# The Linear Theory of Fluctuations Arising from Diffusional Mechanisms-An Attempt at a Theory of Contact Noise

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The spectral density is calculated for the electrical resistance when it is linearly coupled to a diffusing medium (particles or heat) undergoing thermally excited fluctuations. Specific forms of the spectral density are given for several types of coupling which are simple and physically reasonable. The principal objective is the understanding of the frequency dependence of the resistance fluctuations in contacts, rectifying crystals, thin films, etc.

#### 1. Introduction

WHEN a direct current is passed through a granular resistance such as a carbon microphone or a metallic-film grid leak, or through a single contact, there is produced a voltage fluctuation possessing a component called contact noise which is differentiated from the familiar thermal noise component by the fact that its r.m.s. value in any frequency band is roughly proportional to the magnitude of the average applied voltage, and is differentiated from shot noise by the strong frequency dependence of its spectral density. One may regard this component of the voltage fluctuation as arising from the spontaneous resistance fluctuations of the element in question if one is willing to allow the resistance to have a slight voltage dependence. This effect has been the subject of numerous experimental investigations, 1-8 among which we mention in particular that of Christensen and Pearson<sup>9</sup> on granular resistance elements. These authors (henceforth abbreviated as CP) arrived at an empirical formula, to be discussed presently, connecting the contact noise power per unit frequency band with the applied voltage, the resistance, and the frequency for several types of granular resistance. Their measurements covered a range of frequency from 60 to 10,000 cps, and involved the variation of several other parameters, i.e., pressure. More recently, Wegel and Montgomery<sup>10</sup> have measured the noise power arising

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<sup>10</sup> Private communication.

from single contacts and have obtained results in agreement with the CP empirical formula down to frequencies of the order of  $10^{-1} - 10^{-2}$  cps.

Significant theoretical work upon this problem has not been attempted until recently. G. G. Macfarlane11 has advanced a theory based upon a non-linear mechanism containing one degree of freedom which seems to be in agreement with the CP law. W. Miller12 has worked out a general theory of noise in crystal rectifiers. His theory is linear, contains essentially an infinite number of degrees of freedom, and is equivalent in many respects to the theory discussed in this paper; however, he has not succeeded in obtaining agreement with the experimental data on crystal rectifiers (which satisfy approximately the CP law) for any of the specific models he used.

The purpose of this paper is the calculation of the spectral density of the fluctuations of the electrical resistance when it is linearly coupled to a diffusing medium (particles or heat), or, mathematically speaking, is equal to a linear function of the concentration deviations of this diffusing medium. This diffusing medium undergoes thermally excited fluctuations and thereby causes fluctuations in the resistance. The motive behind this investigation was the understanding of the frequency dependence of contact noise discussed in the following paragraphs, but at the present time it is apparent that this treatment in addition may apply to rectifying crystals, thin films, transistors, etc. The quantitative details of the coupling between the resistance and the diffusing medium are not considered here; in consequence of which, this work can hardly pretend to give a complete explanation of contact noise. However, important results are given concerning the relation between the spectral density of the resistance, on one hand, and the geometry of the coupling and the dimensionality of the diffusion field on the other.

Now let us consider the CP empirical formula in detail. Let  $\bar{R}$  be the average resistance13 of the contact (we will henceforth consider only contacts and will regard a granular resistance as a contact assemblage) and let  $R_1$ (t) be the instantaneous deviation from the average. By theorems I-3 of Appendix I, we can express the m.s. value of  $R_1$  as a sum of the m.s. values of  $R_1$  in each frequency interval as follows:

$$\overline{R_1^2} = \int_0^\infty S(\omega) \ d\omega, \tag{1.1}$$

12 To be published.

<sup>&</sup>lt;sup>11</sup> G. G. Macfarlane, Proc. Phys. Soc. 59, Pt. 3, 366-374 (1947).

<sup>&</sup>lt;sup>13</sup> The resistance of a contact is composed of two parts: the "gap resistance" and the "spreading resistance." The term "gap resistance" is self-explanatory. The "spreading resistance" is the resistance involved in driving the electric current through the body of resistance is the resistance involved in driving the electric current current current current body of the body of the contact material along paths converging near the area of lowest gap resistance. The measured contact resistance is the sum of these two parts. In some of the particular physical models considered in Section 5,  $\bar{R}$  is taken to be the gap resistance necessitating ad hoc arguments relating gap resistance and total resistance.

where  $S(\omega)$  is called the spectral density of  $R_1$  and  $\omega$  is the frequency in radians per second. Now in our notation the CP formula may be expressed

$$S(\omega) = KV^{a-2} \bar{R}^{b+2}/\omega, \qquad (1.2)$$

where V is the applied d-c voltage across the contact, K is a constant depending upon the temperature and the nature of the contact, and a and b are constants having values of about 1.85 and 1.25 respectively. CP state that the constant K is equal to about 1.2  $\times$  10<sup>-10</sup> in the case of a single carbon contact at room temperature.

In this paper we will regard the nonvanishing of a-2 as arising from a non-linear effect which should become negligible at a sufficiently low voltage, although this interpretation does not seem completely justified on the basis of the work of CP. Consequently we assume that  $a \to 2$  as  $V \to 0$  in such a way that  $V^{a-2} \to 1$ . This is in keeping with the idea that the resistance fluctuations are truly spontaneous—at least for small applied voltages.

Although Eq. (1.2) may represent the observations over a large range of frequency it must break down at very high and very low frequencies in order that the noise power be finite (or, in other words, in order that the integral (1.1) converge).

One has several clues to be considered in looking for an underlying mechanism of the resistance fluctuations. First of all, the mechanical action of the thermal vibrations in the solid electrodes of the contact seems to be unimportant because of the following reasons: (1) there are no resonance peaks in  $S(\omega)$  at the lowest characteristic frequencies of mechanical vibration of the contact assembly; (2)  $S(\omega)$  becomes very large far below the lowest characteristic frequency; and (3), according to CP,  $\overline{R_1^2}$  is strictly proportional to  $V^2$  when the fluctuations are produced by acoustic noise vibrating the contact, whereas  $\overline{R_1^2}$  is proportional to  $V^{a-2}$ ,  $a \sim 1.85$ , when the fluctuations arise from the dominant mechanism existing in the macroscopically unperturbed contact. One of the obvious mechanisms left is a diffusional mechanism. Such a mechanism does not violate any of the observations to date and, furthermore, possesses a sufficient density of long relaxation times to give large contributions to  $S(\omega)$  near zero frequency.

Evidence that diffusion of atoms (or ions) can be important in modulating a current is provided by the "flicker effect" in which the emission of electrons from a heated cathode is caused to fluctuate by the fluctuations in concentration of an adsorbed layer. We might suppose that contact noise is a different manifestation of the basic mechanism involved in the flicker effect.

In view of these considerations it seems worthwhile to investigate in a

general way a large class of models involving resistance fluctuations arising from diffusional mechanisms. In the next section we propose a general mathematical model embracing a class of linear diffusional mechanisms. In Sections 3 and 4 the consequences of the general mathematical model are obtained by the "Fourier" and "Smoluchowski" methods, respectively, these alternative methods leading to identical results. In Section 5, the general results are specialized to several physical cases, some of which are introduced only for the purpose of providing some insight into the relations between the possible physical mechanisms and the resultant resistance fluctuations, and one of which along with its refinement is a successful<sup>14</sup> attempt to provide a theory of Eq. (1.2). Section 6 is a summary.

## 2. The General Mathematical Model

The physical models which we consider in this paper are concerned with the fluctuations of contact resistance arising from a diffusional process. We are consequently led to consider the following general mathematical model embracing a rather extensive class of the physical models as special cases: Let us consider the instantaneous contact resistance R(t) to be related to the intensity  $c(\mathbf{r}, t)$  of some diffusing quantity as follows<sup>15</sup>:

$$G(R(t)) = \int F(\mathbf{r}, c(\mathbf{r}, t)) d\mathbf{r}, \qquad (2.1)$$

where r is a vector in two or three dimensional space depending on whether the diffusion takes place on a surface or in a volume, and dr is correspondingly a differential area or volume. The intensity c(r, t) may be either a concentration (in the case of diffusion of material in two or three dimensions) or a temperature (in the case of heat flow in three dimensions). In writing Eq. (2.1) we have evidently assumed that the contact resistance R(t) is independent of the applied voltage. Eq. (2.1) may of course allow a dependence on voltage through the quantity c; however, we will consider no processes involving a dependence of c on the voltage. These restrictions, strictly speaking, make the model applicable only in the limit of low applied voltages.

Before proceeding further let us limit the treatment to the case in which the deviations of R and c from their average values are sufficiently small for higher powers of these deviations to be neglected. Let

$$R(t) = \bar{R} + R_1(t),$$
 (2.2)

$$c(\mathbf{r},t) = \bar{c} + c_1(\mathbf{r},t), \tag{2.3}$$

<sup>&</sup>lt;sup>14</sup> That is, successful in so far as agreement with the form of Eq. (1.2) is concerned. <sup>16</sup> A relation more general than R(t) = SF(r, c(r, t)) dr is required as one can see from considering the special case of a total resistance composed of a parallel array of resistive elements.

where  $\overline{R}$  and  $\overline{c}$  are the average<sup>16</sup> values of R(t) and c(r, t) respectively. Evidently,  $\overline{R_1(t)} = 0$ , and  $\overline{c_1(r, t)} = 0$ . Introducing the expressions (2.2) and (2.3) into Eq. (2.1), expanding in terms of  $c_1(r, t)$ , and neglecting terms of the order of  $c_1^2$ , we get

$$R_1(t) = \int f(\mathbf{r}) c_1(\mathbf{r}, t) d\mathbf{r},$$
 (2.4)

where

$$f(\mathbf{r}) = \left[\frac{\partial F(\mathbf{r}, c)}{\partial c}\right]_{c=\tilde{c}} / \left[\frac{dG(R)}{dR}\right]_{R=\overline{R}}$$

The function f(r) defines the linear coupling between  $R_1$  and  $c_1$  and depends upon the specific physical model used. The non-linear terms neglected in Eq. (2.4) may be of importance under some conditions; however, we will not consider them here. Nevertheless, non-linear effects in the behavior of  $c_1$  itself are possibly important in determining the form of the power spectrum of  $R_1(t)$  in the neighborhood of zero frequency.

#### 3. The Fourier Series Method of Solution

In this section we consider the state of the diffusing system to be defined by the Fourier space-amplitudes  $c_k(t)$  of  $c_1(\mathbf{r}, t)$ . The time behavior of  $c_k(t)$  will be described by an infinite set of ordinary differential equations containing random exciting forces according to the conventional theory of Brownian motion.<sup>17</sup> This method yields the spectral density of  $R_1(t)$  directly.

Now the diffusion process is assumed to occur in a rectangular area  $A_2 = L_1 \times L_2$  or in a rectangular parallelopiped of volume  $A_3 = L_1 \times L_2 \times L_3$ . In regions of the above types, if we apply periodic boundary conditions  $^{18}$ ,  $c_1(\mathbf{r}_0t)$  may be expanded in Fourier space-series as follows:

$$c_1(\mathbf{r},t) = \sum_{\mathbf{k}}' c_{\mathbf{k}}(t)e^{i\mathbf{k}\cdot\mathbf{r}}$$
 (3.1)

where the components of k take the values

$$k_i = 2\pi n_i/L_i, i = 1, \dots, \nu,$$
 (3.2)

in which  $n_i$  are integers and  $\nu$  is the number of dimensions. The prime on the summation indicates that the term for k=0 is to be omitted. This is required by the equivalence of the time and space averages of  $c_1$  (true for  $A_r$ , sufficiently large) and by the vanishing of the time average of  $c_1$  (by definition).

<sup>16</sup> The average values here may be considered as either time or ensemble averages but not space averages.

See Wang and Uhlenbeck, Rev. Mod. Phys., 17, 323-342 (1945).
 If the final results are given by integrals over k-space they will be insensitive to the boundary conditions.

Before proceeding to the solution itself let us consider what it is that we wish to know about  $c_k(t)$ . Expanding the function f(r) of Eq. (2.4) in a Fourier space-series in the region  $A_r$ ,

$$f(\mathbf{r}) = \sum_{\mathbf{k}} f_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{3.3}$$

we can write Eq. (2.4) in the form

$$R_1(t) = A_{\nu} \sum_{k}' f_{k}^* c_{k}(t)$$
 (3.4)

where  $f_k^*$  is the conjugate of  $f_k$ .

The spectral density  $S(\omega)$  of  $R_1(t)$  is then

$$S(\omega) = A_{\nu}^{2} \sum_{\mathbf{k}\mathbf{k}'} C_{\mathbf{k}\mathbf{k}'}(\omega) f_{\mathbf{k}}^{*} f_{\mathbf{k}'}, \qquad (3.5)$$

where  $C_{kk'}(\omega)$  is the spectral density matrix for the set  $c_k(t)$  given by

$$C_{kk'}(\omega) = 2\pi \lim_{\tau \to \infty} \frac{1}{\tau} \left[ c_k(\omega, \tau) c_{k'}^*(\omega, \tau) + c_k(-\omega, \tau) c_{k'}^*(-\omega, \tau) \right]$$
(3.6)

in which

$$c_{k}(\omega, \tau) = \frac{1}{2\pi} \int_{-\tau/2}^{+\tau/2} c_{k}(t)e^{-i\omega t} dt.$$
 (3.7)

For a full discussion of spectral densities and spectral density matrices see Appendix I. Consequently our objective in this section is the calculation of the maxtix  $C_{kk'}$  ( $\omega$ ) defined by Eq. (3.6).

Now we assume that  $c_1(\mathbf{r}, t)$  satisfies the diffusion equation

$$\frac{\partial}{\partial t} c_1(\mathbf{r}, t) = D_{\nabla}^{-2} c_1(\mathbf{r}, t) + g(\mathbf{r}, t)$$
 (3.8)

where D is a constant,  $\nabla^2$  is the Laplacian operator in two or three dimensions, and where  $g(\mathbf{r},t)$  is a random source function, whose Fourier space-amplitudes  $g_k(t)$  possess statistical properties to be discussed presently. The random source function g is required for exciting  $c_1$  sufficiently to maintain the fluctuations given by equilibrium theory. In the case of material diffusion the random source function g may be discarded in favor of a random force term of the form  $-D/\chi T \cdot \vec{\nabla} \cdot [f(\bar{c} + c_1)]$ , where  $\nabla \cdot f = divf$ ,  $\chi$  is the Boltzmann constant, T is the temperature, and f is the random force; however, in the linear approximation these two procedures will give identical final results. In the case of heat flow it is understood that the diffusion constant is  $D = K/\rho C$  where K is the thermal conductivity,  $\rho$  the density, and C the specific heat. Eq. (3.8) as written is valid only for D a constant and  $c_1$  small.

Introducing the expansion (3.1) and the expansion

$$g(\mathbf{r}, t) \sum_{\mathbf{k}}' g_{\mathbf{k}}(t)e^{i\mathbf{k}\cdot\mathbf{r}}$$
 (3.9)

into Eq. (3.8) we obtain the infinite set of ordinary differential equations

$$\frac{d}{dt} c_{\mathbf{k}}(t) = -Dk^2 c_{\mathbf{k}}(t) + g_{\mathbf{k}}(t), \qquad (3.10)$$

$$k = |\mathbf{k}|.$$

describing the time behavior of the Fourier space-amplitudes  $c_{\mathbf{k}}(t)$ . The Fourier space-amplitudes  $g_{\mathbf{k}}(t)$  are assumed to be random functions of t possessing a white (flat) spectral density matrix  $C_{\mathbf{k}\mathbf{k}}$ , independent of frequency. Multiplying Eq. (3.10) by  $\frac{1}{2\pi}e^{-i\omega t}$ , integrating with respect to time from  $-\frac{1}{2}\tau$  to  $+\frac{1}{2}\tau$ , and neglecting the transients at the end points of the  $\tau$ -interval, we obtain

$$c_{\mathbf{k}}(\omega, \tau) = \frac{g_{\mathbf{k}}(\omega, \tau)}{i\omega + Dk^2} \tag{3.11}$$

where  $c_k(\omega, \tau)$  is given by Eq. (3.7) and  $g_k(\omega, \tau)$  is given by an analogous equation. Forming the spectral density matrices we get for the diagonal elements

$$C_{kk'}(\omega) = \frac{G_{kk}}{\omega^2 + D^2k^4} \tag{3.12}$$

The matrix  $G_{kk'}$  can now be evaluated by the thermodynamic theory of fluctuations (See Appendix II). This theory gives

$$c_{\mathbf{k}}(l)c_{\mathbf{k}'}^{*}(l) = \frac{\chi \delta_{\mathbf{k}\mathbf{k}'}}{A_{\nu}s''}$$
 (3.13)

where

$$\delta_{kk'} = \begin{cases} 1 \text{ if } k = k' \\ 0 \text{ otherwise,} \end{cases}$$

$$s'' = -\left\{ \frac{\partial^2 s}{\partial c^2} \right\}_{c=\bar{c}} + \frac{1}{\bar{T}} \left\{ \frac{\partial^2 e}{\partial c^2} \right\}_{c=\bar{c}}$$
(3.14)

s and e being the entropy and energy, respectively, per unit area or volume,  $\overline{T}$  the average temperature, and  $\chi$  the Boltzmann constant. In the case where c is the concentration of particles whose configurational energy is constant,  $s'' = \chi/\bar{c}$ . If c be the temperature T then  $s'' = C/\overline{T}^2$  where C

is the heat capacity per unit area or volume. Now by a general theorem concerning spectral density matrices (see Appendix I) we have

$$\overline{c_{\mathbf{k}}(t)c_{\mathbf{k}'}^*(t)} = \int_0^\infty C_{\mathbf{k}\mathbf{k}'}(\omega) \ d\omega,$$

giving finally by combination with (3.12) and (3.13),

$$G_{kk'} = \frac{2}{\pi} \frac{\chi Dk^2}{A_{\nu} s''} \delta_{kk'}, \qquad (3.15)$$

and

$$C_{kk'}(\omega) = \frac{2}{\pi} \frac{\chi D}{A \cdot s''} \frac{k^2 \delta_{kk'}}{\omega^2 + D^2 k^4}.$$
 (3.16)

The spectral density  $S(\omega)$  of  $R_1(t)$  then becomes

$$S(\omega) = \frac{2}{\pi} \frac{\chi A_{\nu} D}{s''} \sum_{k} \frac{k^2 |f_k|^2}{\omega^2 + D^2 k^4}.$$
 (3.17)

If we are concerned with frequencies greater than a characteristic frequency

$$\omega_0 = 4\pi^2 D/L^2 \tag{3.18}$$

where L is the smallest of  $L_i$ ,  $i = 1, \dots, \nu$ , then the summation in (3.17) may be replaced by an integration giving

$$S(\omega) = 2^{\nu+1} \pi^{\nu-1} \frac{\chi D}{s''} \int \frac{|f(\mathbf{k})|^2 k^2 d\mathbf{k}}{\omega^2 + D^2 k^4}$$
(3.19)

where

$$f(\mathbf{k}) = \frac{1}{(2\pi)^{\nu}} \int_{A_{\nu}} f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r}.$$
 (3.20)

The integration in Eq. (3.19) is carried out over the entire  $\nu$ -dimensional k-space. If the range of the function f(r) is sufficiently small compared with the region  $A_{\nu}$ , or if we let  $A_{\nu}$  become indefinitely large, then the integration in Eq. (3.20) may be extended to all of  $\nu$ -dimensional r-space.

It is perhaps revealing to rephrase Eqs. (3.17) and (3.19) in terms of distributions of relaxation times. In the theory of dielectrics we speak of the real part of the dielectric constant being equal to a series of terms summed over a distribution of relaxation times:  $\sum_i a_i \tau_i/(1 + \tau_i^2 \omega^2)$ , if the distribution is discrete, or  $\int_0^\infty a(\tau)\tau d\tau/(1 + \tau^2\omega^2)$ , if the distribution is continuous. In the above,  $a_i$  is the weight for the relaxation time  $\tau_i$ , and, in

the case of a continuous distribution,  $a(\tau)d\tau$  is the weight for the relaxation times in the range  $d\tau$  containing  $\tau$ . In these terms Eq. (3.17) becomes

$$S(\omega) = \sum_{\mathbf{k}} \frac{a_{\mathbf{k}} \tau_{\mathbf{k}}}{1 + \tau_{\mathbf{k}}^2 \omega^2}, \qquad (3.17a)$$

where

$$a_{k} = \frac{2}{\pi} \frac{\chi}{s''} |f_{k}|^{2}.$$
 (3.17b)

Eq. (3.19) becomes

$$S(\omega) = \int_0^\infty \frac{a(\tau)\tau \, d\tau}{1 + \tau^2 \, \omega^2} \tag{3.19a}$$

where

$$a(\tau) = \frac{2^{\nu} \pi^{\nu-1} \chi}{s'' D^{\nu/2} \tau^{\nu/2+1}} \int |f(l/\sqrt{D\tau})|^2 d\Omega_{\nu}$$
 (3.19b)

in which l is the unit vector in the direction of k,  $d\Omega_r$  is the differential "solid" angle in the  $\nu$ -dimensional k-space, and the integration is over the total solid angle  $(2\pi$  in 2 dimensions, or  $4\pi$ , in 3).

It is of interest to calculate the self-covariance  $R_1(t)R_1(t+u)$ . In Appendix I, it is shown that the self-covariance above is related to the spectral density  $S(\omega)$  as follows:

$$\overline{R_1(t)R_1(t+u)} = \int_0^\infty S(\omega) \cos u\omega \, d\omega. \tag{3.21}$$

Using  $S(\omega)$  in the form (3.17), Eq. (3.21) gives

$$\overline{R_1(t)R_1(t+u)} = \chi A_{\nu}/S'' \cdot \sum_{k} |f_k|^2 e^{-Duk^2}, \qquad (3.22)$$

whereas with  $S(\omega)$  in the form (3.19) we get

$$\overline{R_1(t)R_1(t+u)} = (2\pi)^{\nu} \chi/s^{\nu} \int |f(\mathbf{k})|^2 e^{-Duk^2} d\mathbf{k}.$$
 (3.23)

The method of the next section yields the self-covariance directly.

#### 4. Smoluchowski Method of Solution

We call the procedure employed in this section the "Smoluchowski method" because it is based on an equation very closely analogous to the well-known Smoluchowski equation forming the basis of the theory of Markoff processes.<sup>19</sup> We set out directly to calculate the self-covariance for  $R_1(t)$  which, by Eq. (2.4), is given by

$$\overline{R_1(t)R_1(t+u)} = \iint f(\mathbf{r}')f(\mathbf{r})\overline{c_1(\mathbf{r}'t)c_1(\mathbf{r}',t+u)} d\mathbf{r}' d\mathbf{r}. \tag{4.1}$$

Thus the problem is now reduced to the calculation of  $c_1(\mathbf{r}', t)c_1(\mathbf{r}, t+u)$ .

The quantity  $c_1(\mathbf{r}', t)c_1(\mathbf{r}, t+u)$  is calculated in two steps. First we find  $c_1(\mathbf{r}, t+u)$ , the average value of  $c_1$  at the point  $\mathbf{r}$  at the time t+u with the restriction that  $c_1$  is known at every point  $\mathbf{r}'$  with certainty to be  $c_1(\mathbf{r}', t)$  at the time t (assuming, of course, that t is given by multiplying the above  $c_1(\mathbf{r}, t+u)$  by  $c_1(\mathbf{r}', t)$  and averaging over-all values of  $c_1(\mathbf{r}, t)$  at time t; thus:

$$\overline{c_1(\mathbf{r}',t)\overline{c_1(\mathbf{r},t+u)}^{(t+u)}}^{(t)} = \overline{c_1(\mathbf{r}',t)c_1(\mathbf{r},t+u)}. \tag{4.2}$$

Now we assume that  $c_1(r, t + u)$  is related to c(r', t) by an integral equation, analogous to the Smoluchowski equation, as follows:

$$\overline{c_1(\mathbf{r},t+u)}^{(t+u)} = \int \rho(|\mathbf{r}-\mathbf{r}'|,u)c_1(\mathbf{r}',t) d\mathbf{r}'. \tag{4.3}$$

In the case that c represents a concentration as in the diffusion of particles,  $\rho(|r-r'|, u) dr$  is the probability that a particle be in the  $\nu$ -dimensional volume element dr at time t+u when it is known with certainty to be at r' a time t. Now the number of particles in dr' at r' at time t is evidently  $[\bar{c} + c_1(r', t)] dr'$ ; consequently, the probable number of particles in dr at time t+u which were in dr' at time t is  $\rho(|r-r'|, u)[c+c_1(r', t)] dr dr'$ . Integration over dr' gives the total probable number

Integration over 
$$d\mathbf{r}'$$
 gives the total probable number  $(\bar{c} + c_1(\mathbf{r}, t + u)) d\mathbf{r}$  of particles in  $d\mathbf{r}$  equal to  $\left(\int \rho(|\mathbf{r} - \mathbf{r}'|, u)[\bar{c} + c_1(\mathbf{r}', t)] d\mathbf{r}'\right) d\mathbf{r}$  which reduces to  $(\bar{c} + \int \rho(|\mathbf{r} - \mathbf{r}'|, u)c_1(\mathbf{r}', t) d\mathbf{r}') d\mathbf{r}$ . Division by  $d\mathbf{r}$  and subtraction of  $\bar{c}$ 

from both sides of the equality leads directly to Eq. (4.3). For the case of heat flow in crystal lattices the above picture can be used approximately if one uses the concept of phonons.<sup>20</sup> For a diffusional process  $\rho(|r-r'|, u)$  is the normalized singular solution of the diffusion equation<sup>21</sup>; thus

$$\rho(|\mathbf{r} - \mathbf{r}'|, u) = \frac{1}{(4\pi Du)^{r/2}} \exp\left[-|\mathbf{r} - \mathbf{r}'|^2/4Du\right]$$
(4.4)

<sup>19</sup> Loc cit.

J. Weigle, Experientia, 1, 99-103 (1945).
 Chandrasekhar, Rev. Mod. Phys. 15, 1 (1943).

where  $\nu$ , as previously defined, is the number of dimensions of the region in which the process occurs.

Combining Eqs. (4.2) and (4.3) we get

$$\overline{c_1(r', t)c_1(r, t + u)} = \int \overline{c_1(r', t)c_1(r'', t)}^{(t)} \rho(|r - r''|, u) dr''. \quad (4.5)$$

Now using the fact that

$$\overline{c_1(\mathbf{r}', t)c_1(\mathbf{r}'', t)}^{(t)} = \overline{c_1(\mathbf{r}', t)c_1(\mathbf{r}'', t)}$$
(4.6)

and using the relation

$$\overline{c_1(\mathbf{r}',\,t)c_1(\mathbf{r}'',\,t)} = \frac{\chi}{s''}\,\delta(\mathbf{r}'\,-\,\mathbf{r}'') \tag{4.7}$$

proved in Appendix II, Eq. (4.5) reduces to

$$\overline{c_1(\mathbf{r}',t)c_1(\mathbf{r},t+u)} = \frac{\chi}{s''} \rho(|\mathbf{r}-\mathbf{r}'|,u). \tag{4.8}$$

Introducing the expression (4.8) into Eq. (4.1) we obtain at once the desired result

$$\overline{R_1(t)R_1(t+u)} = \frac{\chi}{s''} \iint f(\mathbf{r})f(\mathbf{r}')\rho(|\mathbf{r}-\mathbf{r}'|, u) d\mathbf{r} d\mathbf{r}'$$

$$= \frac{\chi}{s''(4\pi Du)^{\nu/2}} \iint f(\mathbf{r})f(\mathbf{r}') \exp\left[-|\mathbf{r}-\mathbf{r}'|/4Du\right] d\mathbf{r} d\mathbf{r}'.$$
(4.9)

For the sake of comparison with Eq. (3.23) it is necessary to write (4.9) in terms of the Fourier space-transforms of the pertinent quantities. We write

$$f(\mathbf{r}) = \int f(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{k}$$

where

$$f(\mathbf{k}) = \frac{1}{(2\pi)^{\nu}} \int f(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r}.$$

Also, we write

$$\rho(|\mathbf{r} - \mathbf{r}'|, u) = \frac{1}{(4\pi Du)^{\nu/2}} \exp\left[-|\mathbf{r} - \mathbf{r}'|^2/4Du\right]$$
$$= \frac{1}{(2\pi)^{\nu}} \int \exp\left[-Duk^2 + i\mathbf{k}\cdot(\mathbf{r} - \mathbf{r}')\right] d\mathbf{k}.$$

After introduction of these expressions into (4.9) a short calculation yields the result

$$\overline{R_1(t)R_1(t+u)} = (2\pi)^{\nu} \frac{\chi}{S''} \int |f(k)|^2 e^{-Duk^2} dk$$
 (4.10)

which is identical with Eq. (3.23) (provided that we let  $A_{\nu} \to \infty$  in the latter). Thus the methods of approach used in Section 3 and in this Section are completely equivalent.

### 5. Special Physical Models

In the previous two Sections we have developed by two different methods the consequences of the general mathematical model discussed in Section 2. Here we apply the general results to some special physical cases. In this task we will be principally concerned with finding the form of the function  $f(\mathbf{r})$  and establishing the number of dimensions  $\nu$  of the diffusion field. The main objective here is to provide some orientation on what mechanisms are or are not reasonable and to find at least one mechanism leading to the observed spectral density (inversely proportional to the frequency).

a. A General Class of Models. Here we consider all at once mechanisms which can be adequately represented by having  $f(\mathbf{r})$  a  $\nu$ -dimensional Gaussian function of the form

$$f(\mathbf{r}) = \prod_{i=1}^{\nu} \frac{b_i}{(2\pi\Delta_i)^{1/2}} e^{-x_i^2/2\Delta_i}, \qquad (5.1)$$

where  $\Delta_i^{1/2}$  is the "width" of the function measured along the *i*-th coordinate  $x_i$ . This form of f(r) can represent approximately several types of localization of the coupling between  $R_1$  and  $c_1$ , as will be seen in the special examples later. Now if we work with  $A_r = \infty$ , we will then have to consider the Fourier space-transform of f(r), which is readily shown to be

$$f(\mathbf{k}) = \frac{1}{(2\pi)^{\nu}} \int f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r}$$

$$= \prod_{i=1}^{\nu} b_i / 2\pi \cdot e^{-\Delta_i k_i^2/2}$$
(5.2)

Inserting this result into Eq. (3.19) we obtain immediately

$$S(\omega) = \frac{2\chi D}{\pi s''} \left( \prod_{i=1}^{\nu} \frac{b_i^2}{2\pi} \right) \int \frac{\exp\left(-\sum_{i=1}^{\nu} \Delta_i \, k_i^2\right) k^2 \, d\mathbf{k}}{\omega^2 + D^2 \, k^4}$$

$$k = \sum_{i=1}^{\nu} k_i^2.$$
(5.3)

Inserting this expression (5.2) into Eq. (3.23) gives

$$\overline{R_1(t)R_1(t+u)} = \frac{\chi}{s''} \prod_{i=1}^{r} \frac{b_i^2}{[4\pi(\Delta_i + Du)]^{1/2}}; \qquad u > 0$$
 (5.4)

In order that (5.3) give the observed  $S(\omega)$   $\alpha$   $1/\omega$  as a result, the integral nust reduce to something proportional to  $\int k \, dk/(\omega^2 + D^2 k^4)$ . It is clearly impossible that any choice of  $\nu$  and any set of  $\Delta_i$  can achieve this result. Furthermore the self-covariance  $\overline{R_1(t)R_1(t+u)}$  corresponding to the observed  $S(\omega)$  should depend explicitly on the way  $S(\omega)$  deviates from  $1/\omega$  as  $\omega$  goes to zero. The expression (5.4) is finite for all u > 0 and does not depend upon any cut-off phenomena in  $S(\omega)$  at low frequencies. Therefore we can exclude any physical mechanisms belonging to the class considered

here. However, since several mechanisms that have been proposed do fall

into this class, we consider them below:

(i) Schiff's Mechanism. Schiff<sup>22</sup> considered tentatively that the fluctuations in contact resistance may be due to the variation in concentration of diffusing ions (atoms, or molecules) in a high resistance region bounded by parallel planes of very small separation. Schiff arrived at a noise spectrum proportional to  $1/\omega$  but at the expense of disagreeing in a fundamental way with thermodynamic fluctuation theory. Here we will show what the correct consequences of this mechanism are.

Consider that the high resistance region is bounded on either side by planes parallel to the  $(x_1, x_2)$ -plane and that the thickness in the  $x_3$ -direction is very small. Now this is obviously a case of the general model just considered in which we take  $\nu = 3$  and

$$\Delta_{1,2} \gg Du, 
\Delta_3 \ll Du,$$
(5.5)

where 1/u is of the order of magnitude of the frequencies of interest. It is then a matter of algebraic manipulation to show that

$$S(\omega) \simeq \frac{\chi D}{s''} \cdot \frac{b_1^2 b_2^2 b_3^2}{2\pi^3 \Delta_1^{1/2} \Delta_2^{1/2}} \cdot \int_0^\infty \frac{k_1^2 dk_1}{\omega^2 + D^2 k_1^4}$$

$$= \frac{\chi}{D^{1/2} s''} \cdot \frac{b_1^2 b_2^2 b_3^2}{2^{6/2} \pi^2 \Delta_1^{1/2} \Delta_2^{1/2}} \frac{1}{\omega^{1/2}}$$
(5.6)

and

$$\overline{R_1(t)R_1(t+u)} = \frac{\chi}{D^{1/2}s''} \cdot \frac{b_1^2 b_2^2 b_3^2}{(4\pi)^{3/2} \Delta_1^{1/2} \Delta_2^{1/2}} \cdot \frac{1}{u^{1/2}} \quad u > 0.$$
 (5.7)

Thus we see that Shiff's mechanism leads to a noise spectrum proportional to  $1/\omega^{1/2}$ , not  $1/\omega$ . The explanation of the singularity of the self-covariance (5.7) at u = 0 lies in the inequalities (5.5).

<sup>22</sup> L. I. Schiff, BuShips Contract NObs-34144, "Tech. Rpt. ∦3", (1946). Before the publishing of this paper, Schiff informed the writer that he has discarded this mechanism.

The above treatment could just as well be applied to the case in which the diffusing quantity is heat instead of ions.

(ii) Resistance of a Localized Contact Disturbed by a Diffusing Surface Layer. Here we consider the case of two conductors covered with diffusing surface layers. It is supposed that the conduction from one conductor to the other is distributed Gaussianly with a width  $\Delta^{1/2}$ . Finally, it is supposed that the conductivity through the above area varies with the surface concentration of the surface layer in that region. This situation is well represented by the above general model by taking  $\nu = 2$ ,  $\Delta_1 = \Delta_2 = \Delta$ , and  $b_1 = b_2 = b$ .

The self-covariance is readily calculated with the result

$$\overline{R_1(t)R_1(t+u)} = \frac{\chi}{s''} \cdot \frac{b^4}{4\pi(\Delta + Du)}$$
 (5.8)

The corresponding spectral density is

$$S(\omega) = \frac{1}{2\pi^2} \frac{\chi b^4}{s''} \int_0^{\infty} \frac{\cos u\omega \, du}{\Delta + Du} = \frac{1}{2\pi^2} \frac{\chi b^4}{s''D} \left[ -\cos (\omega \Delta/D) Ci(\omega \Delta/D) + \sin (\omega \Delta/D) \left( \frac{\pi}{2} - Si(\omega \Delta/D) \right) \right]$$
(5.8a)

where Ci(x) and Si(x) are the cosine and sine integrals<sup>23</sup> respectively. When  $\omega \ll D/\Delta$ 

$$S(\omega) \simeq -\frac{1}{2\pi^2} \frac{\chi b^4}{s''D} \log (\delta \omega \Delta/D),$$
 (5.8b)  
 $\delta = 0.5772,$ 

and when  $\omega \gg D/\Delta$ 

$$S(\omega) \simeq \frac{1}{2\pi^2} \frac{\chi b^4 D}{s'' \Delta^2} \frac{1}{\omega^2}.$$
 (5.8c)

Thus we see that this case does not lead to the experimental form of the spectral density. It must be remarked that here  $S(\omega)$  is very sensitive to the form of the self-covariance for small u.

b. Contact between Relatively Large Areas of Rough Surfaces Covered with Diffusing Surface Layers. We consider this case in detail since it leads to results in agreement with experiment. Furthermore, the more detailed consideration of this case will illustrate more fully the use of the general mathematical model, which may be of use in studying other diffusional mechanisms should they be postulated at some future time. This mechanism does not fall into the class just considered.

23 See Jahnke and Emde, "Tables of Functions," p. 3, Dover (1943).

Suppose that the contact in an idealized form consists of two rough surfaces close together. Let positions on the surfaces be measured with respect to a plane between the surfaces, which we will call the mid-plane. Let the coordinate system be oriented so that the  $x_1$  and  $x_2$  axes lie in the mid-plane. Furthermore let the region in the mid-plane corresponding to close proximity of the rough surfaces be a rectangular area  $A_2 = L_1 \times L_2$ . Now, for convenience, we describe positions on the mid-plane by a two dimensional vector  $\mathbf{r} = (x_1, x_2)$ , and henceforth it will be understood that all vector expressions refer to this two-dimensional space. Let the distance between the upper and lower surfaces at  $\mathbf{r}$  be denoted by  $h(\mathbf{r})$ . The geometry of the above model is illustrated in Fig. 1.

Now suppose that each surface is covered by a diffusing absorbed layer, such that the sum of the concentrations on both surfaces is  $c(\mathbf{r}, t)$  at the time t in the neighborhood of  $\mathbf{r}$ . Now consider the conduction of current between the surfaces. Let us assume that the conductance per unit area (of mid-plane) is a function of the separation h of the surfaces and the total concentration c of absorbate near the point in question, i.e., F(h, c). The total conductance will be the sum of the conductances through each element of area: hence, the instantaneous resistance R(t) at time t will be given by

$$1/R(t) = \int_{A_2} F(h(r), c(r, t)) dr$$
 (5.9)

where  $d\mathbf{r}$  is the differential area on the mid-plane and the integration extends over the rectangle  $A_2 = L_1 \times L_2$ . Behind the above statements lies the tacit assumption that the radii of curvature of the rough surfaces are generally considerably larger than the values of h. However, we will not explicitly concern ourselves with this implied restriction.

At this point it is expedient to imagine that we have an ensemble of contacts identical in all respects except for different variations of the separation h(r). If we have any function of h,  $\psi(h)$  say, which we wish to average with respect to the variations of h, we simply average the function over the above ensemble giving a result which we denote by  $\overline{\psi(h)}^{(e)}$ .

Now let us write

$$h(\mathbf{r}) = \bar{h}^{(e)} + h_1(\mathbf{r}),$$
 (5.10)

and, as before,

$$R(t) = \bar{R} + R_1(t),$$

$$c(r, t) = \bar{c} + c_1(r, t).$$
(5.11)

We assume that the ensemble average  $\bar{h}^{(e)}$  and the time averages  $\bar{R}$  and  $\bar{c}$  are constants independent of r and t. Let us also assume that the integrals

of  $h_1(r)$  and  $c_1(r, t)$  over  $A_2$  vanish. Inserting (5.10) and (5.11) into (5.9) and expanding, we get

$$1/\bar{R} - R_1(t)/\bar{R}^2 + \cdots = A_2 F(\bar{h}^{(\epsilon)}, \bar{c}) + \left(\frac{\partial^2 F}{\partial h \partial c}\right)^{\circ} \int_{A_2} h_1(\mathbf{r}) c_1(\mathbf{r}, t) d\mathbf{r} + \frac{1}{2} \left(\frac{\partial^2 F}{\partial h^2}\right)^{\circ} \int_{A_2} h_1^2(\mathbf{r}) d\mathbf{r} + \cdots,$$
(5.12)

where the super zero on the derivatives indicates that they are evaluated at  $h = h^{(e)}$  and  $c = \bar{c}$ . In accordance with previous approximations in this memorandum we neglect<sup>24</sup> terms of the order of  $c_1^2$  and  $R_1^2$ . We also neglect terms of the order of  $h_1^2$ . After taking the time average of (5.12) and subtracting the result from (5.12) we get

$$R_{1}(t) = \int_{A_{2}} f(\mathbf{r})c_{1}(\mathbf{r}, t) d\mathbf{r},$$

$$f(\mathbf{r}) = \alpha \bar{R}^{2} h_{1}(\mathbf{r}),$$

$$\alpha = -\left(\frac{\partial^{2} F}{\partial h \partial c}\right)^{\circ}$$
(5.13)

Thus we now have a special case of our general mathematical model for the number of dimensions  $\nu=2$ , provided that we assume that the total concentration c on both of the rough surfaces fluctuates in the same manner as the concentration of a single adsorbed layer confined to a plane rectangular surface. The spectral density  $S(\omega)$  of  $R_1(t)$  is then given by Eq. (3.17) which we repeat here

$$S(\omega) = \frac{2}{\pi} \cdot \frac{\chi A_2 D}{s''} \cdot \frac{\Sigma'}{k} \frac{k^2 |f_k|^2}{\omega^2 + D^2 k^4}$$
 (5.14)

where k is a two-dimensional vector whose components take the values  $k_i = 2\pi n_i/L_i$ ,  $n_i = 0, \pm 1, \pm 2, \cdots$ , and where  $f_k$  are the Fourier space-amplitudes of f(r) given by

$$f_{\mathbf{k}} = A_2^{-1} \int_{A_2} f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r}.$$

It may be appropriate at this point to consider the quantity s'' in detail for this particular case. If the energy e per unit area is independent of c, we have  $s'' = -\frac{\partial^2 s}{\partial c^2}$  evaluated at  $c = \bar{c}$  where s is here the entropy of the absorbate per unit area. For the sake of illustration let us consider a single layer of absorbate in which the molecules are non-interacting. If c, the sur-

<sup>24</sup> We neglect these terms not because they are small compared with  $c_1$  or  $h_1c_1$  but, because they are non-fluctuating (in time), are hence to be compared with  $1/\overline{R}$ .

face concentration of the absorbate, be measured in molecules (atoms, or ions) per unit area, then, for the ideal system above, it follows that  $s = -\chi$   $c \log c$  and finally that  $s'' = \chi/\bar{c}$ . However, in the mechanism discussed in this part we have a compound system consisting of two separate layers on the upper and lower surfaces respectively. Nevertheless, a detailed analysis reveals that with c equal to the sum of the concentrations of both layers we still have  $s'' = \chi/\bar{c}$  even though s itself is no longer given by an expression the same as that above. In conclusion let us consider the factor  $\chi/s''$  in Eq. (5.14). This factor is under the above idealization simply equal to  $\bar{c}$ . That is, the spectral density  $S(\omega)$  is directly proportional to the average concentration of absorbed molecules, meaning simply that each molecule makes its contribution to the resistance fluctuations independently of the others. Of course, in any real system this will not be quite true; however, the existence of interactions will be manifested only by making  $\chi/s''$  not equal to  $\bar{c}$  in Eq. (5.14).

The results quoted thus far apply to a system with a given h(r). Now we shall average the right-hand side of Eq. (5.14) over the ensemble of variations of h(r), it being supposed that  $S(\omega)$  itself on the left-hand side will be negligibly affected by this operation. This amounts to replacing  $|f_k|^2$  by  $\overline{|f_k|^2}^{(e)}$ . We then have

$$|f_k|^{2^{(e)}} = \alpha^2 \bar{R}^4 |h_k|^{2^{(e)}}$$
 (5.15)

where  $h_k$  are the Fourier space-amplitudes of  $h_1(r)$ .

We now consider more closely the problem of calculating  $h_k|^2$ . We want to assume that  $h_1(r)$  is a more or less random function of r. If  $h_1(r)$  were a random function of r in the same way that the thermal noise voltage is a random function of the time t, then  $h_1(r)|_{h_k}|^2$  would be a constant independent of r and the self-covariance  $h_1(r)h_1(r')$  would vanish for  $r \neq r'$ . This clearly cannot be so, since the function  $h_1(r)$  with such statistical properties would represent a highly discontinuous type of surface incapable of physical existence. We then fall back upon the more reasonable assumption that the gradient of  $h_1$  possesses statistical properties of the above type. This notion is precisely formulated by means of the following equations:

$$\vec{\nabla} h_1(\mathbf{r}) = \mathbf{p}(\mathbf{r}) \tag{5.16}$$

where

$$\int_{A} p(\mathbf{r}) d\mathbf{r} = 0, \qquad (5.17)$$

and

$$\overline{p(r)p(r')}^{(e)} = \beta \underbrace{1}_{\sim} \delta(r - r'). \tag{5.18}$$

In Eq. (5.18)  $\beta$  is a parameter (with the dimensions of area) characterizing the amplitude of the surface roughness, and 1 is the unit tensor in two dimensions. Expressing (5.16) in terms of the Fourier space-amplitudes  $h_k$  and  $p_k$  of  $h_1$  and p respectively, we have

$$-ik h_k = p_k, (5.19)$$

giving finally

$$\overline{|h_{\mathbf{k}}|^2}^{(e)} = \mathbf{k} \cdot \overline{p_{\mathbf{k}} p_{\mathbf{k}}^*}^{(e)} \cdot \mathbf{k} / k^4$$
 (5.20)

Expressing (5.18) in terms of Fourier space-amplitudes we get

$$\overline{p_k b_{k'}^*}^{(e)} = \beta A_2^{-1} \underbrace{1}_{kk'}^{\delta_{kk'}}, \tag{5.21}$$

which, when inserted into (5.20) gives the following desired result:

$$\overline{|h_{k}|^{2}}^{(e)} = \beta A_{2}^{-1} k^{-2}. \tag{5.22}$$

Now replacing  $|f_k|^2$  by  $|f_k|^2$  in Eq. (5.14) and substituting the expression (5.22) with the use of Eq. (5.15), we obtain

$$S(\omega) = \frac{2}{\pi} \cdot \frac{\chi D}{s''} \cdot \beta \alpha^2 \overline{k}^4 \sum_{k} \frac{1}{\omega^2 + D^2 k^4}$$
 (5.23)

If the frequencies of interest are larger than a certain characteristic frequency  $\omega_0 = 4\pi^2 \ D/L^2$  where L is the smaller of  $L_1$  and  $L_2$ , the summation in (5.23) may be replaced by an integration giving finally

$$S(\omega) = \chi D/\pi^2 s'' \cdot \alpha^2 \beta \bar{R}^4 A_2 \cdot \int_0^\infty \frac{k \ dk}{\omega^2 + D^2 k^4}$$

$$= \chi/4\pi s'' \cdot \beta \alpha^2 \bar{R}^4 A_2 \cdot 1/\omega$$
(5.24)

This result is in agreement with experiment in most respects. The dependence on frequency is, of course, that experimentally observed by all investigators. The non-dependence on the voltage applied across the contact is implied by the basic assumptions common to all of the mechanisms considered here, and is in approximate agreement with the results of Christensen and Pearson (see Eq. (1.2)). For our result to agree with the results of CP as regards the dependence on the average resistance<sup>25</sup>  $\bar{R}$ , the factor  $c^2\bar{R}^4$   $A_2$  must be proportional to  $\bar{R}^{2+b}$  where  $b \sim 1.25$ . These authors also imply that some of

 $<sup>^{25}</sup>$  It must be remembered that the resistance  $\bar{R}$  in the CP formula is the total contact resistance equal to sum of the gap resistance and the spreading resistance, whereas the  $\bar{R}$  in our theory evidently should be considered the gap resistance. For the purposes of comparison we make the ad hoc assumption that the gap resistance is proportional to the total contact resistance.

the parameters necessary to complete the description of a contact between given substances at a given temperature show up implicitly only through  $\bar{R}$ . According to our theory the factor  $\alpha^2 \bar{R}^4 A_2$  does not depend in any unique way upon  $\bar{R}$ ; it matters by what means  $\bar{R}$  is varied. If the resistance  $\bar{R}$  is changed by altering the contact area  $A_2$ , keeping other parameters fixed, we would find that  $\bar{R}A_2$  is constant so that the factor in question would be proportional to  $\bar{R}^3$ , that is, b=1. However, if  $\bar{R}$  is changed by varying the contact pressure, the effect would show up through the factor  $\alpha^2$ , ( $\beta$  also, to some extent, perhaps) and, since one would expect  $\alpha$  to increase somewhat with pressure whereas  $\bar{R}$  decreases with pressure, the factor of interest would probably depend upon some power of  $\bar{R}$  between 3 and 4, that is, 1 < b < 2.

The theory formulated here suffers from the difficulty that the integral of the power spectrum with respect to frequency is logarithmically divergent at 0 and  $\infty$ , that is

$$\int_{\omega_1}^{\omega_2} S(\omega) \ d\omega \doteq \int_{\omega_1}^{\omega_2} d\omega \ \omega = \log \left( \omega_2 / \omega_1 \right) \to \infty \text{ as } \omega_1 \to 0 \text{ and } \omega_2 \to \infty.$$

The divergence at  $\infty$  does not bother us as much as the divergence at 0 since, with only a divergence at  $\infty$ , the self-covariance  $\overline{R_1(t)R_1(t+u)}$  exists for all non-vanishing values of u; whereas, with a singularity at 0, the self-covariance does not exist for any value of u. For this reason we cannot consider the self-covariance here. In Part c of this Section we consider a possible way of removing the divergence at 0, and consequently, then, we are able to calculate the self-covariance for non-vanishing values of u.

c. Refinement of the Theory of Part b. Here we propose a simple modification of the model of Part b, removing the divergence of the integral of  $S(\omega)$  at  $\omega = 0$ . The modification considered here, although it is one of several possibilities any one of which is sufficient for removing the divergence (See Section 6.), is perhaps the only one that is sufficiently simple to treat in a memorandum of this scope. The results of this section are thus intended to be only provisional and suggestive.

Let us reconsider the statistics of the function h(r) giving the separation between the surfaces near a point r on the mid-plane. The distribution of h's considered in the last section is open to several criticisms: (1) it possesses no characteristic length parallel to the mid-plane; and (2) the self-covariance  $\overline{h_1(r)h_1(r')}^{(e)}$  does not exist for any value of r-r'.

To correct partially for these difficulties we replace Eq. (5.22) by

$$\overline{|h_k|^2}^{(e)} = \frac{\beta \ell^2}{A_2(1 + \ell^2 k^2)}, \qquad (5.25)$$

where  $\ell$  is a new characteristic length. The self-covariance  $\overline{h_1(r)h_1(r')}^{(e)}$  based upon (5.25) now exists for all values of r - r' except 0. Thus we still

have the objection that the variance  $\overline{h_1^2}^{(e)}$  is infinite; however, this will cause us no trouble.

With Eq. (5.25) instead of (5.22) the spectral of density  $R_1$  takes the form

$$S(\omega) = \chi D/\pi^{2} s'' \cdot \beta \alpha^{2} \, \overline{R}^{4} A_{2} \cdot \int_{0}^{\infty} \frac{\ell^{2} \, k^{2}}{1 + \ell^{2} \, k^{2}} \cdot \frac{k \, dk}{\omega^{2} + D^{2} \, k^{4}}$$

$$= (\chi/4\pi s'') \cdot \beta \alpha^{2} \, \overline{R}^{4} A_{2} \cdot 1/\omega \cdot Q(y),$$

$$Q(y) = \frac{y \left(y - \frac{2}{\pi} \log y\right)}{1 + y^{2}}, \quad y = \ell^{2} \omega/D.$$
(5.26)

In obtaining the above equation we have made the usual assumption that the frequencies of interest are larger than  $\omega_0 = 4\pi^2 D/L^2$ , and have replaced the original sum by an integral. The function Q(y) has the following properties:

$$Q(y) \simeq -\frac{2}{\pi} y \log y \quad \text{for} \quad y \ll 1$$

$$Q(y) \simeq 1 \quad \text{for} \quad y \gg 1$$
(5.27)

Hence for  $\omega \ll D/\ell^2$ ,  $S(\omega) \propto \log \omega$ , the integral of which converges as  $\omega \to 0$ ; whereas, for  $\omega \gg D/\ell^2$ ,  $S(\omega)$  differs negligibly from that given by the unrefined theory (Eq. (5.24)).

The self-covariance  $\overline{R_1(t)} R_1(t+u)$  now exists for all non-vanishing u and is given by

$$\overline{R_{1}(t)R_{1}(t+u)} = (\chi/2\pi s'') \cdot \beta \alpha^{2} \, \overline{R}^{4} A_{2} \cdot \int_{0}^{\infty} \frac{\ell^{2} e^{-Duk^{2}kdk}}{1+\ell^{2} \, k^{2}} \\
= (\chi/4\pi s'') \cdot \beta \alpha^{2} \, \overline{R}^{4} A_{2} \cdot e^{Du/\ell^{2}} [-Ei(-Du/\ell^{2})] \qquad (5.28)$$

where

$$-Ei(-x) = \int_{x}^{\infty} e^{-v} dv/v,$$

$$\simeq -\log \gamma x \text{ for } x \ll 1,$$

$$\simeq \frac{e^{-x}}{x} \text{ for } x \gg 1,$$

$$\gamma = 0.5772.$$

Thus for  $u \ll \ell^2/D$ ,  $\overline{R_1(t)R_1(t+u)} \propto -\log(\gamma Du/\ell^2)$  and for  $u \gg \ell^2/D$ ,  $\overline{R_1(t)R_1(t+u)} \propto 1/u$ .

Thus we have illustrated how one modification of the model has removed the divergence at  $\omega = 0$ .

It appears from the treatment here and in part b that roughness and diffusion in two dimensions are essential (at least in a linear treatment) features in obtaining  $S(\omega)\alpha 1/\omega$ . In the case of a non-linear coupling (to be considered in a later paper) a "self-induced" roughness effect may occur without introducing roughness ab initio as an intrinsic feature of the model.

#### 6. Summary

(a) If the resistance deviation  $R_1(t)$  is related to the concentration deviation  $c_1(r, t)$  of a diffusing medium (particles or heat) by the *linear* functional

$$R_1(t) = \int_{A_r} f(r)c_1(r, t) dr, \qquad (6.1)$$

where r is a vector and dr a volume element in a  $\nu$ -dimensional space of volume  $A_{\nu}$ , then the spectral density  $S(\omega)$  of  $R_1(t)$  is

$$S(\omega) = \frac{2}{\pi} \frac{\chi A_{\nu} D}{s''} \frac{k^2 |f_{\mathbf{k}}|^2}{\omega^2 + D^2 k^4}, \tag{6.2}$$

where D is the diffusion constant, s'' is defined by Eq. (3.14),  $\chi$  is the Boltzmann constant,  $\omega$  is the frequency (in radians per sec.), k is the wave number vector in  $\nu$ -dimensional k-space limited to a discrete lattice of points (defined by Eq. (3.2)) over which the summation is taken, and  $f_k$  is the kth Fourier component of f(r) (Eq. (3.3)).

(a) If the important terms in (6.2) vary slowly between lattice points in k-space (true if  $\omega > \omega_0$  given by Eq. (3.18)), then (6.1) can be replaced by the integral

$$S(\omega) = 2^{\nu+1} \pi^{\nu-1} \frac{\chi D}{s''} \int \frac{|f(\mathbf{k})|^2 k^2 d\mathbf{k}}{\omega^2 + D^2 k^4}, \qquad (6.3)$$

where the integration extends over the entire k-space and where f(k) is given by (Eq. 3.20)

$$f(\mathbf{k}) = \frac{1}{(2\pi)^{\nu}} \int_{A_{\nu}} f(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r}. \tag{6.4}$$

(b) Let  $\omega'$  be a frequency in the middle of a wide range. Suppose  $|f(\mathbf{k})|^2$  averaged over the total solid angle in  $\nu$ -dimensional  $\mathbf{k}$ -space is proportional to  $\mathbf{k}^{2n}$ , where n is an integer, in a wide range of k with  $k = \sqrt{\omega'/D}$  in its middle. It follows then that  $S(\omega) \propto D^{-n-\nu/2} \omega^{-1+n+\nu/2}$  as long as  $-1 < 2n + \nu + 1 < 3$ . As a consequence, we see that with n an integer (as is true for the simple cases considered in Section 5)  $\nu$  must be 2—the only even di-

mensionality—in order that  $S(\omega)$  be inversely proportional to  $\omega$  in agreement with experiment. In this case the only allowed value of n is -1.

- (c) From (b) we have the interesting result that  $S(\omega)$  is independent of D when it is inversely proportional to  $\omega$ . This means that very slowly diffusing substances can contribute as much to contact noise as rapidly diffusing substances. This result can be derived on quite dimensional grounds and is not dependent upon the special assumptions underlying our treatment.
- (d) A system comprising a high resistance layer modulated by the three-dimensional diffusion of particles or heat gives  $S(\omega) \propto \omega^{-1/2}$ . See Case a.(i) in Section 5.
- (e) In a system composed of a localized contact disturbed by a diffusing surface layer (See Case a.(ii), Section 5), the self-covariance  $R_1(t)R_1(t+u)$  is inversely proportional to  $\Delta + Du$  where  $\Delta$  may be considered the contact area. We have  $S(\omega) \propto -\log a + \text{const.}$  for  $\omega \ll D/\Delta$  and  $S(\omega) \propto \omega^{-2}$  for  $\omega \gg D/\Delta$ .
- (f) In a system involving the contact between relatively large areas of rough surfaces covered with diffusing surface layers (Cases b. and c., Section 5), we have been successful in obtaining  $S(\omega) \propto \omega^{-1}$ , and also in obtaining a reasonable dependence upon the average resistance.

#### APPENDIX I

# SPECTRAL DENSITY AND THE SELF-COVARIANCE

Here we consider in detail the spectral density, the self-covariance, and the relation between these two quantities, first for the case of a single random variable. The treatment is subsequently extended to the case of a set of random variables which necessitates the consideration of the spectral density matrix and the covariance matrix.

Let y(t) be a real random variable whose time average vanishes, y(t) = 0. Now the m.s. value of y can be defined

$$\overline{y^2(t)} = \lim_{\tau \to \infty} \frac{1}{\tau} \int_{-\infty}^{+\infty} y^2(t, \tau) dt$$
 (I-1)

where  $y(t, \tau) = y(t)$  in the interval  $-\frac{\tau}{2} \le t < \frac{\tau}{2}$  and vanishes outside this interval. Evidently  $y(t, \tau)$  can be expressed by the Fourier integral

$$y(t, \tau) = \int_{-\infty}^{+\infty} z(\omega, \tau) e^{i\omega t} d\omega$$
 (I-2)

where

$$z(\omega, \tau) = \frac{1}{2\pi} \int_{-(\tau/2)}^{+(\tau/2)} y(t)e^{-i\omega t} dt,$$

By Parseval's theorem we obtain

$$\int_{-\infty}^{+\infty} y^2(t,\tau) dt = 2\pi \int_{-\infty}^{+\infty} |y(\omega,\tau)|^2 d\omega,$$

which, when combined with (I-1), gives finally the desired result (using the fact that  $|y(\omega, \tau)|^2$  is an even function of  $\omega$ )

$$\overline{y^2(t)} = \int_0^\infty Y(\omega) \ d\omega \tag{I-3}$$

where

$$Y(\omega) = 4\pi \lim_{\tau \to \infty} \frac{1}{\tau} |y(\omega, \tau)|^2$$
 (I-4)

is the spectral density.

By a procedure not very different from the preceding, one can show that

$$\overline{y(t)y(t+u)} = \int_0^\infty Y(\omega) \cos \omega u \, d\omega, \tag{I-5}$$

$$Y(\omega) = \frac{2}{\pi} \int \overline{y(t)y(t+u)} \cos \omega u \, du. \tag{I-6}$$

The quantity  $\overline{(y(t)y(t+u))}$  is called the self-covariance.

Now let us suppose that we have a set of random variables  $y_i(t)$  which are in general complex and whose time averages vanish. We are then led to consider, instead of (I-3), relations of the form

$$\overline{y_{i}(l)y_{i}^{*}(l)} = \int_{0}^{\infty} Y_{ij}(\omega) \ d\omega$$
 (I-7)

where now

$$Y_{ij}(\omega) = 2\pi \lim_{\tau \to \infty} \frac{1}{\tau} \left[ y_i(\omega, \tau) y_j^*(\omega, \tau) + y_i(-\omega, \tau) y_j^*(-\omega, \tau) \right] \quad \text{(I-8)}$$

in which

$$y_i(\omega, \tau) = \frac{1}{2\pi} \int_{-(\tau/2)}^{+(\tau/2)} y_i(t) e^{i\omega t} dt.$$

Instead of self-covariances like y(t)y(t+u) we have to consider a covariance matrix of the form  $y_i(t)y_j^*(t+u)$ . Since we shall not have occasion in this paper to consider the relation between the spectral density matrix and the covariance matrix we will not consider the derivation of the analogue of Eq. (I-5).

#### APPENDIX II

## THERMODYNAMIC THEORY OF FLUCTUATIONS

The value of the quantity  $c_1(r, t)c_1(r^1, t)$  or  $(c_k(t)c_{k^1}(t))$  is determined from equilibrium considerations. Before going into the above continuum problem let us first consider the problem for the case of a system described by a finite set of variables. More specifically let us suppose that the state of the system subject to certain restraints (i.e. fixed total mass and energy) is described by the set of variables  $x_1, \dots, x_n$ . Let the equilibrium state be given by the values  $x_1^0, \dots, x_n^0$ , and let

$$x_i = x_i^0 + \alpha_i. (II-1)$$

If the system is constrained to constant average energy E, the entropy of the non-equilibrium state  $S = S^0 + \Delta S$  will be less than  $S^0$ , the entropy of the equilibrium state, by an amount

$$\Delta S = -\frac{1}{2} \sum_{ij} S_{ij} \alpha_i \alpha_j, \qquad (\text{II-2})$$

where

$$S_{ij} = -\left(\frac{\partial^2 S}{\partial x_i \partial x_j}\right)_{x_k = x_k^0} + \frac{1}{T^0} \left(\frac{\partial^2 E}{\partial x_i \partial x_j}\right)_{x_k = x_k^0}$$

Obviously,  $\Delta S$  must be the negative of a positive definite quadratic form, otherwise the equilibrium state would not be a state of maximum entropy. The probability distribution<sup>26</sup> for the  $\alpha$ 's is given by

$$P(\alpha_1, \cdots, \alpha_n) = Ne^{\Delta S/\chi}$$
 (II-3)

where N is a normalization factor and  $\chi$  is the Boltzmann constant. Averaging the products  $\alpha_i \alpha_i$  we find that

$$\sum_{i} S_{ij} \overline{\alpha_{i} \alpha_{k}} = \chi \delta_{ik}. \tag{II-4}$$

Multiplying (II-4) by the arbitrary set  $\gamma_i$  and summing over i we get

$$\sum_{ij} \gamma_i S_{ij} \overline{\alpha_j \alpha_k} = \chi \gamma_k.$$
 (II-5)

The generalization to a system described by a continuous set of variables is not difficult on the basis of (II-5). Now suppose that, in a  $\nu$ -dimensional space  $A_{\nu}$ , we have a system whose state at time t is defined by the continuous set of values of the variable  $c(\mathbf{r}, t) = \bar{c} + c_1(\mathbf{r}, t)$ ; we have

$$\Delta S = -\frac{1}{2} \int_{A_{r}} s'' c_{1}^{2}(\mathbf{r}, t) d\mathbf{r}$$
 (II-6)

<sup>26</sup> H. B. G. Casimir, Rev. Mod. Phys. 17, Nos. 1 and 3, 343-4 (1945).

where

$$s^{\prime\prime} = -\left(\frac{\partial^2 s}{\partial c^2}\right)_{c=\tilde{c}} + \frac{1}{T^0} \left(\frac{\partial^2 e}{\partial c^2}\right)_{c=\tilde{c}}$$

when s and e are the entropy and energy, respectively, per unit volume (of  $\nu$ -dimensional space). In calculating (II-6) it was assumed that

$$\int_{A_{\nu}} c_1(\mathbf{r}, t) d\mathbf{r} = 0,$$

expressing the fact that the system is closed. In order to put (II-6) into a form strictly analogous to (II-2) we write it

$$\Delta S = -\frac{1}{2} \int_{A_{r}} \int_{A_{r}} s'' \, \delta(\mathbf{r} - \mathbf{r}') c_{1}(\mathbf{r}, t) c_{1}(\mathbf{r}', t) \, d\mathbf{r} \, d\mathbf{r}'. \quad (II-7)$$

We see that the equation analogous to Eq. (II-5) must be

$$\int_{A_r} \int_{A_r} \gamma(\mathbf{r}') s'' \, \delta(\mathbf{r} - \mathbf{r}'') \overline{c_1(\mathbf{r}'', t) c_1(\mathbf{r}, t)} \, d\mathbf{r} \, d\mathbf{r}' = \chi \gamma(\mathbf{r}) \quad \text{(II-8)}$$

where  $\gamma(r)$  is an arbitrary function. Integrating (II-8) with respect to r'' and using the fact that the delta function is defined by

$$\int \gamma(\mathbf{r}')\delta(\mathbf{r}' - \mathbf{r}) d\mathbf{r}' = \gamma(\mathbf{r})$$

we readily arrive at the result

$$\overline{c_1(r,t)c_1(r',t)} = \frac{\chi}{\varsigma''} \delta(r-r'). \tag{II-9}$$

Using the Fourier space-expansions of  $C_1$  and  $\delta(r)$ 

$$c_1(\mathbf{r}, t) = \sum_{\mathbf{k}}' c_{\mathbf{k}}(t) e^{i\mathbf{k}\cdot\mathbf{r}},$$

$$\delta(\mathbf{r}) = \frac{1}{A_{\nu}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}},$$

in the region  $A_{\nu} = L_1 \times \cdots \times L_{\nu}$  with  $k_i = 2\pi n_i/L_i$ , we can write (II-9) over into the equivalent expression:

$$\overline{c_{\mathbf{k}}(t)c_{\mathbf{k}}^{*}(t)} = \frac{\chi}{A_{\nu}s^{\prime\prime}} \,\delta_{\mathbf{k}\mathbf{k}^{\prime}} \,, \tag{11-10}$$

where

$$\delta_{kk'} = \begin{cases} 1 & \text{if } k = k', \\ 0 & \text{otherwise.} \end{cases}$$