

and V. F. Gantmakher, *Zh. Eksp. Teor. Fiz.* **56**, 1232 (1969) [*Sov. Phys. JETP* **29**, 663 (1969)], some incorrectly {for example: Yu. N. Chiang, V. V. Eremenko, and O. G. Shevchenko, *Zh. Eksp. Teor. Fiz.* **54**, 1321 (1968) [*Sov. Phys. JETP* **27**, 706 (1968)]; for a further treatment of this problem see J. W. Ekin and B. W. Maxfield, *Phys. Rev. B* **2**, 4805 (1970)}. In each case, however, only a speculative interpretation was possible.

<sup>3</sup>See, for example, J. M. Ziman, *Proc. Roy. Soc., Ser. A* **226**, 436 (1954); M. Baily and H. Brooks, *Bull. Amer. Phys. Soc.* **1**, 300 (1956); M. Baily, *Phys. Rev.* **120**, 381 (1960).

<sup>4</sup>The samples were free-mounted polycrystalline wires extruded from potassium obtained from Mine Safety Appliances Ltd., Callery, Pa. All were annealed one week at room temperature and then slowly cooled to liquid-helium temperatures. A dc four-terminal resistance-measurement technique was employed; separation between current and potential contacts was greater than 5 sample diameters. The error in the absolute resistivity values, which were determined assuming a resistivity at 22°C of 7.19  $\mu\Omega$  cm, is about 2%. The relative error is about 0.02%. Temperatures were measured with an absolute accuracy of 0.01°K.

<sup>5</sup>Of course, at lower RRR values, significant deviations from Matthiessen's rule were observed. A more detailed treatment of this and other aspects of the data, as well as a tabulation of the complete experimental and theoretical results, will be given in a later article: J. W. Ekin and B. W. Maxfield, to be published.

<sup>6</sup>A. Hasegawa, *J. Phys. Soc. Jap.* **19**, 504 (1964).

<sup>7</sup>T. M. Rice and L. J. Sham, *Phys. Rev. B* **1**, 4546 (1970).

<sup>8</sup>The pseudopotentials used were those of J. Bardeen, *Phys. Rev.* **52**, 688 (1937); N. W. Ashcroft, *Phys. Lett.* **23**, 48 (1966); M. J. G. Lee and L. M. Falicov,

*Proc. Roy. Soc., Ser. A* **304**, 319 (1968). In the latter case, two sets of values were used corresponding to the extremal values reported by these authors; for details see Rice and Sham, Ref. 7.

<sup>9</sup>R. A. Cowley, A. D. B. Woods, and G. Dolling, *Phys. Rev.* **150**, 487 (1966).

<sup>10</sup>We have defined an umklapp process to be one in which  $\vec{k}_f - \vec{k}_i = \vec{q} + \vec{G}$ , with  $\vec{G} \neq 0$ , where  $\vec{k}_i$  and  $\vec{k}_f$  are, respectively, the initial and final electron wave vectors,  $\vec{q}$  the phonon wave vector, and  $\vec{G}$  a reciprocal lattice vector; for a normal process,  $\vec{G} = 0$ .

<sup>11</sup>Not all of these assumptions have been justified theoretically; in particular, phonon-drag effects may become important at those very low temperatures where umklapp processes are frozen out. The agreement with experiment, however, seems not to be affected appreciably by these assumptions, at least at temperatures above 1.5°K. For further details of the calculation as well as a theoretical treatment of the role of electron-phonon umklapp processes in ultrasonic attenuation see P. N. Trofimenkoff and J. W. Ekin, to be published.

<sup>12</sup>D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, *Proc. Roy. Soc., Ser. A* **256**, 334 (1960).

<sup>13</sup>It is understood that this simplified exponential form is only an approximation valid over limited temperature ranges since it neglects other slower-varying functions of temperature as well as the fact that different phonon polarizations decay at different rates.

<sup>14</sup>W. E. Lawrence, thesis, Cornell University, 1970 (unpublished).

<sup>15</sup>Vice versa, the large and rapidly changing electron-phonon umklapp component at higher temperatures might readily explain the previous lack of experimental evidence of any electron-electron scattering in the alkalis; see, for example, the Letter by Garland and Bowers, Ref. 2.

## Short-Wavelength Cyclotron Waves and Electron Correlations in Potassium

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(Received 27 April 1971)

Observations of the Azbel'-Kaner cyclotron resonance spectra in thick slabs of high-purity potassium metal have revealed additional oscillations on the high-field side of the fundamental resonance. These anomalies in the surface impedance correspond to turning points of the cyclotron-wave dispersion curve at wavelengths which are short compared to orbit radii. The behavior of the signals agrees quite well with free-electron theory and confirms the extremely oscillatory character of the extraordinary mode. The absence of deviations due to Fermi-liquid effects in our experiments is investigated, and in particular we show under what circumstances the Fermi-liquid parameter  $A_1$  can be determined from these signals.

Weak surface-resistance anomalies associated with turning points of the dispersion curves<sup>1</sup> for cyclotron or high-frequency waves have been studied under Azbel'-Kaner cyclotron-resonance

(AKCR) conditions in the case of metallic potassium where the absence of band-structure effects makes comparison with theory particularly interesting. As in Henningsen's recent discovery of

the phenomenon in silver,<sup>2</sup> these signals give information about wave propagation at wavelengths which are small compared to cyclotron-orbit dimensions, whereas previous experiments have dealt almost entirely with the behavior at longer wavelengths. These new data confirm the predicted oscillatory character of the dispersion relations, which is particularly marked in the "extraordinary" polarization.<sup>3</sup> The overall agreement with a free-electron description of the conduction electrons is quite good—a rather disappointing result in view of the expectation that modifications due to many-body effects might prove appreciable in the range of wavelengths investigated.<sup>1,2</sup> However, numerical evaluation of the dispersion relations for an electron fluid via the Fermi-liquid theory of Landau<sup>4</sup> and Silin<sup>5</sup> shows that in the frequency range of the present experiments the absence of appreciable deviations from free-electron behavior is consistent with the magnitude of electron correlation effects expected on the basis of previous observations at smaller wave vector<sup>1</sup> and of microscopic theory.<sup>6</sup>

The essential experimental distinction between earlier studies of cyclotron-wave propagation at long wavelengths and the work of Henningsen with silver, as well as the present investigation of potassium, lies in the use of metal samples sufficiently thick as to attenuate all propagating excitations. The resulting elimination of interference spectra<sup>7</sup> permits observation of rather weak changes in microwave surface resistance resulting from turning points of the dispersion curves. In the experiments reported here distilled potassium of resistivity ratio  $\rho(295^\circ\text{K})/\rho(1.4^\circ\text{K}) \sim 15000$  was extruded in a purified argon atmosphere and permanently pressed between thin glass plates to form a very smooth and flat sample whose surfaces had a brilliant, mirrorlike appearance. The dimensions of the polycrystalline slab were  $12 \times 6 \times 0.6 \text{ mm}^3$ , a thickness sufficient to yield essentially semi-infinite behavior. The sample was placed inside a rectangular microwave cavity to obtain increased sensitivity to surface resistance changes<sup>8</sup> which were monitored in reflection by a conventional homodyne bridge spectrometer. The field derivative of the cavity loss was recorded versus the slowly varying magnetic field lying in the sample plane, either parallel (ordinary mode) or perpendicular (extraordinary mode) to the linearly polarized microwave surface currents. The experiments were performed at  $1.4^\circ\text{K}$  and at 12.2 and 17.8 GHz.

As shown in Fig. 1(a) the derivative spectrum

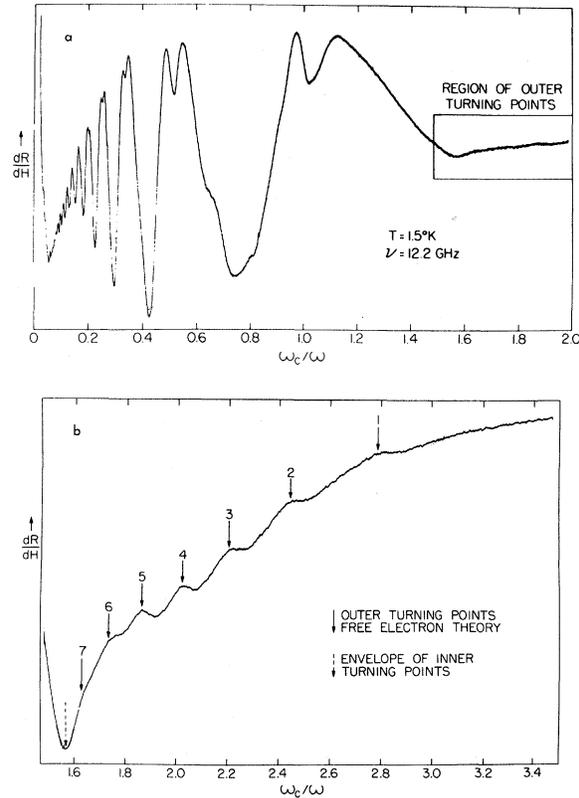


FIG. 1. (a) The AKCR derivative spectra in the extraordinary-mode polarization. (b) Same as (a) with the gain increased by 10 to display the turning-point anomalies. The arrows are at the locations of the turning-point singularities of the free-electron, infinite- $\omega\tau$  dispersion relation.

in the extraordinary polarization at 12.2 GHz is dominated by a series of AKCR single-particle resonance peaks occurring at  $\omega_c = \omega/n$ ,  $n = 1, 2, \dots$ , where  $\omega$  is the microwave angular frequency and  $\omega_c = eH/m^*c$  is the swept cyclotron frequency, inversely proportional to the isotropic effective mass  $m^* = 1.21m_e$ .<sup>9</sup> The fundamental and first few subharmonic peaks are slightly split, which may reflect lack of perfect sample flatness, residual misalignment ( $<0.1^\circ$ ) of the magnetic field with respect to the sample plane, and/or possibly the onset of wave propagation. Extremely weak oscillations may be observed at high gains and long noise integration times corresponding to vestiges of transmission of long-wavelength cyclotron waves.<sup>7</sup>

More easily detected, however, in the range  $1.5 < \omega_c/\omega < 2.8$ , as shown in Fig. 1(b), are a sharp dip and a series of seven peaks, the latter lying on a rising background. As indicated by arrows in the figure these resolved features cor-

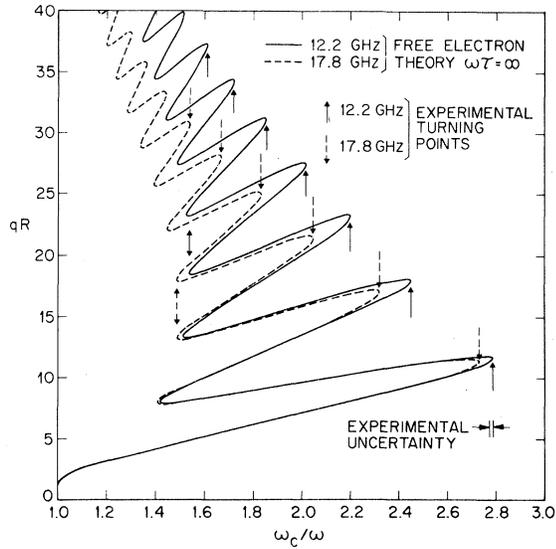


FIG. 2. Free-electron dispersion relations for the extraordinary-mode cyclotron wave at 12.2 and 17.8 GHz. The dimensionless quantity  $qR$  is plotted where  $q$  is the wave vector and  $R = v_f/\omega$ , the cyclotron radius at cyclotron resonance. The arrows locate the experimental peaks and dips at both frequencies.

respond to singular aspects of the extraordinary mode propagation at wavelengths which are short compared to the cyclotron orbit radii. Referring to Fig. 2, where the free-electron cyclotron-wave dispersion curves for infinite  $\omega\tau$  are plotted, it is evident that the peaks in  $dR/dH$  occur at outer turning points of the dispersion curve and the single dip lies at the high-field edge of the envelope of inner turning points. This identification proves empirically valid in that increasing the frequency to 17.8 GHz causes shifts in the observed resistance anomalies which are matched by the appropriate dispersion curve. With increasing temperature the peaks rapidly broaden but do not shift in position, thus making appropriate our comparison with the infinite  $\omega\tau$  dispersion curves. The turning points associated with the fundamental ordinary mode and the first few subharmonics of the extraordinary mode contribute to anomalies of their own in the surface impedance, but lie too close together to be individually resolved in our present experiments.

We are unable to advance any physical argument as to why the onset and cessation of wave propagation should result in dips and peaks respectively in  $dR/dH$ . Henningsen<sup>2</sup> has evaluated the surface impedance for a spherical Fermi surface in the approximation of specular reflection and does find anomalies, but their intensity and shape do not agree with our observations.

For present purposes, therefore, we will use the empirical identification as a means of mapping the short-wavelength behavior of the cyclotron waves.

Before discussing the implication of our experiments for the many-body description of conduction electrons it may be desirable to comment on the concept of *discrete waves*. This was originally advanced theoretically by Kaner and Skobov<sup>10</sup> and by Blank and Kaner<sup>11</sup> and was recently invoked by Henningsen as the basis for the initial interpretation of his results in silver. The highly oscillatory character of the extraordinary-wave dispersion relation results from a near matching of extremal cyclotron-orbit diameters with an integral number of wavelengths.<sup>3</sup> This closely resembles the physical picture of discrete waves proposed by Kaner and co-workers. The analogy led Henningsen to interpret the surface impedance anomalies in silver as due to propagation of discrete waves. More recently he has recognized, as did we, that the anomalies really correspond to turning points of the cyclotron-wave dispersion relation. That these are *not* compatible concepts is shown by the numerical computations of Fredkin, Wilson, and Dunifer<sup>12</sup> including collisional damping, where it is found that the turning points are regions of maximal rather than minimal damping and cannot therefore correspond to discrete waves, whose unambiguous experimental existence remains to be established.

To incorporate electron-electron correlations in the cyclotron-wave dispersion relations we utilize the Landau Fermi-liquid theory.<sup>4,5</sup> From Maxwell's equations the dispersion relations can be readily written in the form

$$(qR)^2 = (\omega_p v_f / \omega c)^2 f(qR, \omega_c / \omega), \quad (1)$$

where  $q$  is the wave vector;  $R = v_f/\omega$ ;  $\omega_p$  the plasma frequency; and  $f$  a universal function of  $qR$  and  $\omega_c/\omega$ , which is made up of a particular set of the conductivity components depending upon the mode in question. The physics of the problem is contained within these components of the conductivity tensor whose exact form depends upon the nature of the electron-electron correlations. As shown earlier<sup>1</sup> the conductivity is modified by the Fermi-liquid parameters  $A_n$ ,  $n = 0, 1, 2, \dots$ , which are coefficients of a Legendre polynomial expansion of the spin-independent part of the Landau correlation function. For large wave vector ( $qR \approx 1000$ ), the Kaner-Skobov regime,<sup>13</sup> all the Fermi-liquid parameters dis-

appear from the cyclotron-wave dispersion curve.<sup>14</sup> For small wave vector ( $qR \lesssim 1$ )  $A_0$  and  $A_1$  do not appear:  $A_0$  because it is associated with net charge density which cannot be developed at microwave frequencies much less than the plasma frequency ( $\omega_p \approx 10^{16} \text{ sec}^{-1}$ ), and  $A_1$  because it is associated with net current flow which also vanishes for small-wave-vector excitations.

The nature of the disappearance of  $A_1$  from the small-wave-vector dispersion relation can be seen from Eq. (1). At small wave vector both  $(qR)^2$  and  $f(qR, \omega_c/\omega)$  are  $\sim 1$ , while the coefficient  $(\omega_p V_f/\omega c)^2$  is  $\sim 10^5$  at X-band frequencies. Consequently under these conditions the dispersion relation is quite accurately written  $f(qR, \omega_c/\omega) = 0$ . However, for both the ordinary and the extraordinary mode  $f$  is comprised of just the proper combination of conductivity components which when set equal to zero automatically sets the net current equal to zero also—hence  $A_1$  cannot appear in the dispersion relations.

For  $A_1$  to become important the two sides of Eq. (1) must be comparable. Since  $f \sim 1/qR$  at large wave vector this requires

$$qR \gtrsim (\omega_p v_f/\omega c)^{2/3}. \quad (2)$$

For potassium metal at X-band frequencies this gives  $qR \gtrsim 50$ , which is not far removed from the seventh-turning-point anomaly we have resolved. The frequency dependence of the dispersion relation seen in Fig. 2 is in fact evidence that Eq. (2) is roughly satisfied at the largest wave vectors. From Eq. (2) it is clear that there must be a frequency below which  $A_1$  cannot modify the dispersion relation significantly at any wave vector, i.e., that frequency for which Eq. (2) is not satisfied before entering the Kaner-Skobov regime. However, with increasing frequency we should expect the importance of  $A_1$  to grow and to become discernable at smaller wave vectors.

Exact numerical calculations for the extraordinary-mode dispersion relation have been carried out using finite values for  $A_1$  and  $A_2$ . For potassium we have  $A_2 \sim -0.03$  from previous measurements at small vector<sup>1</sup> and  $A_1 \sim 0.05-0.1$  from microscopic theory.<sup>6,15</sup> Using these numbers we find that the shifts of the turning points due to  $A_1$  are completely negligible ( $\lesssim 10^{-3}$ ) and those due to  $A_2$  are  $< 0.01$  in  $\omega_c/\omega$  at X-band frequencies—in agreement with our observations. The modifications introduced by finite  $A_2$  diminish with increasing frequency and increasing wave vector and are always negligible at the turning points for all frequencies  $\gtrsim 10$  GHz.

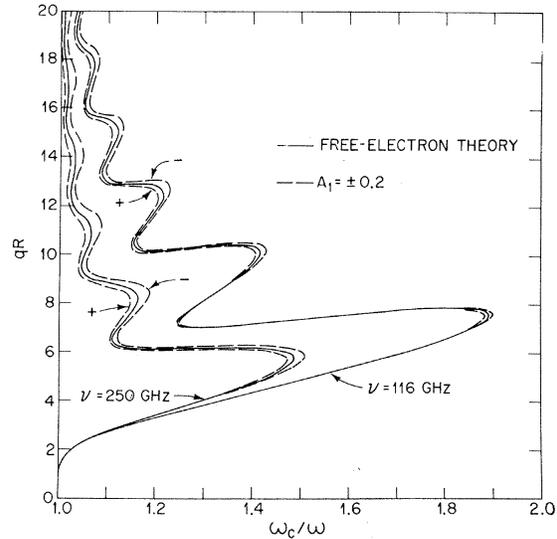


FIG. 3. The extraordinary-mode dispersion relation for infinite  $\omega \tau$  with  $A_1 = 0, \pm 0.2$  at frequencies of 116 and 250 GHz.

The effects of  $A_1$  at higher frequencies can be seen in Fig. 3 where we have plotted the dispersion relations at frequencies of 116 and 250 GHz for  $A_1 = \pm 0.2$  (somewhat larger than anticipated). At 250 GHz the largest shift occurs at the second outer turning point and is 2.4% in  $\omega_c/\omega$ . At 116 GHz the maximum shift is at the third outer turning point and is only 1.3% in  $\omega_c/\omega$ . Thus the shifts are larger and occur at smaller wave vectors with increasing frequency as expected.

In conclusion, we have seen how the dispersion relation for the extraordinary-mode cyclotron waves can be studied for short wavelength by observing surface impedance anomalies associated with the turning points of the dispersion curve. Useful measurements of the Fermi-liquid parameter  $A_1$  appear possible from such observations carried out at frequencies  $\gtrsim 200$  GHz.

Helpful conversations with P. M. Platzman and T. M. Rice are gratefully acknowledged.

<sup>1</sup>P. M. Platzman, W. M. Walsh, Jr., and E-Ni Foo, Phys. Rev. **172**, 689 (1968).

<sup>2</sup>J. O. Henningsen, Phys. Rev. Lett. **24**, 823 (1970), and Physics Laboratory I, The Technical University of Denmark, Report No. 77, 1970 (to be published).

<sup>3</sup>E-Ni Foo, Phys. Rev. **182**, 674 (1969).

<sup>4</sup>L. D. Landau, Zh. Eksp. Teor. Fiz. **30**, 1058 (1956) [Sov. Phys. JETP **3**, 920 (1957)].

<sup>5</sup>V. P. Silin, Zh. Eksp. Teor. Fiz. **33**, 495 (1957) [Sov. Phys. JETP **6**, 387 (1958)].

<sup>6</sup>T. M. Rice, *Phys. Rev.* **175**, 858 (1968).

<sup>7</sup>W. M. Walsh, Jr., P. M. Platzman, P. S. Peercy, L. W. Rupp, Jr., and P. H. Schmidt, in *Proceedings of the Twelfth International Conference on Low Temperature Physics*, Tokyo, Japan, 1970 (to be published).

<sup>8</sup>W. M. Walsh, Jr., L. W. Rupp, Jr., and P. H. Schmidt, *Phys. Rev.* **142**, 414 (1966). That this two-sided excitation merely enhanced sensitivity without distorting the spectra via transmission effects was checked by making a cavity in which a thicker potassium plate formed a conventional end wall.

<sup>9</sup>C. C. Grimes and A. F. Kip, *Phys. Rev.* **132**, 1991

(1963).

<sup>10</sup>E. A. Kaner and V. G. Skobov, *Physics* (Long Is. City, N. Y.) **2**, 165 (1966).

<sup>11</sup>A. Ya. Blank and E. A. Kaner, *Phys. Status Solidi* **22**, 47 (1967).

<sup>12</sup>D. R. Fredkin, A. R. Wilson, and G. L. Dunifer, unpublished results.

<sup>13</sup>E. A. Kaner and V. G. Skobov, *Fiz. Tverd. Tela.* **6**, 1104 (1964) [*Sov. Phys. Solid State* **6**, 851 (1964)].

<sup>14</sup>M. Ya. Azbel', *Zh. Eksp. Teor. Fiz.* **34**, 766 (1958) [*Sov. Phys. JETP* **7**, 527 (1958)].

<sup>15</sup>T. M. Rice, private communication.

## Kinematic Low-Energy Electron-Diffraction Intensities from Averaged Data: A Method for Surface Crystallography\*

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(Received 1 March 1971)

It is demonstrated that the interference of the low-energy-electron multiply scattered amplitudes can be in large part eliminated by averaging data taken with different diffraction geometry but at fixed momentum transfer. This essentially yields a measurement of the kinematic intensities, which are most directly useful for surface crystallography.

The most important potential of low-energy electron diffraction is the unambiguous determination of the atomic structure of the outermost few atomic planes of a crystal, but thus far no structure has been determined using this technique in the sense that bulk structures have been determined using x-ray crystallography. The periodicity of the structure parallel to the surface requires that the parallel component of the electron momentum be conserved to within a reciprocal-net vector. This enables one to determine the size and shape of the unit mesh, but to determine the crystal structure one must be able to interpret the diffracted intensities. Low-energy electron-diffraction intensities are very complicated because of important multiple-scattering contributions to the scattered amplitude. Although recent theoretical results reproduce the features of measured intensities, these require extensive computation and have generally been

limited to simple systems.<sup>1</sup> Calculations of the intensities for assumed structures are far more complex in a dynamic than in a kinematic theory, and the inverse problem of determining the autocorrelation function from measured intensities has not been attempted in the framework of the dynamic theory.

Information about the atomic structure is most readily obtained from data which can be interpreted properly by a kinematic, or single-scattering, theory: Thus one would like to extract this first-order contribution from the low-energy electron-diffraction data. It is the purpose of this Letter to present an experimental demonstration that by properly averaging the data one can indeed eliminate the interference of the multiply scattered amplitudes and extract what is essentially the kinematic intensity.

The procedure is motivated by the following argument. The scattered intensity, including the multiple scattering explicitly to second order, is

$$I(\vec{S}) = AA^* = \sum_{i,m} f_i f_m^* \alpha_i \alpha_m \exp[i\vec{S} \cdot (\vec{r}_i - \vec{r}_m)] + \sum_{\substack{i \neq j \\ m \neq n}} f_{ij} f_{mn}^* \alpha_{ij} \alpha_{mn} \\ \times \exp\{i[-\vec{k}_0 \cdot \vec{r}_{im} + \vec{k} \cdot \vec{r}_{jn} - |\vec{k}|(|\vec{r}_{ij}| - |\vec{r}_{mn}|)]\} + \dots, \quad (1)$$

where  $\vec{k}$  and  $\vec{k}_0$  are the propagation vectors of the scattered and incident beams, respectively, and  $\vec{S} = \vec{k} - \vec{k}_0$ .  $\alpha_{ij}$  accounts for the attenuation of the ray scattered successively by atoms  $i$  and  $j$ , and  $f_{ij}$  is the product of the appropriate scattering factors. Equation (1) and the arguments that follow can easily be extended to higher orders. The first sum,  $I^{(1)}$ , is the single scattering and depends only on