Thermal Fluctuations in a Nonlinear System

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As an example of a nonlinear system, an electric circuit containing a voltage-dependent resistance $R(V)$ is studied. The resistance is in contact with a heat bath and generates current fluctuations. A general method is given to find the spectral density of these fluctuations. For two special forms of $R(V)$, explicit results are obtained by means of a perturbation calculation. The result differs from the Nyquist formula mainly in that new terms appear, corresponding to relaxation times $\frac{1}{2}R_0C$, $\frac{1}{3}R_0C$, etc.

1. INTRODUCTION

IN spite of the vast amount of work on Brownian movement and statistical fluctuations in general,[1] very little attention has been devoted to fluctuations in nonlinear systems. Take the case of an electric circuit with a resistor in equilibrium with a heat bath. It is always supposed that the resistance may be considered constant in a range of the size of the current fluctuations. Undoubtedly it is not easy to find an experimental situation in which this is not true. From a theoretical point of view, however, it is unsatisfactory that the usual treatments are so essentially confined to this linear case. In fact, the Langevin approach, using a random force obeying the relations of Einstein and Nyquist, does not seem to lend itself to a generalization to the nonlinear case. On the other hand, it can be concluded from statistical mechanics that the Fokker-Planck equation is of very general validity for the description of the macroscopic behavior of systems with many degrees of freedom.[2,3] Once this equation is adopted, the remaining task is twofold. Firstly, the two functions occurring in the general Fokker-Planck equation must be determined; this is done in Secs. 2 and 3. Secondly, the relevant properties of the fluctuations, namely the spectral density, must be found. This is done by an approximation method in Sec. 5, after some general relations have been derived in Sec. 4. Finally the results are applied to a few simple cases in Sec. 6.

It should be emphasized that our present problem is different from the problem of noise passing through a nonlinear device.[4] In that case the statistical properties of the input noise are given, and the problem to find the properties of the output noise is a mathematical one. Our problem, however, is to find the spectral density of the noise that is generated inside a nonlinear resistor. This cannot be done by purely mathematical arguments; the physics enters through the use of the fundamental equation (3).

2. FORMULATION OF THE PROBLEM

For simplicity we shall confine ourselves in this paper to the following simple system, previously discussed by MacDonald.[5] An electric circuit consists of a condenser C and a resistance R, in contact with a heat bath. It will be supposed that R is not constant but depends on the potential difference V, so that the current depends on V in a nonlinear way. Let the probability for the charge on C have a value between q and $q+dq$ be denoted by $P(q,t)dq$. The time dependence of $P(q,t)$ obeys a Fokker-Planck equation of the general form

$$\frac{\partial P}{\partial t}=\frac{\partial^2}{\partial q^2}[\xi(q)P]+\frac{\partial}{\partial q}[\eta(q)P], \tag{1}$$

where $\xi(q)$ and $\eta(q)$ are two as yet undetermined functions, which characterize the properties of the irreversible process that takes place in the resistance. Let $G(q)$ be the equilibrium distribution,

$$G(q)=(2\pi kTC)^{-\frac{1}{2}}\exp(-q^2/2kTC), \tag{2}$$

where T is the temperature of the heat bath and k is Boltzmann's constant. It can then be derived from the principle of detailed balance[3] that $\xi(q)$ and $\eta(q)$ are related by

$$\eta(q)=-\frac{1}{G(q)}\frac{d}{dq}[\xi(q)G(q)].$$

(In case ξ is a constant, this equation reduces to the Einstein relation for Brownian movement.) With the aid of this relation η can be eliminated from (1), with the result

$$\frac{\partial P}{\partial t}=\frac{\partial}{\partial q}\left[\xi G\frac{\partial}{\partial q}\left(\frac{P}{G}\right)\right]. \tag{3}$$

This equation determines both the large scale phenomenological behavior of the system and the small

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[1] See, e.g., *Selected Papers on Noise and Stochastic Processes*, edited by N. Wax (Dover Publications, New York, 1954); in particular, M. C. Wang and G. E. Uhlenbeck, Revs. Modern Phys. **17**, 323 (1945); S. O. Rice, Bell System Tech. J. **23**, 282 (1944); **25**, 46 (1945). Also: A. van der Ziel, *Noise* (Prentice Hall, Inc., New York, 1954); R. Becker, *Theorie der Wärme* (Springer-Verlag, Berlin, 1955).

[2] N. G. van Kampen, Physica **20**, 603 (1954); Fortschr. Physik **4**, 405 (1956).

[3] N. G. van Kampen, Physica **23**, 707 (1957).

[4] For references see D. M. Middleton, J. Appl. Phys. **22**, 1143 and 1153 (1951).

[5] D. K. C. MacDonald, Phil. Mag. **45**, 63 (1954).

scale fluctuations. In principle the function $\xi(q)$ is determined by the physical mechanism that is responsible for the dissipation of energy, in this case the scattering of electrons in the resistor. On the other hand, by extracting the large scale features of (3), the connection between $\xi(q)$ and the phenomenological function $R(V)$ can be found. This is done in the next section.

3. PHENOMENOLOGICAL EQUATION

The average charge on the condenser is

$$\langle q\rangle_t=\int_{-\infty}^{+\infty} qP(q,t)dq.$$

The rate of change is, according to (3) and (2),

$$\frac{d}{dt}\langle q\rangle=\int\frac{P}{G}\frac{d}{dq}(\xi G)dq$$
$$=-\langle q\xi(q)\rangle/kTC+\langle\xi'(q)\rangle, \tag{4}$$

where the prime denotes the derivative with respect to the argument. This is to be compared with the equation for the macroscopic charge Q,

$$\frac{d}{dt}Q=-\frac{V}{R(V)}=-\frac{Q}{CR(Q/C)}. \tag{5}$$

Of course, Q is to be identified with $\langle q\rangle$.[6] If R were a constant, Eqs. (4) and (5) could be identified by putting

$$\xi/kT=1/R. \tag{6}$$

We shall show that this relation remains true if R, and hence ξ, are not constant.

If ξ is not constant, it is tempting to write for (4)

$$(d/dt)\langle q\rangle=-\langle q\rangle\xi(\langle q\rangle)/kTC+\xi'(\langle q\rangle). \tag{7}$$

This approximation assumes that the fluctuations are small compared to an interval in which ξ varies appreciably. In order to assess the validity of this assumption, first note that the function ξ is characteristic of the properties of the resistor. Hence ξ will depend on $V=q/C$, rather than on q and C separately. Now the fluctuations of V are of the order $(kT/C)^{\frac{1}{2}}$, and can therefore be made arbitrarily small by taking C large. Hence (7) is certainly valid for sufficiently large C. Moreover,

$$\xi'(q)\equiv d\xi/dq=C^{-1}(d\xi/dV)$$

is of order $1/C$, so that the last term in in (7) should be dropped. Comparing the result with (5), one sees that (6) remains true, even if ξ and R depend on V. For further discussion see Appendix I.

Of course, whenever one uses the phenomenological Eq. (5), it is tacitly assumed that the fluctuations are small. If the voltage fluctuations are large, owing to a small C, then (3) and (4) are still true, but (7) is incorrect while (5) becomes meaningless. Again, even if the fluctuations are small, (5) and (7) only describe the large scale behavior correctly, but cannot be used to describe the fluctuations themselves. Instead one will have to use (3); this will be done in the next sections. We only needed the large-scale behavior to find the identity (6).

4. FLUCTUATIONS

Let $P(q_0|q,t)$ denote the solution of (3) that reduces to $\delta(q-q_0)$ for $t\rightarrow 0$; it may be regarded as the probability of a transition from q_0 to q in time t. The average charge at time t under the condition that the charge at $t=0$ was q_0 is

$$\langle q(t)\rangle_{q_0}=\int P(q_0|q,t)qdq.$$

The correlation between the values of the charge at two different instants with interval t is, in the equilibrium state,

$$\langle q(0)q(t)\rangle_{\text{eq}}=\int G(q_0)q_0dq_0\int P(q_0|q,t)qdq. \tag{8}$$

This is the autocorrelation function of the stochastic variable q. The spectral density of the charge fluctuations is obtained from this by using the Wiener-Khintchin theorem[1]

$$S(\omega)=(2/\pi)\int_0^{\infty}\langle q(0)q(t)\rangle_{\text{eq}}\cos\omega t dt. \tag{9}$$

It is more customary to express results in terms of the spectral density W_f of current fluctuations in the frequency scale $(f=\omega/2\pi)$; one has

$$W_f=2\pi\omega^2S(\omega).$$

Of course, it is not possible to solve (3) for arbitrary ξ; in fact, it seems that only for $\xi=$constant an explicit solution can be found. It is therefore useful to reduce the equation to an eigenvalue problem to make it amenable to standard perturbation methods.

Write (3) in the form

$$\partial P/\partial t=\Xi P, \tag{10}$$

where Ξ is a differential operator, acting in the space of real functions $P(q)$. Let $1/G(q)$ be used as a weight function in this space, so that the scalar product is defined by

$$(P_1,P_2)=\int_{-\infty}^{+\infty}[P_1(q)P_2(q)/G(q)]dq=(P_2,P_1).$$

It can then easily be checked that Ξ is self-adjoint:

[6] Although the definition of the phenomenological quantity Q is not precise enough to distinguish between $\langle q\rangle$ and, for instance, $\langle q^2\rangle^{\frac{1}{2}}$.

$(P_1,\Xi P_2)=(P_2,\Xi P_1)$. In addition Ξ is negative definite:

$$(P,\Xi P)=-\int \xi G[(d/dq)(P/G)]^2 dq\leq 0.$$

The value 0 obtains only for the equilibrium distribution $P=G$. Now put in (10)

$$P(q,t)=e^{-\lambda t}P_\lambda(q),$$

so that P_λ is to be solved from the eigenfunction equation

$$\Xi P_\lambda=-\lambda P_\lambda. \tag{11}$$

Moreover $P_\lambda(q)$ must vanish sufficiently fast for $q\rightarrow\pm\infty$, so that the norm (P_λ,P_λ) is finite.

One solution is of course $\lambda=0$, $P_0(q)=G(q)$. If one has found a complete set of normalized eigenfunctions P_λ, the completeness relation

$$\textstyle\sum_\lambda P_\lambda(q)P_\lambda(q')=G(q)\delta(q-q')$$

holds. Consequently

$$P(q_0|q,t)=\textstyle\sum_\lambda e^{-\lambda t}P_\lambda(q_0)P_\lambda(q)/G(q_0). \tag{12}$$

The autocorrelation (8) becomes

$$\langle q(0)q(t)\rangle_{\text{eq}}=\sum_\lambda e^{-\lambda t}\left[\int P_\lambda(q)qdq\right]^2, \tag{13}$$

and the spectrum of charge fluctuations

$$S(\omega)=\frac{2}{\pi}\sum_\lambda \frac{\lambda}{\lambda^2+\omega^2}\left[\int P_\lambda(q)qdq\right]^2. \tag{14}$$

5. PERTURBATION CALCULATION

We put $\xi(q)=\xi(0)+\xi^{(1)}(q)$ and assume $\xi^{(1)}(q)$ small. First the unperturbed problem with $\xi=\xi(0)=\text{constant}$ has to be solved. It is convenient to choose the unit of charge such that $kTC=1$, and the unit of time such that $\xi(0)=1$. Then the eigenfunction equation (11) takes the form

$$P_\lambda''+P_\lambda'+(\lambda+1)P_\lambda=0.$$

This is the differential equation for Hermite functions. The eigenvalues are $\lambda=n$ ($n=0, 1, 2, \cdots$) and the normalized eigenfunctions

$$h_n(q)=[(2\pi)^{\frac{1}{2}}n!]^{-\frac{1}{2}}\exp(-\tfrac{1}{2}q^2)H_n(q).$$

Here $H_n(q)$ is the nth Hermite polynomial.[7] Substitution in (12) yields

$$P(q_0|q,t)=\exp(-\tfrac{1}{2}q^2)\sum_{n=0}^{\infty}[(2\pi)^{\frac{1}{2}}n!]^{-1}H_n(q_0)H_n(q)e^{-nt}.$$

[7] We use the modified definition, denoted by He_n in W. Magnus and F. Oberhettinger, *Formulas and Theorems for the Special Functions of Mathematical Physics* (Chelsea Publishing Company, New York, 1949), p. 80.

From this one finds, for example,

$$\langle q(t)\rangle_{q_0}=q_0e^{-t},$$

which is the regression equation for this linear case. Furthermore, one has

$$\langle q(0)q(t)\rangle_{\text{eq}}=\langle q(0)^2\rangle_{\text{eq}}e^{-t}=e^{-t}.$$

This autocorrelation function gives for the spectrum

$$S(\omega)=(2/\pi)(1+\omega^2)^{-1}, \tag{15}$$

or

$$W_f=\frac{4kT}{R}\left[\frac{(RC\omega)^2}{1+(RC\omega)^2}\right],$$

which is usually derived as a corollary of the Nyquist formula.[1]

Next put $\xi(q)=1+\xi^{(1)}(q)$, and accordingly $\Xi=\Xi^{(0)}+\Xi^{(1)}$. Equation (11) becomes

$$(\Xi^{(0)}+\Xi^{(1)}+n+\lambda_n^{(1)}+\cdots)(h_n+P_n^{(1)}+\cdots)=0.$$

The standard formulas of perturbation theory then give for the shift of the nth eigenvalue, to first order,

$$\begin{aligned}\lambda_n^{(1)}&=-(h_n,\Xi^{(1)}h_n)\\ &=(2\pi)^{-\frac{1}{2}}\int \xi^{(1)}\exp(-\tfrac{1}{2}q^2)\left[\frac{d}{dq}(h_n/G)\right]^2dq\\ &=[(2\pi)^{\frac{1}{2}}n!]^{-1}\int \xi^{(1)}\exp(-\tfrac{1}{2}q^2)(nH_{n-1})^2dq.\end{aligned}$$

To find $P_n^{(1)}$, let it be expanded in the unperturbed eigenfunctions h_m:

$$P_n^{(1)}(q)=\textstyle\sum c_{nm}h_m(q).$$

One then has, for $m\neq n$,

$$\begin{aligned}(n-m)c_{nm}&=-(h_m,\Xi^{(1)}h_n)\\ &=(2\pi n!m!)^{-\frac{1}{2}}\int \xi^{(1)}\exp(-\tfrac{1}{2}q^2)nH_{n-1}\\ &\qquad\times mH_{m-1}dq.\end{aligned}$$

We shall also need c_{nn}, although it is of the second order in the perturbation. c_{nn} is determined by the condition that $h_n+P_n^{(1)}$ should be normalized:

$$(1+c_{nn})^2=1-\sum_{m\neq n}c_{nm}^2.$$

Finally we need the second-order shift of the eigenvalue,

$$\lambda_n^{(2)}=\sum_{m\neq n}(n-m)^{-1}(h_n,\Xi^{(1)}h_m)^2=\sum_m(n-m)c_{nm}^2.$$

Using these results, one finds for the autocorrelation function (13)

$$\langle q(0)q(t)\rangle_{\text{eq}}=(1+c_{11})^2e^{-\lambda_1 t}+\sum_{n\neq 1}c_{n1}^2e^{-\lambda_n t},$$

which yields the spectrum

$$S(\omega)=\frac{2}{\pi}\left\{(1+c_{11})^2\left(\frac{\lambda_1}{\lambda_1^2+\omega^2}\right)+\sum_{n\neq 1}c_{n1}^2\left(\frac{\lambda_n}{\lambda_n^2+\omega^2}\right)\right\}.$$

This is the modified spectral density to second order. The only first-order modification arises from the first-order shift of λ_1. The interesting feature, however, is the $\sum$ term, which is of second order. Obviously in this sum the λ_n may be replaced with $\lambda_n^{(0)}=n$. Each of the terms in the sum has the same shape as the unperturbed spectrum (15), but for the fact that the relaxation time R_0C is replaced in the successive terms by $\frac{1}{2}R_0C$, $\frac{1}{3}R_0C$, $\cdots$. These terms in the spectrum will show up as low broad wings on both sides. In addition, the original peak is modified (broadened or narrowed according as $\lambda_1>\lambda_1^{(0)}$ or $\lambda_1<\lambda_1^{(0)}$). The integral of the total spectrum is of course not altered, because it is the mean square fluctuation of q in equilibrium; indeed,

$$\int_0^\infty S(\omega)d\omega=(1+c_{11})^2+\sum_{n=2}^{\infty}c_{n1}^2=1.$$

(Because $c_{1n}=-c_{n1}$, and $c_{n0}=0$.)

6. APPLICATION TO TWO SPECIAL CASES

Usually the resistance is symmetrical with respect to the direction of the current; the simplest nonlinearity of this type is $R(V)=R_0+R_2V^2$. This amounts to putting $\xi(q)=1+\gamma q^2$, where $\gamma=-(kTR_0/R_2C)$. The equations of the previous section yield for this case

$$c_{n,1}=0 \quad \text{unless} \quad n=3 \text{ or } 1,$$
$$c_{3,1}=(\tfrac{3}{2})^{\frac{1}{2}}\gamma,$$
$$(1+c_{11})^2=1-\tfrac{3}{2}\gamma^2,$$
$$\lambda_1=1+\gamma-3\gamma^2.$$

Hence we find for the spectral density of the current fluctuations

$$W_f=4\left\{(1-\tfrac{3}{2}\gamma^2)\frac{\lambda_1\omega^2}{\lambda_1^2+\omega^2}+\tfrac{1}{2}\gamma^2\frac{\omega^2}{1+(\frac{1}{3}\omega)^2}\right\}. \tag{16}$$

A second example is obtained by putting $R(V)=R_0+R_1V$, which amounts to $\xi(q)=1+\beta q$ with $\beta=-(R_1/R_0)(kT/C)^{\frac{1}{2}}$. As an even function of V, this case may be regarded as a simple rectifier. The equations of the previous section yield

$$c_{n,1}=0 \quad \text{unless} \quad n=2 \text{ or } 1,$$
$$c_{2,1}=\beta\sqrt{2},$$
$$(1+c_{11})^2=1-2\beta^2,$$
$$\lambda_1=1-2\beta^2.$$

Hence the spectral density of the current fluctuation is

$$W_f=4\left\{\frac{(1-2\beta^2)^2\omega^2}{(1-2\beta^2)^2+\omega^2}+\frac{\beta^2\omega^2}{1+(\frac{1}{2}\omega)^2}\right\}.$$

7. DISCUSSION OF EARLIER LITERATURE

Kramers[8] studied Brownian movement in a field of force. This is different from our problem, because his nonlinearity is due to the external, nonrandom force, whereas the random force is still the same as in the linear Brownian movement. Our problem, on the other hand, amounts to finding the statistical properties of the random force in case the friction of the moving particle is no longer proportional to the velocity. However, Polder[9] has noticed that, if in Kramers' problem the friction coefficient is allowed to depend on the position of the particle, the resulting Fokker-Planck equation is also valid for the current fluctuations in a nonlinear electric circuit. The position of the particle is then the charge q on the condenser, the external force corresponds to a dependence of C on q, and the mass of the particle corresponds to a self-induction.

MacDonald[5] formulated the problem of finding the spectral density of the fluctuations in an electric circuit of the type treated here. He introduced the hypothesis that (7) (without the last term) not only describes the large scale behavior, but also describes correctly the regression of small scale fluctuations. On the basis of this assumption he found for the spectral density, in the first case of our Sec. 6,

$$W_f=4(1-\tfrac{3}{2}\gamma)\frac{\omega^2}{1+\omega^2}+\tfrac{1}{2}\gamma\frac{\omega^2}{1+(\frac{1}{3}\omega)^2}. \tag{17}$$

The remarkable resemblance with our formula (16) is deceptive, as (17) contains the first power of γ where (16) has γ^2. The first order terms in (16) do not at all correspond to (17).

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APPENDIX I

Since the derivation of the relation (6) connecting ξ and R is a very crucial step, we want to discuss it more fully. Essentially three arguments enter into it.

(i) *The same Fokker-Planck equation (3) describes both small scale and large scale phenomena.* The idea is that these phenomena are merely different aspects of one and the same diffusion-like process in the space of observable quantities ("a space," see reference 3).

(ii) *The function $\xi(q)$ must not vary too rapidly.* Firstly, for $q\rightarrow\pm\infty$ it must not increase very strongly, certainly not like $1/G(q)\sim\exp(\frac{1}{2}q^2)$. Secondly, $\xi(q)$ must not oscillate rapidly about some average trend, $\bar{\xi}(q)$ say. Then the large-scale phenomena would be determined by $\bar{\xi}$ rather than by ξ, whereas the fluctuations are determined by ξ. Actually both these conditions will be satisfied for sufficiently large C.

[8] H. A. Kramers, Physica **7**, 284 (1940).
[9] D. Polder, Phil. Mag. **45**, 69 (1954).

(iii) *The system can be decomposed into two parts*, the condenser determining the function G, and the resistance determining ξ. This made it possible to stipulate that ξ is a function of $V=q/C$ only [although *a priori* ξ refers to the system as a whole, and may therefore be an arbitrary function $\xi(q,C)$ of two variables]. Subsequently it is possible to go the limit of large C, in order to compare ξ with R.

APPENDIX II

Recently MacDonald[10] studied the same problem again. He now also gives the Fokker-Planck equation (1), but considers $\xi(q)$ and $\eta(q)$ as two mutually unrelated functions. Actually there is a relation (Sec. 2), which in MacDonald's notation takes the form

$$G(q)=F(q)-kTCF'(q)/q. \qquad (18)$$

The equilibrium distribution (7) of MacDonald then reduces to the ordinary Gauss distribution. MacDonald proved on thermodynamic grounds that $\langle q^2\rangle_{eq}$ must be independent of the function $R(V)$, but in fact the whole distribution function of q turns out to be independent of the resistance. It seems to me that this fact can be generally postulated on statistical grounds. This postulate would then conversely lead to (18).

[10] D. K. C. MacDonald, Phys. Rev. **108**, 540 (1957).

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Optical Properties of Hexagonal ZnS Single Crystals

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The optical transmission of hexagonal zinc sulfide crystals has been measured in the spectral range from 0.32 to 15 μ. From the spacing of interference maxima, the dependence of index of refraction on wavelength has been determined. Measurements of the effects of temperature from 20°C to 120°C and of pressure up to 1700 atmos on the width of the forbidden energy band indicate an increase in band gap of 9×10^{-6} ev/atmos with pressure and a decrease of 7×10^{-4} ev/C° with temperature for both ordinary and extraordinary rays. Approximately one-fifth of the shift with temperature is the result of dilatation of the lattice. The origin of the larger portion of the shift is discussed.

INTRODUCTION

Studies of hexagonal ZnS single crystals have included the effect of temperature on the wavelength of the absorption edge[1] and determination of the energy of the band gap by optical absorption and electrical conductivity measurements.[2] In this paper we report further measurements of the optical properties of hexagonal ZnS crystals grown from the vapor phase. The effect of pressure up to 1700 atmospheres on the absorption edge is used to determine what fraction of the temperature shift of the absorption edge is due to lattice dilatation. These measurements were made for both the ordinary and the extraordinary rays in the crystal.

The well-defined interference fringes observed in the transmission measurements make possible the determination of the dependence of the index of refraction on wavelength in the spectral range 0.34 to 2.0 μ. A thick crystal was used to check the index of refraction in the range from 2 to 15 μ.

EXPERIMENTAL

The optical system employed for the transmission measurements is shown in Fig. 1. A Beckman hydrogen discharge or a tungsten incandescent source was focused on the sample by a spherical mirror which acts as the limiting aperture of the apparatus. This arrangement minimized loss of radiation by scattering or refraction through small angles. The light-collecting optics have been described in a previous publication.[3] The clear areas of the crystals used were not less than 2 mm square and the beam entering the monochormator

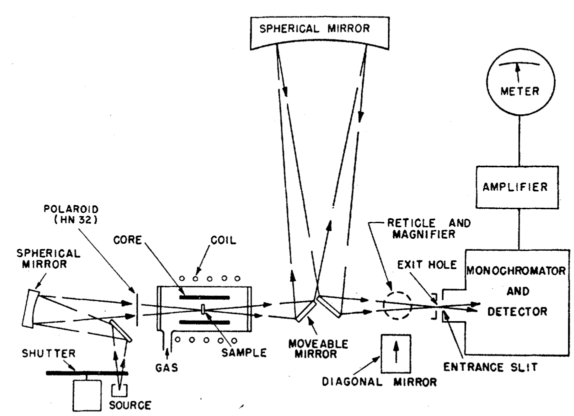

Fig. 1. Optical arrangement for transmission measurements.

[1] C. Z. Van Doorn, Physica **20**, 1155 (1954).
[2] W. W. Piper, Phys. Rev. **92**, 23 (1953).
[3] D. T. F. Marple, J. Opt. Soc. Am. **46**, 490 (1956).