

## Low-Frequency Magnetoplasma Resonance in Metals\*

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(Received 14 November 1962)

This paper concerns a low-frequency magnetoplasma resonance which is related to the "whistler" of ionosphere physics and the "helicon" proposed by Aigrain. A sensitive method for observing this resonance is described. It involves the use of a diode ring demodulator as a phase-sensitive detector. The method has been used to study the resonance quantitatively in Cu, Ag, Au, and Pb; resonances were also observed in Sn, Zn, Cd, and Hg. Analysis of the data yields values for the Hall coefficient and resistivity as a function of magnetic field. The results obtained for the noble metals and Pb by this probeless method are in agreement with those obtained using conventional methods.

### I. INTRODUCTION

THE very low frequency magnetoplasma resonance has previously been studied in detail only in those high-purity metals with a small magnetoresistance.<sup>1-4</sup> In this paper we describe improvements in the measuring techniques which make it possible to study the effect quantitatively in a large class of metals. Detailed results are given for Cu, Ag, Au, and Pb. Resonances have also been observed in Sn, Zn, Cd, and Hg. Analysis of these results gives information concerning the Hall coefficient and the resistivity as a function of magnetic field in these metals.

The experiments to be described were carried out using thin-plate specimens. The theoretical treatment of the magnetoplasma resonance in thin plates with the large magnetic field perpendicular to the plate<sup>2</sup> predicts that the height of the resonance is proportional to the tangent of the Hall angle,  $u = RB/\rho(B)$ , where  $R$  is the Hall coefficient and  $\rho(B)$  is the total resistivity in the magnetic field  $B$ . This total resistivity  $\rho(B)$  is the sum of  $\rho(0)$ , the resistivity in the absence of the field (residual resistance) and  $\Delta\rho(B)$ , the increase in resistivity due to the applied field  $B$ . Theory also predicts that the  $Q$  of the resonance is given by  $(1+u^2)^{1/2}/2$ . Hence, the observability of the resonance is determined by the magnitude of  $u$ . For a given metal,  $u$  is limited by the resistivity  $\rho(B)$ . The highest values of  $u$  can be obtained with those very pure metals [small  $\rho(0)$ ] which have either a small magnetoresistance (Na and K)<sup>1-3</sup> or a saturating magnetoresistance (In and Al).<sup>2,4</sup> Values of  $u$  of the order of 10 ( $B \approx 10\,000$  G) can be obtained in these metals and the resonance is very easily observed. Our recent work has been with metals in which the magnitude of  $u$  has been one or two orders of magnitude smaller.

### II. METHOD OF MEASUREMENT

The configuration of the driving and pickup coils is essentially the same as that reported in reference 3. The improvement in measuring technique was achieved with the use of a phase sensitive detector. This permits the separation of the pickup coil signal into two components, one in-phase and the other in quadrature with respect to the driving current; this differs from the earlier measurements<sup>1-4</sup> which examined only the amplitude of the signal voltage. With the phase-sensitive detector, the resonant frequency is determined by a zero crossing, which can be measured very precisely. This can also be done using a Hartshorn bridge to separate the real and imaginary parts of the mutual inductance between the driving and pickup coils.<sup>2</sup> The phase-sensitive detector avoids the tediousness of point-by-point bridge measurements. The circuit is shown in Fig. 1; the crucial element is the diode ring modulator. The reference signal is the voltage across a resistor in series with the driving circuit and is of such a magnitude that it will always keep one side of the diode bridge conducting in the forward direction. The component of the signal voltage which is in phase with the driving current is obtained by applying the amplified pickup coil voltage directly to the diode bridge. The signal voltage in quadrature with the driving current is ob-

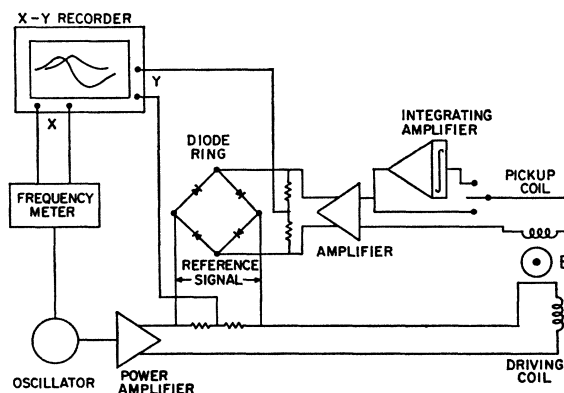


FIG. 1. Schematic circuit diagram, the integrating amplifier consists of a wide-band amplifier with a 2-sec  $RC$  circuit on the output.

\* This work was supported by the U. S. Atomic Energy Commission and the Advanced Research Projects Agency.

<sup>1</sup> R. Bowers, C. Legendy, and F. E. Rose, *Phys. Rev. Letters* **7**, 339 (1961).

<sup>2</sup> R. G. Chambers and B. K. Jones, *Proc. Roy. Soc. (London)* **A270**, 417 (1962).

<sup>3</sup> F. E. Rose, M. T. Taylor, and R. Bowers, *Phys. Rev.* **127**, 1122 (1962).

<sup>4</sup> P. Cotti, P. Wyder, and A. Quattropiani, *Phys. Letters* **1**, 50 (1962).

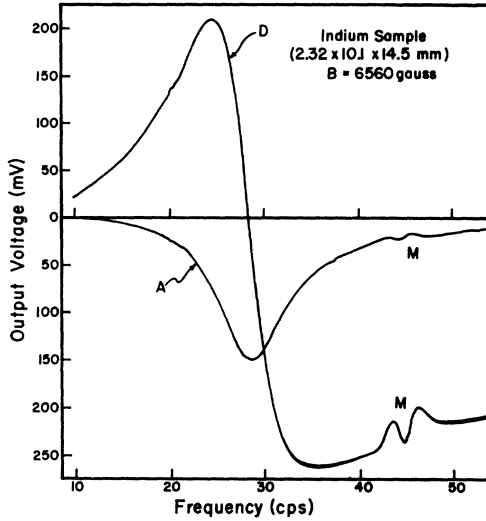


FIG. 2. Magnetoplasma resonance in an In sample with dimensions  $2.32 \text{ mm} \times 10.1 \text{ mm} \times 14.5 \text{ mm}$  for a magnetic field of 6560 G. The sample was in a 2000-turn coil. *A*—absorption curve amplified by a factor of  $10^4$  and with the integration time constant 2 sec. *D*—dispersion curve amplified by factor by  $10^2$ . *M* is a mechanical resonance (frequency independent of field).

tained by integration using an RC circuit with a 2 sec time constant. A convenient consequence of this integration is the fact that the height of the resonance is directly proportional to  $u$  and does not contain a factor  $\omega (=2\pi \times \text{frequency})$  which enters the total signal amplitude method. The output from the phase-sensitive detector is applied to the *Y* input of an *X-Y* recorder while a dc voltage proportional to frequency is applied to the *X* input. Typical resonances are shown in Figs. 2, 3, and 5; these are discussed in Sec. IV.

In order to reduce voltages induced in the pickup coil by mechanical vibration, a similar empty coil was mounted near the pickup coil but in series opposition. The coils had several thousand turns of No. 38 Cu wire. The metal specimens were rectangular thin plates approximately  $15 \text{ mm} \times 15 \text{ mm} \times 2 \text{ mm}$ . The coils had a rectangular cross section to achieve a maximum filling factor.

### III. ANALYSIS OF DATA

We have used the theory of Jones and Chambers<sup>2</sup> in order to analyze our data. For metals with a large  $u$ , the value of  $u$  can be determined from the  $Q$  of the resonance [ $Q = (1+u^2)^{1/2}/2$ ] but for metals with  $u < 1$ ,  $Q$  tends to  $\frac{1}{2}$  and, therefore,  $u$  cannot be determined accurately from  $Q$ . The absolute height of the resonance ( $=4u/\pi^2$ ) can be used to determine  $u$  with more precision when  $u$  is small. This requires either the measurement of the absolute height of the resonance or calibration using a metal for which  $u$  is known. We calibrated using an In sample of approximately the same size and shape as the other specimens to be measured; the value of  $u$  for this specimen had been determined (to 5%) from the  $Q$  of the resonance.

In order to obtain the Hall coefficient we require the resonant frequency ( $\nu_\infty$ ) in the limit of infinite  $u$  (i.e., negligible dissipation); the correction to be made to the observed frequency  $\nu_u$ , at finite  $u$ , is given by<sup>5</sup>

$$\nu_\infty = \nu_u [u / (1+u^2)^{1/2}]. \quad (1)$$

When  $u$  is small, the correction is large. Consequently,  $u$  must be determined as accurately as possible. (There is a direct analogy between this situation and a resonant LCR circuit with large dissipation.) The Hall coefficient

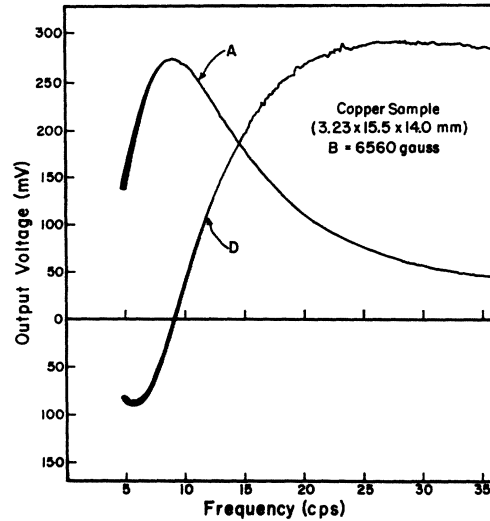


FIG. 3. Magnetoplasma resonance in a Cu sample with dimensions  $3.23 \text{ mm} \times 15.5 \text{ mm} \times 14.0 \text{ mm}$  for a magnetic field of 6560 G. The sample was in a 3000 turn coil. *A*—absorption curve amplified by a factor of  $10^5$  and with the integration time constant 3 sec. *D*—dispersion curve amplified by a factor  $10^2$ . Note the reversal in the sign of the signal compared with those of In and Pb.

is related to the corrected frequency  $\nu_\infty$  by

$$R = 8d^2(\nu_\infty/B) \times 10^{-7} \text{ m}^2/\text{A sec}, \quad (2)$$

where  $d$  is the thickness of the plate in m and  $B$  is the magnetic induction in webers/m<sup>2</sup>. The resistivity is given by

$$\rho(B) = [8d^2\nu_u / (1+u^2)^{1/2}] \times 10^{-7} \Omega \text{ m}. \quad (3)$$

Thus, the resonant frequency, when  $u \ll 1$ , is a direct measure of the resistivity. For the case of thin plates,  $\rho(B)$  in Eq. (3) is the transverse magnetoresistivity.

In practice, the calibration sample does not have exactly the same dimensions as the samples to be measured; therefore, a correction to the value of  $u$  was made for the difference in filling factor. The filling factor was taken to be proportional to the volume of the specimen. The validity of this correction was supported by our observation that the amplitude of the resonance was proportional to the sample volume. The corrections made for this variation of filling factor never exceeded

<sup>5</sup>  $\nu_\infty$  and  $\nu_u$  correspond to  $\nu_d$  and  $\nu_{1r}$ , respectively, in the notation of Jones and Chambers in reference 2.

50% and were more typically 10%. Corrections (typically 4%) were made for any variation of the drive current with frequency; the height of the resonance is proportional to the amplitude of the drive current. The input impedance of the  $X$ - $Y$  recorder (Moseley Autograph 2D) is a function of its sensitivity, and with the particular instrumentation used, the output impedance of the phase sensitive detector became comparable with the  $X$ - $Y$  recorder input impedance when used at the higher sensitivities ( $>20\text{mV/in.}$ ). The correction for this can be calculated from the impedances of the  $X$ - $Y$  recorder and the diode bridge; it can also be determined by measuring the effect of substitution of known resistances between the bridge and the  $X$ - $Y$  recorder. Both these methods gave consistent results. The magnitude of the correction was as follows: For the metals Cu and Ag 2%, whereas for Au and Pb the correction was variously 10 and 25%.

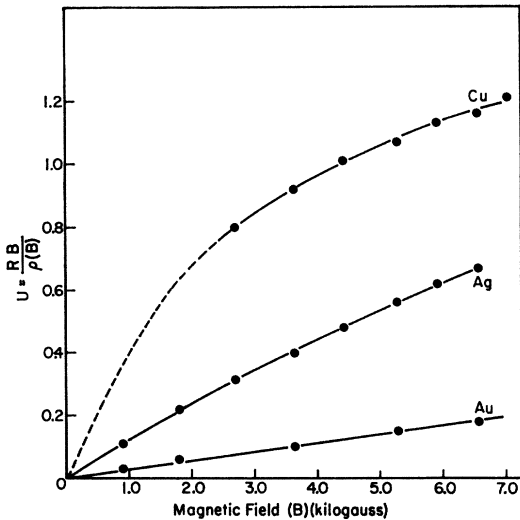


FIG. 4. The variation of the tangent of the Hall angle with magnetic field for the noble metals.

## IV. RESULTS

### A. Indium

The In calibration sample was made from Johnson and Matthey materials. Figure 2 shows a typical resonance in this material for a field of 6560 G. The dispersion curve is asymmetrical because it is the product of the true dispersion curve and frequency. The resonance marked  $M$  is a mechanical resonance of the coil support, such resonances are distinguished by the fact that the resonant frequency is independent of magnetic field; this is shown quite clearly in Fig. 5.

The Hall coefficient of In was determined during each helium run; the values obtained were all within 2% of the mean value  $+17.3 \times 10^{-11} \text{ m}^3/\text{A sec}$  which demonstrates the consistency of the method. This value is

TABLE I. Summary of residual resistance ratios and Hall coefficients.

Metal	$\rho_{RT}/\rho_4^\circ K^a$	$R_{\text{exptl}}$ ( $10^{-11} \text{ m}^3/\text{As}$ )	Exptl. error (%)	$R_{\text{theo}}^b$ ( $10^{-11} \text{ m}^3/\text{As}$ )
Cu	800	-7.8	12	-7.45
Ag	180	-11.4	16	-10.65
Au	100	-7.8	20	-10.60
Pb	5200	+14.9	20	...

<sup>a</sup> Average of values obtained using eddy current decay and values obtained from this experiment by extrapolation to zero field.

<sup>b</sup> Assuming 1 electron/atom.

slightly larger than the value  $+16.8 \times 10^{-11} \text{ m}^3/\text{A sec}$  obtained by Jones and Chambers, but the difference is within our total experimental error which is approximately 6% for this material. The  $n$  for our In sample has a typical value of 5 in 6000 G.

### B. Noble Metals

The primary material was American Smelting and Refining high-purity metal; the residual resistance ratios are given in Table I. The samples used in the resonance work were thin plates obtained by pressing the bulk material in a hydraulic press. After annealing for approximately 4 h at  $1200^\circ\text{K}$ , the plates were highly polycrystalline having a grain size of the order of  $\frac{1}{2} \text{ mm}$ .

Figure 3 shows a resonance in Cu for a magnetic field of 6560 G. This is typical of resonances observed in the noble metals, except that the resonances had a smaller

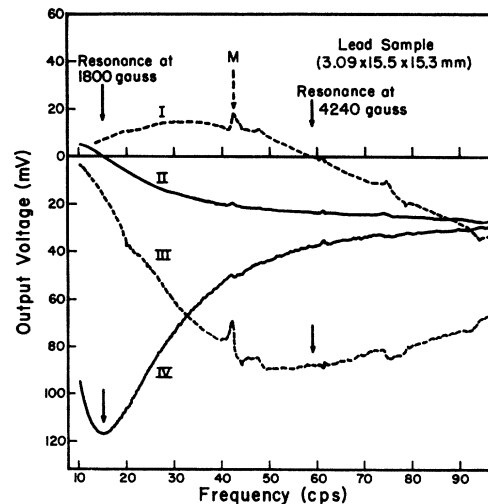


FIG. 5. Magnetoplasma resonances in a Pb sample with dimensions  $3.09 \text{ mm} \times 15.5 \text{ mm} \times 15.3 \text{ mm}$  for two magnetic fields. The solid curves are for 1800 G and the dashed curves for 4240 G. The sample was in a 2000-turn coil. The absorption curves III and IV were amplified by a factor  $10^5$  and had an integration time of 2 sec. The dispersion curves I and II were amplified by a factor  $10^2$ .  $M$  is a mechanical resonance with frequency independent of field. Note the decrease in amplitude of the resonance with increase in field.

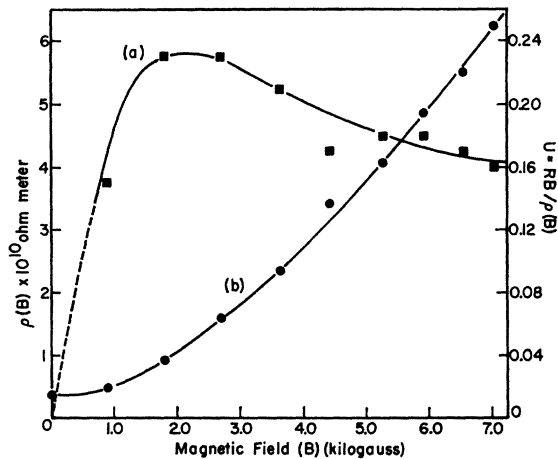


FIG. 6. The tangent of the Hall angle [curve (a)] and the magnetoresistivity [curve (b)] as functions of the magnetic field for Pb.

amplitude in Ag and Au. The change in sign of this resonance with respect to the In resonance (Fig. 2) demonstrates the difference in sign of the Hall coefficients of In and Cu. The results for the noble metals are presented together because they behave qualitatively in the same manner, i.e., the Hall coefficient was found to be independent of the magnetic field between 1000 G and the maximum field available, 7000 G; also the Hall angle increased with increasing magnetic field.

The parameter  $u$  for Au was approximately linear with magnetic field, showing that there was very little magnetoresistance. This was due to the high residual resistance of the gold; in fact the maximum value obtained for  $u$  was only 0.20 at 7000 G. For Cu,  $u$  appears to be tending towards saturation, demonstrating the presence of appreciable magnetoresistance. Ag shows a variation of  $u$  with field which is intermediate between that of Cu and Au; see Fig. 4.

The Hall coefficients obtained are given in Table I; the values are the average of several measurements on at least two samples. The measurements are consistent to within 5%. The absolute accuracy is discussed below. The correction for thermal contraction is of the order of 0.3% and was neglected. The coefficients for Cu and Ag are close to the free electron values of  $-7.45 \times 10^{-11}$  and  $-10.65 \times 10^{-11}$  m<sup>3</sup>/A sec, respectively, whereas the Au value is much smaller than  $-10.60 \times 10^{-11}$  m<sup>3</sup>/A sec as has also been found by other workers.<sup>6,7</sup>

### C. Lead

The material used was American Smelting and Refining high-purity metal. The metal was pressed into thin plates; the resulting samples were then annealed for an hour at 450°K.

The parameter  $u$  shows a variation with field which is quite different from that found in the noble metals and from all previously reported results for the magnetoplasma resonance.<sup>1-4</sup> Figure 5 shows two resonances in a Pb sample for magnetic fields of 1800 and 4240 G; note that the height of the resonance is smaller for the larger field. The parameter  $u$  first increases with increasing field, reaches a maximum and then decreases; see Fig. 6(a). This behavior is due to the large magnetoresistance of this metal [see Fig. 6(b)] which varies as  $B^2$  for fields below 4000 G.<sup>7</sup> The residual resistance ratio of this material was found to be 5200 by extrapolation of the total resistivity  $\rho(B)$  to zero field.

The Hall coefficient was found to be independent of the magnetic field for fields greater than 3000 G; this value is  $+14.9 \times 10^{-11}$  m<sup>3</sup>/A sec. The mean deviation from this value was 6%. Again the correction for thermal contraction (order of 1%) was neglected. This value for the Hall coefficient is in agreement with the value obtained for the same field range by Borovik.<sup>7</sup>

### D. Other Metals

Preliminary measurements have also been made upon the resonances observed in thin plates of Sn, Zn, Cd, and Hg. The Sn, Zn, and Cd all show behavior qualitatively similar to that found in Pb. The Hg shows an increasing  $u$  for increasing field similar to that found for Ag.

### E. Errors

The error in the Hall coefficient obtained by this method is very dependent upon the magnitude of  $u$  when  $u < 3$ . For  $u \gg 3$ , the Hall coefficient can be obtained from the observed resonant frequency with very small error.<sup>2,3</sup>

We estimate that the thickness of the samples is known to 1%, the magnetic field to 3%, and frequency to 2%. All of these quantities could be determined more precisely, but this would require substantial improvement in instrumentation. Since our objective was an exploratory study of the resonance in many metals, we considered the above precision to be adequate for this purpose.

The error in  $u$  obtained by the calibration technique was approximately 12%. The effect of this error upon the Hall coefficient depends upon the magnitude of  $u$ , as can be seen from Eqs. (1) and (2). Thus the estimated total error in the Hall coefficient is very much a function of the metal as can be seen from Table I.

### V. CONCLUSION

We have shown that the increase in sensitivity resulting from the use of a phase-sensitive detector makes

<sup>6</sup> R. G. Chambers, Proc. Roy. Soc. (London) A238, 344 (1956).

<sup>7</sup> E. S. Borovik, Zh. Eksperim. i Teor. Fiz. 27, 355 (1954).

it possible to study the magnetoplasma resonance in a much broader class of metals than has been previously studied. The work shows the potential and the limitations of this probeless technique for measuring the Hall coefficient and magnetoresistivity.

ACKNOWLEDGMENTS

The authors are indebted to R. D. Deslattes for suggesting the use of a diode ring as a phase-sensitive detector and J. M. Goodman for helpful discussions. We are grateful to S. Tallman for contributions to the experiment.

Anharmonic Contributions to the Debye-Waller Factor

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(Received 26 July 1962)

Cubic- and quartic-anharmonic contributions to the Debye-Waller factor of a Bravais lattice have been obtained in the classical limit. The contributions are of two types: One is proportional to the square of the absolute temperature, and the other is proportional to the cube of the temperature. This is in contrast to the linear dependence on the absolute temperature of the Debye-Waller factor in the harmonic approximation. The first type of anharmonic contributions represents just the anharmonic corrections to the mean square amplitude of vibration of an atom, while the second type of term is related to the thermal average of the fourth power of an atomic displacement. These results have the consequence that with the anharmonic contributions included, the Debye-Waller factor for a cubic crystal is no longer isotropic in the components of the scattering vector, although it possesses cubic symmetry. The anharmonic contributions are evaluated numerically for a fcc crystal with nearest-neighbor, central-force interactions. The anharmonic contributions to the intensity of x rays scattered by one-phonon processes are discussed briefly.

I. INTRODUCTION

THE intensity of x rays scattered by the thermal vibrations of a monatomic Bravais lattice is proportional to the sum<sup>1</sup>

$$I = \sum_{l,l'} \exp\{i\boldsymbol{\kappa} \cdot [\mathbf{x}(l) - \mathbf{x}(l')] + i\boldsymbol{\kappa} \cdot [\mathbf{u}(l) - \mathbf{u}(l')]\}, \quad (1.1)$$

where  $\mathbf{x}(l)$  is the position vector of the  $l$ th unit cell in the crystal and  $\mathbf{u}(l)$  is the displacement of the  $l$ th atom from its equilibrium position. The vector  $\boldsymbol{\kappa}$  is given by

$$\boldsymbol{\kappa} = (2\pi/\lambda)(\mathbf{s} - \mathbf{s}_0), \quad (1.2)$$

where  $\mathbf{s}_0$  and  $\mathbf{s}$  are unit vectors in the directions of the normals to the wave fronts of the incoming and scattered x rays, respectively, while  $\lambda$  is the wavelength of the x rays.

The displacements  $\{\mathbf{u}(l)\}$  are time dependent so that the expression (1.1) gives the instantaneous intensity. The observed intensity can be regarded as the average of (1.1) over a time long compared with the period of the atomic vibrations but short on a macroscopic scale. However, it is usually easier in statistical mechanical problems to replace time averages by ensemble averages, and this is the procedure we follow here. Thus, to obtain the expression for the observed

scattered intensity we must evaluate the thermal average,

$$\langle \exp\{i\boldsymbol{\kappa} \cdot [\mathbf{u}(l) - \mathbf{u}(l')]\} \rangle = \frac{\int \exp\{-\beta H + i\boldsymbol{\kappa} \cdot [\mathbf{u}(l) - \mathbf{u}(l')]\} d\Omega}{\int \exp(-\beta H) d\Omega}, \quad (1.3)$$

where  $H$  is the lattice Hamiltonian and  $d\Omega$  is the appropriate volume element of phase space.

The expression given by Eq. (1.3) represents the thermal average calculated in the classical or "high-temperature" limit. We have chosen to work in this limit because the effects we are studying in this paper are expected to be largest at high temperatures.

In the case that the lattice Hamiltonian is that appropriate to a harmonic crystal, in which case we denote it by  $H_0$ , the average given by Eq. (1.3) was evaluated first in 1914 by Debye,<sup>2</sup> whose analysis was subsequently corrected by Faxén<sup>3</sup> and by Waller.<sup>4</sup> However, no crystal is truly harmonic and the effects of anharmonic terms in the lattice Hamiltonian on all thermal properties of solids become more important as

<sup>1</sup> R. W. James, *The Optical Principles of the Diffraction of X-Rays* (G. Bell and Sons, London, 1954), Chap. V. To obtain the intensity in electron units the sum  $I$  must be multiplied by  $|f_0|^2$ , where  $f_0$  is the atomic scattering factor.

<sup>2</sup> P. Debye, *Ann. Physik* **43**, 49 (1914).

<sup>3</sup> H. Faxén, *Ann. Physik* **54**, 615 (1918).

<sup>4</sup> I. Waller, *Z. Physik* **17**, 398 (1923).