first- and higher-order contributions, respectively.

From Eq. (12) we see that $\tilde{d}(k_\parallel,\omega)$ in $P^0(\omega)$ has the following δ dependence:

$$\tilde{d}(k_\parallel,\omega)=\frac{4\pi}{W_\parallel(k_\parallel,\omega)}[-1+B(k_\parallel,z_m)\delta^2]^{-1} \ , \qquad (16)$$

where $B(k_\parallel,z_m)$ is independent of δ. Thus the nn' element of $P^0(\omega)$ has the following δ dependence:

$$P^0_{nn'}(\omega)=\pi a^2\delta^2\int\frac{dk_\parallel}{2\pi}k_\parallel C_{nn'}(k_\parallel)\left|\frac{4\pi}{W_\parallel(k_\parallel,\omega)}\right|^2\times\left|\frac{1}{-1+B(k_\parallel,z_m)\delta^2}\right|^2 , \qquad (17)$$

where $C_{nn'}$ is independent of δ.

The integrand in Eq. (17) has large values on the dispersion curves of the surface plasmon polaritons (SPP's) in the junction. This is because the relation

$$W_\parallel(k_\parallel,\omega)=0 \qquad (18)$$

determines the dispersion curves of the SPP's. There are two modes of SPP in the Al-Ox-Ag junction with a four-layered structure: the fast mode SPP that is localized at the top surface, and the slow mode SPP that is localized across the oxide layer. A numerical investigation shows that the integrand in Eq. (17) has larger values on the dispersion curve of the slow mode SPP than on the dispersion curve of the fast mode SPP for any location of the roughness ($m=1$, 2, or 3). Thus Eq. (17) is approximately written

$$P^0_{nn'}(\omega)\cong\pi a^2\delta^2\left|\frac{1}{-1+B(k_{\parallel\text{slow mode}},z_m)\delta^2}\right|^2\times\int\frac{dk_\parallel}{2\pi}k_\parallel C_{nn'}(k_\parallel)\left|\frac{4\pi}{W_\parallel(k_\parallel,\omega)}\right|^2 , \qquad (19)$$

where $k_{\parallel\text{slow mode}}$ is the absolute value of the wave vector of the slow mode SPP at frequency ω, and z_m is the z position of the mth averaged interface plane.

Now we can understand the δ dependence of $P^0_{nn'}(\omega)$ and $I_{n\text{th}}$. When $|\delta^2B(k_{\parallel\text{slow mode}},z_m)|\ll 1$ we see from Eq. (19) that $P^0_{nn'}(\omega)$ is proportional to δ^2. Thus as seen from Eq. (15) the contribution from the higher-order terms to $I_{n\text{th}}$ becomes significant when δ increases. For the opposite case of $|\delta^2B(k_{\parallel\text{slow mode}},z_m)|\gg 1$, $P^0_{nn'}(\omega)$ is proportional to δ^{-2}. Thus the higher-order terms in Eq. (15) decrease with the increase in δ. Then $I_{n\text{th}}$ approaches $I_{1\text{st}}$ for large δ.

From the definition of $B(k_\parallel,z_m)$ in Eq. (16) we see that $B(k_{\parallel\text{slow mode}},z_m)$ is proportional to the electric fields of the upward and downward propagating slow modes of SPP's,

$$\frac{\mathbf{e}^>(k_{\parallel\text{slow mode}},\omega|z_{m+})}{\left[\dfrac{W_\parallel(k_{\parallel\text{slow mode}},\omega)}{4\pi}\right]^{1/2}}$$

and

$$\frac{\mathbf{e}^<(k_{\parallel\text{slow mode}},\omega|z_{m-})}{\left[\dfrac{W_\parallel(k_{\parallel\text{slow mode}},\omega)}{4\pi}\right]^{1/2}} \ .$$

Since the slow mode SPP is localized across the oxide layer, $|B(k_{\parallel\text{slow mode}},z_m)|$ for $m=1$ and 2 have values larger than that for $m=3$. Thus the condition $|\delta^2B(k_{\parallel\text{slow mode}},z_m)|\sim 1$ is satisfied around $\delta=1$ nm for $m=1$ and 2, and the emission intensity due to the higher-order terms $(I_{n\text{th}}-I_{1\text{st}})$ decreases above $\delta=1$ nm. On the other hand, for $m=3$, $|\delta^2B(k_{\parallel\text{slow mode}},z_3)|\ll 1$ in the relevant δ region, because of the weak electric-field strength of the slow mode SPP at the top surface of the junction. Thus $(I_{n\text{th}}-I_{1\text{st}})$ increases monotonically as δ increases.

The origin of the decrease of the light intensity for large δ due to the higher-order terms for $m=1$ and 2 can be understood in a different manner. The one-photon Green's function for an n-layered structure with roughness at the mth interface is

$$d_{\mu\nu}(\mathbf{k}_\parallel,\omega|z_{m-},z_{m+})=\tilde{d}(k_\parallel,\omega)e^<_\mu(\mathbf{k}'_\parallel,\omega|z_{m-})e^>_\nu(\mathbf{k}'_\parallel,\omega|z_{m+}) \ . \qquad (20)$$

This expression is a generalization of Eq. (35) in Ref. 14. By substituting Eq. (16) into Eq. (20) we obtain

$$d_{\mu\nu}(\mathbf{k}_\parallel,\omega|z_{m-},z_{m+})=\left\{\frac{4\pi}{W_\parallel(k_\parallel,\omega)}e^<_\mu(\mathbf{k}'_\parallel,\omega|z_{m-})e^>_\nu(\mathbf{k}'_\parallel,\omega|z_{m+})\right\}\times\left\{\frac{1}{-1+B(k_\parallel,z_m)\delta^2}\right\} . \qquad (21)$$

We see that the right-hand side consists of two factors: the electromagnetic Green's function for the n-layered structure with flat interfaces [the first curly bracket of the right-hand side of Eq. (21)] and the δ-dependent factor (the second curly bracket) which shows the same δ dependence as the root of $P^0_{nn'}(\omega)$. Since the one-photon Green's function is a mathematical representation of SPP's, the δ-dependent term in Eq. (21) represents the dissipation of SPP's due to scattering from roughness. The cross section of the scattering that is determined by $B(k_\parallel,z_m)$ takes large values for $m=1$ and 2. Then the frequent scattering of the slow mode SPP from the roughness at the metal-oxide interfaces results in a large dissipation of SPP's, and this large dissipation causes the decrease of the light-emission intensity due to the higher-order terms $(I_{n\text{th}}-I_{1\text{st}})$ for $m=1$ and 2.

V. CONCLUSION

We have investigated the multiple-scattering effect of SPP's due to the roughness at the metal-oxide interfaces in the light-emission process from the *M-I-M* tunnel junction. We found that the light-emission intensity due to the higher-order terms $(I_{n\text{th}}-I_{1\text{st}})$ first increases, takes a maximum value around a half of that obtained by the first-order perturbation theory $I_{1\text{st}}$, and then decreases with increasing δ. The large dissipation of the slow mode SPP due to scattering from the roughness at the metal-oxide interfaces causes this decrease.

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APPENDIX

$E_\mu^<(\mathbf{k}_\parallel,\omega|z)$ and $E_\mu^>(\mathbf{k}_\parallel,\omega|z)$ are two linearly independent solutions of the homogeneous wave equation corresponding to Eq. (2). They can be written

$$E_\mu^{>,<}(\mathbf{k}_\parallel,\omega|z)=\{A_\mu^{>,<}(\mathbf{k}_\parallel,\omega|i)\exp[ik_z(i)(z-z_{i-1})]+B_\mu^{>,<}(\mathbf{k}_\parallel,\omega|i)\exp[-ik_z(i)(z-z_{i-1})]\}\exp[i(\mathbf{k}_\parallel\cdot\mathbf{x}-\omega t)]\ ,$$

where

$$z_i>z>z_{i-1}\ ,$$

and

$$k_z(i)=\left[\frac{\varepsilon_i\omega^2}{c^2}-\mathbf{k}_\parallel^2\right]^{1/2}\ .$$

A matrix method for obtaining $A_\mu^{>,<}(\mathbf{k}_\parallel,\omega|i)$ and $B_\mu^{>,<}(\mathbf{k}_\parallel,\omega|i)$ for an arbitrary n-layered structure is presented in Ref. 13.

$W(\mathbf{k}_\parallel,\omega)$, $e_\mu^>(\mathbf{k}_\parallel,\omega|z)$, $e_\mu^<(\mathbf{k}_\parallel,\omega|z)$, f_{nm}^0, and g_{nm}^0 are obtained by a simple extension of the results in Ref. 10. Thus we present only the final results here. They have different forms for p- and s-polarized light.

For p-polarized light, we obtain

$$W(k_\parallel^{(0)},\omega)\equiv W_\parallel(k_\parallel^{(0)},\omega)$$
$$=-i\frac{2\varepsilon_n^{1/2}\omega\cos\theta_0}{c\sin^2\theta_0}B_z^<(k_\parallel^{(0)},\omega|n)\ ,$$

$$\mathbf{e}^{>,<}(k_\parallel^{(0)},\omega|z)=E_x^{>,<}(k_\parallel^{(0)},\omega|z)\hat{\mathbf{k}}_\parallel+iE_z^{>,<}(k_\parallel^{(0)},\omega|z)\hat{\mathbf{z}}\ ,$$

where $\hat{\mathbf{k}}_\parallel$ and $\hat{\mathbf{z}}$ are the unit vectors parallel to $\mathbf{k}_\parallel$ and the z axis, respectively,

$$f_{4h+1,m}^0(k_\parallel^{(0)},\omega|2)$$
$$=\exp(-\tfrac{1}{4}a^2k_\parallel^{(0)2})E_z^<(k_\parallel^{(0)},\omega|z_{m+})^*\times E_z^<(k_\parallel^{(0)},\omega|z_{m+})\left[\frac{1}{\sqrt{2}}ak_\parallel^{(0)}\right]^{2h}\ ,$$

$$f_{4h+2,m}^0(k_\parallel^{(0)},\omega|2)$$
$$=i\exp(-\tfrac{1}{4}a^2k_\parallel^{(0)2})E_z^<(k_\parallel^{(0)},\omega|z_{m+})^*\times E_x^<(k_\parallel^{(0)},\omega|z_{m+})\left[\frac{1}{\sqrt{2}}ak_\parallel^{(0)}\right]^{2h+1}\ ,$$

$$f_{4h+3,m}^0(k_\parallel^{(0)},\omega|2)$$
$$=i\exp(-\tfrac{1}{4}a^2k_\parallel^{(0)2})E_x^<(k_\parallel^{(0)},\omega|z_{m+})^*\times E_z^<(k_\parallel^{(0)},\omega|z_{m+})\left[\frac{1}{\sqrt{2}}ak_\parallel^{(0)}\right]^{2h+1}\ ,$$

$$f_{4h+4,m}^0(k_\parallel^{(0)},\omega|2)$$
$$=\exp(-\tfrac{1}{4}a^2k_\parallel^{(0)2})E_x^<(k_\parallel^{(0)},\omega|z_{m+})^*\times E_x^<(k_\parallel^{(0)},\omega|z_{m+})\left[\frac{1}{\sqrt{2}}ak_\parallel^{(0)}\right]^{2h}\ .$$

$f_{n,m}^0(k_\parallel,\omega|1)$ is given by replacing $E_\mu^<(k_\parallel^{(0)},\omega|z_{m+})$ with $E_\mu^>(k_\parallel,\omega|z_{m+})$ in $f_{n,m}^0(k_\parallel,\omega|2)$.

$$g_{4h+1,m}^0(Q_\parallel,\omega|4)$$
$$=\exp(-\tfrac{1}{4}a^2Q_\parallel^2)E_z^>(Q_\parallel,\omega|z_{m-})^*\times E_z^>(Q_\parallel,\omega|z_{m-})\left[\frac{1}{\sqrt{2}}aQ_\parallel\right]^{2h}\ ,$$

$$g_{4h+2,m}^0(Q_\parallel,\omega|4)$$
$$=i\exp(-\tfrac{1}{4}a^2Q_\parallel^2)E_z^>(Q_\parallel,\omega|z_{m-})^*\times E_x^>(Q_\parallel,\omega|z_{m-})\left[\frac{1}{\sqrt{2}}aQ_\parallel\right]^{2h+1}\ ,$$

$$g_{4h+3,m}^0(Q_\parallel,\omega|4)$$
$$=i\exp(-\tfrac{1}{4}a^2Q_\parallel^2)E_x^>(Q_\parallel,\omega|z_{m-})^*\times E_z^>(Q_\parallel,\omega|z_{m-})\left[\frac{1}{\sqrt{2}}aQ_\parallel\right]^{2h+1}\ ,$$

$$g_{4h+4,m}^0(Q_\parallel,\omega|4)$$
$$=\exp(-\tfrac{1}{4}a^2Q_\parallel^2)E_x^>(Q_\parallel,\omega|z_{m-})^*\times E_x^>(Q_\parallel,\omega|z_{m-})\left[\frac{1}{\sqrt{2}}aQ_\parallel\right]^{2h}\ .$$

$g^0_{n,m}(Q_\parallel,\omega|3)$ and $g^0_{n,m}(Q_\parallel,\omega|2)$ are given by replacing $E_x^>(Q_\parallel,\omega|z_{m-})$ with $E_x^<(Q_\parallel,\omega|z_{m-})$, and replacing $E_x^>(Q_\parallel,\omega|z_{m-})^*$ with $E_x^<(Q_\parallel,\omega|z_{m-})^*$, respectively, in $g^0_{n,m}(Q_\parallel,\omega|4)$. $g^0_{n,m}(Q_\parallel,\omega|1)$ is given by replacing $E_x^>(Q_\parallel,\omega|z_{m-})^*$ and $E_x^>(Q_\parallel,\omega|z_{m-})$ by $E_x^<(Q_\parallel,\omega|z_{m-})^*$ and $E_x^<(Q_\parallel,\omega|z_{m-})$, respectively, in $g^0_{n,m}(Q_\parallel,\omega|4)$.

For *s*-polarized light,

$$W(k_\parallel^{(0)},\omega)\equiv W_\perp(k_\parallel^{(0)},\omega)=i\frac{2\varepsilon_0^{1/2}\omega\cos\theta_0}{c}B_y^<(k_\parallel^{(0)},\omega|n)\ ,$$

$$\mathbf{e}^{>,<}(k_\parallel^{(0)},\omega|z)=E_y^{>,<}(k_\parallel^{(0)},\omega|z)(\hat{\mathbf{k}}_\parallel\times\hat{\mathbf{z}})\ ,$$

$$f^0_{4h+1,m}(k_\parallel^{(0)},\omega|2)=f^0_{4h+2,m}(k_\parallel^{(0)},\omega|2)=f^0_{4h+3,m}(k_\parallel^{(0)},\omega|2)=0\ ,$$

$$f^0_{4h+4,m}(k_\parallel^{(0)},\omega|2)=\exp(-\tfrac{1}{4}a^2k_\parallel^{(0)2})E_y^<(k_\parallel^{(0)},\omega|z_{m+})^*\times E_y^<(k_\parallel^{(0)},\omega|z_{m+})\left[\frac{1}{\sqrt{2}}ak_\parallel^{(0)}\right]^{2h}\ ,$$

$$g^0_{4h+1,m}(Q_\parallel,\omega|4)=g^0_{4h+2,m}(Q_\parallel,\omega|4)=g^0_{4h+3,m}(Q_\parallel,\omega|4)=0\ ,$$

$$g^0_{4h+4,m}(Q_\parallel,\omega|4)=\exp(-\tfrac{1}{4}a^2Q_\parallel^2)E_x^>(Q_\parallel,\omega|z_{m-})^*\times E_x^>(Q_\parallel,\omega|z_{m-})\left[\frac{1}{\sqrt{2}}aQ_\parallel\right]^{2h}\ .$$

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