

Approximate Calculation of the Anisotropy of the Relaxation Time of the Conduction Electrons in the Noble Metals

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The ratio of the relaxation times on the "belly" and on the "necks" of the Fermi surface is estimated numerically by very crude methods. It is shown that the relative amount of *s*-wave to *p*-wave scattering by impurities is important and that Umklapp processes play a major role in phonon scattering. The ratio depends on impurity type and on temperature in just the right way to explain qualitatively the variation of the Hall coefficient in the metals and their alloys.

INTRODUCTION

IT has been shown elsewhere¹ that it is impossible to get good agreement between theory and experiment for such properties as the Hall effect, magnetoresistance, thermoelectric power, optical absorption, etc., in the noble metals unless we assume that the relaxation time of the electrons is different at different points on the Fermi surface. From these transport properties we can infer the general way in which τ must behave, but we still lack a direct calculation starting from the electronic band structure of the metal and the type of scattering. This is the problem which is studied here.

Let it be said at once that the full problem is of extreme complexity. Even supposing that we knew exactly the shape of the Fermi surface, the electron velocity for each value of \mathbf{k} , the matrix elements for all scattering processes, the lattice spectrum, and all, we should still be faced with the very difficult mathematical problem of solving the Boltzmann equation—an integral equation over the Fermi surface. Indeed, we should then find that a relaxation time could not be defined uniquely at each point on the Fermi surface; it would depend on the direction of the electric field, the existence of thermal gradients, magnetic fields, etc. This can easily be shown by an inspection of the integral equation and the consideration of special cases.

Nevertheless, we must not escape in this way from all responsibility towards this problem. Experience and physical intuition tell us that there must be something very like a relaxation time for the state $|\mathbf{k}\rangle$ inversely proportional to the probabilities of transitions into and out of this state weighted to allow for the big effect on an electric current of scattering through large angles. We are bound to find an expression similar to the exact solution in the standard isotropic case.²

$$\frac{1}{\tau(\mathbf{k})} = \frac{2\pi}{\hbar} \int (1 - \cos\theta) |\langle \mathbf{k}, \mathbf{k}' \rangle|^2 \frac{dS'}{\hbar v'}, \quad (1)$$

where $\langle \mathbf{k}, \mathbf{k}' \rangle$ is the matrix element for the transition

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¹ J. M. Ziman, *Advances in Phys.*, **10**, 1 (1961).

² J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960), Sec. 7.4.

from $|\mathbf{k}\rangle$ to the state $|\mathbf{k}'\rangle$ in the region dS' of the Fermi surface, where the velocity is v' . We assume here elastic scattering.

In the free-electron case, θ is just the angle between the wave vectors \mathbf{k} and \mathbf{k}' . Our more complex system does not have this solution; even the plausible guess that θ might be the angle between the electron velocities at \mathbf{k} and \mathbf{k}' does not generate a better formal solution of the integral equation, and is not easy to deal with mathematically. But the main thing is that the transition probability in the integral should be strongly weighted for processes in which the electron velocity makes a large change—for example, when it is scattered right across the Fermi surface. So we shall assume that the formula (1) is valid, with θ the angle between \mathbf{k} and \mathbf{k}' . This is our first approximation.

IMPURITY SCATTERING

To proceed with the calculation, we need a formula for the Fermi surface. We shall use the "8-cone" model^{1,3} in which the wave function of the state $|\mathbf{k}\rangle$ is expressed in the form

$$|\mathbf{k}\rangle = \alpha_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{r}) + \beta_{\mathbf{k}} \exp[i(\mathbf{k} - \mathbf{g}) \cdot \mathbf{r}]. \quad (2)$$

The coefficients $\alpha_{\mathbf{k}}$, $\beta_{\mathbf{k}}$ can be calculated at each point of the Fermi surface from the standard formula for the mixing of plane waves. More exactly, we should use orthogonalized plane waves (OPW's) in place of the simple waves, but the difference is not relevant to our present purposes.

For the matrix element for scattering by an impurity, we then write

$$\langle \mathbf{k}, \mathbf{k}' \rangle = \alpha_{\mathbf{k}}^* \alpha_{\mathbf{k}'} U_{\mathbf{k}' - \mathbf{k}} + \beta_{\mathbf{k}}^* \beta_{\mathbf{k}'} U_{\mathbf{k}' - \mathbf{g}' - \mathbf{k} + \mathbf{g}} + \alpha_{\mathbf{k}}^* \beta_{\mathbf{k}'} U_{\mathbf{k}' - \mathbf{g}' - \mathbf{k}} + \beta_{\mathbf{k}}^* \alpha_{\mathbf{k}'} U_{\mathbf{k}' - \mathbf{k} + \mathbf{g}}, \quad (3)$$

where $U_{\mathbf{k}' - \mathbf{k}}$ is a matrix element for the scattering of a single plane wave (or single OPW) between \mathbf{k} and \mathbf{k}' . This formula is not exact, since it uses the Born approximation, but it ought to give a reasonable semi-quantitative description of the form of the scattering probability. In particular, it takes care of the interference between the two waves in (2) which is, as we

³ J. M. Ziman, *Proc. Roy. Soc. (London)* **A252**, 63 (1959).

shall see, very important in the final result. But for an exact calculation we should need a better theory of the functions $U_{\mathbf{k}'-\mathbf{k}}$.

Even now, the integration of (1) is extremely heavy work. We make two further approximations, which depend mainly on the fact that $U_{\mathbf{k}'-\mathbf{k}}$ is a slowly-varying function of $|\mathbf{k}'-\mathbf{k}|$ —that is, of the angle θ . We suppose that the variation of $U(\theta)$ over a single "cone" of the zone can be neglected. Then $\mathbf{k}-\mathbf{g}$ will always lie in the cone that is opposite to the cone of \mathbf{k} , and we can assume, on the average, that it is equal to $-\mathbf{k}$. Thus, we write

$$U_{\mathbf{k}'-\mathbf{k}+\mathbf{g}} \approx U_{\mathbf{k}'-(-\mathbf{k})} = U(\pi-\theta), \text{ etc.} \quad (4)$$

When we have put (4) and (3) into (1), and used the normalization condition, $\alpha^2 + \beta^2 = 1$, we can reduce our formula into the following expression:

$$\begin{aligned} \frac{1}{\tau(\mathbf{k})} = & \frac{2\pi}{\hbar^2} \int (1 - \cos\theta) \\ & \times \left\{ \frac{1}{2} [U^2(\theta) + U^2(\pi-\theta)] + 2\alpha'\beta' [U(\theta)U(\pi-\theta)] \right. \\ & + (\alpha^2 - \beta^2)(\alpha'^2 - \beta'^2) \frac{1}{2} [U^2(\theta) - U^2(\pi-\theta)] \\ & + 2\alpha\beta \{ U(\theta)U(\pi-\theta) \\ & \left. + 2\alpha'\beta' \frac{1}{2} [U^2(\theta) + U^2(\pi-\theta)] \} \right\} \frac{dS'}{v'}. \quad (5) \end{aligned}$$

The coefficients α and β are functions of \mathbf{k} , and can be taken out of the integral. But α' , β' , and θ are all functions of \mathbf{k}' . We approximate again by noting that α' and β' go through the whole range of their values on a single "cone," where $U(\theta)$ may be treated as more or less constant. Thus, we replace $\alpha'\beta'$ and $\alpha'^2 - \beta'^2$ by their averages over a cone, and integrate the remaining explicit functions of θ over a whole solid angle.

With these assumptions we write

$$1/\tau(\mathbf{k}) = (T_t + \langle 2\alpha'\beta' \rangle_{\text{av}} T_U) + (\alpha^2 - \beta^2) \langle (\alpha'^2 - \beta'^2) \rangle_{\text{av}} \times (T_r - T_t) + 2\alpha\beta (T_U + \langle 2\alpha'\beta' \rangle_{\text{av}} T_t), \quad (6)$$

where

$$T_t = \frac{4\pi^2}{\hbar^2} \int (1 - \cos\theta) \times \frac{1}{2} [U^2(\theta) + U^2(\pi-\theta)] \sin\theta d\theta, \quad (7)$$

$$T_U = \frac{4\pi^2}{\hbar^2} \int (1 - \cos\theta) U(\theta) U(\pi-\theta) \sin\theta d\theta, \quad (8)$$

$$T_r - T_t = \frac{4\pi^2}{\hbar^2} \int (1 - \cos\theta) \times \frac{1}{2} [U^2(\theta) - U^2(\pi-\theta)] \sin\theta d\theta, \quad (9)$$

$$\begin{aligned} \langle 2\alpha'\beta' \rangle_{\text{av}} = & \frac{1}{4\pi} \int 2\alpha'\beta' \frac{dS'}{v'}; \\ \langle \alpha'^2 - \beta'^2 \rangle_{\text{av}} = & \frac{1}{4\pi} \int (\alpha'^2 - \beta'^2) \frac{dS'}{v'}. \end{aligned} \quad (10)$$

All these expressions have already turned up in previous calculations,³ and can be evaluated very easily. With Cu, or Au, as our model, we find

$$\langle 2\alpha'\beta' \rangle_{\text{av}} \sim \pm \frac{3}{4}, \quad \langle \alpha'^2 - \beta'^2 \rangle_{\text{av}} \sim \frac{1}{2}. \quad (11)$$

We can now use our standard formulas to find α and β at any chosen spot on the Fermi surface, and hence estimate the local value of τ . But all we need for our present analysis is some measure of the anisotropy of τ . A convenient parameter is the ratio of the extreme values of τ , i.e., on the belly and on the neck. At these points

$$\begin{aligned} 2\alpha\beta & \approx \pm \frac{1}{2}, \quad \alpha^2 - \beta^2 \approx \frac{7}{8} \text{ on the belly;} \\ 2\alpha\beta & = \pm 1, \quad \alpha^2 - \beta^2 = 0 \text{ on the neck.} \end{aligned} \quad (12)$$

These numbers are not to be supposed very accurate, but they clearly indicate that α is much larger than β on the belly, where a single plane wave or OPW is a good representation of the wave function. On the neck, however, we must have $|\alpha| = |\beta|$. There is still the relative *sign* of the coefficients to be fixed. This depends on the symmetry of the wave function at the zone boundary. It now seems fairly certain⁴ that the p state lies lowest in Cu, and perhaps also in the other noble metals; this makes the sign of $2\alpha\beta$ in (11) and (12) always negative.

We now need only values at the various weighted scattering cross sections, (7), (8), and (9). These are easily estimated from the phase shifts for scattering by charged impurities, e.g., in the work of Blatt.⁵ But instead of using such numbers, let us suppose that only two partial waves are important—the s wave and the p wave—and that the corresponding phase shifts, δ_0 and δ_1 , are both small enough to write $\sin\delta_0 \approx \delta_0$, etc. We then have (dropping constant factors like \hbar and π)

$$\begin{aligned} T_t & = \sum_{l=0}^{\infty} (2l+1) \sin^2 \delta_l \approx \delta_0^2 + 3\delta_1^2, \\ T_U & = \sum_{l=0}^{\infty} (2l+1) (-1)^l \sin^2 \delta_l \approx \delta_0^2 - 3\delta_1^2, \\ T_r & = \sum_{l=1}^{\infty} l \sin^2 (\delta_{l+1} - \delta_l) = (\delta_0 - \delta_1)^2, \end{aligned} \quad (13)$$

whence

$$\frac{\tau(\text{belly})}{\tau(\text{neck})} \approx \frac{84(\delta_1/\delta_0)^2}{1 - 7(\delta_1/\delta_0) + 63(\delta_1/\delta_0)^2}. \quad (14)$$

This relation, plotted in Fig. 1, does not, of course, really give precise numerical estimates of the ratio of the extreme values of the relaxation time on the Fermi surface. The calculation is obviously much too crude. Nevertheless, it shows some important qualitative features. If we look at Blatt's data, we find $0.5 < \delta_1/\delta_0 < 1$ for most of the charged impurities. This would make the

⁴ B. Segall (private communication).

⁵ F. J. Blatt, Phys. Rev. **108**, 285 (1957).

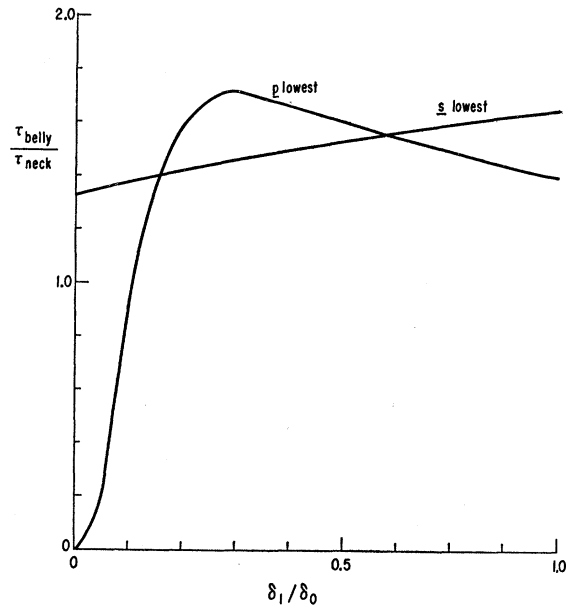


FIG. 1. Anisotropy of τ , as a function of the ratio of the phase shifts, for impurity scattering.

ratio (14) lie somewhere between 1.4 and 1.6. That is, τ is rather smaller on the neck than on the belly—which is just what we need to explain the values of the Hall coefficient in alloys of the noble metals.¹

But we also note that when $\delta_1 \ll \delta_0$, the ratio (14) tends to zero. In other words, the relaxation time on the neck is then very much greater than on the belly. It is quite easy to see why this should be so. The ratio δ_1/δ_0 measures the relative proportions of p -wave to s -wave scattering by the impurity. On the neck the electrons are in pure p states. If the scattering of a plane wave is s -like, then these electrons will not be scattered at all, and their relaxation time will be very long. That is, when the scattering of a single plane wave is isotropic, there will be complete destructive interference of the partial waves and no scattering.

This is just the sort of condition that might occur when the impurity, for example Ag, has the same valency as the parent metal, for example, Au. These elements both have the same atomic volume so that the effect of the Ag on a conduction electron in the Au will be due only to minor differences in the effective potential deep down in the ion core. We then expect the scattering of a single plane wave (actually, a single OPW) to be equivalent to the scattering by a very localized perturbation, almost a δ function. Such scattering is well known to be s -like, i.e., nearly isotropic. It is significant that the Hall effect in the Ag—Au behaves as if $\tau(\text{belly})$ were very much less than $\tau(\text{neck})$, in agreement with our analysis. In the Cu—Au system, however, the difference of atomic volumes gives rise to substantial phase shifts for the p wave⁵ and the ratio of relaxation times can then be much the same as for

charged impurities. This is also indicated by the experiments on the Hall effect.

PHONON SCATTERING

The above calculation gives us some hope that we may be able to calculate directly the anisotropy of τ for different types of impurity, and encourages us to plunge into the more complicated problem of the effect of phonon scattering on the electrons. We make our attempt as follows.

First we note that (3) is still valid. In calculating the electron-phonon interaction we only use first-order perturbation theory, so that the effect on a mixed wave can be expressed as a matrix element combining contributions from the separate simple plane waves. It is true that when these are OPW's there are some corrections to the simple Bardeen formula and the whole theory of screening is much more complex, but Collins⁶ has shown that these are not large effects. Thus, we may write⁷

$$U_{\mathbf{k}'-\mathbf{k}} = (\mathbf{k}'-\mathbf{k}) \cdot \mathbf{e}_q C(|\mathbf{k}'-\mathbf{k}|). \quad (15)$$

To avoid difficulties with the lattice spectrum, we shall assume (quite illegitimately, of course) that *longitudinal and transverse waves have the same velocity*. From the three degenerate lattice modes of wave number \mathbf{g} , we can then construct a single mode whose polarization vector \mathbf{e}_q is parallel to $(\mathbf{k}'-\mathbf{k})$, and this is the one that scatters; the other two will be polarized normal to $(\mathbf{k}'-\mathbf{k})$ and need not be counted. This device keeps the counting of transitions correct, but tends to underweight certain types of electron-phonon Umklapp process where the scattering is mainly by the low-frequency transverse modes.

For the functional form of the rest of (15) we assume

$$C(K) \propto G(Kr_s) = \frac{\sin(Kr_s) - (Kr_s) \cos(Kr_s)}{(Kr_s)^3}, \quad (16)$$

where r_s is, as usual, the radius of an atomic sphere. This ignores a screening factor, and another complicated and incalculable factor depending on the effect of displacing the ion core; these factors tend to cancel one another to some extent, but ought to be included in a proper calculation. It is reasonable to take

$$|\mathbf{k}'-\mathbf{k}| = 2k_F \sin \frac{1}{2}\theta, \quad (17)$$

where k_F is, say, the radius of a simple Fermi sphere.

We must still allow for a most important effect—the part of the transition probability that depends on the occupation number of the phonon states involved in the scattering and on the conservation of energy in the process. This is a mechanical calculation⁸ in which we include certain extra factors in (1), and integrate over all

⁶ J. G. Collins (to be published).

⁷ J. M. Ziman, see reference 2, Sec. 5.8.

⁸ J. M. Ziman, see reference 2, Sec. 9.5.

phonon wave vectors and over the energy of the final state $|\mathbf{k}'\rangle$. In the case of electrical conductivity (but not of thermal conductivity nor of thermoelectric power), these integrations can easily be done; they tell us that we shall get the proper answer if we include under the integral sign in (1) the factor

$$\frac{1}{4} \text{csch}^2(\hbar\nu/2kT), \quad (18)$$

where ν is the frequency of the phonon involved in the process of scattering from $|\mathbf{k}\rangle$ to $|\mathbf{k}'\rangle$.

We naturally assume a Debye model, in which

$$\hbar\nu/kT = (q/Q_D)(\theta/T), \quad (19)$$

with Q_D the Debye cutoff wave number. For N -processes we can very easily calculate the contribution, putting $\mathbf{q} = \mathbf{k}' - \mathbf{k}$. This holds for small values of θ , until q reaches its maximum, Q_D . But then we go over to electron-phonon Umklapp processes, where the relation between q and θ is much more complicated. There is only one simple general rule; when θ is somewhere near π , q will have reached a minimum value, corresponding to a transition from \mathbf{k} to the nearest point on the Fermi surface in the repeated zone (Fig. 2). This minimum value of q is really the most significant parameter in the theory of U -processes, since it provides a lower cutoff frequency, a minimum temperature, below which U -processes involving the state \mathbf{k} are no longer possible. As we shall see, the value of τ at \mathbf{k} depends strongly on this parameter.

To give arithmetical expression to this behavior, we use the following formula:

$$\begin{aligned} q/Q_D &= \sqrt{2} \sin \frac{1}{2}\theta \quad \text{for } 0 < \theta < \frac{1}{2}\pi, \\ q/Q_D &= \sin[\frac{1}{2}(\pi + \chi - \theta)] / \sin(\frac{1}{4}\pi + \frac{1}{2}\chi) \quad \text{for } \frac{1}{2}\pi < \theta < \pi, \end{aligned} \quad (20)$$

where the angle χ is defined so that

$$q_{\min}/Q_D = \sqrt{2}/(1 + \cot \frac{1}{2}\chi), \quad (21)$$

i.e., so that $q = q_{\min}$ when $\theta = \pi$. This function is continuous as we pass from N -processes to U -processes (a necessary property of any electron-phonon interaction theory; the zone boundary in the phonon reciprocal space is as arbitrary as it is in electron \mathbf{k} space) and provides a reasonable interpolation formula between the limits that we know; obviously it can only be a very crude approximation to the true function.

For a point \mathbf{k} on the neck it is easy to see that $q_{\min} = 0$. Truly, here the distinction between N -processes and U -processes is artificial, but we must retain it formally so as to include all terms in the integral. But on the belly, in the $[100]$ direction say, the point \mathbf{k} is quite a long way from the zone boundary, and further still from points on the Fermi surface in the next zone. A rough estimate, from the geometry of the surfaces in Cu, gives

$$q_{\min}/Q_D \approx \frac{1}{2}. \quad (22)$$

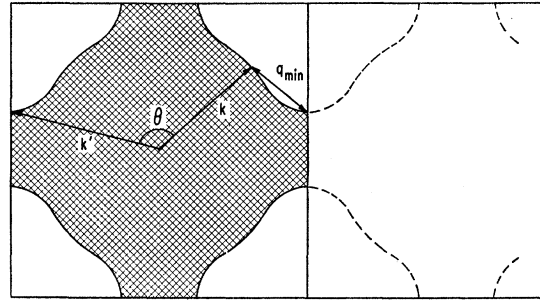


FIG. 2. The minimum phonon wave vector for U -processes.

We now have enough data to carry out the calculation. We put (15), (16), and (17) into (7), (8), and (9), including under the integral sign also the factor (18) with (19), (20), and (21). The result is of the form of (6), except that the cross sections depend on \mathbf{k} (through the choice of q_{\min}), and everything depends on the temperature. I have carried out the integrations on a desk calculator, and find the following results:

T/θ	0.2	0.5	1
$\tau(\text{belly})/\tau(\text{neck})$	2.2	1.1	0.9

First we observe that at "high" temperatures ($T \approx \theta$) the relaxation times are about equal, on belly and on neck. Thus, phonon scattering at high temperatures partakes a little of the s -wave behavior [we can check this by looking at (15) in detail] and does not give so much anisotropy as scattering by charged impurities. This is what we want for the Hall effect in alloys, although to explain the magnitude of n^* we want $\tau(\text{belly})$ still to be a bit greater than $\tau(\text{neck})$ at room temperature.

But we notice that the anisotropy of τ increases rapidly as we go to lower temperatures; at $T = 0.2\theta$ it has already greatly exceeded the typical ratio for charged impurities. If we look at the temperature dependence of the Hall effect,¹ we find direct evidence of this—a rapid drop in n^* at around 100°K in Cu. This behavior stems directly from the limit (15). U -processes for states on the belly are effectively "frozen out," and the relaxation time there, governed only by N -processes, increases rapidly. On the neck there is no lower limit to q , and U -processes contribute down to the lowest temperatures.

The above calculations, both for impurity and phonon scattering, are extremely uncouth; it is to be hoped that no one will be misled into treating these numerical ratios as if they were exact. Nevertheless, without straining the algebra or arithmetic, we find a pattern which agrees qualitatively with the experimental evidence. This pattern would, for example, be entirely different if we had s -states at the zone boundary; for impurity scattering the anisotropy ratio would then be between 1.3 and 1.6, (depending on the ratio δ_1/δ_0) and would not become small when $\delta_1/\delta_0 \ll 1$, while for phonon

scattering it would be much larger—something like 3, and not so sensitive to temperature. The fact that all three noble metals show the same behavior in their Hall effects—both with alloying and with temperature—is good evidence for the p -state lying lowest in all three.

It would be interesting to apply the same arguments to the alkali metals, where the Fermi surface is not so distorted, and where, it seems, the s -state moves below the p -state as we pass from Na, through K, to Rb and Cs.⁹ But we lack serious data on the Hall effect, and other manifestations of the anisotropy of τ , so that we should have little chance of making comparison with experiment. Another effect that might be calculated in

⁹ F. S. Ham, *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 9.

the way we indicate here is the thermoelectric power, since this depends strongly on the way that $\tau(\mathbf{k})$ varies as we change the energy of \mathbf{k} . For example, is $\partial\tau(\epsilon)/\partial\epsilon$ negative on the belly simply because increasing the length of \mathbf{k} reduces the value of q_{\min} , and thus allows many more U -processes? These, and many other questions in this field, remain to be answered.

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Infrared Lattice Bands of Quartz

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The infrared lattice bands of α quartz have been investigated at 297°K from 5 to 37 μ in reflection and transmission with polarized light. Previously published measurements of the optical constants do not agree in this spectral range. It is shown that dispersion theory can fit the data within experimental error throughout the range, and accurate values of the dispersion parameters and the optical constants are obtained. This is the first accurate dispersion analysis of a complex spectrum. A study was made of the accuracy of the Kramers-Kronig method of analysis on this spectrum. The strength, width, and frequency of 14 optically active lattice vibrations are given, 4 of which have not previously been established. From a consideration of published Raman data, 10 of the resonances are assigned according to symmetry type as fundamental vibrations.

I. INTRODUCTION

QUARTZ was one of the first materials to be extensively investigated in the infrared. Because of its scientific and technological importance it has continued to be a frequent subject of investigation. Yet, its optical properties in the infrared remain largely unknown or uncertain. From an inspection of the existing literature one cannot obtain consistent values for the optical constants. The present work was undertaken with the purpose of providing a comprehensive and consistent account of the optical properties of α quartz in the region of its strong lattice absorption bands, 5 to 37 microns (μ). In order to achieve this purpose it was necessary to analyze the optical data according to classical dispersion theory. The success of this analysis demonstrates the applicability of this theory to very complicated lattice spectra such as that of quartz.

The reflectivity of α quartz (hereafter written simply "quartz") at room temperature (297°K) was measured in the range 5–37 μ for the ordinary (O) ray and extraordinary (E) ray with plane polarized light. The

transmission was measured in the range 7–27 μ for the O and E rays with polarized light. The sample thickness for the transmission was 0.00262 cm, which was sufficiently thin to afford considerable transmission between the main absorption bands. The range 5–37 μ includes all of the strong lattice absorption bands. Since a major aim of this work was to obtain a high degree of accuracy in the measurements, they were taken with great care and in many cases rechecked several times. Particular attention was paid to eliminating errors due to scattered light and incomplete polarization. A quantitative dispersion analysis was carried out with the aid of an IBM 704 computer on the reflectivity for both the O and E rays. From this analysis the optical constants for both rays were obtained over the entire spectral region studied. The extinction coefficients so obtained compare well in the regions between the strong absorption bands with the transmission measurements. The primary results of the dispersion analysis are the values of the dispersion parameters which describe the optically active lattice oscillators. The measurements and analysis are com-