

Interference Effects in High-Energy Bremsstrahlung from Crystals*

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An attempt is made to understand the negative results of Panofsky and Saxena and the positive results of Frisch and Olson in terms of the theory of the interference effects in high-energy bremsstrahlung from crystals worked out by Überall. Several of the theoretical approximations are examined in detail: the validity of the Born approximation, the calculation of temperature effects, the validity of the closure approximation for the crystal lattice, and the use of the Debye form for the lattice vibration spectrum. It is concluded that all of these are justified, except that a partial failure of the Born approximation may be responsible for the nonappearance of the central minimum in the Frisch-Olson experiment. Crystal imperfections and multiple and inelastic scattering of the primary electrons are also considered briefly, but are found to be unimportant. No explanation is found for the Panofsky-Saxena results. The interference should be enhanced by making the primary electron energy as large as possible and the ratio of photon to electron energy as small as possible. In the case of a diamond-type crystal the [110] direction for the electron beam is to be preferred to either the [100] or the [111] direction. Little advantage is to be gained from cooling the crystal.

I. INTRODUCTION

THE interference effects to be expected in the production of bremsstrahlung by high-energy electrons in single crystal targets have been discussed in several papers,¹ most recently and in greatest detail by Überall.^{2,3} Two experiments have been performed; by Panofsky and Saxena,⁴ and by Frisch and Olson.⁵ The first of these does not show a significant intensity variation as the angle between the primary electron beam and the crystal axis is varied, although an increase of about 25% is expected for very small angles. The second experiment shows an increase of a factor 2, which is reported to be smaller than expected. The present paper is an attempt to understand the relation between the experimental results and the theoretical predictions.

The next four sections are devoted to a detailed examination of some of the approximations made by Überall: the use of the Born approximation (Sec. II); the computation of the effects of thermal vibrations of the crystal lattice under the assumption that any change in the phonon occupation numbers is permissible (we refer to this as the closure approximation) (Sec. III); the validity of the closure approximation (Sec. IV); the use of the simple Debye form for the vibration spectrum of the lattice (Sec. V). Finally, in Sec. VI, we summarize these theoretical results, and discuss their relation to the experiments.

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¹ E. J. Williams, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **13**, No. 4 (1935); B. Ferretti, Nuovo cimento **7**, 118 (1950); M. L. Ter-Mikaelyan, Zhur. Eksptl. i Teoret. Fiz. **25**, 289, 296 (1953); F. J. Dyson and H. Überall, Phys. Rev. **99**, 604 (1955).

² H. Überall, Phys. Rev. **103**, 1055 (1956).

³ H. Überall, CERN Report 58-21, September, 1958 (unpublished); also private communications to W. K. H. Panofsky.

⁴ W. K. H. Panofsky and A. N. Saxena, Phys. Rev. Letters **2**, 219 (1959).

⁵ O. R. Frisch and D. N. Olson, Phys. Rev. Letters **3**, 141 (1959).

II. VALIDITY OF THE BORN APPROXIMATION

It is customary to use the Born approximation in calculating bremsstrahlung when the atomic number Z of the atom that scatters the electron is not too large. However, even for small Z , the phase change of the wave function of an electron in traversing a crystal can be very large, and the validity of an approximation that uses plane waves for the incident and scattered electrons should be questioned. The effect of the periodic potential $V(\mathbf{r})$ of the crystal lattice on the electron can be taken into account by using Bloch wave functions $u(\mathbf{r}) \exp(i\mathbf{k} \cdot \mathbf{r})$ in place of plane waves, where $u(\mathbf{r})$ has the periodicity of the lattice and $\exp(i\mathbf{k} \cdot \mathbf{r})$ satisfies periodic boundary conditions at the edges of the crystal. This substitution has the effect of replacing matrix elements of the form

$$\int \exp(-i\mathbf{k}_2 \cdot \mathbf{r}) V(\mathbf{r}) \exp(i\mathbf{k}_1 \cdot \mathbf{r}) d^3r \quad (1)$$

by

$$\int \exp(-i\mathbf{k}_2 \cdot \mathbf{r}) [\bar{u}_2(\mathbf{r}) V(\mathbf{r}) u_1(\mathbf{r})] \exp(i\mathbf{k}_1 \cdot \mathbf{r}) d^3r. \quad (2)$$

Now the interference effects in crystal bremsstrahlung are expected to arise from a combination of two factors. (a) Owing to the lattice periodicity, the electron-scattering matrix elements can be large only when the momentum transfer vector \mathbf{q} is very close to a vector of the reciprocal lattice.⁶ For an expression of the form (1), $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$, where \mathbf{k}_1 and \mathbf{k}_2 are related to the initial and final electron momenta \mathbf{k}_0 and \mathbf{k}_f and the radiated photon momentum \mathbf{k}_γ , in such a way that

$$\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_f - \mathbf{k}_\gamma. \quad (3)$$

(b) Owing mainly to the requirement of energy conservation, the radiation probability is large only when

⁶ We normalize the reciprocal lattice vectors \mathbf{g} so that their scalar products with vectors of the primitive space lattice are integer multiples of 2π .

\mathbf{q} lies in a thin pancake-shaped region that has the following properties²: it is oriented with its plane perpendicular to the direction \mathbf{k}_0 of the primary electron; its radius in this plane is of order the electron mass μ ; the perpendicular distance from the origin to the nearer surface of the region is equal to $\mu^2 k_\gamma / 2k_0 k_f$, and the thickness of the region is also of this order of magnitude. We choose units such that $\hbar = c = 1$.

These two factors imply that the intensity of the radiation will depend on the number of reciprocal lattice points that are contained in the pancake-shaped region, which in turn depends on the orientation of the region and hence on the direction of the primary electrons with respect to the crystal lattice. There are now two ways in which we can show that the substitution of (2) for (1) has no substantial effect on the intensity. We can say that the square bracket in (2) has the same lattice periodicity as $V(\mathbf{r})$ in (1), so that the requirement that \mathbf{q} be very close to a reciprocal lattice vector is not affected by the replacement of plane waves by Bloch wave functions. Or we can say that $u(\mathbf{r})$ can be written as a Fourier series in reciprocal lattice vectors, so that the Bloch wave function is a superposition of plane waves whose \mathbf{k} vectors differ from the original \mathbf{k} by reciprocal lattice vectors. From the latter point of view, any of these \mathbf{k} 's can be used to specify the electron momentum, so that \mathbf{k}_0 , \mathbf{k}_f , and hence \mathbf{q} are not defined within a reciprocal lattice vector. But such a shift in the origin of the pancake-shaped region has no effect on the number of reciprocal lattice points it contains. We therefore expect no qualitative change in the interference effects to arise from the use of Bloch wave functions in place of plane waves in the matrix elements.⁷

There is, however, another effect to be considered. The energy of a free electron is related to its momentum through $E = (k^2 + \mu^2)^{1/2}$; this $E-k$ relation will be altered for an electron in a periodic potential. If we replace the free electron relation by

$$E = (k^2 + \mu^2)^{1/2} + \epsilon, \quad (4)$$

where ϵ is expected to be of the order of the lattice potential V , we find that the location of the pancake-shaped region described in (b) above is changed. The distance from the origin to its nearest surface is changed from

$$\mu^2 k_\gamma / 2k_0 k_f \quad (5)$$

to

$$(\mu^2 k_\gamma / 2k_0 k_f) - (\epsilon_0 - \epsilon_f), \quad (6)$$

where ϵ_0 and ϵ_f are the quantities defined by (4) for the initial and final electrons, respectively. Now an ϵ is expected to be largest when its corresponding \mathbf{k} approaches the boundary of a Brillouin zone. In the

⁷ This lack of definiteness of the electron momentum vectors is related to the extinction effect known in connection with the diffraction of x rays by crystals. The writer is indebted to F. Bloch for discussion of the extinction effect.

experiments^{4,5} the direction of \mathbf{k}_0 is accurately oriented along or very close to a crystal axis. However, the electron beam would have to have its energy defined within a few thousand electron volts, and its direction defined within about 10^{-5} radian, in order that its \mathbf{k}_0 not cross a zone boundary. Thus we can expect that in practice the second parenthesis in (6) will have a range of values. If this range is comparable with the reciprocal lattice spacing, it is apparent that the accompanying shift in the pancake-shaped region will tend to wash out the interference effect. If the range is comparable with (5), which is much smaller than the reciprocal lattice spacing in the situations of interest, the smeared-out pancake-shaped region will contain the reciprocal lattice points that lie in the plane through the origin perpendicular to \mathbf{k}_0 even when \mathbf{k}_0 is along a crystal axis; this will have the effect of washing out the minimum predicted by Überall when the angle between \mathbf{k}_0 and a crystal axis is precisely zero, but will still leave an interference maximum for small values of this angle. Finally, if the range is much smaller than (5), the Born approximation will be valid.

We therefore proceed to estimate the quantity ϵ that appears in (4). The second-order Dirac equation may be written

$$(E - V)^2 \psi = (-\nabla^2 + \mu^2) \psi + i(\boldsymbol{\alpha} \cdot \text{grad} V) \psi. \quad (7)$$

We wish to keep the terms of order EV , but not of order V^2 , on the left side. The last (spin) term on the right side is of order $\text{grad} V$, and hence small in comparison with EV for the high-energy case of interest here. We thus approximate (7) by

$$(E^2 - 2EV) \psi = (-\nabla^2 + \mu^2) \psi, \quad (8)$$

and ignore the spinor character of ψ . For a particular state i of energy E_i , ψ_i may be expanded in terms of plane waves with momenta \mathbf{k}_j that differ from each other by a reciprocal lattice vector:

$$\psi_i = \sum_j A_{ij} \exp(i\mathbf{k}_j \cdot \mathbf{r}). \quad (9)$$

What we wish to do is diagonalize the energy for a group of n plane-wave states that are connected to each other by matrix elements of V and whose unperturbed energies differ by amounts that are less than or of order V . These plane waves may be thought of as having \mathbf{k} vectors that cluster about the nominal vector \mathbf{k}_0 for the group (which we have already seen is not defined within a reciprocal lattice vector), and whose displacements from \mathbf{k}_0 are a small number of reciprocal lattice unit vectors. We associate an unperturbed E_j^0 with each vector \mathbf{k}_j , and E_0 with \mathbf{k}_0 , in accordance with the energy-momentum relation for a free electron.

Substitution of (9) into (8) yields, to an approximation that is consistent with the neglect of the V^2 term in (7), the following set of equations for the

energy eigenvalues of (8):

$$(E_i - E_0)A_{ij} = (E_j^0 - E_0)A_{ij} + \sum_l A_{il}V_{lj}, \quad (10)$$

where i, j, l run from 1 to n ; each term in these equations is of order V , and

$$V_{ij} = \Omega^{-1} \int V(\mathbf{r}) \exp[i(\mathbf{k}_i - \mathbf{k}_j) \cdot \mathbf{r}] d^3r, \quad (11)$$

where Ω is the volume of the entire crystal. It follows from (10) that $(E_i - E_0 - V_0)$ are the eigenvalues of the matrix $(E_j^0 - E_0 - V_0)\delta_{ij} + V_{ij}$, where

$$V_0 = V_{ii} = \Omega^{-1} \int V(\mathbf{r}) d^3r.$$

Thus by taking traces of the matrix and its square we find that

$$\sum_i (E_i - E_0 - V_0) = \sum_i (E_i^0 - E_0), \quad (12)$$

$$\sum_i (E_i - E_0 - V_0)^2 = \sum_i (E_i^0 - E_0)^2 + \sum_i \sum_{j \neq i} |V_{ij}|^2. \quad (13)$$

If we start from a set of unperturbed states that have nearly the same energy, the right side of (12) will be very small, and the average of the perturbed energies E_i will be very close to $E_0 + V_0$. The first term on the right side of (13) will also be very small in this case, and the mean square deviation of E_i from its average will be very close to

$$n^{-1} \sum_i \sum_{j \neq i} |V_{ij}|^2 \approx \sum_{j \neq i} |V_{ij}|^2. \quad (14)$$

The right side of (14) is obtained from the left side by noting that the i -summation results approximately in a factor n . The shift V_0 in the average E_i is not significant, since it appears in both the initial and the final electron states, and hence drops out of (6). Thus (14) provides a simple estimate for the square of one of the e 's that appear in (6).

The lattice potential $V(\mathbf{r})$ may be written

$$V(\mathbf{r}) = \sum_{\mathbf{L}} v(\mathbf{r} - \mathbf{L}),$$

where $v(\mathbf{r})$ is the atomic potential and \mathbf{L} is the vector position of any atom (for a composite lattice, \mathbf{L} includes the atoms within the unit cell as well as the primitive space lattice vectors). Thus, with $\mathbf{g} = \mathbf{k}_i - \mathbf{k}_j$, which is a reciprocal lattice vector, (11) becomes:

$$V_{ij} = \Omega^{-1} \sum_{\mathbf{L}} \exp(i\mathbf{g} \cdot \mathbf{L}) \int v(\mathbf{r}) \exp(i\mathbf{g} \cdot \mathbf{r}) d^3r. \quad (15)$$

For a diamond lattice (the experiments were performed with silicon and germanium) the summation over \mathbf{L} may be expressed as the resultant of a summation over the eight atoms in a fundamental cube of edge length

a , and a summation over the primitive space lattice which is simple cubic. In this case, the \mathbf{g} 's form a simple cubic lattice with edge length $2\pi/a$. If there are N atoms in the crystal, the second part of the \mathbf{L} -sum is equal to $N/8$, while the first part yields the structure factor $S(\mathbf{g})$. Since $N/8\Omega = 1/a^3$, (15) may be written:

$$V_{ij} = S(\mathbf{g})a^{-3} \int v(\mathbf{r}) \exp(i\mathbf{g} \cdot \mathbf{r}) d^3r. \quad (16)$$

It is easily verified that $S(\mathbf{g})$ vanishes except when \mathbf{g} lies on either of two parallel lattices: (1) a face-centered cubic lattice of edge length $8\pi/a$ with a lattice point at the origin, for which $S(\mathbf{g}) = 8$; (2) a simple cubic lattice of edge length $4\pi/a$ located such that the origin is at the center of a lattice cube, for which $S(\mathbf{g}) = 4(1 \pm i)$ at alternate lattice points (this may also be described as a sodium chloride lattice of edge length $8\pi/a$). A convenient approximate form for $v(\mathbf{r})$ is $-(Ze^2/r) \times \exp(-r/b)$, with $b = \hbar^2/mc^2Z^3$; in the case of silicon this is quite close to the potential (self-consistent without exchange) computed by Mullaney⁸ for the important region of small r and large v . Then the integral in (16) becomes:

$$\int v(\mathbf{r}) \exp(i\mathbf{g} \cdot \mathbf{r}) d^3r = -4\pi Ze^2 b^2 / (1 + g^2 b^2). \quad (17)$$

In computing the right side of (14) we are only interested in including those states whose unperturbed energies differ by amounts that are less than or of order V_{ij} . From (16) and (17) we find that V_{ij} is less than or of order 5 eV for silicon and germanium. Now if \mathbf{g} is parallel to \mathbf{k}_0 the unperturbed energy difference is of the order of g , which is at least several thousand electron volts; on the other hand if \mathbf{g} is perpendicular to \mathbf{k}_0 the unperturbed energy difference is roughly $g^2/2k_0$, which is of order a few hundredths of an electron volt. Thus we include in the j -summation of (14) only the \mathbf{g} 's that lie in a plane perpendicular to \mathbf{k}_0 . For an order-of-magnitude estimate we replace the sum by an integral over the plane:

$$\sum_{j \neq i} |V_{ij}|^2 \approx [2\pi\sigma(4\pi Ze^2 b^2)^2 / a^6] \int_0^\infty g dg / (1 + g^2 b^2)^2 = 16\pi^3 \sigma Z^2 e^4 b^2 / a^6. \quad (18)$$

Here σ is defined so that $2\pi g \sigma dg$ is the average sum of $|S(\mathbf{g})|^2$ over the annular ring of radius g and width dg ; the infinite upper limit can be used since the integral converges. The value of σ evidently depends on the direction of \mathbf{k}_0 with respect to the crystal axes; for \mathbf{k}_0 along the [100] axis $\sigma = 2(a/\pi)^2$, for \mathbf{k}_0 along the 110 axis $\sigma = 8^3(a/\pi)^2$, and for \mathbf{k}_0 along the [111] axis $\sigma = (4/3^3)(a/\pi)^2$.

In the Panofsky-Saxena experiment, silicon was used

⁸ J. F. Mullaney, Phys. Rev. 66, 326 (1944).

and the electron beam was along the [100] axis.⁴ Then $Z=14$, $a=5.43 \times 10^{-8}$ cm and $b=2.20 \times 10^{-9}$ cm, so that the square root of (18) is about 13 ev. For comparison, the quantity (5), with $k_0=600$ Mev and $k_\gamma=235$ Mev, is about 140 ev. Thus we cannot account for the absence of interference in this experiment in terms of a failure of the Born approximation.

In the Frisch-Olson experiment germanium was used and the electron beam was along the [110] axis.⁵ Then $Z=32$, $a=5.66 \times 10^{-8}$ and $b=1.67 \times 10^{-9}$ cm, so that the square root of (18) is about 29 ev. For comparison the quantity (5), with $k_0=1$ Bev and $k_\gamma=100$ Mev, is about 15 ev; the distance between adjacent reciprocal lattice planes is $8\frac{1}{2}\pi/a=3100$ ev. Thus the inadequacy of the Born approximation in this case is expected to remove the central minimum, as is observed, but not to wash out the interference maximum.

III. TEMPERATURE EFFECTS ASSUMING THE CLOSURE APPROXIMATION

The basic process under consideration is the transition from a state in which the electron has momentum \mathbf{k}_0 and the crystal lattice has a set of phonon occupation numbers m_i to a state in which the electron has momentum \mathbf{k}_f , the photon has momentum \mathbf{k}_γ , and the lattice has phonon occupation numbers n_i . We assume in this section that the values of the n_i are not observationally significant, so that all possible sets can be summed over. We call this the closure approximation, and inquire into its validity in the next section.

The basic matrix element may be written

$$\langle n_1, n_2, \dots | \sum_{\mathbf{L}} \exp i\mathbf{q} \cdot (\mathbf{L} + \mathbf{u}_{\mathbf{L}}) | m_1, m_2, \dots \rangle \times M(\mathbf{k}_0, \mathbf{k}_f, \mathbf{k}_\gamma), \quad (19)$$

where \mathbf{q} is given by (3), $\mathbf{u}_{\mathbf{L}}$ is the displacement of the atom at the lattice site \mathbf{L} from its equilibrium position, and M is the radiative matrix element for a single atom. The closure approximation consists in neglecting the dependence of M on the energy change of the lattice, and summing the squared matrix element over the n_i for fixed \mathbf{q} . We therefore have to calculate the expectation value of

$$|\sum_{\mathbf{L}} \exp i\mathbf{q} \cdot (\mathbf{L} + \mathbf{u}_{\mathbf{L}})|^2 \quad (20)$$

for the m_i that correspond to the temperature T . Physically this approximation is equivalent to the assumption that the bremsstrahlung can be calculated from atoms at fixed positions ($\mathbf{L} + \mathbf{u}_{\mathbf{L}}$), and the resulting intensity then averaged over the temperature distribution of the $\mathbf{u}_{\mathbf{L}}$.

For a large crystal containing N atoms the expectation value of (20) may be written

$$\sum_{\mathbf{L}} \sum_{\mathbf{L}'} \exp i\mathbf{q} \cdot (\mathbf{L} - \mathbf{L}') \langle \exp i\mathbf{q} \cdot (\mathbf{u}_{\mathbf{L}} - \mathbf{u}_{\mathbf{L}'}) \rangle_T = N \sum_{\mathbf{l}} f_T(\mathbf{l}) \exp i\mathbf{q} \cdot \mathbf{l}, \quad (21)$$

where $\mathbf{l} = \mathbf{L} - \mathbf{L}'$ and the dependence of

$$f_T(\mathbf{l}) = \langle \exp i\mathbf{q} \cdot (\mathbf{u}_{\mathbf{L}} - \mathbf{u}_{\mathbf{L}'}) \rangle_T \quad (22)$$

on \mathbf{L} and \mathbf{L}' separately (surface effect) is neglected. The computation of (22) proceeds by expanding $\mathbf{u}_{\mathbf{L}}$ in terms of quantized waves (phonons) of propagation vector \mathbf{k} . In actuality, there are $N/8$ such independent vectors that fill uniformly a cell of volume $(2\pi/a)^3$ in reciprocal lattice space, and for each \mathbf{k} there are 24 independent modes that arise from the three coordinates of the eight atoms in the fundamental cube of edge length a .⁹ However, we shall only require rather general properties of f_T that are independent of the crystal structure. It is, then, sufficient for our computation to use a simple cubic lattice instead of a diamond lattice, and this results in a considerable simplification. The space lattice constant is then $a/2$, and there are N independent phonon propagation vectors \mathbf{k} that fill uniformly a cell of volume $(4\pi/a)^3$ in reciprocal lattice space. The lattice frequency spectrum $\omega(\mathbf{k})$ determines the phonon occupation numbers for the particular value of T .

Several calculations related to the evaluation of (22) appear in the literature, in connection with ferromagnetism,¹⁰ the diffuse spots in x-ray diffraction,¹¹⁻¹³ and slow neutron capture¹⁴ and scattering.¹⁵ We therefore omit the details here, and simply quote the result:

$$f_T(\mathbf{l}) = \exp - \sum_{\mathbf{k}} \left\{ [q^2/2MN\omega(\mathbf{k})](1 - \cos \mathbf{k} \cdot \mathbf{l}) \times \left[1 + \frac{2}{\exp[\omega(\mathbf{k})/\kappa T] - 1} \right] \right\}. \quad (23)$$

Here κ is Boltzmann's constant and M is the atomic mass.

In the remainder of this section we use the simple Debye form

$$\omega(\mathbf{k}) = vk \quad (24)$$

for the vibrational spectrum of the lattice, where v is the velocity of sound in the crystal, and approximate the summation in (23) by an integration, $N(a/4\pi)^3 \int d^3k$, over a sphere of radius

$$k_m = \omega_m/v = (4\pi/a)(3/4\pi)^{\frac{1}{3}} = \kappa\Theta/v, \quad (25)$$

where Θ is the Debye temperature. The summation in (23) then becomes

$$-\ln f_T(\mathbf{l}) = A_0 q^2 \left\{ \left[1 - \frac{\sin^2(k_m l/2)}{(k_m l/2)^2} \right] + 4(T/\Theta)^2 \times \int_0^{\Theta/T} \left[1 - \left(\frac{\sin \lambda y}{\lambda y} \right) \right] \frac{y dy}{\exp y - 1} \right\}, \quad (26)$$

⁹ See for example R. E. Peierls, *Quantum Theory of Solids* (Oxford University Press, New York, 1955), Chap. I.

¹⁰ F. Bloch, *Z. Physik* **74**, 295 (1932).

¹¹ H. Ott, *Ann. Physik* **23**, 169 (1935).

¹² M. Born and K. Sarginson, *Proc. Roy. Soc. (London)* **A179**, 69 (1942).

¹³ H. A. Jahn, *Proc. Roy. Soc. (London)* **A179**, 320 (1942).

¹⁴ W. E. Lamb, Jr., *Phys. Rev.* **55**, 190 (1939).

¹⁵ A. C. Zemach and R. J. Glauber, *Phys. Rev.* **101**, 118 (1956).

where $A_0 = 3/4M\kappa\Theta$ and $\lambda = k_m T/\Theta$. The corresponding expression obtained by Überall² is contained in his Eqs. (35) and (36), and differs from our Eq. (26) because of approximations made by him. Now the $f_T(\mathbf{l})$ given by (26) is equal to unity for $\mathbf{l}=0$, and approaches $\exp(-Aq^2)$ as \mathbf{l} becomes very large, where

$$A = A_0 \left[1 + 4(T/\Theta)^2 \int_0^{\Theta/T} \frac{y dy}{\exp y - 1} \right]. \quad (27)$$

Überall approximates further by using this asymptotic value of $f_T(\mathbf{l})$ for all \mathbf{l} different from zero. Since $f_T(\mathbf{l})$ actually lies between unity and its asymptotic value for moderate values of \mathbf{l} , it is apparent that Überall's calculation underestimates the correlation between the motions (zero-point and thermal) of near neighbor atoms in the crystal, and hence underestimates the interference effect in bremsstrahlung.

The additional correlation may be estimated in the following way. While $f_T(\mathbf{l})$ only has physical meaning when \mathbf{l} is a space lattice vector, Eq. (23) or (26) defines it as a continuous function of \mathbf{l} . The right side of (21) can therefore be rewritten:

$$\begin{aligned} N \sum_{\mathbf{l}} f_T(\mathbf{l}) \exp(i\mathbf{q} \cdot \mathbf{l}) \\ &= N \int d^3l' f_T(\mathbf{l}') \exp(i\mathbf{q} \cdot \mathbf{l}') \sum_{\mathbf{l}} \delta(\mathbf{l} - \mathbf{l}') \\ &= N \int d^3l' f_T(\mathbf{l}') \exp(i\mathbf{q} \cdot \mathbf{l}') \sum_{\mathbf{l}} (2\pi)^{-3} \int d^3k \exp i\mathbf{k} \cdot (\mathbf{l} - \mathbf{l}') \\ &= N \int d^3k \sum_{\mathbf{l}} \exp(i\mathbf{k} \cdot \mathbf{l}) \phi_T(\mathbf{q} - \mathbf{k}), \end{aligned}$$

where

$$\phi_T(\mathbf{k}) = (2\pi)^{-3} \int d^3l f_T(\mathbf{l}) \exp(i\mathbf{k} \cdot \mathbf{l})$$

is the Fourier transform of $f_T(\mathbf{l})$. In evaluating the \mathbf{l} -summation it is necessary to use the true crystal structure, since it is at this point that the dependence of the pancake-shaped region on its orientation with respect to the reciprocal lattice arises. It is sufficient to evaluate it for an infinitely large crystal, remembering that $\mathbf{l} = \mathbf{L} - \mathbf{L}'$ is itself a space lattice vector:

$$\sum_{\mathbf{l}} \exp(i\mathbf{k} \cdot \mathbf{l}) = (2\pi/a)^3 \sum_{\mathbf{g}} S(\mathbf{g}) \delta(\mathbf{k} - \mathbf{g}),$$

where $S(\mathbf{g})$ is the structure factor defined below Eq. (16). Thus the right side of (21) becomes:

$$N \sum_{\mathbf{l}} f_T(\mathbf{l}) \exp(i\mathbf{q} \cdot \mathbf{l}) = N (2\pi/a)^3 \sum_{\mathbf{g}} S(\mathbf{g}) \phi_T(\mathbf{q} - \mathbf{g}). \quad (28)$$

Now if $f_T(\mathbf{l})$ were equal to unity for all \mathbf{l} (complete correlation, so that the $\mathbf{u}_{\mathbf{l}}$ all equal each other), $\phi_T(\mathbf{k})$ would be equal to $\delta(\mathbf{k})$, and the interference would be that corresponding to a perfect crystal with no zero-point or thermal motion of the atoms. If $f_T(\mathbf{l})$ were to

approach zero for large \mathbf{l} , $\phi_T(\mathbf{k})$ would have a finite maximum at $\mathbf{k}=0$, and fall off to zero for large \mathbf{k} . Actually, as we have seen, $f_T(\mathbf{l})$ approaches $\exp(-Aq^2)$ for large \mathbf{l} ; thus $\phi_T(\mathbf{k})$ can be written

$$\phi_T(\mathbf{k}) = \delta(\mathbf{k}) \exp(-Aq^2) + \phi_T'(\mathbf{k}), \quad (29)$$

where $\phi_T'(\mathbf{k})$ has a finite maximum at $\mathbf{k}=0$ and falls off to zero for large \mathbf{k} .

As pointed out near the beginning of Sec. II, interference arises because of the inclusion of varying numbers of reciprocal lattice points within the pancake-shaped region as the direction of the primary electron is varied. Überall's approximations are equivalent to the assumption that $\phi_T'(\mathbf{k})$ in (29) is nearly constant over a reciprocal lattice spacing. Since it is actually peaked at $\mathbf{k}=0$, a portion of it should be included with the δ -function in computing the change in intensity as the primary electron direction is varied. While it is difficult to make a quantitative computation of the magnitude of this effect, it is readily seen that it is quite small. It can only be significant when the damping factor $\exp(-Aq^2)$ is appreciably smaller than unity, since only then can it matter if part of the continuum associated with $\phi_T'(\mathbf{k})$ is included with the δ -function. However, the bremsstrahlung cross section falls off with increasing q because of the atomic form factor, and q values much greater than the reciprocal of the screening radius¹⁶ ($111/\mu Z^3$) do not contribute in an important way. For this value of q the damping factor is equal to $\exp -A(\mu Z^3/111)^2 = 0.87$ for silicon at room temperature ($Z=14$, $A=280/\mu^2$). A rough numerical estimate indicates that the theoretically expected interference effect found by Überall should be increased by only a few percent. This is too small to be significant in connection with the present experiments.

IV. VALIDITY OF THE CLOSURE APPROXIMATION

The closure approximation is expected to be valid if the energy

$$\epsilon = \sum_i (n_i - m_i) \omega_i, \quad (30)$$

transferred to the crystal in the basic process described by the matrix element (19) is too small to be observationally significant. Since this ϵ appears in the same way as the quantity $(\epsilon_0 - \epsilon_f)$ in (6), it must be at least of order 10 eV before it can be of importance for the present experiments. To find the distribution with respect to ϵ we multiply the squared matrix element (19) by

$$\begin{aligned} \delta[\epsilon - \sum_i (n_i - m_i) \omega_i] \\ &= (2\pi)^{-1} \int_{-\infty}^{\infty} dt \exp i[\epsilon - \sum_i (n_i - m_i) \omega_i] t \end{aligned}$$

before summing over the n_i .¹⁴ The summation over the n_i , and the calculation of the expectation value of the

¹⁶ L. I. Schiff, Phys. Rev. **83**, 252 (1951).

result for the m_i that correspond to the temperature T , can then be carried through by making use of an elegant formalism developed by Zemach and Glauber.¹⁵ This formalism replaces the factor $\exp(-i\sum_i n_i \omega_i t)$ by $\exp(-iHt)$ operating on the final state, and $\exp(i\sum_i m_i \omega_i t)$ by $\exp(iHt)$ operating on the initial state, where H is the Hamiltonian for the lattice vibrations. The amplitudes of the lattice normal modes can then be replaced by t -dependent amplitudes.

The result of the calculation is that the quantity $f_T(\mathbf{l})$ defined by (22) is replaced by

$$f_T(\epsilon, \mathbf{l}) = (2\pi)^{-1} \int_{-\infty}^{\infty} dt \exp(i\epsilon t) \times \exp - \sum_{\mathbf{k}} [q^2/2MN\omega(\mathbf{k})] \cdot \left\{ [1 - \cos\omega(\mathbf{k})t \cos\mathbf{k} \cdot \mathbf{l}] \times \left[1 + \frac{2}{\exp[\omega(\mathbf{k})/\kappa T] - 1} \right] + i \sin\omega(\mathbf{k})t \cos\mathbf{k} \cdot \mathbf{l} \right\}. \quad (31)$$

It is apparent that $\int_{-\infty}^{\infty} f_T(\epsilon, \mathbf{l}) d\epsilon$ is equal to $f_T(\mathbf{l})$ given in (23), as of course it must be.

Equation (31) has a quite complicated structure, even when the Debye approximation for $\omega(\mathbf{k})$ is made. Its general dependence on ϵ can, however, be inferred by inspection. Thus if all $\omega(\mathbf{k})$ were zero, $f_T(\epsilon, \mathbf{l})$ would be proportional to $\delta(\epsilon)$. The actual range of $\omega(\mathbf{k})$ determines the scale of the dependence of the second exponential on t , through the factors $\cos\omega(\mathbf{k})t$ and $\sin\omega(\mathbf{k})t$. The smallest scale of the t -dependence is of order $1/\omega_{\max}$, so that the largest range of the ϵ -dependence of $f_T(\epsilon, \mathbf{l})$ is of order ω_{\max} . From (25), $\omega_{\max} \approx \kappa\Theta$, which for silicon and germanium is somewhat less than 0.1 ev. Thus the spread of the ϵ -dependence of $f_T(\epsilon, \mathbf{l})$ about $\epsilon=0$ is so small that it can be ignored, and the closure approximation is excellent in this situation.

It might seem at first that the \mathbf{k} -summation in (31) would increase the effective value of $\omega(\mathbf{k})$ to a much larger value, perhaps even as large as $N\omega(\mathbf{k})$. However, the \mathbf{k} -summation over $3N$ lattice modes is balanced by the factor N in the denominator, so that it is actually the individual mode frequencies that are significant. A more physical way of seeing this is to note that the effective mass of a lattice mode oscillator is not of the order of the atomic mass M , but rather of the order of the entire crystal mass NM .¹⁴ This means that the transition probability for emission or absorption of n phonons is proportional to N^{-n} . Thus the important transitions for large N are those in which n different modes each emit or absorb a single phonon, not those in which any one mode emits or absorbs more than one phonon. The number of combinations of n modes is $(3N)!/n!(3N-n)!$, from which it follows that the expectation value of the energy transfer must be of order $\omega(\mathbf{k})$, as was inferred from inspection of (31).

V. LATTICE VIBRATION SPECTRUM

It is generally recognized that the vibrational spectra of silicon and germanium differ significantly from the simple Debye form.^{3,17-21} The spectrum may be expressed in terms of the number of lattice modes $n(\omega)d\omega$ that have angular frequencies between ω and $\omega+d\omega$. A knowledge of $n(\omega)$, together with the assumption of isotropy, makes it possible to reduce (23) to a form analogous to that given in (26) for the Debye spectrum.

Integration over the polar angles of \mathbf{k} in (23) leads to

$$-\ln f_T(\mathbf{l}) = (q^2 a^3 / 32\pi^2 M) \int_0^{k_m} \left(1 - \frac{\sin kl}{kl} \right) \times \left[1 + \frac{2}{\exp(\omega/\kappa T) - 1} \right] (k^2/\omega) dk, \quad (32)$$

where $k_m = (4\pi/a)(3/4\pi)^{1/3}$. If we assume there is a single-valued relation between ω and k , this relation can be determined from the condition

$$n(\omega)d\omega = 4\pi k^2 dk, \quad (33)$$

which may be integrated to give

$$\int_0^{\omega} n(\omega') d\omega' = 4\pi \int_0^k k'^2 dk' = 4\pi k^3/3. \quad (34)$$

We make use of (33) to change the variable of integration in (32) from k to ω :

$$-\ln f_T(\mathbf{l}) = (q^2 a^3 / 128\pi^3 M) \int_0^{\omega_m} \left(1 - \frac{\sin kl}{kl} \right) \times \left[1 + \frac{2}{\exp(\omega/\kappa T) - 1} \right] n(\omega) d\omega/\omega, \quad (35)$$

where k is given in terms of ω by (34). The normalization of $n(\omega)$ can also be found from (34):

$$\int_0^{\omega_m} n(\omega) d\omega = 4\pi k_m^3/3 = (4\pi/a)^3. \quad (36)$$

In order to estimate the effect of replacing the Debye form for $n(\omega)$ by a more realistic spectrum, we compute the asymptotic value of (35) when $\mathbf{l} \rightarrow \infty$. This means that the quantity A in (27) is to be replaced by

$$B = \frac{\int_0^{\omega_m} \left[1 + \frac{2}{\exp(\omega/\kappa T) - 1} \right] n(\omega) d\omega/\omega}{2M \int_0^{\omega_m} n(\omega) d\omega}, \quad (37)$$

¹⁷ H. Cole and E. Kineke, Phys. Rev. Letters **1**, 360 (1958).

¹⁸ J. C. Phillips, Phys. Rev. **113**, 147 (1959).

¹⁹ Flubacher, Leadbetter, and Morrison, Phil. Mag. **4**, 273 (1959); the writer is indebted to J. C. Phillips for calling his attention to this paper and its relevance for the numerical computations of Sec. V.

²⁰ B. N. Brockhouse, Phys. Rev. Letters **2**, 256 (1959).

²¹ Palevsky, Hughes, Kley, and Tunkelo, Phys. Rev. Letters **2**, 258 (1959).

which reduces to A when $n(\omega) = C\omega^2$. It is apparent that a redistribution of the normal mode frequencies that favors small ω will make B larger than A , and hence make $f_T(\mathbf{I}) = \exp(-Bq^2)$ smaller than Überall's asymptotic value for the same quantity. There are two physical reasons why low-frequency modes are especially effective in reducing interference. (a) The squared amplitude of the displacement of a quantum oscillator of given mass is inversely proportional to the frequency. Thus the low-frequency modes give rise to relatively large displacements of the atoms, and hence to the factor $1/\omega$ in the integrand of the numerator of B . (b) The excitation energies of the low-frequency modes are relatively small in comparison with κT , and hence are favored in the T -dependent part of this integrand.

The numerical values of B for silicon and germanium at room temperature are easily obtained from the moments of the frequency spectrum given by Flubacher, Leadbetter and Morrison.¹⁹ These moments are expressed in the form:

$$\mu_n = (3N)^{-1} \int_0^\infty \nu^n G(\nu) d\nu, \quad (38)$$

where $\nu = \omega/2\pi$, $G(\nu)d\nu = n(\omega)d\omega$, and $\int_0^\infty G(\nu)d\nu = 3N$ is the total number of lattice modes in the crystal; there is, of course, no reason why the finite upper limits in the integrals of (37) cannot be replaced by infinity, as in (38), if $n(\omega)$ and $G(\nu)$ are modified accordingly. For temperatures that are not too small the following expansion is useful:

$$(\exp x - 1)^{-1} = x^{-1} (1 - \frac{1}{2}x + a_1 x^2 - a_2 x^4 + a_3 x^6 - a_4 x^8 + \dots),$$

where

$$1/a_1 = 12, 1/a_2 = 720, 1/a_3 = 30\,240, 1/a_4 = 1\,209\,600.$$

The corresponding series for B is:

$$B = (2\pi M)^{-1} [(\kappa T/h)\mu_{-2} + a_1(h/\kappa T) - a_2(h/\kappa T)^3 \mu_2 + a_3(h/\kappa T)^5 \mu_4 - a_4(h/\kappa T)^7 \mu_6 + \dots]; \quad (39)$$

note that h , not \hbar , appears here since the moments are defined in terms of ν rather than ω . For room temperature ($T = 300^\circ\text{K}$), successive terms of the series (39) decrease very rapidly, and numerical substitution gives $B = 400/\mu^2$ for silicon and $B = 465/\mu^2$ for germanium; the units of B are those defined near the beginning of Sec. II; $\hbar = c = 1$, and μ is the electron mass.

In order to see the effect of the departure from the Debye spectrum, we may calculate A from (27), using Phillips' values for Θ at room temperature.¹⁸ For silicon $\Theta = 645^\circ\text{K}$, so that $A = 280/\mu^2$; for germanium $\Theta = 354^\circ\text{K}$, so that $A = 330/\mu^2$. As expected, the enhancement of the low frequencies makes B much larger than A in both cases.

VI. SUMMARY AND DISCUSSION

We have considered several of the approximations made by Überall² in relation to the two experiments that have been performed thus far.^{4,5} In Sec. II it was shown that the use of the Born approximation tends to overestimate the interference effect. A more correct treatment has no significant effect on the Panofsky-Saxena (PS) experiment, while it can account for the failure to observe the predicted central minimum in the Frisch-Olson (FO) experiment. In Sec. III it was shown that a more correct treatment of temperature effects tends to enhance the expected interference. With the assumption of the Debye spectrum and the validity of the closure approximation, this enhancement is too small to affect the comparison between the theory and either experiment. In Sec. IV it was shown that the closure approximation is an exceedingly good one. In Sec. V it was shown that the departures from the Debye spectrum are expected to change the damping factor from $\exp(-Aq^2)$ to $\exp(-Bq^2)$, where B is much larger than A .

In order to evaluate this last change we proceed as at the end of Sec. III, and find the corresponding change in the damping factor when q is set equal to the reciprocal screening radius. Now PS analyzed their experiment by suitably scaling Überall's calculated curves for copper at $T = 0^\circ\text{K}$ and a primary electron energy of 1 Bev for the differences in primary energy and lattice constant. Thus we must not use $A = 280/\mu^2$ for silicon at room temperature, but rather $A = 121/\mu^2$ for copper at zero temperature, and replace this by $B = 400/\mu^2$. This changes the damping factor from $\exp[-121(Z^3/111)^2] = 0.95$ to $\exp[-400(Z^3/111)^2] = 0.83$. A slightly better estimate indicates that the expected interference should be reduced by 15 to 20%. It follows that even though the Debye spectrum is a rather poor approximation, use of the correct spectrum does not resolve the discrepancy between the PS experiment and the theory, nor will it have much effect in the case of the FO experiment.

A few other effects may be considered briefly. It might be possible that imperfections in their silicon crystal could account for the failure of PS to observe interference. They quote an angular width for an x-ray rocking curve run on this crystal which can be roughly translated into a finite smearing-out size of each reciprocal lattice "point." This size turns out to be of the order of a few ev; it is much smaller than the thickness of the pancake-shaped region, and hence is unable to account for the negative result. PS also state that multiple scattering of the primary electrons in the target, which they measured directly, is too small to have an effect. Another possibility is that excitation or ionization of silicon atoms by the electrons occurs frequently enough to destroy the coherence of the primary electrons which is essential for interference.

The energy loss per unit length is

$$(2\pi N_0 Z e^4 / \mu) \ln(k_0^3 / 2I^2 Z^2 \mu),$$

where N_0 is the number of atoms per unit volume, IZ is the average energy loss per collision, and $I=11$ to 14 ev.²² If we divide this by $N_0 IZ$ we obtain an average inelastic collision cross section. The result is actually an overestimate, since polarization of the solid silicon reduces the logarithm in the stopping power formula by reducing the small-energy-loss, large-impact-parameter collisions; by the same token it increases the average energy loss per collision above the free-atom value IZ . The cross section obtained in this way corresponds to a free path for inelastic collisions of about 850 atom distances in silicon, and this number, as just remarked, must be an underestimate. A coherence distance of the order of this free path has roughly the same effect as a lack of perfection of the crystal over a comparable distance, and this smears out the reciprocal lattice "points" to a size of about 1/850 times their distance apart, or about 10 ev. Since we have overestimated this size, the effect is unimportant.

We are forced to conclude, then, that no theoretical explanation has yet been devised for the absence of interference in the PS experiment.²³ This makes it especially interesting to make a comparison between the two experiments that have been performed. As compared to the PS experiment, FO (1) used germanium instead of silicon, (2) directed the primary electron beam along the [110] axis of the crystal instead of the [100] axis, (3) used $k_0=1$ Bev and $k_\gamma \approx 100$ Mev instead of $k_0=600$ Mev and $k_\gamma=235$ Mev, and (4) collimated the observed radiation to a cone of half-angle about one milliradian. From the present point of view, silicon and germanium differ with respect to their lattice constants and their values for the parameter B . However, both of these differences are small enough so that point (1) above cannot be expected to account for the striking difference in the results of the two experiments. The difference in choice of crystal axis

means that the effective density of reciprocal lattice points in the perpendicular plane is different in the two cases. When account is taken of the structure factor $S(\mathbf{g})$, these effective densities turn out to be in the ratio $1:2^{3/2}:(4/3)^{3/2}=1:1.414:1.155$ for the [100], [110], and [111] directions. Thus point (2) leads to a 41% greater interference effect in the FO than in the PS experiment. Point (3) is certainly the most important difference between the two experiments. As mentioned at the end of Sec. II, the FO experiment has a minimum q value of 15 ev, as compared with 140 ev for PS. In accordance with Überall's numerical results, the curve of radiation intensity as a function of angle between primary electron and crystal axis is compressed to smaller angles and has a larger maximum value when q_{\min} is decreased. Finally, point (4) seems to be quite unimportant. The characteristic radiation angle is of order $\mu/k_0 = \frac{1}{2}$ milliradian and almost independent of k_γ ,¹⁶ so that any larger angle must arise from multiple scattering of the primary electrons in the target²⁴ or from their lack of parallelism before they strike the target.

The fact that the FO experiment gives less interference than expected might be caused by divergence or multiple scattering of the primary electrons, which were not measured by them, or by the same unknown factor that leads to the negative result of the PS experiment. Indeed, it can be said, considering points (2) and (3) above, that the two experiments are consistent with each other if divergence and multiple scattering are unimportant for FO. We conclude by remarking that the interference effect can be enhanced by making $q_{\min} = \mu^2 k_\gamma / 2k_0 k_f$ as small as possible and, in the case of a diamond-type crystal, using the [110] direction for the primary electrons. On the other hand, since the interference is insensitive to the value of B , little advantage is to be expected from cooling the crystal. It also seems likely that the central minimum in the interference pattern will be difficult to establish experimentally, since for the most favorable case of small q_{\min} , partial failure of the Born approximation tends to fill it in.

²² See for example W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, New York, 1954), third edition, p. 370.

²³ Y. H. Ichikawa and M. Yamamoto (to be published) have suggested that the small- q processes are inhibited by a screening that arises from the phonon field. However, phonon effects were calculated essentially correctly by Überall assuming the closure approximation (Sec. III), and the closure approximation was found to be excellent (Sec. IV). Thus we cannot accept a phonon-screening effect as an explanation of the PS experiment.

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²⁴ L. I. Schiff, *Phys. Rev.* **70**, 87 (1946).