

Intensity Fluctuations of the Radiation from a Dispersive Blackbody

R. K. OSBORN AND A. Z. AKCASU

The University of Michigan, Department of Nuclear Engineering, Ann Arbor, Michigan

(Received 25 April 1967; in final form 26 June 1967)

Statistical properties of the output fluctuations of a photon detector measuring the intensity of radiation at a given frequency emitted by a dispersive blackbody are investigated using Langevin's technique. The variance-to-mean ratio of the accumulated counts, and the power spectral density of the count rate are obtained in terms of properties of the emitting medium. The possibility and limitations of obtaining information about the emitter by measuring these quantities are discussed. It is found in particular that the temperature of the emitting medium can be determined in principle by observing the intensity fluctuations of the radiation at a single frequency. The photon fluctuations in a microwave cavity are also discussed in the framework of the present formalism and compared to previous work.

1. INTRODUCTION

The purpose of this paper is to investigate the intensity fluctuations in radiation emitted by a dispersive blackbody. The experiment which we intend to analyze is illustrated in Fig. 1. The photons from the emitter are detected by a photon detector, e.g., photocathode, whose output, $Z(t)$, is assumed to be in the form of an electric voltage, and proportional to the number of photons absorbed per second in its active volume. Hence, $Z(t)$ can be identified as the "instantaneous count rate." The detector is visualized as a uniform absorbing medium described by the microscopic absorption rate per photon, $r_D(\mathbf{k})$, for photons with wavevector \mathbf{k} .

The emitter is assumed to be a homogeneous finite medium in which the atoms are in thermal equilibrium at a temperature, θ (units of energy). The medium is characterized by $\alpha(\mathbf{k})$ and $\epsilon(\mathbf{k})$ which are the absorption and emission rates, respectively, of photons with wavevector, \mathbf{k} . The scattering of photons in the medium is neglected. The medium is allowed to be dispersive with a photon speed $v(\mathbf{k})$ which is different than the speed, c , in vacuum.

Let $N(\mathbf{x}, \mathbf{k}, t)$ denote the instantaneous number of randomly polarized photons at time t per unit volume about \mathbf{x} in configuration space, and per unit volume about \mathbf{k} in wavevector space. We shall denote a point in the six-dimensional space by $\mathbf{r} = (\mathbf{x}, \mathbf{k})$ to compress the notation. The instantaneous value of the photon density $N(\mathbf{r}, t)$ is a fluctuating function of time with a stationary mean value $\langle N(\mathbf{r}) \rangle$. These fluctuations are due to the statistical nature of the absorption and emission processes. We shall denote the fluctuating part of $N(\mathbf{r}, t)$ by $n(\mathbf{r}, t)$, i.e., $n(\mathbf{r}, t) \equiv N(\mathbf{r}, t) - \langle N(\mathbf{r}) \rangle$. The aim of this paper will be to investigate some of the statistical properties of $n(\mathbf{r}, t)$ in terms of the observed fluctuations $z(t)$ in the count rate (or absorption rate) of the detector, i.e., $z(t) = Z(t) - \langle Z(t) \rangle$. In particular we shall consider the autocorrelation functions and the power spectral density associated with $z(t)$ which are defined by

$$\Phi_z(\tau) \equiv \langle z(t)z(t+\tau) \rangle, \tag{1}$$

and

$$G_z(\omega) \equiv \int_{-\infty}^{+\infty} \exp(-i\omega\tau) \Phi_z(\tau) d\tau. \tag{2}$$

We shall also discuss the variance-to-mean ratio of the accumulated counts,

$$C(T) = \int_0^T z(t) dt, \tag{3}$$

as a function of the gate time, T , which is defined as¹

$$\begin{aligned} \eta(T) &\equiv \langle C^2(T) \rangle / T \langle Z \rangle \\ &= 2 / \langle Z \rangle \int_0^T (1-\tau/T) \Phi_z(\tau) d\tau. \end{aligned} \tag{4}$$

The interest in these particular statistical quantities stems from the fact that they are the quantities which are usually measured in fluctuation experiments.

The fluctuations in the count rate can be expressed as

$$z(t) = \int_D d^3r z(\mathbf{r}, t), \tag{5}$$

where $z(\mathbf{r}, t)$ is the fluctuating part of the "count-rate density" at \mathbf{r} in the phase space, i.e., $z(\mathbf{r}, t) = r_D n(\mathbf{r}, t) + s^D(\mathbf{r}, t)$, where s^D is an appropriate noise source associated with the removal of photons by the detection process. The autocorrelation function $\Phi_z(\tau)$ can be expressed in terms of $z(\mathbf{r}, t)$ as

$$\Phi_z(\tau) = \int_D d^3r \int_D d^3r' \langle z(\mathbf{r}, t) z(\mathbf{r}', t+\tau) \rangle. \tag{6}$$

Both, in Eqs. (5) and (6) the integrations are performed in the active volume of the detector in the phase space. It follows from Eq. (6) that the statistical quantities $\Phi_z(\tau)$, $G_z(\omega)$ and $\eta(T)$ can readily be obtained once the correlation function

$$\phi_z(\mathbf{r}, \mathbf{r}', \tau) \equiv \langle z(\mathbf{r}, t) z(\mathbf{r}', t+\tau) \rangle, \tag{7}$$

¹ Use

$$\int_0^T dt \int_0^T dt' \phi_z(t-t') = \int_{-T}^T (T-|\tau|) \phi_z(\tau) d\tau.$$

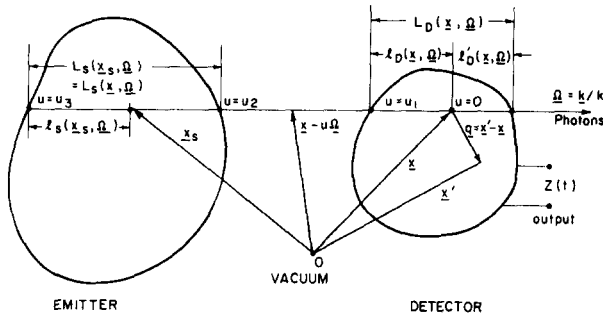


FIG. 1. Intensity-fluctuation experiment.

associated with the count-rate density is determined. Thus, our main task will be to calculate $\phi_z(\mathbf{r}, \mathbf{r}', \tau)$ in terms of the parameters $\alpha(\mathbf{k})$, $\epsilon(\mathbf{k})$, and $r_D(\mathbf{k})$ describing the optical source and the emitter. In order to achieve this, we must first determine the correlation function.

$$\phi_n(\mathbf{r}, \mathbf{r}', \tau) \equiv \langle n(\mathbf{r}, t)n(\mathbf{r}', t+\tau) \rangle, \quad (8)$$

appropriate to the photon-density fluctuations and then use the relations between ϕ_n and ϕ_z as discussed in Sec. 3, by Eq. (31).

2. CORRELATIONS IN PHOTON-DENSITY FLUCTUATIONS

The mean photon density, $\langle N(\mathbf{r}, t) \rangle$, in an inhomogeneous, dispersive medium satisfies, when the photon scattering is neglected, the following approximate transport equation

$$[(\partial/\partial t) + \mathbf{\Omega} \cdot \nabla v(\mathbf{r}) + \sigma(\mathbf{r})] \langle N(\mathbf{r}, t) \rangle = \epsilon(\mathbf{r})\rho(\mathbf{k}), \quad (9)$$

where $\sigma(\mathbf{r}) = \alpha(\mathbf{r}) - \epsilon(\mathbf{r})$, $\rho(\mathbf{k}) =$ density of photon states $= 1/4\pi^3$, and $\mathbf{\Omega} = \mathbf{k}/k$. We consider the optical source, vacuum and the detector as a single inhomogeneous medium. Then, both $\alpha(\mathbf{r})$ and $\epsilon(\mathbf{r})$ are zero in the vacuum, and $\alpha(\mathbf{r}) = r_D(\mathbf{k})$ within the detector.

Equation (9) is a simple statement of photon balance. The right-hand side represents the spontaneous emission, the term $\epsilon(\mathbf{r}) \langle N(\mathbf{r}, t) \rangle$ denotes the rate of stimulated emission, and the terms $\mathbf{\Omega} \cdot \nabla v(\mathbf{r}) \langle N(\mathbf{r}, t) \rangle$ and $\alpha(\mathbf{r}) \langle N(\mathbf{r}, t) \rangle$ account for the rates of loss of photons by streaming and absorption. A careful derivation of Eq. (9) can be found elsewhere.²

In a stationary system, as we assume here, the mean photon density is independent of time. Therefore, Eq. (9) reduces to

$$\mathbf{\Omega} \cdot \nabla F(\mathbf{r}) + \Sigma(\mathbf{r})F(\mathbf{r}) = S_0(\mathbf{r}), \quad (10)$$

where we have defined

$$F(\mathbf{r}) = v(\mathbf{r}) \langle N(\mathbf{r}) \rangle, \quad (11a)$$

$$\Sigma(\mathbf{r}) = \sigma(\mathbf{r})/v(\mathbf{r}), \quad (11b)$$

$$S_0(\mathbf{r}) = \epsilon(\mathbf{r})\rho(\mathbf{k}). \quad (11c)$$

Clearly, $F(\mathbf{r})$ is the mean photon flux. Equation (10) can readily be solved by the method of characteristics to obtain

$$F(\mathbf{r}) = \int_0^\infty du S_0(\mathbf{x} - \mathbf{\Omega}u, \mathbf{k}) p(u, \mathbf{r}), \quad (12a)$$

where

$$p(u, \mathbf{r}) \equiv \exp \left[- \int_0^u \Sigma(\mathbf{x} - u'\mathbf{\Omega}, \mathbf{k}) du' \right]. \quad (12b)$$

The photon density $\langle N(\mathbf{r}) \rangle$ in various regions can be obtained from Eq. (12) as:

(a) In the emitter

$$\langle N(\mathbf{r}_S) \rangle = S_0(\mathbf{k}) \{ 1 - \exp[-\Sigma(\mathbf{k})l_S(\mathbf{x}_S, \mathbf{\Omega})]/\sigma(\mathbf{k}) \}. \quad (13)$$

(b) In the detector

$$\langle N(\mathbf{r}) \rangle = S_0(\mathbf{k}) \frac{v(\mathbf{k})}{c} \frac{1 - \exp[-\Sigma(\mathbf{k})L_S(\mathbf{x}_S, \mathbf{\Omega})]}{\sigma(\mathbf{k})} \times \exp[-\Sigma_D(\mathbf{k})l_D(\mathbf{x}, \mathbf{\Omega})], \quad (14)$$

where the distances $l_S(\mathbf{x}_S, \mathbf{\Omega})$, $L_S(\mathbf{x}, \mathbf{\Omega})$ and $l_D(\mathbf{x}, \mathbf{\Omega})$, as well as $L_D(\mathbf{x}, \mathbf{\Omega})$ which will be used later, are indicated in Fig. 1.

We shall now investigate the fluctuations, $n(\mathbf{r}, t)$, in the photon density by means of Langevin's technique. The discussion of this technique as a method for investigating fluctuation phenomena in other physical systems can be found elsewhere,³ and will not be reproduced here in detail. This approach, which is somewhat phenomenological, is adopted in this work for its simplicity as opposed to the more deductive kinetic approach.⁴ The Langevin technique starts with the "stochastic" equation

$$[(\partial/\partial t) + \mathbf{\Omega} \cdot \nabla v(\mathbf{r}) + \sigma(\mathbf{r})]n(\mathbf{r}, t) = s(\mathbf{r}, t), \quad (15)$$

where $s(\mathbf{r}, t)$ is a random source introduced to account for the fluctuations about the mean photon density. Since $\langle n(\mathbf{r}, t) \rangle = 0$ by definition, one requires that

$$\langle s(\mathbf{r}, t) \rangle = 0. \quad (16)$$

Physically, $s(\mathbf{r}, t)$ represents the natural fluctuations in the rate of absorption, stimulated emission and spontaneous-emission processes. In addition to Eq. (16), one attributes the following statistical property to $s(\mathbf{r}, t)$ as a postulate:

$$\langle s(\mathbf{r}, t')s(\mathbf{r}', t') \rangle = Q_S(\mathbf{r})\delta(\mathbf{r}' - \mathbf{r})\delta(t' - t), \quad (17)$$

where

$$Q_S(\mathbf{r}) = [\alpha(\mathbf{r}) + \epsilon(\mathbf{r})] \langle N(\mathbf{r}) \rangle + S_0(\mathbf{r}). \quad (18)$$

³ A. Z. Akcasu and R. K. Osborn, Nucl. Sci. Eng. **26**, 13 (1966). For a more general discussion see Melvin Lax, Rev. Mod. Phys. **38**, 541 (1966).

⁴ R. K. Osborn and M. Natelson, J. Nucl. Energy, Part A/B, **19**, 916 (1965).

² E. H. Klevans, Ph.D. thesis, University of Michigan (1962).

Equation (17) implies that the fluctuations in the noise-equivalent source are uncorrelated in phase space and in time. Equation (18) is based on the assumptions that (a) absorption, stimulated emission, and spontaneous emission of photons are statistically independent, and (b) each of these processes is described by a Poisson distribution.

The foregoing statistical properties, i.e., Eqs. (16) and (17), are sufficient to investigate the space, time, and energy correlations of the photon-density fluctuations, viz., $\phi_n(\mathbf{r}, \mathbf{r}', \tau) = \langle n(\mathbf{r}, t)n(\mathbf{r}', t+\tau) \rangle$. In order to obtain ϕ_n , we must first solve Eq. (15) for $n(\mathbf{r}, t)$. Defining

$$f(\mathbf{r}, t) \equiv v(\mathbf{r})n(\mathbf{r}, t), \quad (19)$$

and taking the Fourier transform of Eq. (15), one finds

$$\mathbf{\Omega} \cdot \nabla \tilde{f}(\mathbf{r}, i\omega) + [i\omega/v(\mathbf{r}) + \Sigma(\mathbf{r})] \tilde{f}(\mathbf{r}, i\omega) = \tilde{s}(\mathbf{r}, i\omega), \quad (20)$$

where \tilde{f} and \tilde{s} denote the Fourier transforms of the respective time functions. This equation is readily solved in a similar fashion to Eq. (10) to obtain

$$\tilde{f}(\mathbf{r}, i\omega) = \int_0^\infty du \tilde{s}(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}, i\omega) \times \exp\left[-i\omega \int_0^u \frac{du'}{v(\mathbf{x}-u'\mathbf{\Omega}, \mathbf{k})}\right] p(u, \mathbf{r}). \quad (21)$$

The inverse Fourier transform yields

$$n(\mathbf{r}, t) = [v(\mathbf{r})]^{-1} \int_0^\infty du \times s\left[\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}, t - \int_0^u \frac{du'}{v(\mathbf{x}-u'\mathbf{\Omega}, \mathbf{k})}\right] p(u, \mathbf{r}), \quad (22)$$

which is the desired solution of Eq. (15).

We are now in a position to evaluate $\phi_n(\mathbf{r}, \mathbf{r}', t)$ in terms of the assumed statistical properties of $s(\mathbf{r}, t)$. Substituting Eq. (22) into the definition of ϕ_n in Eq. (8), one finds

$$\begin{aligned} \phi_n(\mathbf{r}, \mathbf{r}', \tau) &= \int_0^\infty du \int_0^\infty du' p(u, \mathbf{r}) p(u', \mathbf{r}') \\ &\times Q_S(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}) \delta(\mathbf{k}'-\mathbf{k}) \delta[\mathbf{x}'-\mathbf{x}-u'\mathbf{\Omega}'+u\mathbf{\Omega}] \\ &\times [v(\mathbf{r})v(\mathbf{r}')]^{-1} \delta\left[\tau - \int_0^{u'} \frac{dq}{v(\mathbf{x}'-q\mathbf{\Omega}', \mathbf{k}')} \right. \\ &\quad \left. + \int_0^u \frac{dq}{v(\mathbf{x}-q\mathbf{\Omega}, \mathbf{k})}\right], \quad (23) \end{aligned}$$

where we have used Eqs. (17) and (18) to evaluate $\langle s(\mathbf{r}, t)s(\mathbf{r}', t') \rangle$. This equation can be simplified by making use of $\mathbf{k}'=\mathbf{k}$, $\mathbf{\Omega}'=\mathbf{\Omega}$ and $\mathbf{x}'=\mathbf{x}+(u'-u)\mathbf{\Omega}$ in

appropriate terms:

$$\begin{aligned} \phi_n(\mathbf{r}, \mathbf{r}', \tau) &= \delta(\mathbf{k}'-\mathbf{k}) [v(\mathbf{r})v(\mathbf{r}')]^{-1} \\ &\times \int_0^\infty du Q_S(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}) p^2(u, \mathbf{r}) \int_0^\infty du' \\ &\times \exp\left[-\int_0^{u'-u} \Sigma(\mathbf{x}+q\mathbf{\Omega}, \mathbf{k}) dq\right] \delta[\mathbf{x}'-\mathbf{x}-(u'-u)\mathbf{\Omega}] \\ &\times \delta\left[\tau - \int_0^{u'-u} \frac{dq}{v(\mathbf{x}+q\mathbf{\Omega}, \mathbf{k})}\right]. \quad (24) \end{aligned}$$

We shall now perform the integration on u' assuming that $\tau > 0$. The case of $\tau < 0$ need not be considered separately because the following relation holds

$$\phi_n(\mathbf{r}, \mathbf{r}', -|\tau|) = \phi_n(\mathbf{r}', \mathbf{r}, |\tau|), \quad (25)$$

which can be verified easily using the definition of $\phi_n(\mathbf{r}, \mathbf{r}', \tau)$ in Eq. (8).

The last delta function in Eq. (24) can be replaced by⁵

$$\delta(u'-u-r_0) v[\mathbf{x}+r_0\mathbf{\Omega}, \mathbf{k}], \quad (26)$$

where $r_0=r_0(\mathbf{r})$ is a number obtained by

$$\tau = \int_0^{r_0} dq/v(\mathbf{x}+q\mathbf{\Omega}, \mathbf{k}). \quad (27)$$

In the case of a constant speed, Eq. (27) yields $r_0=v\tau$. The integration on u' can now be performed easily yielding

$$\begin{aligned} \phi_n(\mathbf{r}, \mathbf{r}', \tau) &= \delta(\mathbf{k}'-\mathbf{k}) \delta[\mathbf{x}'-\mathbf{x}-\mathbf{\Omega}r_0] \\ &\times \exp\left[-\int_0^{r_0} \Sigma(\mathbf{x}+\mathbf{\Omega}q, \mathbf{k}) dq\right] v^{-1}(\mathbf{r}) \int_0^\infty du \\ &\quad \times p^2(u, \mathbf{r}) Q_S(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}), \quad (28) \end{aligned}$$

where $Q_S(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k})$ and $p(u, \mathbf{r})$ were defined before in Eqs. (17) and (12b), respectively.

We shall now specialize Eq. (28) to the case of a uniform emitter, vacuum, and detector as indicated in Fig. 1. We shall be interested in the values of $\phi_n(\mathbf{r}, \mathbf{r}', \tau)$ within the detector. One can see that

$$\begin{aligned} Q_S(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}) &= \tau_D(\mathbf{k}) \langle N(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}) \rangle \quad \text{for } 0 < u < u_1 \\ &= [\alpha(\mathbf{k}) + \epsilon(\mathbf{k})] \langle N(\mathbf{x}-u\mathbf{\Omega}, \mathbf{k}) \rangle + S_0(\mathbf{k}) \\ &\quad \text{for } u_2 < u < u_3 \\ &= 0 \quad \text{elsewhere.} \end{aligned}$$

Hence, the integration on u will be performed only in the intervals $(0, u_1)$ and (u_2, u_3) . On the other hand Eq. (27) gives $r_0=\tau c$ because $v(\mathbf{x}+q\mathbf{\Omega}, \mathbf{k})=c$ in the detector and in vacuum.

After some lengthy but elementary calculations, one

⁵ Use $\delta[\phi(x)] = \sum_i [\delta(x-x_i)/|\phi'(x_i)|]$, where the x_i are the roots of $\phi(x)=0$ which are assumed to be simple, and $\phi'=d\phi/dx$.

finds

$$\phi_n(\mathbf{r}, \mathbf{r}', \tau) = \delta(\mathbf{k}' - \mathbf{k}) \delta(\mathbf{x}' - \mathbf{x} - \Omega \tau c) \\ \times \exp[-r_D \tau] \langle N(\mathbf{r}) \rangle [1 + Q(\mathbf{r})] \quad \text{for } \tau > 0, \quad (29a)$$

where

$$Q(\mathbf{r}) \equiv [\epsilon(\mathbf{k}) / \sigma(\mathbf{k})] \exp[-\Sigma_D(\mathbf{k}) l_D(\mathbf{x}, \Omega)] \\ \times \{1 - \exp[-\Sigma(\mathbf{k}) L_S(\mathbf{x}, \Omega)]\}, \quad (29b)$$

and where the distances l_D and L_S are already indicated in Fig. 1.

When $\tau < 0$, one obtains from Eqs. (25) and (29)

$$\phi_n(\mathbf{r}, \mathbf{r}', \tau) = \delta(\mathbf{k}' - \mathbf{k}) \delta(\mathbf{x}' - \mathbf{x} + \Omega |\tau| c) \\ \times \exp[-r_D |\tau|] \langle N(\mathbf{r}') \rangle [1 + Q(\mathbf{r}')] \quad \text{for } \tau < 0. \quad (30)$$

3. FLUCTUATIONS IN THE COUNT RATE

This section is devoted to the calculation of the count-rate correlation function defined previously in Eq. (7). It is shown in Ref. 3 that $\phi_z(\mathbf{r}, \mathbf{r}', \tau)$ can be expressed in terms of $\phi_n(\mathbf{r}, \mathbf{r}', \tau)$ as

$$\phi_z(\mathbf{r}, \mathbf{r}', \tau) = \langle N(\mathbf{r}) \rangle r_D(\mathbf{k}) \delta(\mathbf{r}' - \mathbf{r}) \delta(\tau) \\ + r_D^2(\mathbf{k}) \{ \phi_n(\mathbf{r}, \mathbf{r}', \tau) - \langle N(\mathbf{r}) \rangle g_1(\mathbf{r}, \mathbf{r}', \tau) \} \\ \text{for } \tau > 0, \quad (31)$$

where $g_1(\mathbf{r}, \mathbf{r}', \tau)$ is the Green's function which satisfies the photon transport equation in the detector, i.e.,

$$[(\partial/\partial\tau) + c\Omega' \cdot \nabla_{\mathbf{x}'} + r_D(\mathbf{k})] g_1(\mathbf{r}, \mathbf{r}', \tau) = \delta(\mathbf{r}' - \mathbf{r}) \delta(\tau). \quad (32)$$

Here we have used explicitly the fact that the detector medium is nondispersive and a pure photon absorber.

The solution of Eq. (32) is readily found to be

$$g_1(\mathbf{r}, \mathbf{r}', \tau) = \delta(\mathbf{k}' - \mathbf{k}) \delta(\mathbf{x}' - \mathbf{x} - \Omega \tau c) \exp[-r_D(\mathbf{k}) \tau]. \quad (33)$$

Substituting Eqs. (29a) and (33) into Eq. (31), one obtains

$$\phi_z(\mathbf{r}, \mathbf{r}', \tau) = \langle N(\mathbf{r}) \rangle r_D(\mathbf{k}) \delta(\mathbf{k}' - \mathbf{k}) \{ \delta(\mathbf{x}' - \mathbf{x}) \delta(\tau) \\ + r_D(\mathbf{k}) Q(\mathbf{r}) \delta(\mathbf{x}' - \mathbf{x} - \Omega \tau c) \exp[-r_D \tau] \} \quad \tau > 0. \quad (34)$$

The Autocorrelation Function

The autocorrelation of the detector output, defined by Eq. (6), is now obtained by integrating Eq. (34) on \mathbf{r} and \mathbf{r}' . The result is

$$\Phi_z(\tau) = \int_D d^3 r r_D(\mathbf{k}) \langle N(\mathbf{r}) \rangle \{ \delta(\tau) + r_D(\mathbf{k}) \exp[-|\tau| r_D] \\ \times Q(\mathbf{r}) U[l_D'(\mathbf{x}, \Omega) - |\tau| c] \}, \quad (35)$$

where $U[x]$ is the unit step function arising from⁶

$$\int_V d^3 x' \delta(\mathbf{x}' - \mathbf{x} - \Omega |\tau| c) = \int_V dq \delta(q - |\tau| c) \\ = U[l_D'(\mathbf{x}, \Omega) - |\tau| c].$$

Observe also that we have replaced τ by $|\tau|$ since after the integration over \mathbf{r} and \mathbf{r}' , Eq. (34) becomes an even function of the time lag τ .

THE POWER SPECTRAL DENSITY

Fourier transforming Eq. (35) with respect to τ yields the power spectral density associated with the count rates:

$$G_c(\omega) = \int_D d^3 r r_D(\mathbf{k}) \langle N(\mathbf{r}) \rangle [1 + Q(\mathbf{r}) I(\mathbf{r}, \omega)], \quad (36)$$

where

$$I(\mathbf{r}, \omega) \equiv \int_{-\infty}^{+\infty} d\tau \exp[-r_D(\mathbf{k}) |\tau|] U[l_D'(\mathbf{x}, \Omega) - |\tau| c] \exp(i\omega\tau) r_D(\mathbf{k}) \\ = 2 \frac{\exp[-\Sigma_D l_D'] [(\omega/r_D) \sin(\omega l_D'/c) - \cos(\omega l_D'/c)] + 1}{1 + (\omega/r_D)^2}. \quad (37)$$

The Variance-to-Mean Ratio

The variance-to-mean ratio, $\eta(T)$, was defined in Eq. (4). We first note that the mean count rate is

$$\langle Z \rangle = \int_D d^3 r r_D(\mathbf{k}) \langle N(\mathbf{r}) \rangle. \quad (38)$$

We then perform the integral indicated in the right-hand side of Eq. (4). The final result is found to be

$$\eta(T) = 1 + \left[2 \int_D d^3 r r_D(\mathbf{k}) Q(\mathbf{r}) \langle N(\mathbf{r}) \rangle \mathfrak{J}(\mathbf{k}, T) / \langle Z \rangle \right], \quad (39)$$

where

$$\mathfrak{J}(\mathbf{k}, T) \equiv r_D(\mathbf{k}) \int_0^T (1 - u/T) \exp[-r_D u] \\ \times U[l_D'(\mathbf{x}, \Omega) - uc] \\ = [1 - \exp(-l_D' \Sigma_D)] - (T r_D)^{-1} [1 - (1 + l_D' \Sigma_D) \\ \times \exp(-l_D' \Sigma_D)], \quad Tc > l_D'(\mathbf{x}, \Omega), \quad (40a) \\ = \{1 - [1 - \exp(-r_D T)/r_D T]\}, \\ Tc < l_D'(\mathbf{x}, \Omega). \quad (40b)$$

⁶ Use $\delta(\mathbf{x}' - \mathbf{x} - \Omega |\tau| c) = \delta(q - |\tau| c) \delta(\Omega q - \Omega) / q^2$, where $q = \mathbf{x}' - \mathbf{x}$.

The limit of large gate time, i.e., $Tc \gg L_D(\mathbf{x}, \Omega)$ and $Tr_D \gg 1$, is of particular interest because large gate times are easier to work with experimentally. In this case, the second term in Eq. (40a) can be ignored as compared to the first one, and Eq. (39) reduces to

$$\eta(T \rightarrow \infty) = 1 + \left[2 \int_D d^3r r_D(\mathbf{k}) Q(\mathbf{r}) \langle N(\mathbf{r}) \rangle \times \{1 - \exp[-\Sigma_D l_D'(\mathbf{x}, \Omega)]\} / \langle Z \rangle \right]. \quad (41)$$

We shall now apply these general results to a particular geometry in order to gain more insight into the intensity-fluctuation phenomenon.

4. SLAB GEOMETRY

We consider an experiment indicated in Fig. 2. The source is an infinite slab of thickness a , and the detector is a cylinder of length l , and cross section A . We look at the photons with wavevector, \mathbf{k}_0 , which is parallel to the axis of the detector. The relative positions of the detector and the source are shown in Fig. 2.

The following quantities are needed to calculate the autocorrelation function, power spectral density, and variance-to-mean ratio:

$$L_D = l, \quad l_D' = l - x, \quad l_D = x,$$

$$L_S = a,$$

$$Q(x) = (\epsilon/\sigma) \exp(-\Sigma_D x) [1 - \exp(-\Sigma a)],$$

$$\langle N(x) \rangle = (v/c) (\epsilon\rho/\sigma) [1 - \exp(-\Sigma a)] \exp(-\Sigma_D x),$$

$$\langle Z \rangle = v(\epsilon\rho/\sigma) [1 - \exp(-\Sigma a)] [1 - \exp(-\Sigma_D l)] A.$$

We shall discuss only the power spectral density and the variance-to-mean ratio, because the autocorrelation function is usually used only to obtain the power spectral density. Equations (36) and (41) reduce in the present case to

$$G_z(\omega) = \langle Z \rangle \left\{ 1 + \epsilon/\sigma \frac{[1 - \exp(-\Sigma a)]}{1 + (\omega^2/r_D^2)} \times \frac{1 + \exp(-2\Sigma_D l) - 2 \exp(-\Sigma_D l) \cos(l\omega/c)}{1 - \exp(-\Sigma_D l)} \right\}, \quad (42)$$

and

$$\eta(T \rightarrow \infty) = 1 + (\epsilon/\sigma) [1 - \exp(-\Sigma a)] [1 - \exp(-\Sigma_D l)]. \quad (43)$$

Two limiting cases are of particular interest:

$$(a) \quad \Sigma a \gg 1, \quad \Sigma_D l \gg 1.$$

These conditions imply that the emitter is optically thick and that all the photons entering the detector are detected. One finds

$$G_z(\omega) = (vA) (\epsilon\rho/\sigma) \{1 + (\epsilon/\sigma) / [1 + (\omega/r_D)^2]\}, \quad (44)$$

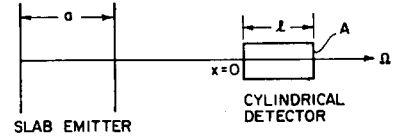


FIG. 2. Slab geometry.

and

$$\eta(T \rightarrow \infty) = 1 + (\epsilon/\sigma). \quad (45)$$

$$(b) \quad \Sigma a \gg 1, \quad \Sigma_D l \ll 1.$$

This case differs from the previous one in that the detector is now optically thin:

$$G_z(\omega) = (v/c) r_D (\epsilon\rho/\sigma) V \left[1 + \epsilon/\sigma (\Sigma_D l)^2 \left(\frac{\sin(l\omega/2c)}{(l\omega/2c)} \right)^2 \right], \quad (46)$$

and

$$\eta(T \rightarrow \infty) = 1 + (\epsilon/\sigma) (\Sigma_D l). \quad (47)$$

where $V = Al$, i.e., is the detector volume.

5. DISCUSSION

It is observed from Eqs. (44) and (46) that the break frequency in the power spectral density, i.e., r_D or (c/l) depending on the value of $\Sigma_D l$, is related to the characteristics of the detector only, and hence does not yield any information about the optical properties of the source medium. This implies that one does not gain more information by measuring $G_z(\omega)$ at different frequencies. However, one can measure (ϵ/σ) , which is related to the source, by determining $G_z(\omega)$ for very small $[\omega \ll \min(c/l, r_D)]$ and very large $(\omega \gg r_D)$ values of ω . Indeed, one finds from Eq. (42) that

$$\begin{aligned} & [G_z(0) - G_z(\infty)] / G_z(\infty) \\ &= (\epsilon/\sigma) [1 - \exp(-\Sigma a)] [1 - \exp(-\Sigma_D l)] \\ &= \eta(T \rightarrow \infty) - 1. \end{aligned} \quad (48)$$

As already indicated in Eq. (48) this quantity (ϵ/σ) is also obtainable from the variance-to-mean experiment. The choice between these two experimental techniques may depend on the mode of operation of the detector. If the detector is operated in the current mode, i.e., the output $Z(t)$ is a continuous fluctuating voltage, the power spectral density can be determined by conventional techniques. If the detector is operated in the pulse mode, i.e., $Z(t)$ is a train of pulses, then the variance-to-mean experiment is the natural choice.

Consider now the information contained in ϵ/σ , assuming that it is measured. If we are looking at a blackbody system in which the atoms are in thermal equilibrium at a temperature T , one can show that

$$\alpha = \epsilon \exp(\hbar\omega_0/KT), \quad (49)$$

where K is Boltzmann's constant and $\hbar\omega_0$ is the energy

of the observed photons. Recalling that $\sigma = \alpha - \epsilon$, one finds

$$\begin{aligned} \epsilon/\sigma &= [\exp(+\hbar\omega_0/KT) - 1]^{-1} \\ &= KT/\hbar\omega_0, \quad (\hbar\omega_0/KT) \ll 1 \\ &= \exp[-\hbar\omega_0/KT], \quad (\hbar\omega_0/KT) \gg 1. \end{aligned} \quad (50)$$

Thus, one can obtain the temperature of a black-body source by observing the fluctuation in the intensity of radiation at a *single* frequency.⁷

According to Eq. (50), it appears that it would be preferable to measure the fluctuations of the low-energy photons ($\hbar\omega_0/KT \ll 1$). However, it must be borne in mind that both the power spectral density and the variance are proportional to the average photon density which is, in turn, proportional to the ratio of the photon speed in the emitting medium and the speed of light in a vacuum. In at least some circumstances this fact presents a significant limitation on the feasibility of such measurements at very low frequencies. To illuminate this point a little, consider the case in which the emitter is a plasma—at least insofar as its dispersive properties are concerned. According to Ref. 1, the speed, v , which appears in Eq. (9) is defined to be

$$v = c^2/(\omega/k) = c\eta, \quad (51)$$

where η is the index of refraction and ω as a function of k is given by the dispersion relation for the propagation of transverse electromagnetic waves in the medium. In the plasma case, at a sufficiently low order of approximation, this dispersive relation is

$$\omega^2 = \omega_0^2 + \omega_e^2, \quad (52)$$

where ω_e is the plasma frequency and ω_0 is the vacuum frequency of the photons observed. Thus, for this example, one finds that

$$v/c = [1 + (\omega_e/\omega_0)^2]^{-1/2}. \quad (53)$$

Therefore, if one were to attempt to measure the fluctuations in the emergent photon distribution at frequencies for which $\omega_0/\omega_e \ll 1$, one must expect that both the variance and the power spectral density will be proportional to this small quantity and that hence the feasibility of a statistically significant measurement is called into question. Thus the desirability of measuring the variance at frequencies such that $\hbar\omega_0/KT \ll 1$ is in competition with the feasibility of any measurement at all if the corresponding index of refraction is small.

Finally, it must be pointed out that the present theory is predicated upon the assumption that the principal mechanisms for the emission and absorption of radiation involve free-free particle transitions only, i.e., that particle states of short lifetime do not play a significant role in the observations discussed

above. The point is that Eq. (15), which purported to describe the fluctuating photon density, has been derived with the explicit use of a coarse graining, or averaging, in time which implies a loss of information regarding the fluctuations in time intervals of the order of, or less than, the lifetime of atomic-bound states. Consequently the effects of emission and absorption on Eq. (15) cannot be represented simply by a term of the form, σn . Instead there should be resonant elastic and inelastic scattering terms, as well as fission-like terms which describe the multiple production of photons following the absorption of a single one. In such an event, the equation describing the fluctuating photon density is not simply solvable as is the case in the present example. Furthermore, because of the possibility that two or more photons may have common ancestors due to the fission-like processes, an entirely new mechanism for correlation is introduced into the system (quite analogous to the correlations observed in neutron-counting experiments in multiplying media). The contribution to the variance, or spectral density due to this latter mechanism may be as important—or even more so—than the one investigated in this work.

The present approach based on the stochastic Eq. (15) has been applied also to the study of photon fluctuations in a microwave cavity as a special case in which the transport effects can be neglected. The description of the physical system and the results are given in Appendix A. The results agree exactly with those obtained by McCombie⁸ using the master equation.

ACKNOWLEDGMENTS

This work was supported in part by the Advanced Research Projects Agency, Project Defender, ARPA Order No. 675, and was monitored by the United States Army Research Office (Durham) under Contract DA-31-124-ARO-D-403.

APPENDIX A: FLUCTUATIONS IN A CAVITY

The system under consideration consists of a microwave cavity, a source and two detectors. The source and the detectors are located at the center of the cavity, which supports a radiation mode of frequency, ω_0 . The dimensions of the source and the detectors are small as compared to the wavelength of the radiation. This assumption eliminates transport effects which play an important role in the general problem treated in the text. The source is described by $\epsilon(\mathbf{k}_0)$ and $\alpha_s(\mathbf{k}_0)$ which are the emission and absorption rates. The detectors consist of atoms which absorb photons. The re-emission of photons by the detector atoms is neglected. Thus, the detectors are described only by the absorption rate

⁷ R. Hanbury Brown and R. Q. Twiss, Proc. Roy. Soc. (London) **243A**, 291 (1958).

⁸ C. W. McCombie, *Fluctuation, Relaxation and Resonance in Magnetic Systems*, Scottish Universities Summer School, 1961, D. Ter Haar, Ed., (Plenum Press, Inc., New York, 1962), p. 193.

$r_D(\mathbf{k}_0)$. Dropping the arguments in $\epsilon(\mathbf{k}_0)$, $\alpha_s(\mathbf{k}_0)$, and $r_D(\mathbf{k}_0)$, and defining $\alpha = 2r_D + \alpha_s$, one obtains the stochastic Eq. (15) in present case as

$$[(\partial/\partial t) + \sigma]n(t) = s(t). \quad (\text{A1})$$

The mean number of photons in the mode under consideration, \bar{N} , is obtained from Eq. (10), by dropping the streaming term, as

$$\bar{N} = \epsilon\rho/\sigma, \quad (\text{A2})$$

where ρ is the density of photon states, and is unity if there is only one mode with frequency, w_0 . If there are degenerate modes with the same frequency, then ρ will denote the number of such modes. The correlation function of the noise source follows from Eqs. (17) and (18) as

$$\langle s(t)s(t') \rangle = 2\alpha\bar{N}\delta(t-t'). \quad (\text{A3})$$

The correlation function associated with the photon density, $n(t)$, is obtained by solving the stochastic Eq. (A1) as

$$n(t) = \int_0^\infty du \exp(-\sigma u) s(t-u),$$

and forming $\langle n(t)n(t+\tau) \rangle$. The result is

$$\langle n(t)n(t+\tau) \rangle = (\alpha\bar{N}/\sigma) \exp(-\sigma|\tau|). \quad (\text{A4})$$

We now consider the statistical properties of the count rates and the accumulated counts. The mean count rates of the detectors (1) and (2) are equal and given by $\langle Z \rangle = r_D\bar{N}$.

The stochastic equation for the count rate is $z(t) = r_D n(t) + s^D(t)$, where $s^D(t)$ is the noise-equivalent source accounting for the statistical fluctuations in the counting process, and has the following statistical properties: $\langle s^D(t) \rangle = 0$ and $\langle s^D(t)s^D(t') \rangle = +r_D\bar{N}\delta(t-t')$. Since a detector removes a photon, the noise-equivalent sources, $s(t)$ and $s^D(t)$, are correlated, i.e., $\langle s^D(t)s(t') \rangle = -r_D\bar{N}\delta(t-t')$.

The cross correlations between the count rates of the detectors, i.e., $z_1(t)$ and $z_2(t)$, consists of four terms:

$$\begin{aligned} \langle z_1(t)z_2(t+\tau) \rangle &= r_D^2 \langle n(t)n(t+\tau) \rangle + r_D \langle n(t)s_2^D(t+\tau) \rangle \\ &+ r_D \langle s_1^D(t)n(t+\tau) \rangle + \langle s_1^D(t)s_2^D(t+\tau) \rangle. \end{aligned} \quad (\text{A5})$$

The last term vanishes because the fluctuations in the absorption process in the detectors are uncorrelated. The second and third terms are zero, respectively, when $\tau > 0$ and $\tau < 0$. When they are not zero, they both are equal to

$$\langle n(t)s_2^D(t+\tau) \rangle = -r_D\bar{N} \exp(-\sigma|\tau|). \quad (\text{A6})$$

Combining these results, we obtain the count-rate cross-correlation functions as

$$\langle z_1(t)z_2(t+\tau) \rangle = (\langle Z \rangle^2/\rho) \exp(-\sigma|\tau|), \quad (\text{A7})$$

which is a special case of Eq. (34).

Finally, the cross correlation between the accumu-

lated counts of the detectors in a collection time, T , is obtained from Eq. (4) as

$$\langle C_1(T)C_2(T) \rangle = 2 \int_0^T (T-\tau) \langle z_1(t)z_2(t+\tau) \rangle, \quad (\text{A8})$$

which yields

$$\begin{aligned} \langle C_1(T)C_2(T) \rangle &= 2\langle C(T) \rangle^2 \\ &\times \{ [1 + \sigma T - \exp(-\sigma T)] / (\sigma T)^2 \rho \}, \end{aligned} \quad (\text{A9})$$

where $\langle C(T) \rangle = T\langle Z \rangle$, the mean number of counts in one of the detectors in T . For large gate times, i.e., $\sigma T \gg 1$, this reduces to

$$\langle C_1(T)C_2(T) \rangle = 2\langle C(T) \rangle^2 / T\sigma\rho, \quad (\text{A10})$$

which is identical to Eq. (4.14) of Ref. 8, apart from differences in notations.

APPENDIX B: THE RELATION BETWEEN COUNT RATE CORRELATIONS AND PHOTON CORRELATIONS

Though described in some detail in Ref. 3 (by Akcasu and Osborn), the derivation of Eq. (31) in the text is sketched here for completeness. Recall that the count-rate correlation function was defined by [Eq. (7)],

$$\phi_z(\mathbf{r}, \mathbf{r}', \tau) \equiv \langle z(\mathbf{r}, t)z(\mathbf{r}', t+\tau) \rangle, \quad (\text{B1})$$

with the fluctuating count rate given by

$$z(\mathbf{r}, t) = r_D n(\mathbf{r}, t) + s^D(\mathbf{r}, t). \quad (\text{B2})$$

In Eq. (B2), r_D is the probability per unit time per photon for the occurrence of a detection event, and s^D is the detection-rate noise source. In general, r_D may be a function of the photon wavevector. Inserting Eq. (B2) into Eq. (B1); and recalling the definition, Eq. (8), in the text; we find

$$\begin{aligned} \phi_z(\mathbf{r}, \mathbf{r}', \tau) &= r_D(\mathbf{k})r_D(\mathbf{k}')\phi_n(\mathbf{r}, \mathbf{r}', \tau) \\ &+ \langle s^D(\mathbf{r}, t)s^D(\mathbf{r}', t+\tau) \rangle + r_D(\mathbf{k})\langle n(\mathbf{r}, t)s^D(\mathbf{r}', t+\tau) \rangle \\ &+ r_D(\mathbf{k}')\langle s^D(\mathbf{r}, t)n(\mathbf{r}', t+\tau) \rangle. \end{aligned} \quad (\text{B3})$$

According to Eq. (17), the second term on the right-hand side of Eq. (B3) reduces to

$$\langle s^D(\mathbf{r}, t)s^D(\mathbf{r}', t+\tau) \rangle = r_D(\mathbf{k})\langle N(\mathbf{r}) \rangle \delta(\mathbf{r}-\mathbf{r}')\delta(\tau) \quad (\text{B4})$$

within the detector.

The fluctuating photon distribution is described by Eq. (15), which, in the detector, becomes

$$[(\partial/\partial t) + B]n(\mathbf{r}, t) = s(\mathbf{r}, t), \quad (\text{B5})$$

where

$$B = c\Omega \cdot \nabla + r_D(\mathbf{k}). \quad (\text{B6})$$

Thus

$$n(\mathbf{r}, t) = \int_0^\infty dx \exp(-xB)s(\mathbf{r}, t-x), \quad (\text{B7})$$

and the third and fourth terms on the right-hand side

of Eq. (B3) become

$$r_D(\mathbf{k}) \int_0^\infty dx \exp(-xB) \langle s(\mathbf{r}, t-x) s^D(\mathbf{r}', t+\tau) \rangle \\ + r_D(\mathbf{k}') \int_0^\infty dx \exp(-xB') \langle s^D(\mathbf{r}, t) s(\mathbf{r}', t+\tau-x) \rangle. \quad (\text{B8})$$

As discussed in Ref. 3 (by Akcasu and Osborn), the cross-correlation between the noise sources for photon loss and detected particle gain in the detector reduces to

$$\langle s(\mathbf{r}, t) s^D(\mathbf{r}', t') \rangle = -r_D(\mathbf{k}) \langle N(\mathbf{r}) \rangle \delta(\mathbf{r}-\mathbf{r}') \delta(t-t'). \quad (\text{B9})$$

Inserting Eq. (B9) into Eq. (B8), we find for $\tau > 0$ that Eq. (B3) becomes

$$\phi_z(\mathbf{r}, \mathbf{r}', \tau) = r_D^2(\mathbf{k}) \phi_n(\mathbf{r}, \mathbf{r}', \tau) + r_D(\mathbf{k}) \langle N(\mathbf{r}) \rangle \\ \times \delta(\mathbf{r}-\mathbf{r}') \delta(\tau) - r_D^2(\mathbf{k}) \langle N(\mathbf{r}) \rangle \exp(-\tau B') \delta(\mathbf{r}-\mathbf{r}'). \quad (\text{B10})$$

Here we have used Eq. (B4), and the fact that $\phi_n(\mathbf{r}, \mathbf{r}', \tau)$ contains the factor, $\delta(\mathbf{k}-\mathbf{k}')$. Recognizing that $g_1(\mathbf{r}, \mathbf{r}', \tau)$ defined in Eq. (32), is given by

$$g_1(\mathbf{r}, \mathbf{r}', \tau) = \exp(-\tau B') \delta(\mathbf{r}-\mathbf{r}'), \quad \tau > 0, \quad (\text{B11})$$

it is seen that Eq. (B10) and Eq. (31) of the text are the same.

Orientation Inversions in Polycrystalline CdS Bulk Crystals and Thin Films

G. A. ROZGONYI AND N. F. FOSTER

Bell Telephone Laboratories, Incorporated, Murray Hill, New Jersey

(Received 10 August 1967)

A study of the crystallographic orientation of the single-crystal grains of large (8.5-cm-diam) polycrystals of CdS and of thin films of CdS has been made. Chemical and sputter-etching observations, x-ray crystallographic techniques, and piezoelectric polarity measurements have been used to classify the growth habits of the thin films as a whole and of the individual grains of the bulk crystals.

Three types of grains have been identified in the bulk CdS polycrystals. These are type I: c axis normal to a Cd-rich growth face; type II: c axis greater than 15° from the normal to an S-rich growth face; and inverted type II: c axis greater than 15° from the normal to a Cd-rich growth face. Type I and inverted type II grains exhibit negative compression piezovoltages, while type II grains are positive. Type I grains are identical to the type I single crystals previously discussed by Reynolds, and type II grains are similar to the 14° misoriented type II single crystals of Reynolds.

Similar results were obtained from the evaporated CdS thin films which could be classified as type I and type II, with the normal to the type II films about 20° off the c axis, indicating a close parallelism between the processes occurring in evaporated thin films and in bulk crystal growth. Sputtered CdS films were always type I. It is suggested that the variable efficiencies that have been observed in piezoelectric thin-film transducers with similar crystallographic orientations are related to orientation inversions in the films.

INTRODUCTION

In many binary semiconducting compounds it is possible to orient a single crystal such that the opposite faces of the same sample exhibit different electrical, chemical and physical properties. Basically it is the lack of inversion symmetry in the $\langle 111 \rangle$ directions of the cubic III-V compounds and $\langle 00.1 \rangle$ directions of the hexagonal II-VI compounds which gives rise to the differences observed. Gatos and co-workers,¹ on the basis of x-ray and chemical studies, have been able to characterize the influence of crystal orientation on surface properties for many compound materials. These techniques have also been used by Brafman *et al.*² on studies of the polar properties of ZnS platelets grown from the vapor phase. It is also possible to distinguish the polar faces of piezoelectric materials by the sign of the piezovoltage. Berlincourt *et al.*,³ using an elementary

electroelastic model theory, found that compression along the polar axis gives rise to a negative charge on the (111) Zn face of ZnS and a negative charge on the (00.1) Cd face of CdS.

Reynolds and Greene⁴ have identified two types of CdS crystals grown from the vapor phase: (a) type I: crystals with the growth surface parallel to the (00.1) plane, and (b) type II: crystals with the growth surface making an angle of approx 14° with the (00.1) plane. In a later paper, Reynolds and Czyzak⁵ determined that the growing surface of type I crystals, when etched in HCl, always exhibited hexagonal etch pits, indicative of the (00.1) Cd face,⁶ whereas type II crystals grew with either polarity.

In the present study, chemical, crystallographic, and

⁴ D. C. Reynolds and L. C. Greene, *J. Appl. Phys.* **29**, 559 (1958).

⁵ D. C. Reynolds and S. J. Czyzak, *J. Appl. Phys.* **31**, 94 (1960).

⁶ The hexagonal etch pits were initially believed to be on the (00.1) S face, but this has been shown to be in error: G. A. Wolff, J. J. Frawley, and J. R. Heitanen, *J. Electrochem. Soc.* **111**, 22 (1964).

¹ H. C. Gatos, *Science* **137**, 311 (1962), and references therein.

² O. Brafman, E. Alexander, B. S. Fraenkel, Z. H. Kalman, and D. T. Steinberger, *J. Appl. Phys.* **35**, 1855 (1964).

³ D. Berlincourt, H. Jaffe, and L. R. Shiozawa, *Phys. Rev.* **129**, 1009 (1963).