

## Overbias Light Emission due to Higher-Order Quantum Noise in a Tunnel Junction

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Understanding tunneling from an atomically sharp tip to a metallic surface requires us to account for interactions on a nanoscopic scale. Inelastic tunneling of electrons generates emission of photons, whose energies intuitively should be limited by the applied bias voltage. However, experiments [G. Schull *et al.*, Phys. Rev. Lett. 102, 057401 (2009)] indicate that more complex processes involving the interaction of electrons with plasmon polaritons lead to photon emission characterized by overbias energies. We propose a model of this observation in analogy to the dynamical Coulomb blockade, originally developed for treating the electronic environment in mesoscopic circuits. We explain the experimental finding quantitatively by the correlated tunneling of two electrons interacting with a *LRC* circuit modeling the local plasmon-polariton mode. To explain the overbias emission, the non-Gaussian statistics of the tunneling dynamics of the electrons is essential.

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Light emission of electrons tunneling from a scanning tunneling microscope (STM) to a metallic surface has already been studied for many years [1]. The basic mechanism leading to light emission has been identified as the interaction of the tunneling electrons with a localized surface plasmon-polariton (SPP) mode [2,3]. In a simple picture, one expects that the emitted light spectrum is limited by the bias voltage,  $\hbar\omega < eV$ . This is a consequence of the Fermi seas in the electronic leads, which prohibit inelastic tunneling with higher energy exchange due to the Pauli principle. The SPP resonance, which is finally responsible for the photon emission, acts as a frequency filter, and, hence, the measured spectrum is essentially the SPP resonance cut off at a frequency  $eV/\hbar$ . This picture has been confirmed in numerous experimental [4] and theoretical [5] studies. However, a closer look at some experiments [6–8] reveals the unexpected feature that, in addition, light with energy  $\hbar\omega > eV$  is emitted that shows a spectrum which is also reminiscent of the SPP modes. Several models explaining this overbias emission have been proposed, ranging from black-body radiation [9] and smearing of the electrodes' Fermi distributions [10] to nonlinear electronic processes such as hot-hole formation [6–8,11,12] and Auger-like processes [6]. In addition, fluorescence with overbias emission has been reported for molecular films [13–16]. However, understanding the detailed electron processes is an ongoing task [17].

In this Letter, we will develop a theoretical model of light emission by a tunnel contact based on the powerful method of dynamical Coulomb blockade. The idea is that on a short time scale, tunneling is not adequately described by incoherent electrons but by a coherent two-electron tunneling process in which each electron contributes an energy  $\lesssim eV$  creating an overbias SPP excitation that finally leads to the overbias light emission [see Figs. 1(a) and 1(b)]. It is essential that the coupled electron-SPP system is treated as a

quantum coherent entity since intermediate virtual states are involved. Considering a single sufficiently damped SPP resonance, we can quantitatively reproduce the experimentally observed bias voltage-dependent emission spectrum. Furthermore, it is absolutely essential that the tunnel junction is described as a non-Gaussian quantum noise emitter, which at the same time means that the observed overbias emission is a new manifestation of the nontrivial statistics of quantum transport [18,19]. Using the method of Ref. [20], we calculate the emission spectrum for all energies up to second order in the tunnel conductance and find quantitative agreement with experimental results of Ref. [7]. Hence, we present for the first time a unified description of plasmon-enhanced light emission in the sub-bias as well as in the overbias regime [see, e.g., Fig. 1(d)].

As a first step, we develop a model of the SPP-mediated light emission of a tunnel junction inspired by the theory of environmental Coulomb blockade developed more than two decades ago in mesoscopic physics [21]. In this picture, tunneling in a junction is strongly modified because the electronic environment leads to fluctuations of the voltage difference across the junction showing up as, e.g., a zero-bias anomaly in the differential conductance [22,23]. Considering that nonsymmetrized current fluctuations induced by the combined tunnel-junction-environment-impedance system can be seen as light emission [24–26], we suggest to model the SPP resonance as an electromagnetic resonator with appropriate damping interacting with the tunneling electrons of the STM (see Fig. 1).

In the second step, to theoretically address the problem, we will make use of a recent observation in Ref. [20] stating that phase fluctuations in a coherent conductor-environment system lead to a subtle interference effect between two-photon processes and two-electron processes that may be identified by a strongly coupled quantum tunneling detector. We will adapt the formalism of Ref. [20] to the quantum

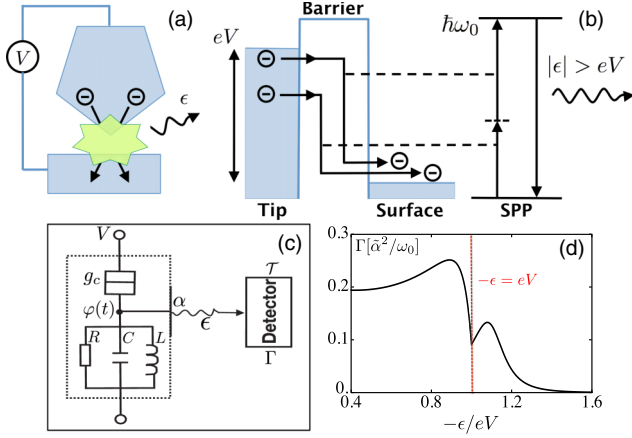


FIG. 1 (color online). (a) A STM contact with bias voltage  $V$  showing a correlated two-electron tunneling process. The electrons interact via a SPP mode (green), which finally leads to light emission from the junction. (b) The electron tunneling process in energy space shows how two electrons excite a SPP via a virtual state. The SPP decays by emitting a photon with an energy  $|\epsilon| > eV$ . (c) The electromagnetic model circuit: a  $LRC$  resonant circuit mimics the (damped) SPP, and the photons emitted from the coupled tunnel junction are captured by the detector. (d) The photon detection rate  $\Gamma$  reflects the emission spectrum with energy  $|\epsilon|$  showing a sharp kink at  $eV$  and a substantial overbias emission. The parameters are the resonance frequency  $\omega_0 = 1.1 eV$ , the broadening  $\eta = 0.2 eV$ , and the coupling parameter  $g_c z_0^2 = 0.3$ . See the text for further explanations of the parameters.

detection of light emission from our coupled junction-resonator system, but since experimentally the detector is far away from the junction and the emission efficiency is only  $\lesssim 10^{-4}$  [7], it is sufficient to work in lowest order in the detector coupling,  $\alpha$ , in our model; see Fig. 1(c).

We will start by showing how we intend to model the interaction between the tunneling current and the SPP using methods of environmental Coulomb blockade theory [21,22]. According to standard theory [21,22], we model the tunneling from the STM tip to the surface in an electromagnetic environment as the circuit diagram depicted in Fig. 1(c). We consider a tunnel conductor with a dimensionless conductance  $g_c = R_Q/R_c$  with  $R_Q = h/2e^2$  and  $R_c$  being the quantum and tunneling resistances, respectively. The junction is coupled to a damped  $LC$  circuit, which we model by an impedance  $z_\omega = iz_0\omega\omega_0/(\omega_0^2 - \omega^2 + i\omega\eta)$ , where  $\omega_0 = 1/\sqrt{LC}$  is the resonance frequency of the SPP mode,  $\eta = 1/RC$  models the damping, and  $z_0 = \sqrt{L/C}/R_Q$ . We will later determine these parameters from the experiment [7]. The interaction between the tunnel junction and the SPP occurs in this model via the dynamical voltage fluctuations on the node between the tunnel junction and the  $LRC$  circuit, which can be expressed by the phase variable  $\varphi(t) = (e/\hbar) \int_{-\infty}^t dt' V(t')$ .

To model the emission detection of the photons, we follow the standard path and model the detector as a two-level

system, in which the emitted photons trigger transitions between states characterized by an energy difference  $\epsilon$  and a matrix element  $\mathcal{T}$ . We introduce a coupling constant  $\alpha$  between the voltage fluctuations and the energy level of the detector, viz.,  $\epsilon \rightarrow \epsilon + \alpha eV(t)$ . Finally, we will take the interaction to be weak, since the photon detectors in the real experiments are far away from the junction. Using Fermi's golden rule and setting  $\hbar = 1$ , the detection rate at energy  $\epsilon$  due to the fluctuations of  $\alpha\varphi(t)$  [20–22] is

$$\Gamma(\epsilon) = |\mathcal{T}|^2 \int dt \langle e^{i\alpha\varphi(t)} e^{-i\alpha\varphi(0)} \rangle e^{i\epsilon t}. \quad (1)$$

To calculate  $\langle e^{i\alpha\varphi(t)} e^{-i\alpha\varphi(0)} \rangle$ , we employ the path integral method, in which the real fields  $\varphi^\pm(t)$  are defined on the forward and backward Keldysh contours, respectively. The dynamics of the coupled SPP-electron system is determined by the Keldysh actions of the conductor  $\mathcal{S}_c$  and the circuit,  $\mathcal{S}_e$ . The correlator can then be represented as

$$\langle e^{i\alpha\varphi(t)} e^{-i\alpha\varphi(0)} \rangle = \int \mathcal{D}[\Phi] \exp\{-i\mathcal{S}_e[\Phi] - i\mathcal{S}_c[\Phi] + i\alpha[-\varphi^+(0) + \varphi^-(t)]\}, \quad (2)$$

where  $\Phi = ((\varphi^+ + \varphi^-)/2, \varphi^+ - \varphi^-)^T$ . The action of the  $LRC$  circuit, i.e., the damped  $LC$  oscillator acting as the environment on the tunnel conductor, is quadratic in the fields and at zero temperature given by [27,28]

$$\mathcal{S}_e = \int d\omega \Phi_{-\omega}^T A_\omega \Phi_\omega, \quad A_\omega = -\frac{i}{2} \begin{pmatrix} 0 & -\frac{\omega}{z_\omega} \\ \frac{\omega}{z_\omega} & |\omega| \text{Re}\left\{\frac{1}{z_\omega}\right\} \end{pmatrix}.$$

The action  $\mathcal{S}_c$  can be expressed in terms of Keldysh Green's functions  $\check{G}_{L,R}$  for the free electrons on the left ( $L$ ) and right ( $R$ ) sides of the tunneling barrier:

$$\mathcal{S}_c = \frac{i}{8} g_c \int dt dt' \text{Tr}\{\check{G}_L(t, t'), \check{G}_R(t' - t)\}. \quad (3)$$

With the equilibrium Keldysh Green's function

$$\check{G}(\omega) = \begin{pmatrix} 1 - 2f(\omega) & 2f(\omega) \\ 2[1 - f(\omega)] & 2f(\omega) - 1 \end{pmatrix}$$

containing the Fermi function  $f(\omega) = [\exp(\beta\omega) + 1]^{-1}$ , we can write  $\check{G}_R(\omega) = G(\omega - eV)$ . We introduce  $\check{G}_L(t, t') = \check{U}^\dagger(t) \check{G}(t - t') \check{U}(t')$  with the counting fields [29] as

$$\check{U}(t) = \begin{pmatrix} e^{-i\varphi^+(t)} & 0 \\ 0 & e^{-i\varphi^-(t)} \end{pmatrix}.$$

This concludes the description of our theoretical formalism. The rate cannot be calculated exactly since the action of the

conductor is non-Gaussian, and we need an approximation scheme.

A first approximation is considering only the Gaussian part of the conductor action. In this case, the whole path integral becomes Gaussian and corresponds to the well-known results from  $P(E)$  theory. The quadratic part of the conductor action reads

$$\mathbb{S}_c^G = \int d\omega \Phi_{-\omega}^T B_\omega \Phi_\omega, \quad B_\omega = -\frac{i}{2} \begin{pmatrix} 0 & -\omega g_c \\ \omega g_c & S_c(\omega) \end{pmatrix},$$

with the symmetrized quantum noise of a tunnel contact  $S_c(\omega) = g_c[|\omega| + Y(|\omega| - eV)]$  using  $Y(\omega) = -\omega\theta(-\omega)$ . We will later discuss quantitative limitations of this approximation. However, already now we see that the Gaussian part alone will only lead to single photon emission with sub-bias energies.

Combining all the quadratic parts from both the *LRC* circuit and the conductor in a single matrix  $D_\omega \equiv A_\omega + B_\omega$ , the correlation function  $\langle e^{i\alpha\varphi(t)} e^{-i\alpha\varphi(0)} \rangle \equiv e^{\alpha^2 J(t)}$  can be evaluated. As a result, one finds

$$J(t) = \int d\omega \frac{|\tilde{z}_\omega|^2}{\omega^2} S_t(\omega) (e^{-i\omega t} - 1), \quad (4)$$

where  $S_t(\omega) = S_c(\omega) + g_c\omega + 2Y(-\omega)\text{Re}\{1/z_\omega\}$  is the total noise spectral density and the impedance  $\tilde{z}_\omega = z_\omega/(1 + z_\omega g_c)$ . The renormalized impedance  $\tilde{z}_\omega$  is the parallel connection of the tunnel junction and the environmental impedance as seen by the detector. This means the factor  $g_c$  leads to an increased damping and can be absorbed in a renormalized  $\eta$ . From Eq. (1), in lowest order in  $\alpha^2$ , we find the rate

$$\Gamma_G(\epsilon) = \tilde{\alpha}^2 \frac{|\tilde{z}_\epsilon|^2}{\epsilon^2} \left[ g_c Y(|\epsilon| - eV) + \text{Re}\left(\frac{2}{\tilde{z}_\epsilon}\right) Y(-\epsilon) \right]. \quad (5)$$

Here, we have introduced a prefactor  $\tilde{\alpha}^2 = |\mathcal{T}|^2 \alpha^2$ .

The result (5) matches the simple expectation from the golden rule [25]. The photon emission that is described by energies  $\epsilon < 0$  is only caused by the nonequilibrium electrons of the tunnel junction and is, therefore, limited by the maximum energy  $eV$  available for inelastic transitions. As the emission requires interaction with the environmental resonator, the electronic inelastic emission spectrum is filtered by the SPP resonance. This is demonstrated in Fig. 2, which shows the emission rate for different bias voltages. There is always a sharp threshold for  $-\epsilon = eV$ , and the SPP resonance becomes visible if the threshold is larger than the resonance energy,  $\omega_0$ .

We have seen that the Gaussian approximation of the rate does not result in an overbias emission. It is not possible to calculate the non-Gaussian part exactly. Fortunately, we can make use of the limit  $z_\omega^2 g_c \ll 1$  motivated by the experiment by Schull *et al.* [7]. From our fitting later, we can infer that the

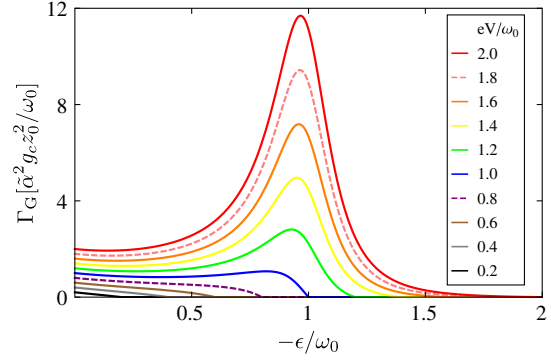


FIG. 2 (color online). The Gaussian contribution to the emission spectrum for different bias voltages. The SPP peak becomes clearly visible as the bias voltage exceeds the resonance energy,  $\omega_0$ . In all cases, the spectrum sharply drops to zero for  $-\epsilon > eV$ . This behavior ascertains that the responsible processes are limited by single-electron tunneling events. The broadening parameter is chosen as  $\eta = 0.3\omega_0$ .

fluctuations of the phase are small due to the dominating Gaussian part governed by the small environmental impedance. Therefore, we can make an expansion of the non-quadratic part of the action  $\mathbb{S} = \mathbb{S}_e + \mathbb{S}_e^G + \mathbb{S}_c^{(3)} + \mathbb{S}_c^{(4)} + O(\Phi^5)$  using  $\exp[-i\mathbb{S}_c^{(3)} - i\mathbb{S}_c^{(4)}] \approx 1 - i\mathbb{S}_c^{(3)} - i\mathbb{S}_c^{(4)}$ . This is possible since the Gaussian part of the action is dominated by the fluctuations of the small impedance of the environment, viz.,  $\Phi^2 < z_\omega/\omega^2$ , and, therefore, the higher-order terms are small by the factor  $g_c z_\omega^2 \ll 1$ .

After the expansion, the remaining path integral is just the Gaussian average of the third and fourth moments. The Gaussian average is then given by

$$\langle\langle \dots \rangle\rangle \equiv \int \mathcal{D}[\Phi] (\dots) e^{\int d\omega \{-i\Phi_{-\omega}^T D_\omega \Phi_\omega + i\alpha b_\omega^T(t) \Phi_\omega\}}, \quad (6)$$

where  $b_\omega(t) = (e^{-i\omega t} - 1, -(e^{-i\omega t} + 1)/2)^T$ . Now, all remaining averages can be calculated using Wick's theorem and, as usual, this gives the sum over all possible pairings of single and double averages. The basic averages in frequency space can be expressed in terms of the building blocks  $D_\omega$  and  $b_\omega(t)$  using  $\tilde{D}_\omega^{-1} = D_\omega^{-1} \exp[\alpha^2 J(t)]/2$ :

$$\langle\langle \Phi_\omega \rangle\rangle = \alpha \tilde{D}_\omega^{-1} b_{-\omega}(t), \quad \langle\langle \Phi_\omega \Phi_{-\omega}^T \rangle\rangle = -i \tilde{D}_\omega^{-1}. \quad (7)$$

Note that these expressions still are valid for an arbitrary value of  $\alpha$ . A drastic simplification arises if we limit ourselves to the experimentally relevant weak detection limit in which  $\alpha \ll 1$ . The leading-order contributions to the detector rate are given by combinations of the type  $\langle\langle \varphi_\omega \rangle\rangle \langle\langle \varphi_{-\omega} \rangle\rangle \langle\langle \varphi_{\omega'} \varphi_{-\omega'} \rangle\rangle$  since single averages are of leading order  $\alpha$ . Contributions of zeroth order in  $\alpha$  are time independent and, therefore, only play a part in the elastic rate characterized by  $\epsilon = 0$ , which is not of interest here. Limiting ourselves to the light emission, i.e.,  $\epsilon < 0$ , we find

$$\Gamma_{\text{nG}}(\epsilon < 0) = \frac{\tilde{\alpha}^2}{8} g_c^2 \frac{|\tilde{z}_\epsilon|^2}{\epsilon^2} \left\{ \int_0^{eV} d\omega \frac{|\tilde{z}_\omega|^2}{\omega^2} (eV - \omega) [\xi(\omega + \epsilon) + \xi(\omega - \epsilon) + 2\epsilon - \xi(\epsilon)] + \frac{2Y(-eV - \epsilon)}{\epsilon} \right. \\ \times \int_0^\infty \frac{d\omega}{\omega} (\text{Re}\{\tilde{z}_\epsilon\} \text{Re}\{\tilde{z}_\omega\} [\xi(\omega + \epsilon) - \xi(\omega - \epsilon) - 2\epsilon - \xi(\epsilon)] \\ \left. + \text{Im}\{\tilde{z}_\epsilon\} \text{Im}\{\tilde{z}_\omega\} [4eV + \xi(\omega + \epsilon) + \xi(\omega - \epsilon) - 2\xi(\omega) - 2\xi(\epsilon)]) \right\} \quad (8)$$

with  $\xi(\omega) = |\omega + eV| + |\omega - eV|$ . This is the main result of our work and describes the influence of the non-Gaussian contribution to the light emission in the whole energy range. Note that it can be further simplified in the overbias regime for  $eV < -\epsilon < 2eV$  and takes the same form as in Ref. [20] to order  $\alpha^2$ . We also see that the overbias emission rate is  $\sim g_c^2$ , which signals the fact that a correlated two-electron tunneling process is responsible.

The non-Gaussian rate (8) explains the emission of photons with energies  $-\epsilon > eV$ . The detailed behavior of this rate as a function of energy is shown in Fig. 3 for different values of  $eV$ . We observe that the rate has a distinct kink for  $-\epsilon = eV$ , which can be seen as a signature of the sharp Fermi edge. This leads, for  $eV < \omega_0$ , to a two-peak structure with peaks of comparable heights above and below the threshold voltage. For higher voltages, only a single peak at the resonance frequency remains.

To compare our theoretical model with the experimental data [7], we have to take the Gaussian as well as the non-Gaussian rates into account. As mentioned above, the two rates differ parametrically by a factor of  $g_c z_0^2$ . We determine this parameter from the experimental results. In the inset of Fig. 4, we show the total rate  $\Gamma = \Gamma_G + \Gamma_{\text{nG}}$  for two different bias voltages. These rates have to be compared to the results presented in Fig. 2(a) of Ref. [7]. From the relative scaling of the two curves by a factor of 300 and the width of the resonance, we determine the parameters  $g_c z_0^2 \approx 0.1$  and  $\eta \approx 0.3\omega_0$ , respectively. Note that the experimental

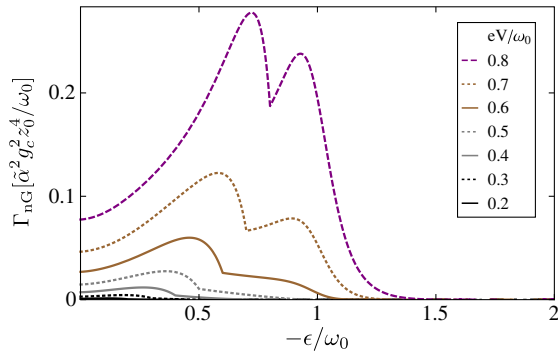


FIG. 3 (color online). The non-Gaussian emission spectrum for different bias voltages. The spectrum is clearly induced by the SPP resonance and shows a kink at the bias voltage. The overbias emission rate is distinctly visible, and the scaling with  $g_c^2$  shows that this effect is due to two-electron tunneling processes. The broadening of the resonance is  $\eta = 0.3\omega_0$ .

results depend on the detailed surroundings of the STM tip's position. We show the voltage- and energy-dependent emission rate in the main panel of Fig. 4. The comparison to Fig. 1(b) of Ref. [7] is striking, although the resonance parameters in the experiment are different. We clearly observe the threshold behaviors at  $-\epsilon = eV$  and  $-\epsilon = 2eV$ . Recently, the light in the  $2eV$  energy range has been investigated in more detail experimentally [12], but a confirmation of a well-developed threshold behavior still needs more evidence. We should add that experimentally the data are cut for  $-\epsilon < 1.2eV$ , which is attributed to the detector sensitivity. Finally, we should emphasize that the experimental finding that the one- (two-)electron rate scales approximately with  $g_c (g_c^2)$  is correctly reproduced by our theoretical model.

In conclusion, motivated by the experimental observation of photons with overbias energies emitted by tunnel junctions, we have developed a model of electron-SPP interaction based on dynamical Coulomb blockade. The interplay between the non-Gaussian statistics of the

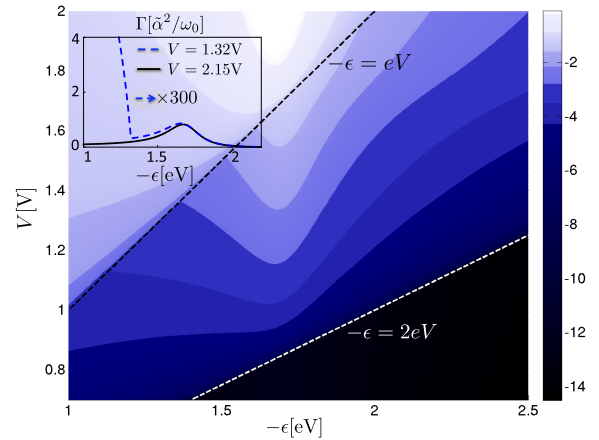


FIG. 4 (color online). Main: Light emission spectrum on a logarithmic scale as a function of bias voltage. The SPP resonance energy is taken to be on the order of the experimental value  $\omega_0 = 1.7$  eV,  $g_c z_0^2 = 0.1$ , and the broadening is taken to be  $\eta = 0.2\omega_0$ . The one- and two-electron thresholds at  $-\epsilon = eV$  and  $-\epsilon = 2eV$  are indicated by dashed lines. Inset: To extract the coupling parameter  $g_c z_0^2 = 0.1$ , we compare the peak values at  $-\epsilon = \omega_0$  for two different bias voltages:  $V = 2.15$  V (solid line);  $V = 1.32$  V (dashed line). By scaling the low-bias curve by a factor of 300, we find curves similar to those of Fig. 2(a) in Ref. [7]. Note that we have taken  $\eta = 0.3\omega_0$  to achieve a better agreement of the resonance shape with the experiment.

tunneling process and the resonant excitations of the SPP leads to a pronounced emission spectrum in which the SPP spectrum is overlaid with the sharp quantum threshold behavior determined by the bias voltage  $eV$ . Furthermore, the theory reproduces the experimentally observed emission with energies larger than the single-particle energy  $eV$ . A comparison of our model calculation to the experimental spectrum reveals a quantitative agreement of both the spectrum of the SPP resonance and the quantum thresholds. Our work enables a new level of modeling electron-SPP interaction in nanosize contacts. Furthermore our calculation shows that the overbias emission can be used to experimentally probe higher-order quantum fluctuations from a tunnel junction. Open questions concern going beyond the tunneling approximation and the weak coupling regime [30] or considering the effect of molecules in the junction [15,31,32]. Finally, more experimental and theoretical effort is needed to consolidate our model for describing overbias light emission in a STM junction. This is an important step since having a correct description of the comparably simple case of a STM junction is crucial for studying more complicated junctions incorporating, e.g., molecular films.

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