Spectral analysis of adsorbate induced field-emission flicker noise

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Spectral analysis of field-emission flicker noise is developed for a probe current originating from a portion of a single (*hkl*) plane of the emitter. The current noise is related to equilibrium absorbate density fluctuations in the probed region with the total number of net plane adatoms constant. The noise spectrum factors as $S(\omega)=S_\infty(\omega)+S_B(\omega)$ when the finite size of the net plane is accounted for. The spectrum due to unbounded diffusion $S_\infty(\omega)$ is proportional to one derived in Burgess's model of semiconductor contact noise. The boundary-effect contribution $S_B(\omega)$ increases $dS(\omega)/d\omega$ at low frequency, whereas anisotropic diffusion decreases it.

I. INTRODUCTION

Numerous mechanisms have been proposed to explain flicker ($1/f$) noise. Theories have developed spectral-density functions from nonstationary processes,[1] motion in a random potential,[2] and diffusion-mediated two-site switching.[3,4] Investigations of contact noise in semiconductors by Richardson,[5] MacFarlane,[6] and van der Ziel[7] led to the development of flicker spectra from diffusion processes. These analyses were, in turn, based on Smoluchowski's theory of density fluctuations.[8,9] Since then, many studies have developed autocorrelation functions that are proportional to Smoluchowski's probability after-effect factor

$$P_s=1-A_p^{-1}\int_{A_p}d\mathbf{r}_1\int_{A_p}d\mathbf{r}_2 G(\mathbf{r}_1,t;\mathbf{r}_2)\ ,$$

where the density or temperature is taken as the random variable, $G(\mathbf{r}_1,t;\mathbf{r}_2)$ is the Green function for the diffusion equation, and A_p is the probed region.[10–20] These theories implicitly assume fluctuations occur at equilibrium and are linear and time reversible on a microscopic scale.

The present study develops the spectral analysis of field-emission flicker noise associated with adsorbate diffusion over a single (*hkl*) plane of the emitter, including the effect of net plane boundaries and anisotropic diffusion. Because equilibrium density fluctuations induce the observed current noise, this analysis has close connections with the above-mentioned studies based on a diffusion mechanism. These systems belong to a canonical ensemble, here meaning they are closed with respect to the number of net plane adatoms. Only totally reflecting boundaries are consistent with the assumption of equilibrium density fluctuations occurring on a finite-size net (*hkl*) plane. To maintain equilibrium with absorbing or partially reflecting boundaries requires additional sources in the diffusion equation. Introducing these fundamentally changes the processes assumed to cause the noise, e.g., an additional mechanism such as evaporation-condensation would then have to be included. Such modifications are not considered here.

Factors related to the Fowler-Nordheim (FN) equation, and thus specific to the field-emission process, are considered in Sec. II, where an integral equation for $R(t)$ is developed similar to ones derived by Lax[15] and by van Vliet and Fassett.[16] The system is assumed not to be dominated by critical fluctuations, and thus the noise spectrum $S(\omega)$ can be evaluated using $G(\mathbf{r}_1,t;\mathbf{r}_2)$ and a simplified version of the pair covariance $C(\mathbf{r}_1,\mathbf{r}_2)$. In this regime interactions affect diffusivity but do not alter the time dependence of $G(\mathbf{r}_1,t;\mathbf{r}_2)$. Therefore, a dimensionless expression for $S(\omega)$ can be written that is not explicitly dependent on the interactions.

The influence of probe—net plane geometry on the structure of $R(t)$ and $S(\omega)$ is investigated in Sec. III. Certain geometries simplify the expressions for $R(t)$ and $S(\omega)$ and also determine the form the total noise power takes. In general, finding the explicit representation is difficult. A closed solution is derived for $S(\omega)$ including the effect of the finite size of the net (*hkl*) plane when the probed and net plane regions are concentric circles. When a square net plane and probe geometry is assumed, $R(t)$ reduces to an expression derived by Gomer and co-workers.[13,17] It is shown in Sec. IV that the total noise power must vanish when the probe and net (*hkl*) areas are equal independent of their particular geometries. This condition is characteristic of the canonical ensemble.

In the absence of net plane boundaries the autocorrelation functions for both circular and square probe geometries are reduced to one proportional to the after-effect factor P_s. The solution for P_s was originally stated by Smoluchowski for the case of a circular probed region.[8] An explicit derivation of P_s is provided in Sec. V to eliminate confusion resulting from a misstated formula given by Chandrasekhar[9] that was later adopted by MacFarlane in his analysis of the corresponding $S_\infty(\omega)$.[6] MacFarlane's $S_\infty(\omega)$ has also been used by Saitou *et al.* in their study of field-emission noise.[21] Excluding the prefactor specific to the field-emission process, the correct spectrum for unbounded diffusion, $S_\infty(\omega)$, is similar to one stated by Burgess[10] and subsequently used in the study of Timm and van der Ziel.[11] The $S_\infty(\omega)$ derived in the present work corresponds precisely to the autocorrelation function employed by Gomer and co-workers in their

study of the diffusion coefficient.[12–14,22]

The expression for $S(\omega)$, including the boundary effect, is derived in Sec. VI. Its functional form eliminates criticisms that the diffusion process is inadequate for describing flicker noise of certain adsorbate systems.[4,23] It is noted, however, that early experiments have not provided a stringent test of the theory. More recent spectral-density measurements of K/W(111) using the probe-hole method corroborate the diffusion mechanism.[24]

Anisotropic diffusion is considered in Sec. VII. Here the limiting case of purely one-dimensional motion is solved analytically and found to increase the low-frequency portion of $S(\omega)$.

II. THE FIELD-EMISSION AUTOCORRELATION FUNCTION

Kleint's adsorption-desorption theory[25] attempted to explain the early experimental measurements of field-emission flicker noise by Kleint and Gasse.[26] However, Tim and van der Ziel provided the first spectral analysis of adsorbate induced noise that included the FN equation.[11] They considered diffusive adsorbate motion over an infinite planar surface with site-exclusion interactions only. The use of a probe technique to detect adsorbate density fluctuations on a single (hkl) plane along with a more precise analysis of the FN equation and field-emission noise in the time domain has since been carried out by Gomer and Chen.[12,13] Mazenko, Banavar, and Gomer extended the fluctuation theory to include fully interacting systems.[14] Their study demonstrated that a density-dependent surface-diffusion coefficient $D(n)$ is derivable from the field-emission–current autocorrelation-function, $R(t)$, measurement of equilibrium density fluctuations.

We begin by deriving a general integral equation for the field-emission–current autocorrelation. The field-emission–current density j (amps/cm^2) is described by the Fowler-Nordheim equation

$$j = BF^2 \exp\left[\frac{-6.8\times10^7 v}{F}\phi^{3/2}\right], \tag{1}$$

where F is the electric field in V/cm, v an image-correction term, ϕ the work function in eV,

$$B = 6.2\times10^6(\mu/\phi)^{1/2}/v^2(\phi+\mu),$$

and μ the Fermi level.[27] The factor B is insensitive to work-function fluctuations compared to the exponential term, and henceforth is considered constant.

From Eq. (1) the current fluctuation $\delta i(t)\equiv i(t)-\langle i\rangle$ is given by

$$\frac{\delta i(t)}{\langle i\rangle} = e^{-a[\phi^{3/2}(t)-\langle\phi\rangle^{3/2}]}-1, \tag{2}$$

where $\langle i\rangle=\langle j\rangle A_p$ is the equilibrium current and $a=-6.8\times10^7 v/F$. The work function $\phi=\phi(F,n)$ is dependent on the field F and adatom density $n=N/A_p$, N is the number of adatoms in probed area $A_p=\pi r_p^2$,

$$\phi=\phi_n+2\pi\alpha F n, \tag{3}$$

ϕ_n is the zero-field work function, and α is the polarizability per adatom.

With the work-function fluctuation $\delta\phi\equiv\phi-\langle\phi\rangle$, Eq. (2) is approximated to first order as

$$\frac{\delta i}{\langle i\rangle} = \tfrac{3}{2}a\langle\phi\rangle^{1/2}\delta\phi, \tag{4}$$

assuming $\delta\phi \ll \langle\phi\rangle$. Noting from Eq. (3) that

$$\delta\phi=\delta\phi_n+c_\alpha\,\delta n, \tag{5}$$

where $c_\alpha=2\pi\alpha F$, the relative current fluctuation

$$\frac{\delta i}{\langle i\rangle} = \left[\frac{c_1+c_2(\partial\phi/\partial n)}{A_p}\right]\delta N \tag{6}$$

follows from Eq. (2) with $c_1=c_\alpha c_2$, $c_2=\frac{3}{2}a\langle\phi\rangle^{1/2}$, and $\delta\phi_n=(\partial\phi/\partial n)\delta n$. By using Eq. (6) the relative field-emission–current autocorrelation

$$R(t)\equiv\frac{\langle\delta i(t)\delta i(0)\rangle}{\langle i\rangle^2}$$

becomes

$$R(t)=\left[\frac{c_1+c_2(\partial\phi/\partial n)}{A_p}\right]^2\langle\delta N(t)\delta N(0)\rangle, \tag{7}$$

where $\delta N(t)$ is the equilibrium fluctuation in particle number of the probed area A_p at time t.

Several comments are necessary about the term $\partial\phi/\partial n$. Gomer has noted that since field emission is governed by the potential to $\sim$5.0 Å from the surface, it is possible to have an adatom outside the probed region influence emission within it.[12] This effect can be approximated by

$$\delta\phi=[\bar{w}h(n)+c_\alpha]\delta n, \tag{8}$$

where the factor $\bar{w}$ is the average contribution per adparticle to the potential energy $\sim$5.0 Å above the probed region and $|h(n)|\leq 1$ is a dimensionless function describing dipole depolarization. In Gomer's analysis $\bar{w}$ appears inside the particle-number autocorrelation $\langle N(t)N(0)\rangle$ because it is really a function of position within the probed area A_p. If density fluctuations outside A_p do not affect the probed current, then $\bar{w}$ can be written in the simple form, $\bar{w}=2\pi\mu$, where μ is the permanent dipole moment of the adatom. The approximation becomes exact when the probed radius $r_p \gg 5.0$ Å (Ref. 12) or when the permanent dipole moment is small, in which case the polarizability term dominates. This happens for a physisorbed system such as Xe/W.[22]

In general, $\bar{w}$ influences the value of $D(n)$ by increasing the effective probed radius r_p, but does not change the time dependence of the autocorrelation.[12] Therefore, keeping in mind that a systematic error may exist in the estimation of $D(n)$, Eq. (7) can be written

$$R(t)=C_{\rm FN}\langle\delta N(t)\delta N(0)\rangle, \tag{9}$$

where

$$C_{\rm FN}=\left[\frac{c_1+2\pi\mu h(n)c_2}{A_p}\right]^2.$$

For a fixed value of adsorbate density, $h(n)$ is constant

and does not influence the spectral analysis. Several models relate mutual depolarization to work function, and so an expression for $h(n)$ could be included in Eq. (8).[28,29] Although the density dependence of the autocorrelation function as expressed through $h(n)$ is not of primary importance here, it does induce one notable noise effect. For a chemisorbed system, e.g., an alkali-metal adsorbate on a refractory-metal substrate, $\phi(n)$, exhibits a distinct minimum, i.e., $\partial\phi/\partial n \mid_{\phi_{\min}}=0$ and $h(n_{\min})=0$. Under this condition Eq. (7) implies

$$\frac{\langle(\delta i)^2\rangle}{\langle i\rangle^2}=c_1^2\langle(\delta n)^2\rangle \ .$$

The relative mean-square current fluctuation is often greatly reduced since the polarizability term c_1 is usually much smaller than the other term involving the dipole moment. An example of this reduction in noise power at the work-function minimum can be found in a study of K/W (Fig. 1 of Ref. 30).

The particle number in the probed area A_p, expressed in terms of the local density $n(\mathbf{r},t)$, is

$$N(t)=\int_{A_p} n(\mathbf{r},t)d\mathbf{r} \ . \tag{10}$$

Hence, Eq. (9) becomes

$$R(t)=C_{\rm FN}\int_{A_p}\int_{A_p}\langle\delta n(\mathbf{r}_1,t)\delta n(\mathbf{r}_2,0)\rangle d\mathbf{r}_1 d\mathbf{r}_2 \ , \tag{11}$$

where the integrand of Eq. (11) is the two-point (pair) autocovariance of the density fluctuation. The classical expression is

$$\langle\delta n(\mathbf{r}_1,t)\delta n(\mathbf{r}_2,0)\rangle=\int_{A_T} d\mathbf{r}_3 G(\mathbf{r}_1,t;\mathbf{r}_3)C(\mathbf{r}_2,\mathbf{r}_3) \ , \tag{12}$$

where $G(\mathbf{r}_1,t;\mathbf{r}_3)$ is the Green function for the diffusion equation, the integration is over the net plane area A_T, and

$$C(\mathbf{r}_2,\mathbf{r}_3)\equiv\langle\delta n(\mathbf{r}_2)\delta n(\mathbf{r}_3)\rangle \tag{13}$$

is the pair-covariance function.[15,16] Combining Eqs. (11) and (12) leads to the following general form of the field-emission autocorrelation function:

$$R(t)=C_{\rm FN}\int_{A_p} d\mathbf{r}_1\int_{A_p} d\mathbf{r}_2\int_{A_T} d\mathbf{r}_3 G(\mathbf{r}_1,t;\mathbf{r}_3)C(\mathbf{r}_2,\mathbf{r}_3) \ . \tag{14}$$

III. KARHUNEN-LOÈVE EXPANSION

In this section we evaluate $R(t)$, as expressed by Eq. (14), in terms of its properties as an integral equation. The analysis will show that, excluding critical fluctuations, a correlationless pair covariance always exists. This simplifies the form of $R(t)$, although in practice it is often difficult to find the proper linear transformation. However, a specific example is given in Sec. VI which yields a closed solution for the spectrum $S(\omega)$. The expressions for $R(t)$ and the total noise power are dependent on system geometry.

Consider the following homogeneous Fredholm equation of the second kind,

$$\phi_n(\mathbf{r}_1)=\int_{A_T} d\mathbf{r}_2 C(\mathbf{r}_1,\mathbf{r}_2)\phi_n(\mathbf{r}_2) \ , \tag{15}$$

and note that the kernel $C(\mathbf{r}_1,\mathbf{r}_2)$ as defined by Eq. (13) is real-valued and symmetric, i.e.,

$$C(\mathbf{r}_1,\mathbf{r}_2)=C(\mathbf{r}_2,\mathbf{r}_1) \ . \tag{16}$$

Given that the system is not at the critical point, $C(\mathbf{r}_1,\mathbf{r}_2)$ is continuous. To prove continuity the inequality

$$\langle(\delta n)^2\rangle\geq C(\mathbf{r}_1,\mathbf{r}_2)$$

is used, which is a general property of stationary or homogeneous random functions.[31] By considering $C(\mathbf{r}_1,\mathbf{r}_2)$ to be a linear functional $T[\delta n(\mathbf{r}_1)]$, with parameter $\mathbf{r}_1$, on $\delta n(\mathbf{r}_2)$,

$$T[\delta n(\mathbf{r}_1)]=\langle\delta n(\mathbf{r}_1)\delta n(\mathbf{r}_2)\rangle \ ,$$

and noting the thermodynamic-fluctuation-theory result

$$\langle(\delta n)^2\rangle=\langle n\rangle^2 kT\kappa_T/A_p \ , \tag{17}$$

one can find a constant

$$c=(\langle n\rangle^2 kT\kappa_T/A_T A_p)^{1/2} \ ,$$

such that for all $\delta n(\mathbf{r}_1)$,

$$|T[\delta n(\mathbf{r}_1)]|\leq c||\delta n(\mathbf{r}_1)|| \ , \tag{18}$$

where the norm is defined

$$||\delta n(\mathbf{r}_1)||\equiv\left[\int_{A_T} d\mathbf{r}_2\langle(\delta n)^2\rangle\right]^{1/2}$$
$$=[A_T\langle(\delta n)^2\rangle]^{1/2}$$

and κ_T is the isothermal compressibility. Equation (18) demonstrates that T is bounded as long as κ_T is finite. Using the theorem that boundedness and continuity are equivalent for linear functionals[32] ensures that $C(\mathbf{r}_1,\mathbf{r}_2)$ is continuous.

The kernel is nondegenerate as it is not expressible as a finite sum of separable functions

$$C(\mathbf{r}_1,\mathbf{r}_2)\neq\sum_{i=1}^{m}\alpha_i(\mathbf{r}_1)\beta_i(\mathbf{r}_2) \ .$$

The multivariable density function implicit in $\langle\delta n(\mathbf{r}_1)\delta n(\mathbf{r}_2)\rangle$ is not of this form because for bounded regions $\mathbf{r}_1$ is statistically dependent on $\mathbf{r}_2$ even in the absence of critical fluctuations.

The following theorem can then be applied to the kernel $C(\mathbf{r}_1,\mathbf{r}_2)$:[33] "Every continuous, symmetric kernel that does not vanish identically possesses eigenvalues and eigenfunctions; their number is denumerably infinite if and only if the kernel is nondegenerate. All eigenvalues of a real symmetric kernel are real." Therefore, the set $\{\phi_n\}$ exists, is denumerably infinite, and can serve as a basis for functions defined over the area A_T. Given the existence of a basis set $\{\phi_n\}$ over A_T the Green function in Eq. (14) can be expanded as

$$G(\mathbf{r}_1,t;\mathbf{r}_3)=\sum_{n=1}^{\infty} a_n\phi_n(\mathbf{r}_3) \ . \tag{19}$$

Substituting Eq. (19) into (14) leads to

$$R(t)=C_{\rm FN}\langle(\delta N)^2\rangle A_p^{-1}\int_{A_p} d\mathbf{r}_1\int_{A_p} d\mathbf{r}_2 G(\mathbf{r}_1,t;\mathbf{r}_2) \ . \tag{20}$$

This decomposition is equivalent to a Karhunen-Loève (KL) expansion[31,34,35] of $\delta n(\mathbf{r}_2)$ as shown by writing

$$\delta n(\mathbf{r}_2)=\sum_{n=1}^{\infty}\tilde{b}_n\phi_n(\mathbf{r}_2)\ . \tag{21}$$

The random coefficients $\tilde{b}_n$ have the properties $\langle\tilde{b}_n\rangle=0$ and $\langle|\tilde{b}_n|^2\rangle=1$. They are orthogonal since solutions $\{\phi_n\}$ exist which satisfy Eq. (15).[31] The right-hand side of Eq. (21) converges in the mean-square sense to $\delta n(\mathbf{r}_2)$ if the kernel is of the form[31]

$$C(\mathbf{r},\mathbf{r})=\sum_{n=1}^{\infty}|\phi_n(t)|^2\ . \tag{22}$$

The kernel $C(\mathbf{r}_1,\mathbf{r}_2)$ satisfies Mercer's theorem, which states[33] "If $C(\mathbf{r}_1,\mathbf{r}_2)$ is a definite continuous kernel then the expansion given by Eq. (22) is valid and converges absolutely and uniformly." This theorem ensures the convergence of Eq. (21).

This proves that a reduction of the integral equation for $R(t)$ occurs with a special representation of $G(\mathbf{r}_1,t;\mathbf{r}_2)$. The existence and completeness of the basis set $\{\phi_n\}$ corresponding to this representation is demonstrated by recognizing $C(\mathbf{r}_1,\mathbf{r}_2)$ to be a nondegenerate symmetric kernel of a homogeneous Fredholm integral equation. Representing $G(\mathbf{r}_1,t;\mathbf{r}_2)$ by this particular basis is equivalent to a KL expansion of the density fluctuation $\delta n(\mathbf{r})$. This induces a linear transformation that yields a correlationless pair covariance.

Excluding the constant C_{FN}, Eq. (20) is analogous to Eq. (4) of van Vliet and Chenette,[20] although they assumed that a sufficient condition for its derivation occurs when there is zero correlation for $|\mathbf{r}_1-\mathbf{r}_2|\gg\rho$, where ρ is the correlation length.[16] The above development shows that an additional requirement is that the eigenfunctions used in Eq. (19) correspond to a basis set of a KL expansion of the density, Eq. (21). However, their original analysis remains valid because a circular probe was assumed, and Sec. VI shows that the conditions leading to Eq. (20) are satisfied for this geometry. Note that for square geometries a natural decomposition of $G(\mathbf{r}_1,t;\mathbf{r}_2)$ as a KL series does not occur. This modifies the integral equation involving $R(t)$. These geometry-dependent modifications in $R(t)$ are required to account for the probe-area dependence of the noise power as clarified in the following section. Specifically, the noise power must be zero when the probed and net plane areas are equal. This condition is independent of probe—net plane geometry and characteristic of a closed system.

IV. CORRELATION EFFECTS

The influence of net plane boundaries and critical fluctuations on the pair covariance $C(\mathbf{r}_1,\mathbf{r}_2)$ is now considered, assuming reflecting boundaries exist on the perimeter of the net plane and that they exert a negligible affect on the critical fluctuations. The pair covariance is then written

$$C(\mathbf{r}_1,\mathbf{r}_2)=C_\beta+C_\rho\ , \tag{23}$$

where

$$C_\beta=A_p^{-1}\langle(\delta N)^2\rangle[\delta(\mathbf{r}_1-\mathbf{r}_2)-A_T^{-1}] \tag{24}$$

is the correlation boundary effect.[16] Critical fluctuations are excluded from the present study, which implies $C_\rho=0$. To find the condition under which this assumption is valid, the Fourier transform of the two-dimensional density fluctuation

$$\delta n(\mathbf{r})=\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}\delta n(\mathbf{k}) \tag{25}$$

and the probability density function for the Fourier component $\delta n(\mathbf{k})$ in mean-field approximation[36]

$$f[\delta n(\mathbf{k})]=d_0e^{-(d_1+d_2k^2)|\delta n(\mathbf{k})|^2/2} \tag{26}$$

are used, where d_0 is a normalization constant and d_1,d_2 are coefficients resulting from the density-fluctuation expansion of the thermodynamic potential. Use of the defining relation for the k-space covariance of the density fluctuation and Eq. (26) leads to

$$\langle|\delta n(\mathbf{k})|^2\rangle=(d_1+d_2k^2)^{-1}\ . \tag{27}$$

Equations (17) and (27) yield

$$d_1^{-1}=\langle n\rangle^2kT\kappa_T/A_p\ . \tag{28}$$

The Fourier transform of Eq. (27) is

$$C_\rho(R)=(2\pi d_2)^{-1}K_0(R/\rho)\ , \tag{29}$$

where the area is assumed large enough to replace summation by integration, $R=|\mathbf{r}_1-\mathbf{r}_2|$, K_0 is the zero-order modified Bessel function, and $\rho=(d_2/d_1)^{1/2}$. The asymptotic expansion of Eq. (29) is

$$\lim_{R/\rho\to\infty}C_\rho\sim(\rho/R)^{1/2}e^{-R/\rho}\ .$$

Thus, critical fluctuations are negligible when $A_p\gg\rho^2$. Assuming this holds, then $C_\rho=0$ and substitution of Eq. (24) into (14) yields

$$R(t)=C_{\mathrm{FN}}A_p^{-1}\langle(\delta N)^2\rangle\left[\int_{A_p}d\mathbf{r}_1\int_{A_p}d\mathbf{r}_2\left[G(\mathbf{r}_1,t;\mathbf{r}_2)-A_T^{-1}\int_{A_T}d\mathbf{r}_3G(\mathbf{r}_1,t;\mathbf{r}_3)\right]\right]\ . \tag{30}$$

If the net plane, with reflecting boundary, and probed regions are squares of area $4b^2$ and $4a^2$, respectively, and centered at the origin, then the Green function is

$$G(\mathbf{r}_1,t;\mathbf{r}_2)=\prod_{i=1}^{2}\left[\sum_{n=0}^{\infty}e^{-\lambda^2Dt}\gamma_n(x_{i1})\gamma_n(x_{i2})\right]\ , \tag{31}$$

where $\lambda^2=(\pi n/b)^2$, $\gamma_n(x)=(\epsilon_n/2b)^{1/2}\cos(\pi nx/b)$, and $\epsilon_n\equiv1(2)$ if $n=0$ ($n\neq0$) is the Neumann factor. Combining Eqs. (31) and (30) results in

$$R(t)=C_{\mathrm{FN}}\langle(\delta N)^2\rangle\left[A_p^{-1}\int_{A_p}\int_{A_p}G(\mathbf{r}_1,t;\mathbf{r}_2)d\mathbf{r}_1d\mathbf{r}_2-A_p/A_T\right]\ . \tag{32}$$

Equation (32) is equal to the autocorrelation function Gomer and co-workers obtained by other means.[13,14,17]

The factor A_p/A_T is a result of the boundary effect. Its physical significance is clarified by considering the relative noise power $P \equiv \int_0^\infty S(f)df$, which is also given by

$$P = 2R(0) . \tag{33}$$

Using Eqs. (32) and (33) the relative field-emission flicker-noise power becomes

$$P = 2C_{\mathrm{FN}}\langle(\delta N)^2\rangle(1 - A_p/A_T) . \tag{34}$$

Thus, in the limit $A_p \to A_T$, the noise power approaches zero. The reason is that fluctuations occur in this system only when particles diffuse in or out of the probed region A_p, which is impossible when $A_p = A_T$. This condition holds whenever adsorbate induced flicker noise occurs in a closed system and will be used in Sec. VI to check the validity of the spectral density derived for the case of a circular net plane. The autocorrelation function derived by Reed and Ehrlich does not have this property.[37] That the autocorrelation functions given by Eqs. (20) and (32) are not equivalent is due to the system geometry. The $\gamma_n(x)$ in Eq. (31) constitute a Fourier series, which do not form a basis in a Karhunen-Loève expansion.[31] Thus, one cannot substitute Eq. (31) into (20) and obtain the proper expression for the noise power, Eq. (34), since the factor A_p/A_T would be missing.

It is also noted in passing that the total field-emission noise power is really a sum of Eq. (34) and the shot-noise term $2eI\Delta f_{\mathrm{bw}}$, where Δf_{bw} is the bandwidth of the spectrum analyzer. However, this last term will be dropped from further discussion.

V. UNBOUNDED DIFFUSION SPECTRUM

The noise spectrum $S(\omega)$ is now developed in the absence of net plane boundaries. First, an explicit derivation of the after-effect factor P_s is provided to eliminate confusion over several erroneous formulations.[6,9,21] Then, the correct form of $S(\omega)$ is given when site exclusion occurs for an otherwise noninteracting adsorbate lattice gas. Also, another method is suggested for deriving $D(n)$ from $S(\omega)$ for a fully interacting adsorbate system.

In the limit $A_T \to \infty$, Eqs. (20) and (32) become identical and are related to Smoluchowski's probability after-effect factor since

$$R(t) = C_{\mathrm{FN}}\langle(\delta N)^2\rangle[1 - P_s(t)] .$$

Specifically,

$$R(t) = \frac{C_{\mathrm{FN}}\langle(\delta N)^2\rangle}{A_p 4\pi Dt}\int_{A_p} d\mathbf{r}_1 \int_{A_p} d\mathbf{r}_2 e^{-|\mathbf{r}_1 - \mathbf{r}_2|^2/4Dt} . \tag{35}$$

Equation (35) can be evaluated in closed form by transforming to polar coordinates. The factor P_s becomes

$$P_s(t) = \frac{1}{\alpha^2}\int_{\alpha^2}^\infty dx\, e^{-x}\int_0^{\alpha^2} dw\, e^{-w} I_0(2\sqrt{xw}) , \tag{36}$$

where $\alpha^2 = r_p^2/4Dt$ and I_0 is the zeroth-order modified Bessel function. The integrand of Eq. (36) can be expressed as the Bessel function J_0; hence,

$$P_s = \frac{1}{\alpha^2}\int_{\alpha^2}^\infty dx \int_0^{\alpha^2} dw\, e^{-(x+w)} J_0(2\sqrt{x(-w)}) . \tag{37}$$

Expanding the αth-order Bessel function as a sum of Laguerre polynomials,[38]

$$e^z J_\alpha(2\sqrt{xz}) = (xz)^{\alpha/2}\sum_{n=0}^\infty [\Gamma(n+\alpha+1)]^{-1} L_n^\alpha(x) z^n , \tag{38}$$

casts Eq. (37) in the form

$$P_s = \sum_{n=0}^\infty \frac{(-\alpha^2)^n}{(n+1)!}\int_{\alpha^2}^\infty dx\, e^{-x} L_n(x) . \tag{39}$$

The integral in Eq. (39) is given by

$$\int_x^\infty e^{-y} L_n^\alpha(y)dy = e^{-x}[L_n^\alpha(x) - L_{n-1}^\alpha(x)] . \tag{40}$$

Combining Eqs. (40) and (39) leads to

$$P_s = e^{-\alpha^2}\left[1 + x + \sum_{n=1}^\infty \frac{(-\alpha^2)^n}{(n+1)!}[(n+1)L_n(x) + nL_{n-1}(x) + (x-n)L_n(x)]\right] . \tag{41}$$

With the aid of the recursion relation

$$xL_n^1(x) = nL_{n-1}(x) + (x-n)L_n(x) , \tag{42}$$

Eq. (41) becomes

$$P_s = e^{-\alpha^2}\left[\sum_{n=0}^\infty \frac{(-\alpha^2)^n}{n!} L_n(\alpha^2) + \alpha^2 \sum_{n=0}^\infty \frac{(-\alpha^2)^n}{(n+1)!} L_n^1(\alpha^2)\right] . \tag{43}$$

Substituting Eq. (38) into (43) yields

$$P_s = e^{-2\alpha^2}[I_0(2\alpha^2) + I_1(2\alpha^2)] . \tag{44}$$

As noted in the Introduction, the value of the argument α has been the source of confusion, although Eq. (44) was first stated long ago by Smoluchowski.[8]

Using Eq. (44) the field-emission—current autocorrelation, Eq. (35), can be written

$$R(t) = C_{\mathrm{FN}}\langle(\delta N)^2\rangle\{1 - e^{-2\alpha^2}[I_0(2\alpha^2) + I_1(2\alpha^2)]\} . \tag{45}$$

The asymptotic expansion of Eq. (45) is

$$\lim_{t\to\infty} R(t)\sim C_{\mathrm{FN}}\langle(\delta N)^2\rangle(r_p^2/4Dt) .$$

This expansion, also derivable directly from Eq. (35), determines $D(n)$ when noise measurements are made in the time domain.[12,13,22] Using the Wiener-Khinchin (WK) theorem, the spectral density corresponding to Eq. (45) is

$$S_\infty(\omega)=C_{\mathrm{FN}}\langle(\delta N)^2\rangle\frac{8r_p^2}{D}\left[\frac{\mathrm{ber}_1(u_p)^{1/2}\mathrm{kei}_1(u_p)^{1/2}+\mathrm{bei}_1(u_p)^{1/2}\mathrm{ker}_1(u_p)^{1/2}}{-u_p}\right] . \tag{46}$$

The subscript ∞ is a reminder that the diffusive motion is unbounded, $u_p\equiv\omega r_p^2/D$, and $\mathrm{ber}_1,\mathrm{bei}_1,\mathrm{kei}_1,\mathrm{ker}_1$ are Kelvin functions. The term in curly braces was first given by Burgess[10] and later by van Vliet and Chenette.[20] The Eq. (46) spectrum also corresponds to the field-emission—current autocorrelation function developed by Mazenko, Banavar, and Gomer.[14]

If the adsorbate is represented as a noninteracting lattice gas with site exclusion, then

$$\langle(\delta N)^2\rangle=\langle n\rangle A_p(1-\lambda) , \tag{47}$$

where $\lambda\equiv na_0^2$ and a_0 is the lattice constant. Note that $\lim_{\lambda\to 0}\langle(\delta N)^2\rangle=\langle N\rangle$, which is characteristic of ideal-gas behavior. In this case Eq. (46) yields a spectrum similar to that of Timm and van der Ziel,[11] except they mistakenly inserted an additional factor of λ into Eq. (47).

The low- and high-frequency limits of Eq. (46) are, respectively,

$$\lim_{\omega\to 0} S_\infty(\omega)=C_{\mathrm{FN}}\langle(\delta N)^2\rangle r_p^2 D^{-1}\ln(D/r_p^2\omega) \tag{48}$$

and

$$\lim_{\omega\to\infty} S_\infty(\omega)=2^{3/2}C_{\mathrm{FN}}\langle(\delta N)^2\rangle D^{1/2}r_p^{-1}\omega^{-3/2} . \tag{49}$$

Recent measurements of K/W(111) show a good fit to Eq. (46).[24] Comparison of the theoretical and experimental curves allows a value of $D(n)$ to be obtained in two ways. The most direct method is to compare abscissas, which, assuming r_p is known, leads directly to $D(n)$. The second method is to compare ordinates and introduce a value for $C_{\mathrm{FN}}\langle(\delta N)^2\rangle$, which is obtainable by a measurement of the total noise power P. This determines $D(n)$ as it is the only remaining variable.

VI. BOUNDED DIFFUSION SPECTRUM

In this section a closed-form solution for the spectral-density function including the boundary effect is derived. The net plane and probed region are taken to be concentric circles with radii r_t and r_p, respectively. Physically, the bounded area plane corresponds to the chosen (hkl) plane of the emitter. For this geometry, the form of the Green function is shown to be equivalent to a KL expansion of δn. The spectral density will then be found by applying the WK theorem to Eq. (20):

$$S(\omega)=4C_{\mathrm{FN}}\langle(\delta N)^2\rangle A_p^{-1}\mathrm{Re}\int_{A_p}\int_{A_p} G(\mathbf{r}_1,\omega;\mathbf{r}_2)d\mathbf{r}_1 d\mathbf{r}_2 , \tag{50}$$

where

$$G(\mathbf{r}_1,\omega;\mathbf{r}_2)=\int_0^\infty G(\mathbf{r}_1,t;\mathbf{r}_2)e^{i\omega t}dt .$$

To prove that the representation of $G(\mathbf{r}_1,\omega;\mathbf{r}_2)$ yields a correlationless $C(\mathbf{r}_1,\mathbf{r}_2)$, it is sufficient to show

$$\mathrm{Re}\left[\int_{A_T} d\mathbf{r}_3 G(\mathbf{r}_1,\omega;\mathbf{r}_3)\right]=0 . \tag{51}$$

Then the spectrum derived from Eq. (30) will be equal to that obtained from Eq. (20). The function $G(\mathbf{r}_1,t;\mathbf{r}_2)$ satisfies

$$\left[\frac{\partial}{\partial t}-D\nabla^2\right]G(\mathbf{r}_1,t;\mathbf{r}_2)=\delta(\mathbf{r}_1-\mathbf{r}_2)\delta(t) , \tag{52}$$

and $G(\mathbf{r}_1,\omega;\mathbf{r}_2)$ solves

$$(\nabla^2+k^2)G(\mathbf{r}_1,\omega;\mathbf{r}_2)=-\frac{1}{D}\delta(\mathbf{r}_1-\mathbf{r}_2) , \tag{53}$$

with the Neumann boundary condition

$$\frac{\partial}{\partial n}G(\mathbf{r}_1,\omega;\mathbf{r}_2)=0 , \tag{54}$$

where $k^2=-i\omega/D$ and $\partial/\partial n$ is the normal derivative evaluated at the boundary. Expanding $G(\mathbf{r}_1,\omega;\mathbf{r}_2)$ as

$$G(\mathbf{r}_1,\omega;\mathbf{r}_2)=\frac{1}{2\pi}\sum_{m=-\infty}^{\infty} p_k^{(m)}(r_1;r_2)e^{im(\phi_1-\phi_2)} \tag{55}$$

and then substituting Eq. (55) into (53) leads to

$$\frac{1}{2\pi}\sum_{m=-\infty}^{\infty} e^{im(\phi_1-\phi_2)}\left[\frac{\partial^2 p_k^{(m)}}{\partial r^2}+\frac{1}{r}\frac{\partial p_k^{(m)}}{\partial r}-\left[\frac{m^2}{r^2}-k^2\right]p_k^{(m)}\right]=\frac{-\delta(r_1-r_2)\delta(\phi_1-\phi_2)}{Dr_1} . \tag{56}$$

Operating on Eq. (56) with $\int_0^{2\pi} d\phi\, e^{-in(\phi_1-\phi_2)}$ and defining

$$p_m(z_1,z_2)=p_k^{(m)}(r_1,r_2) ,$$

where $z_1=kr_1$ and $z_2=kr_2$, results in

$$p_m''+\frac{1}{z_1}p_m'+\left[1-\frac{m^2}{z_1^2}\right]p_m=\frac{-1}{Dkz_1}\delta(r_1-r_2) . \tag{57}$$

Here the single and double primes represent $\partial/\partial z$ and $\partial^2/\partial z^2$, respectively, and p_m satisfies Bessel's equation. The Eq. (55) expansion must also be substituted into the

Eq. (54) boundary condition, which yields

$$\frac{\partial}{\partial n}p_m(z_1,z_2)\bigg|_{r=r_t}=0\ . \tag{58}$$

The reciprocity relation for the Green function requires that the solution to Eq. (57) be written

$$p_m(z_1,z_2)=\frac{-1}{Dr_2W(y_1,y_2)}y_1(kr_<)y_2(kr_>)\ , \tag{59}$$

where $r_<$ ($r_>$) is the lesser (greater) of r_1 and r_2. The Wronskian $W(y_1,y_2)$ is evaluated at $r_1=r_2$, and y_1 and y_2 must satisfy the boundary conditions at $r_<=0$ and $r_>=r_t$. The finiteness of G at $r_<=0$ implies

$$y_1=J_m(kr_<)\ . \tag{60}$$

The function y_2 must be bounded as $r_>\to\infty$, which requires r_2 to have the form

$$y_2(kr_>)=AJ_m(kr_>)+BH_m^{(1)}(kr_>)\ , \tag{61}$$

where $H_m^{(1)}$ is the mth-order Hankel function of the first kind. A and B are determined by the boundary condition $(\partial/\partial r)y_2(r)\,|_{r=r_t}=0$, and so

$$A=-B\left[\frac{kH_{m-1}^{(1)}(kr_t)-(m/r_t)H_m^{(1)}(kr_t)}{kJ_{m-1}(kr_t)-(m/r_t)J_m(kr_t)}\right]\ . \tag{62}$$

The Wronskian is then equal to

$$W(y_1,y_2)=2iB/\pi r\ . \tag{63}$$

Equation (59) becomes

$$p_m(kr_1,kr_2)=\frac{\pi i}{2D}\left[J_m(kr_<)H_m^{(1)}(kr_>)-J_m(kr_<)J_m(kr_>)\left[\frac{kH_{m-1}^{(1)}(kr_t)-(m/r_t)H_m^{(1)}(kr_t)}{kJ_{m-1}(kr_t)-(m/r_t)J_m(kr_t)}\right]\right]\ . \tag{64}$$

Combining Eqs. (55) and (64) leads to

$$\int_{A_T}d\mathbf{r}_3G(\mathbf{r}_1,\omega;\mathbf{r}_3)=\pi/i\omega\ . \tag{65}$$

Thus, Eq. (51) holds, which proves that the expression for $G(\mathbf{r}_1,\omega;\mathbf{r}_2)$, as given by Eqs. (55) and (64), does correspond to a Karhunen-Loève expansion of $\delta n(\mathbf{r})$. Substituting Eqs. (55) and (64) into (50) leads to the following expression for the spectral density,

$$S(\omega)=S_\infty(\omega)+S_B(\omega)\ , \tag{66}$$

where

$$S_\infty(\omega)=\frac{8\pi^2C_{\rm FN}\langle(\delta N)^2\rangle}{A_TD}\mathrm{Re}\left[i\int_0^{r_p}dr_>r_>H_0^{(1)}(kr_>)\int_0^{r_>}dr_<r_<J_0(kr_<)\right] \tag{67}$$

and

$$S_B(\omega)=\frac{8\pi^2C_{\rm FN}\langle(\delta N)^2\rangle}{A_TD}\mathrm{Re}\left[\frac{iH_1^{(1)}(kr_t)}{J_1(kr_t)}\right]\int_0^{r_p}dr_>r_>J_0(kr_>)\int_0^{r_>}dr_<r_<J_0(kr_<)\ . \tag{68}$$

Evaluation of Eq. (67) leads to Eq. (46). Equation (68) describes the $S(\omega)$ boundary effect. The Eq. (66) factorization is a general result independent of the net plane geometry, which arises from the possibility of separating the Green function into a source and a boundary term.[39] Performing the integrations in Eq. (68) yields

$$S_B(\omega)=\left[\frac{4\pi r_p^2C_{\rm FN}\langle(\delta N)^2\rangle}{D}\right]\mathrm{Re}\left[\left[\frac{iH_1^{(1)}(kr_t)}{J_1(kr_t)}\right]\left[\frac{J_1(kr_p)}{kr_p}\right]^2\right]\ . \tag{69}$$

A check on $S_B(\omega)$ is obtained by noting

$$\lim_{r_t\to\infty}S_B(\omega)=0\ . \tag{70}$$

Equation (69) reduces to

$$\begin{aligned}S_B(\omega)=&[8C_{\rm FN}\langle(\delta N)^2\rangle r_p^2u_p^{-1}D^{-1}][\mathrm{ber}_1^2(u_t)^{1/2}+\mathrm{bei}_1^2(u_t)^{1/2}]^{-1}\\&\times\{[\mathrm{ber}_1^2(u_p)^{1/2}-\mathrm{bei}_1^2(u_p)^{1/2}][\mathrm{kei}_1(u_t)^{1/2}\mathrm{ber}_1(u_t)^{1/2}-\mathrm{ker}_1(u_t)^{1/2}\mathrm{bei}_1(u_t)^{1/2}]\\&+2[\mathrm{ber}_1(u_p)^{1/2}\mathrm{bei}_1(u_p)^{1/2}][\mathrm{ker}_1(u_t)^{1/2}\mathrm{ber}_1(u_t)^{1/2}+\mathrm{kei}_1(u_t)^{1/2}\mathrm{bei}_1(u_t)^{1/2}]\}\ ,\end{aligned} \tag{71}$$

where $u_t\equiv\omega r_t^2/D$. Comparing Eqs. (46) and (71) shows that for all frequencies

$$\lim_{r_p\to r_t}S(\omega)=0\ . \tag{72}$$

As discussed earlier with regards to the total noise power, Eq. (72) is a necessary condition that must be obeyed by a closed diffusive system. Furthermore,

$$\lim_{\omega\to0}S_B(\omega)=C_{\rm FN}\langle(\delta N)^2\rangle r_p^2D^{-1}\ln u_T\ . \tag{73}$$

Combining Eqs. (48), (66), and (73) results in

$$\lim_{\omega\to 0} S(\omega)=2C_{\mathrm{FN}}\langle(\delta N)^2\rangle r_p^2 D^{-1}\ln(r_t/r_p)\ . \tag{74}$$

Equation (74) shows that in the low-frequency limit the spectrum for a bounded net plane is constant. This conclusion has also been arrived at by different means for the case of one-dimensional diffusion by van Vliet and Fassett[16] and by van Vliet and van der Ziel,[40] who argued within the general framework of Richardson's theory that a constant low-frequency spectrum would result from placing a lower bound on vectors in k space.

The presence of the boundary removes the low-frequency logarithmic divergence of $S(\omega)$. This negates the argument that a diffusion process cannot produce a constant low-frequency spectrum as is found experimentally for K/W.[4,23] However, it must be noted that a clear refutation of the model was never really provided by these experiments as they did not conform to the assumptions of the theory. Neither study probed a portion of a single-crystal plane, and in one case the adsorbate was in equilibrium with its vapor,[23] which requires analysis using a grand-canonical ensemble.

With the aid of Eqs. (49) and (66), the asymptotic expansion of Eq. (71) yields

$$\lim_{\omega\to\infty} S(\omega)=2^{3/2}D^{1/2}C_{\mathrm{FN}}\langle(\delta N)^2\rangle \times r_p^{-1}\omega^{-3/2}(1-e^{-(r_t-r_p)\sqrt{2\omega/D}})\ . \tag{75}$$

When $u_p \gg 1/2(1-r_t/r_p)^2$, then $S(\omega)\to S_\infty(\omega)$, i.e., Eq. (75) reduces to a $\omega^{-3/2}$ dependence equivalent to Eq. (49)

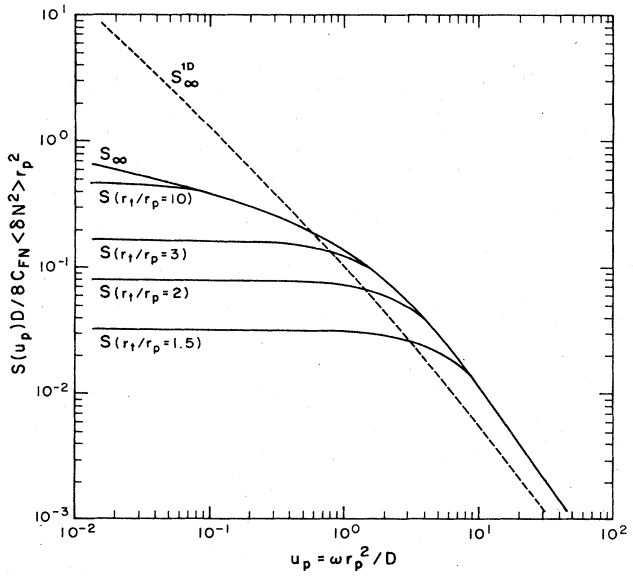

FIG. 1. Graph of the normalized spectra for unbounded diffusion $S_\infty(u_p)$ with probe radius r_p, bounded diffusion $S(u_p; r_t/r_p=1.5,2,3,10)$ with net plane radius r_t, and unbounded one-dimensional diffusion $S^{1D}_\infty(u_p)$ with a square probe of length l_x. The latter quantity is plotted assuming $l_x=\pi r_p^2$. See Eqs. (46), (66), (71), and (78) for the functional forms.

for the unbounded system. Physically, this is because appreciable adatom interaction with the boundary is not allowed on such a small timescale.

Figure 1 graphs the spectra $S_\infty(\omega)$ and $S(\omega)$ given, respectively, by Eqs. (46) and (66). $S(\omega)$ is plotted for several different ratios of the net plane to probed-area radii $r_t/r_p=1.5$, 2, 3, and 10. The presence of the boundary reduces the noise power in the low-frequency regime.

VII. ANISOTROPIC DIFFUSION

The preceding sections assume two-dimensional adsorbate diffusion is isotropic. However, certain substrate systems exist, e.g., bcc (211) planes, that are known to induce anisotropy in the diffusive adatom motion. The relation between a diffusion tensor and the field-emission autocorrelation function has been worked out by Bowman *et al.*[41] This section considers what influence such behavior has on the unbounded spectral density.

If a square region of the net plane is probed, the Eq. (35) autocorrelation can be factored as

$$R^{\square}_\infty(t)=R^x_\infty(t)R^y_\infty(t)\ . \tag{76}$$

The superscript $\square$ is a reminder that a square probe is used,

$$R^i_\infty(t)=\left[\frac{C_{\mathrm{FN}}\langle(\delta N)^2\rangle}{l_i^2 4\pi D_i t}\right]^{1/2} \times\int_0^{l_i}dx_1\int_0^{l_i}dx_2 e^{-(x_1-x_2)^2/4D_i t}\ ,$$

and l_i is the length of the probed region in the ith direction ($i=x,y$). D_i denotes the corresponding diffusion coefficient. The spectrum is then

$$S^{\square}_\infty(\omega)=S^x_\infty(\omega)*S^y_\infty(\omega)\ , \tag{77}$$

where $*$ denotes the convolution product and $S^i_\infty(\omega)$ is the spectrum corresponding to the one-dimensional $R^i_\infty(t)$. In general, this results in a complicated expression for $S^{\square}_\infty(\omega)$. By treating the limit of one-dimensional (1D) diffusion, i.e., $D_y/D_x=0$, with no boundaries, the effect of channeled motion on the noise can be demonstrated. Restricting adatom motion to the x direction reduces Eq. (76) to

$$R^{1\mathrm{D}}_\infty(t)=[C_{\mathrm{FN}}\langle(\delta N)^2\rangle]^{1/2}R^x_\infty(t)\ .$$

The superscript 1D shows that the spectrum results from one-dimensional motion. Application of the Wiener-Khinchin theorem yields

$$S^{1\mathrm{D}}_\infty(\Omega)=C_{\mathrm{FN}}\langle(\delta N)^2\rangle l_x^2 D^{-1}\Omega^{-3}[1-e^{-\Omega}(\cos\Omega+\sin\Omega)]\ , \tag{78}$$

and $\Omega\equiv l_x(\omega/2D)^{1/2}$. The frequency dependence due to one-dimensional diffusion has been given previously by Burgess[10] and Voss and Clarke.[18] Equation (78) is graphed in Fig. 1 assuming $l_x=\pi r_p^2$. This results in equality of the scaled noise power $P/\langle(\delta N)^2\rangle$ for circular and square probed regions. The normalization allows for

possible anisotropy in $\langle(\delta N)^2\rangle$, which has been observed by measuring $R(0)$ with a narrow rectangular probe.[42]

To compare one- and two-dimensional motion using S_∞^{1D} and S_∞, respectively, it is necessary to show that the probe geometry has a negligible effect on the frequency dependence of S_∞, because in the latter case a circular probe was assumed for the calculation. The ambiguity is removed by first noting

$$\lim_{\omega\to 0} S_\infty^{1D}(\omega)=4C_{FN}\frac{\langle(\delta N)^2\rangle l_x}{3(D\omega)^{1/2}} \tag{79}$$

and

$$\lim_{\omega\to\infty} S_\infty^{1D}(\omega)=C_{FN}\langle(\delta N)^2\rangle\left[\frac{8D}{l_x^2\omega^3}\right]^{1/2} . \tag{80}$$

Comparison of Eqs. (49) and (80) yields

$$\lim_{\omega\to\infty}\left[\frac{S_\infty(\omega)}{S_\infty^{1D}(\omega)}\right]=\frac{l_x}{r_p} . \tag{81}$$

Thus, excluding a numerical factor, the high-frequency dependence of the flicker noise is of probe geometry. This is a characteristic property of diffusion processes.[19] The high-frequency noise component is one dimensional in nature.

It is now shown that the low-frequency limit of Eq. (77), i.e., the spectrum corresponding to a square probe, is the same as that obtained in Eq. (48) for the case of a circular probe. For a square probe $A_p=l_x^2$ and Eq. (77) becomes

$$S_\infty^{\square}(\omega)=\frac{C_{FN}\langle(\delta N)^2\rangle}{A_p}\frac{8}{\pi D}\int_0^{l_x}dx\int_0^{x}dz\int_0^{l_x}dy\int_0^{y}dv\,\ker([\omega(z^2+v^2)/D]^{1/2}) . \tag{82}$$

Equation (82) can be written

$$S_\infty^{\square}(\omega)=\frac{C_{FN}\langle(\delta N)^2\rangle}{A_p}\frac{16}{\pi D}\int_0^{l_x}dx\int_0^{l_x}dy\int_0^{\tan^{-1}(y/x)}d\theta\int_0^{x/\cos\theta}dr\,r\,\ker(ar) , \tag{83}$$

where $a\equiv\sqrt{\omega/D}$ and $r\equiv(z^2+v^2)^{1/2}$. For low frequencies Eq. (83) becomes

$$\lim_{\omega\to 0}\frac{S_\infty^{\square}(\omega)D}{8C_{FN}\langle(\delta N)^2\rangle r_p^2}=\frac{1}{4}\ln\left[\frac{4}{\pi}\frac{D}{\omega r_p^2}\right] . \tag{84}$$

Comparing of Eqs. (48) and (84) coupled with Eq. (81) demonstrates that probe geometry has a negligible effect on the spectra, and therefore one can use S_∞^{1D} and S_∞ to represent the limiting cases of one- and two-dimensional diffusion. Figure 1 illustrates that one-dimensional motion increases the noise power in the low-frequency band of the spectrum.

VIII. SUMMARY

The aim of this paper has been to analyze the spectrum of field-emission flicker noise induced by equilibrium adsorbate density fluctuations in a canonical ensemble. The relative field-emission—current autocorrelation function $R(t)$ is a product of two factors. One contains terms related to the Fowler-Nordheim equation, C_{FN}, and the second is the autocorrelation of the total adparticle number within the probed region, $\langle\delta N(t)\delta N(0)\rangle$, Eq. (9). A general expression for $R(t)$ has been derived that contains the Green function for the diffusion equation $G(\mathbf{r}_1,t;\mathbf{r}_2)$ and the pair covariance of the density fluctuations $C(\mathbf{r}_1,\mathbf{r}_2)$, Eq. (14). This expression is simplified using several results from the theory of integral equations, where $C(\mathbf{r}_1,\mathbf{r}_2)$ is taken as the kernel. The procedure is equivalent to a Karhunen-Loève expansion of the density fluctuation $\delta n(\mathbf{r})$, Sec. III. The presence of boundaries and critical fluctuations on $C(\mathbf{r}_1,\mathbf{r}_2)$ has also been discussed, although subsequent analysis assumes the system is not influenced by critical fluctuations, Sec. IV. A form of $R(t)$ studied by Gomer was obtained by considering a square probed region on a square net plane, Eq. (32). This formulation yields an expression for the flicker-noise power that is a product of the constant C_{FN}, the mean-square particle-number fluctuation $\langle(\delta N)^2\rangle$ within the probed region, and the factor $1-A_p/A_T$. The latter term relates the noise power to the ratio of the probed to net plane area, Eq. (34). For unbounded diffusion the autocorrelation function is proportional to Smoluchowski's probability after-effect factor, Eq. (35), and the spectral-density function $S_\infty(\omega)$ is analogous to one derived by Burgess for contact noise in semiconductors, Eq. (46). A closed expression was found for diffusion on a circular net plane with a circular probe. The form of $S(\omega)$ explains several characteristics of field-emission—noise measurements. It was shown that the existence of finite net plane area produces a flat low-frequency spectrum, Eq. (74). The effect of anisotropic diffusion on $S(\omega)$ in the limit of one-dimensional motion was considered in Sec. VII. The qualitative result of such motion is to increase the noise power in the low-frequency band in contrast to the boundary effect, which decreases it. The high-frequency dependence of $S(\omega)$ is affected by neither the finite size of the net plane nor by anisotropic motion.

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