choose between the procedure of applying the exclusion principle in intermediate states and that of ignoring it for such states; both yield the same total numerical results. The former procedure appears as the natural one when an individual scattering problem is approached by

the traditional method of variation of parameters. The latter procedure, on the other hand, corresponds to a simple algorithm for the systematic analysis of the S-matrix, and to a direct and simple statement as to the cause of the scattering.

PHYSICAL REVIEW

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The Mobility of Electrons in Diamond

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The mobility of electrons in diamond has been measured by means of the Hall effect. In agreement with the theory of Seitz, the mobility is inversely proportional to the three-halves power of the absolute temperature. The measured room temperature value of the mobility is 900 cm²/volt-sec as compared with the theoretical estimate of 156 cm²/volt-sec.

I. INTRODUCTION

THEORY of the scattering of electrons by lattice vibrations in nonpolar crystals has been given by Seitz,¹ which predicts that, for a nondegenerate electron gas, the electronic mobility and mean free path vary as $T^{-\frac{1}{2}}$ and T^{-1} , respectively. The experimental data on silicon² and germanium³ are in excellent agreement with the theoretical prediction of the absolute magnitude of the mobility and its temperature dependence.

Diamond is a typical nonpolar crystal and, because of its successful use as a crystal counter, the problem of the electronic mobility in it is of considerable interest. Seitz has estimated the room temperature mobility of diamond to be approximately 156 cm²/volt-sec. The measurements of the Hall effect of Lenz⁴ yield a room temperature mobility of about 200 cm²/volt-sec. On the basis of admittedly scanty evidence, Lenz concluded, however, that the mobility was probably independent of temperature.

Some time ago⁵ we reported a value of 900 cm²/voltsec for the room temperature electronic mobility in diamond and a temperature variation in agreement with Seitz. The purpose of the present paper is to present in detail the Hall effect measurements on which these conclusions were based.

The theory and technique of measurement of the Hall effect are well known.^{6,7} The electronic mobility,

 μ , may be defined in terms of the electronic conductivity, σ , by the equation,

$$\sigma = n e \mu, \tag{1}$$

where n is the concentration of free electrons and e is the charge of an electron. The elementary theory⁸ of a nondegenerate electron gas subjected to mutually perpendicular electric and magnetic fields results in the following equation for the mobility of the electrons:

$$\mu = (8/3\pi)(c/H)(E_h/E_a), \qquad (2)$$

where E_h is the Hall field developed in the crystal when the applied electric and magnetic fields are E_a and H, respectively. The measured Hall and applied potentials are related to their respective fields by the equations $V_h = E_h t$ and $V_a = E_a L$. The length of the specimen measured parallel to the applied field is L, and t is the width of the specimen between the Hall electrodes.

The mobility and mean free path, l, are simply related by the equation,

$$\mu = 4el/3(2\pi mkT)^{\frac{1}{2}},\tag{3}$$

where m is the electronic mass and k is Boltzmann's constant.

II. EXPERIMENTAL

The intrinsic conductivity of diamond at room temperature and below is too small to permit the measurement of the Hall effect. Upon absorption of ultraviolet and visible radiation, diamond exhibits a photo-conductivity which has been the subject of numerous investigations.9-11 Photocurrents of the order of 10-10

Now at the Naval Research Laboratory, Washington, D. C.

<sup>Now at the Internation Research Laboratory, Washington, D. C.
Now at the University of Illinois, Urbana, Illinois.
F. Seitz, Phys. Rev. 73, 549 (1948).
G. L. Pearson and J. Bardeen, Phys. Rev. 75, 865 (1949).
K. Lark-Horovitz, Report NDRC 14-585, OSRD Contract OEMsr-36, 1945, unpublished.
H. Lang, App. Dhysik 82, 041 (1027).</sup> ⁴ H. Lenz, Ann. Physik 83, 941 (1927).

⁶ C. C. Klick and R. J. Maurer, Phys. Rev. 76, 179 (1949).
⁶ F. Seitz, Modern Theory of Solids (McGraw-Hill Book Company, Inc., 1940) p. 192.
⁷ A. H. Wilson, Semi-conductors and Metals (Cambridge University Press, 1939), p. 95.

⁸H. Fröhlich, *Elektronentheorie der Metalle*, (Verlag. Julius Springer, Berlin, 1936) p. 220. ⁹B. Gudden and R. Pohl, Z. Physik 20, 14 (1924).

¹⁰ Robertson, Fox, and Martin, Proc. Roy. Soc. (London) 157, 579 (1936).

¹¹ Sec. Symposium on the Structure and Properties of Diamond, Proc. Indian Acad. Sci. 24A (1946).

amp were produced in the diamond used in the present investigation by absorption of the radiation from a high pressure mercury arc. The incident radiation was limited to the wavelength region from 3000 to 6200A by filters. The diamond was completely opaque to radiation of wavelength less than 2900A.

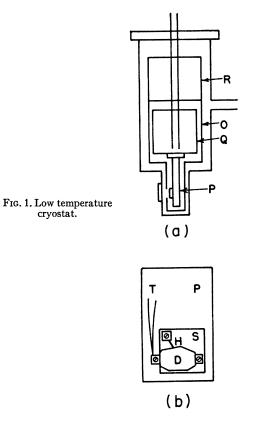
The diamond was mounted inside a vacuum cryostat which is illustrated in Fig. 1(a). It was supported on a vertical plate, P, which was fastened to the bottom of the liquid air reservoir, Q. This reservoir was suspended and thermally insulated from a second liquid air reservoir, R, to which was attached the radiation shield, O. German silver tubes were used to support the reservoirs and introduce the liquid air.

The diamond was a plate 0.22 cm thick. The length of the diamond between electrodes was 0.58 cm and the width of the plate at the Hall electrode was 0.35 cm. The maximum width of the diamond was 0.42 cm. A flat face of the diamond was fixed to a piece of polystyrene, $\frac{1}{16}$ -inch thick, by means of polystyrene cement. The polystyrene sheet S was fastened to the vertical metal plate, P. A sketch of the mounted diamond is shown in Fig. 1(b). Aquadag current electrodes were painted on the diamond, D, and electrical contact made to them by spring brass electrodes. One of these brass electrodes was clamped to the plate, P, in order to cool the diamond effectively. The junction of a copperconstant n thermocouple, T, was placed in contact with this brass electrode in order to determine the temperature of the diamond. The second spring electrode was soldered to an insulated wire, which was wound upon the liquid air reservoirs to prevent heat loss from the diamond. The electrical leads were brought out of the top of the cryostat through individual Kovar-glass seals.

One Hall electrode, H, was used in a circuit originated by Evans.¹² The Hall electrode was a line of Aquadag on one face of the crystal. A thin wire of spring brass, one end of which was imbedded in the polystyrene plate, was pressed against the Aquadag electrode. A thin constantan wire, soldered to the spring brass wire, served as a lead to the electrometer. Polystyrene beads were used to insulate the constantan lead wire, which was brought out of the cryostat top through a double Kovar-glass seal, the outer seal serving as a guard ring. To eliminate a.c. pick-up and insulation problems, the electrometer tube and circuit elements, which were connected to ground through a high impedance, were mounted in a brass can which was an integral part of the cryostat top.

The temperature of the diamond was controlled by providing interchangeable brass and copper spacers between the reservoir, Q, and the vertical plate, P. In addition, the plate, P, and the radiation shield, O, had insulated heating elements wrapped on them.

The interior parts of the cryostat were suspended from a top plate which rested on a rubber gasket to



provide a vacuum-tight seal to the outer jacket. The lower portion of the outer jacket, which surrounded the diamond, was in the form of a rectangular box, $3'' \times 3'' \times \frac{3}{4}''$, which was placed between the poles of the electromagnet. A quartz window, waxed to the cryostat wall, allowed radiation to fall upon the diamond through a hole in the radiation shield. The cryostat was evacuated by a mechanical pump and the pressure determined with a thermocouple gauge.

The optical system is illustrated in Fig. 2. The glass lenses, L, allowed the arc, S, to be placed well outside the field of the electromagnet, M. The rhodium surfaced mirror, R, was used to reflect the radiation, through a right angle, to the diamond, D. The diamond was uniformly illuminated by focusing the image of the rather broad arc upon it. The light source was a 100-watt H4 General Electric mercury arc operated from a voltage regulator. The glass lenses and filters, F, removed radiation of wavelength less than 3000A in order to avoid possible surface photo-emission from the electrodes. The work of Gudden and Pohl indicates that irradiation of diamond with red or infrared light during exposure to ultraviolet radiation results in an enhanced photocurrent. Since this enhanced current may be due to mobile "holes," which would affect the Hall measurements, a one-centimeter thick water cell and Corning No. 4308 filter were used to eliminate radiation of wavelengths greater than 6200A.

A block diagram of the circuit used for the measure-

¹² J. Evans, Phys. Rev. 57, 47 (1940).

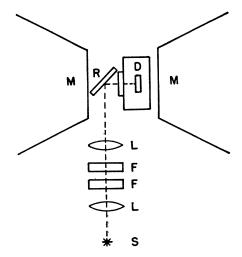


FIG. 2. Optics for illumination of diamond.

ment of the Hall potential and photo-conductive current is illustrated in Fig. 3. In this circuit, originated by Evans,¹² a single Hall electrode is used; and the measured Hall potential difference is one-half that given by the conventional two-electrode circuit, which is inconvenient for use with high resistance materials. With the diamond, D, illuminated, the photocurrent was measured with the high sensitivity ammeter circuit, A. This was a battery operated amplifier using a single VX-41 Victoreen tube and a grid resistance of 10⁹ ohms. The sensitivity of the circuit was 10^{-12} amp/mm deflection of the output galvanometer. The variable tap, T, was adjusted so that the high impedance electrometer circuit, E, indicated that the Hall electrode was at ground potential. After application of the magnetic field, the change in potential of the Hall electrode was compensated by the potentiometer, P. The voltmeter, V, then gave the Hall potential difference.

To reduce a.c. pick-up it was necessary to by-pass all resistors with condensers and to choose with care the proper position for grounding the circuit. The lead from the Hall electrode to the electrometer circuit was shielded by the cryostat jacket. The high impedance electrometer circuit was used only as a null instrument, so that exact calibration was unnecessary. The elec-

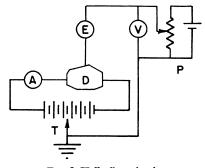


FIG. 3. Hall effect circuit.

trometer consisted of a three-stage battery operated voltage amplifier with an over-all gain of about 40. The effective input impedance of the first stage was raised to better than 10^{13} ohms without increase of the time constant by the use of negative feedback. This large input impedance was necessary because of the high resistance of the diamond.

The electromagnet had five-inch diameter pole pieces, which tapered to a diameter of three inches at the gap. The magnetomotive force was supplied by two coils, each possessing 30,000 turns of small diameter wire and a resistance of 3200 ohms. The maximum magnetic field strength was 5150 oersteds, which was obtained with a coil current of 226 ma. The power supplied to the magnetic coils was current regulated. The emf generated in a small coil placed in the magnet gap and rotated by a synchronous motor was used to determine the dependence of the magnetic field strength upon the magnet coil current.

III. RESULTS

A test of the proportionality between Hall potential and magnetic field strength is shown in Fig. 4. The mag-

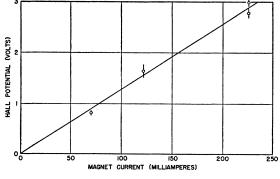


FIG. 4. Hall potential versus magnet current.

netic field was proportional to the magnet coil current, which is used as the independent variable in plotting this data. The potential difference applied to the diamond during this test was 370 volts and the illumination yielded a constant photocurrent of 2×10^{-10} amp.

Within the precision of measurement the Hall potential was found to be proportional to the magnetic field strength. Each point plotted in Fig. 4 (and succeeding graphs) is the average of eight readings with the magnetic field reversed between each pair of readings. In agreement with theory, the magnitude of the Hall voltage was found to be independent of the magnetic field direction, and the sign of the Hall potential changed upon reversal of the magnetic field direction. The relative directions of the applied electric field, magnetic field, and Hall potential were such as to indicate that the photocurrent resulted from a displacement of negatively charged particles.

The Hall potential should be independent of the

magnitude of the photocurrent when the magnetic field strength and applied potential are held constant. The results obtained with a potential of 370 volts applied to the diamond and a magnetic field of 2780 oersteds are shown in Fig. 5. The results of Lenz⁴ are indicated, for comparison, by the crosses of Fig. 6. The photocurrent was varied by changing the intensity of illumination. It is to be observed that in each case the Hall potential decreased and went to zero for small photocurrents but was constant for currents greater than a minimum value which was about 100 times greater in Lenz' work than it was in the present experiment.

It appears probable that the decrease of the Hall potential at small photocurrents is due to the finite input resistance of the electrometers. The data of Fig. 5 are consistent with an electrometer input resistance of about 2×10^{13} ohms, a reasonable value. In making this calculation the resistance of the diamond was taken as the ratio of the applied voltage to the photocurrent. The solid curve of Fig. 6, was calculated with the assumption that the leakage resistance of Lenz' electrometer was 1.8×10^{12} ohms, indicating rather inferior insulation.

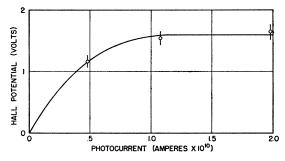
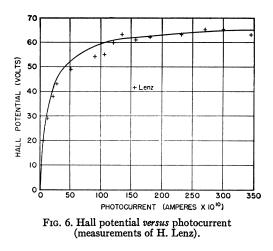


FIG. 5. Hall potential versus photocurrent.

The data of Fig. 7 show the linear dependence of the Hall potential upon the applied potential with constant magnetic field and illumination. The reversal in sign of the Hall potential with reversal of the applied potential is also illustrated. In these experiments the magnitude of the magnetic field was 5150 oersteds and the maximum photocurrent 3×10^{-10} amp. Potentials larger than 600 volts could not be applied to the crystal because of insulation difficulties.

A similar experiment was not performed by Lenz. Figure 8 illustrates his data (solid curve) on Hall potential versus applied potential with the photocurrent maintained constant by proper adjustment of the illumination. In this case, the crystal resistance increases as the voltage is raised. The saturation of the solid curve does not seem to be due, however, to electrometer leakage as has been postulated to explain the data of Figs. 5 and 6. The dashed curve of Fig. 8 was calculated for infinite electrometer resistance on the assumption that the solid curve was taken with an electrometer having a resistance of 1.8×10^{12} ohms.



Although the saturation of Lenz' curves at large potentials is unexplained, the linear form of the curves at moderate potentials indicates that it is possible to define a mobility in this region.

The passage of photocurrent through the diamond resulted in the development of space charge. The photocurrent exhibited a rapid initial decrease, which was accompanied by a change in the potential of the Hall electrode. The rate of change of the potential decreased with time and finally became negligibly small. In a typical experiment, at room temperature, the total applied potential was 225 volts. After a lapse of several minutes the photocurrent reached a steady value and the potential difference between the Hall electrode and the anode was 25 volts with zero applied magnetic field. At 100°K, with the same total potential difference between cathode and anode, the equilibrium Hall electrode potential was 15 volts negative with respect to the anode.

The equilibrium potential distribution inside the

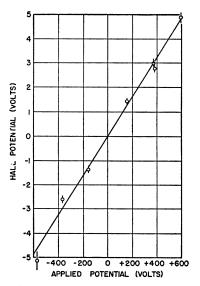


FIG. 7. Hall potential versus applied potential.

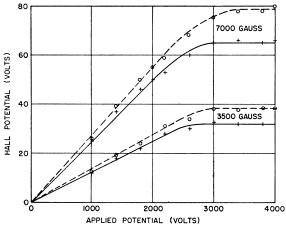


FIG. 8. Hall potential versus applied potential (measurements of H. Lenz).

crystal appears to be similar to that sketched in Fig. 9, with a rapid drop in potential near the cathode. This cathode fall in potential implies a positive space charge adjacent to the cathode. A possible explanation is that electrons cannot enter the crystal from the cathode and that the removal of photo-electrons by the applied field leaves this region of the crystal with a positive space charge of immobile "holes."

In the treatment of the experimental data, the internal electric field has been taken as $E_a = (V_0 - V'')/S$ where V'' is the potential difference between anode and the Hall electrode in zero magnetic field. The distance between the parallel planes containing the anode interface and the Hall electrode is S.

The polarization, defined as (V'/V_0) (Fig. 9) changed slightly but detectably with temperature. Figure 10 is a temperature-polarization curve.

The preliminary observations which have been discussed indicated that the measured Hall potential behaved satisfactorily as a function of magnetic field, photocurrent, and applied potential. The greatest uncertainty in the calculation of a mobility undoubtedly lies in the estimation of the internal electric field, which

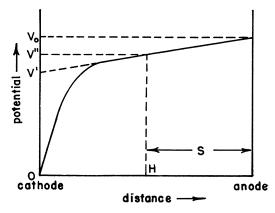


FIG. 9. Probable potential distribution within diamond.

differs from that calculated from the applied potential because of polarization. The results, as a function of temperature are shown in Fig. 11, where the logarithm of the mobility is plotted as a function of the logarithm of temperature. The solid line has the theoretical slope. In these experiments a magnetic field of 2780 oersteds and an applied potential of 265 volts were used. The photocurrent was approximately 7×10^{-10} amp. While the consistency of the data leaves much to be desired, it is clear that an inverse $\frac{3}{2}$ -power dependence of the mobility on the temperature is a much better representation than either an inverse first- or second-power dependence. The room temperature value of the molity is 900 ± 50 cm²/volt-sec.

The present data yield a room temperature mobility of about 200 cm²/volt-sec if the effect of polarization is neglected. There seems to be no justification, however, for this procedure.

The same data which were used to calculate the mobility may be used to compute the mean free path by means of Eq. (3). At room temperature, the mean free path is 6.5×10^{-6} cm.

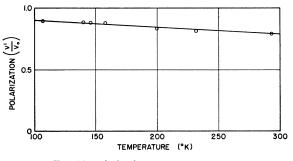


FIG. 10. Polarization versus temperature.

Pearlstein, who has investigated the rise time of the voltage pulses produced in diamond under gammairradiation, has reported¹³ that the room temperature electronic mobility of diamond is greater than 1500 cm²/volt-sec. More recently, he has increased his estimate¹⁴ of the minimum mobility to 4000 cm²/voltsec. It is not immediately evident why our results of the Hall measurements and those of Pearlstein differ so widely; but it is possible that we have overestimated the internal applied field, E_a , because of the presence of an unsuspected abrupt change in potential near the anode.

IV. ADDITIONAL OBSERVATIONS

McKay¹⁵ has reported that positive holes in diamond, produced by electron bombardment, are mobile. Gudden and Pohl⁹ have also observed that the simultaneous illumination of diamond with ultraviolet and infrared radiation results in an enhanced photocurrent,

- ¹⁴ E. Pearlstein, private communication.
 ¹⁵ K. McKay, Phys. Rev. 74, 1606 (1948).

¹³ E. Pearlstein and R. B. Sutton, Phys. Rev. 79, 217 (1950).

which they interpret as being caused by the freeing of positive charges by the long wavelength radiation. High intensities of red and infrared radiation appear to be necessary for the observation of this effect. The diamond used in the present investigation was illuminated simultaneously with radiation from the filtered mercury arc, as previously described, and radiation from a 100-watt tungsten lamp, filtered to remove wavelengths below 5500A. The enhancement of the photocurrent by the radiation from the tungsten lamp was 0.5 percent.

As shown in Fig. 12, the photocurrent excited in the diamond by constant illumination was practically independent of temperature. Lenz found, however, that at 100°K the photocurrent had decreased to 15 percent of its room temperature value. He also noted that the polarization, which was 20 percent at room temperature, increased to 85 percent at 100°K. In the present

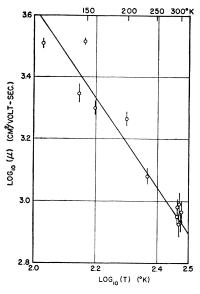


FIG. 11. Mobility versus temperature.

work, the polarization was found to be large (Fig. 10) but, like the photocurrent, practically independent of temperature. The difference between our results and those of Lenz appears to be due to the difference in the nature of the exciting radiation. Lenz used the full radiation of the mercury arc as transmitted by quartz. The diamond used in the present experiments, when illuminated in this manner, exhibited a photocurrent which at 100°K had decreased to 22 percent of its room temperature value.

When the diamond was illuminated with the full radiation of the mercury arc, a pronounced asymmetry in the Hall potential was also observed, the magnitude of the potential depending on the relative directions of the applied electric and magnetic fields. This effect is illustrated by the data of Table I. In Table I are shown typical sets of Hall potential data, taken at 100°K with the diamond excited by unfiltered radiation and by

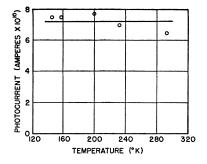


FIG. 12. Photocurrent versus temperature.

radiation filtered in the manner previously described. At room temperature the asymmetry in the Hall potential, as measured with unfiltered radiation, was even larger than at 100°K. When the diamond was excited with unfiltered radiation, the Hall potential assumed a maximum value when the relative direction of the electric and magnetic fields was such as to give the Hall electrode a negative charge. Lenz does not report any asymmetry in the magnitude of the Hall potential, possibly because he used the conventional two-electrode Hall circuit rather than the single Hall electrode used in the present experiments.

The observed asymmetry in the Hall potential may be due to surface photo-emission from the Hall electrode which is produced by the short wavelength radiation of the unfiltered mercury arc. A photocurrent of 10^{-12} amp from the diamond electrodes was observed when unfiltered radiation was used, but the fraction of the current which originated at the Hall electrode was not determined. The photo-conductive process in diamond does appear, however, to depend on the wavelength of the exciting radiation, as evidenced by the behavior of the polarization, the photocurrent, and their temperature dependence. A possible explanation is as follows. The fundamental optical absorption band of diamond begins at 2250A. The long wavelength tail of this band, which may extend into the visible region of the spectrum, is undoubtedly due to impurities. Absorption of radiation in the impurity tail of the fundamental band frees electrons from impurity atoms; the positive

 TABLE I. Asymmetry of Hall potential under full radiation of the mercury arc.

Magnetic field direction	Magnetic field turned	Change in potential of Hall probe (volts) filtered unfiltered radiation radiation	
direction	turnea	radiation	radiation
Forward	on	+0.94	+0.71
	off	-0.82	-0.63
Reversed	on	-0.58	-1.47
	off	+0.70	+1.52
Forward	on	+0.88	+0.68
	off	-0.62	-0.72
Reversed	on	-0.62	-1.51
	off	+0.70	+1.50
	Ave	cage 0.73	

holes are immobile, being bound to the impurity atoms. Under these circumstances the polarization is large and temperature independent. Absorption of radiation in the long wavelength region of the fundamental absorption band is expected to result in the production of excitons. If these are thermally dissociated at room temperature, free electrons and holes are produced, and the polarization is small. At low temperatures, where the excitons are not thermally dissociated, they wander to impurity atoms where dissociation does occur, the electron becoming free but the hole remaining bound to the impurity atom. Under these conditions of excitation, as the temperature is reduced, the polarization increases.

The dissociation energy of an exciton can be estimated from the relation¹⁶

$$E_n = -\pi^2 m e^4 / \mu^4 h^2 n^2 \tag{4}$$

by taking n=1. The index of refraction, μ , of diamond ¹⁶ F. Seitz, Phys. Rev. **76**, 1376 (1949).

PHYSICAL REVIEW

is 2.42 and $E_1 \simeq 0.2$ ev. The lifetime of the exciton is given by

$$\tau = \tau_0 \exp(+E_1/kT). \tag{5}$$

Estimating $\tau_0 \simeq 10^{-12}$ sec, one obtains $\tau(100^{\circ}\text{K}) \simeq 10^{-2}$ sec and $\tau(300^{\circ}\text{K}) \simeq 10^{-9}$ sec. If the cross section for collision with an impurity atom is taken to be 10^{-14} cm², the concentration of impurity atoms of the order of 10^{17} cm⁻³, and the velocity of the exciton as 10^{5} cm/sec, then at 100°K an exciton will make a million collisions with impurity atoms during its thermal lifetime. At room temperature, the corresponding number of collisions is less than unity. The behavior of the exciton is therefore in agreement with the preceeding interpretation.

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Relativistic Quantum Theory for Finite Time Intervals

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If transition probabilities are evaluated for transitions occuring during a finite time interval, additional divergencies occur different from those commonly encountered for infinite time intervals. The expressions obtained can however be made convergent, if an indeterminacy of time is attributed to each epoch of observation. The method is applied to the emission of a photon by a free electron.

I. INTRODUCTION

HE convergent results in the relativistic quantum theory of elementary particles, which have been recently obtained by different authors,¹ apply only to time periods of infinite duration between two observations. If one tries to evaluate transition probabilities for processes which are localized in space-time by a sharply defined boundary (for example two time-like hypersurfaces specifying an initial and final observation), one obtains divergent results. These divergences arise from regions near the boundary, where processes occur without conservation of the momentum-energy component normal to the hypersurface. However, we show here that one can obtain convergent results if diffuse boundaries are introduced. We show in Sec. I that this generalization is possible without affecting the unitarity and causality of the array of probability amplitude forming the S-matrix. In Sec. III, we evaluate, in second-order approximation, the timeindependent probability for the emission of a photon by an electron.

Time, with these unsharp limits, no longer appears as a *parameter*, *t*, whose values t=t' and t=t'' are fixed for the two limits of the *period of evolution*, t''-t'=2T, during which the photon emission takes place. The initial and final epochs themselves, $t\cong t'$ and $t\cong t''$, are now of finite duration, $\Delta t'$ and $\Delta t''$, and must be given in terms of *two probability amplitudes for time*, f'(t) and f''(t), describing the precision with which t' and t'' have been determined. In the probability $dw(\omega)$ that an electron has emitted a photon of frequency between ω and $\omega + d\omega$ during the period considered, the Fourier transforms, $g'(\omega)$ and $g''(\omega)$, of the two probability amplitudes figure as convergence factors for the integral.² We have:

$$dw(\omega) = d\omega(|g''|^2 + |g'|^2)(\omega)n(\omega) = dw''(\omega) + dw'(\omega).$$
(1)

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^{*} Work supported by the Swiss Atomic Energy Commission.

¹S. Tomonaga, Progr. Theor. Phys. 1, No. 2, 27 (1946); J. Schwinger, Phys. Rev. 74, 1439 (1948); 75, 651 (1949); 76, 790 (1949); R. P. Feynman, Phys. Rev. 76, 749, 769 (1949); F. Dyson, Phys. Rev. 75, 486, 1736 (1949).

 $[\]overline{f'(t)} = \exp(i\omega t'); g''(\omega) = \exp(i\omega t')$ correspond the epochs $f'(t) = \delta(t-t')$ and $f''(t) = \delta(t-t')$ of sharply determined time values, for which the integral of Eq. (1) diverges.