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X-Ray Compton Scattering for Aluminum*

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The intensity of the Compton scattering of x-rays by aluminum has been measured for values of $\sin\theta/\lambda$ from 0.3 to 0.6 A^{-1}. The method involved a self-consistent separation into the Compton and thermal diffuse scattering of the total intensities of diffuse scattering measured at 300°K and 5°K. The measured Compton scattering is significantly lower than the intensity calculated theoretically. Limitations of the present theories are discussed.

THE study of the diffuse scattering of x-rays offers a powerful method for investigating various imperfections in crystals, as is evidenced by the recent studies of ordering[1] and precipitation[2] transformations in alloys and of lattice vibrations[3] in monatomic crystals. The coherent scattering arising from such imperfections is always accompanied by the incoherent Compton scattering, which often forms a large part of the total scattering. In order that quantitative information concerning the imperfections be drawn from such measurements, one must have an accurate knowledge of the intensity of this Compton scattering.

THEORY

Expressions for the intensity of the Compton scattering by the electrons of a free atom have been derived both by classical[4,5] and quantum-mechanical[6,7] approaches and for the Thomas-Fermi model of an atom.[8,9] Breit[10] and Dirac[11] have shown that these expressions must be modified by a corrective factor arising from the recoil of the Compton electron. The corrected Compton-Raman-Wentzel equation, the expression usually employed, gives the intensity of this scattering in electron units (e.u.) as

$$I_{\text{e.u.}}=\{Z-\sum_{j=1}^{Z}|f_{jj}|^2\}\left(\frac{\nu'}{\nu}\right)^3, \tag{1}$$

and the corrected Waller-Hartree equation, theoretically the more accurate, is

$$I_{\text{e.u.}}=\{Z-\sum_{j=1}^{Z}|f_{jj}|^2-\sum_{j}\sum_{\substack{k\\ j\neq k}}|f_{jk}|^2\}\left(\frac{\nu'}{\nu}\right)^3, \tag{2}$$

where

$$f_{lm}=\int\psi_l^*(p)\psi_m(p)e^{i\mathbf{k}\cdot\mathbf{r}_p}dv_p.$$

The integration extends over all coordinates of any one electron, p. The Breit-Dirac correction[12] is the multiplicative factor, $(\nu'/\nu)^3$, where ν and ν' are the frequencies of the x-ray photons before and after scattering, respectively.

The extra negative term appearing in the Waller-Hartree equation, due to electron exchange, is made up of a sum over pairs of one-electron wave functions with which electrons of the same spin are associated. Physically, this term arises from the inclusion of the Pauli principle, which forbids transitions involving the passage of electrons to occupied states. Values for this term have been calculated for only two atoms, argon[7] and neon[13]; in practice it is generally neglected. Values for $\sum_j|f_{jj}|^2$ have been calculated by James and Brindley[14] from Hartree wave functions for the light atoms and for a few of the heavy atoms; these are tabulated in Compton and Allison.[15] In the absence of such calculations, one can employ the expressions derived by Heisenberg and Bewilogua from the Thomas-Fermi model of the atom, but the results must be expected to be more inaccurate.

If the atoms are not free but are located on a crystalline lattice, two effects will occur which may modify the above calculations. First, the packing of the atoms onto a lattice should severely perturb the wave functions of the outer electrons. The scattering from such atoms will still be formally described by the above expressions if the proper wave functions are used, but these are generally not available. An approximate expression can be obtained for the scattering from metal atoms by considering separately the scattering from the core and conduction electrons, treating the latter as components of a free electron gas. Zener[16] and Debye[17] have shown

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[1] J. M. Cowley, J. Appl. Phys. **21**, 24 (1950).

[2] C. B. Walker and A. Guinier, Acta Metallurgica **1**, 568 (1953).

[3] E. H. Jacobsen, Phys. Rev. **97**, 654 (1955).

[4] A. H. Compton, Phys. Rev. **35**, 925 (1930).

[5] C. V. Raman, Indian J. Phys. **3**, 357 (1928).

[6] G. Wentzel, Z. Physik **43**, 1 (1927).

[7] I. Waller and D. R. Hartree, Proc. Roy. Soc. (London) **A124**, 119 (1929).

[8] W. Heisenberg, Physik. Z. **32**, 737 (1932).

[9] L. Bewilogua, Physik. Z. **32**, 740 (1932).

[10] G. Breit, Phys. Rev. **27**, 362 (1926).

[11] P. A. M. Dirac, Proc. Roy. Soc. (London) **A111**, 405 (1926).

[12] If the scattered radiation is detected by a counter, so that actually one measures the number of scattered quanta per unit solid angle, the appropriate corrective factor is $(\nu'/\nu)^2$.

[13] Harvey, Williams, and Jauncey, Phys. Rev. **46**, 365 (1934).

[14] R. W. James and G. W. Brindley, Phil. Mag. **12**, 81 (1931).

[15] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), second edition, p. 782.

[16] C. Zener, Phys. Rev. **48**, 573 (1935).

[17] P. Debye, Physik. Z. **38**, 161 (1937).

that the intensity of the scattering from such conduction electrons is

$$I_{\text{e.u.}}=Z_c\left\{\frac{3}{2}\left(\frac{a\sin\theta}{\lambda}\right)-\frac{1}{2}\left(\frac{a\sin\theta}{\lambda}\right)^3\right\},\quad \frac{a\sin\theta}{\lambda}\leq 1,$$

$$I_{\text{e.u.}}=Z_c,\quad \frac{a\sin\theta}{\lambda}>1, \tag{3}$$

where

$$a=\left(\frac{8\pi}{3Z_c}\frac{V}{N}\right)^{\frac{1}{3}}.$$

Z_c is the number of conduction electrons per atom, and N/V is the number of atoms per unit volume. The intensity as calculated from this approximation is smaller than that calculated from the usual expression, Eq. (1), for angles which can extend up to $(\sin\theta)/\lambda \approx 0.3$ A^{-1}.

The drawing together of atoms onto a lattice also affects the distribution of allowed states for the Compton electron, reducing this from a semi-infinite continuum to a succession of allowed energy bands separated by forbidden energy gaps. As Laval[18] has pointed out, the Compton scattering process cannot operate when the recoil electron would be required to occupy a forbidden state. Thus this effect also should lead to a reduction of the intensity of the Compton scattering, the reduction probably being a function of the crystal orientation as well as of the scattering angle. It is extremely difficult even to estimate the order of magnitude of this effect.

There have been relatively few experiments designed to test the accuracy of these theoretical expressions as applied to the crystalline state. Several early experiments[13,19,20] were devoted to investigating the need for the extra negative term in the Waller-Hartree expression, Eq. (2), and other experiments[21–23] have been applied to a study of the Debye-Zener conduction electron approximation, Eq. (3); however, in all these experiments it was assumed that the scattering due to thermal vibrations could be described by the Debye expression,

$$I_{\text{e.u.}}=f^2(1-e^{-2M}),$$

an approximation whose validity is questionable.[24,25] Laval[26] has reported on a few measurements of the scattering from diamond, Al, and KCl, where the correction for the thermal scattering was made by various approximations. The values determined for the Compton scattering showed serious departures, particularly at small angles, from the values calculated from Eq. (1). No estimate was made of the accuracy of the measurements, which were made for only a few values of $(\sin\theta)/\lambda$. In view of Laval's results, it was decided to determine experimentally this scattering for aluminum in conjunction with an x-ray study of its lattice vibrations.

EXPERIMENTAL PROCEDURE

Measurements were made of the intensity of the diffuse x-ray scattering along the [100] axis in reciprocal space for specimen temperatures of 300°K and 5°K. The specimen was a flat single crystal of aluminum of 99.99% purity whose faces were cut parallel to (100) planes. CuKα radiation from a Machlett A-2 tube powered by a full-wave rectified source at 45 kv and 16 ma was monochromated by a toroidally bent LiF crystal so oriented as to focus the diffracted radiation on the face of the specimen. Higher frequency harmonics passed by the monochromator were eliminated from the recorded scattered radiation with a balanced nickel-aluminum filter placed before the Geiger counter detector. Horizontal and vertical divergences of the primary beam were ±1°. The face of the specimen was so oriented as to maintain equal angles with the incident and scattered radiation, thus keeping the absorption correction angularly independent. The average total counting time per point in reciprocal space was approximately 2½ hours. The measured intensities were normalized to absolute units by the standard technique of comparison with the high-angle scattering from amorphous substances. As amorphous materials, paraffin, lucite, and fused silica were used, the interference function of the last named being determined from a separate experiment with MoKα radiation; the three normalizations were internally consistent to better than 1%.

The low-temperature x-ray diffraction apparatus was developed by C. S. Barrett, E. A. Long, L. Meyer, and the author. It consists essentially of a double Dewar cryostat mounted on the center table of a precision spectrometer. The specimen is mounted beneath the inner Dewar and cooled by conduction. Radiation heating of the specimen from the outer walls is minimized by the use of slitted shields held at liquid N_2 and liquid He temperatures. Entrance and exit of the x-ray beam is allowed by a window of 0.003-in. Be foil cemented on with Araldyte cement and made vacuum-tight with a thin baked coating of Glyptal cement. The spectrometer is one of the original double-crystal spectrometers designed by Compton[27] and built by the Société Genevoise.

The measured intensities of the scattering at these two temperatures are shown in Fig. 1. This scattering

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[18] J. Laval, Compt. rend. **215**, 359 (1942).

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[20] G. E. M. Jauncey and J. H. Deming, Phys. Rev. **48**, 577 (1935).

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[22] K. Alexopoulos and S. Peristerakis, Physik. Z. **39**, 688 (1938).

[23] K. Alexopoulos and H. Scouloudi, Phil. Mag. **40**, 115 (1949).

[24] B. E. Warren, Acta Cryst. **6**, 803 (1953).

[25] F. H. Herbstein and B. L. Averbach, Acta Cryst. **8**, 843 (1955).

[26] J. Laval, Compt. rend. **215**, 278 (1942).

[27] A. H. Compton, Rev. Sci. Instr. **2**, 365 (1931).

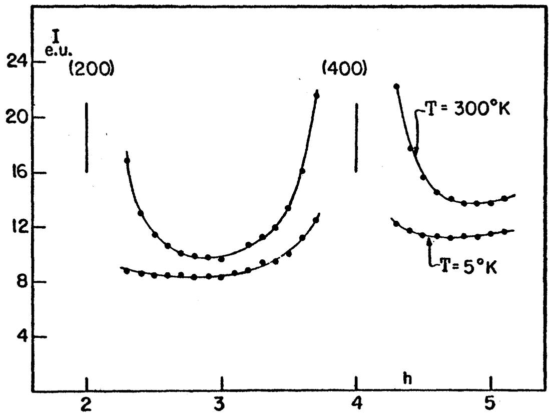

FIG. 1. Intensity of the diffuse scattering along the [100] axis in reciprocal space at 300°K and 5°K. The abscissa represents the distance along this axis in units of the reciprocal cell edge, *b*. The marked vertical lines indicate the positions of the (200) and (400) Bragg reflections.

is made up of the incoherent Compton scattering and the coherent thermal diffuse scattering. The Compton scattering is essentially independent of temperature. The thermal diffuse scattering[28] can be described effectively as the sum of three terms, called the first-, second-, and third-order scattering, of which the latter two can be calculated approximately.[29] The temperature dependence of the first-order thermal scattering along this direction in reciprocal space is given as

$$I(\mathbf{g})_{\text{e.u.}}=\frac{f^2e^{-2M}}{m}\left|\frac{S}{\lambda}\right|^2\frac{h}{\nu}\left[(e^{h\nu/kT}-1)^{-1}+\tfrac{1}{2}\right]. \qquad (4)$$

$I(\mathbf{g})$ is the intensity of this scattering at a point a vector distance $\mathbf{g}$ from the nearest reciprocal lattice point; f is the atomic scattering factor, taken from calculations[14] checked by experiments[30]; e^{-2M} is the Debye factor[31]; and m is the atomic mass. S/λ is the diffraction vector, the difference between the wave vectors of the diffracted and incident x-rays; h is Planck's constant; and ν is the frequency of the longitudinal elastic wave of wave vector $\mathbf{g}$.

The two sets of intensity data, after subtraction of the calculated contributions of the second- and third-order thermal diffuse scattering, allow the determination at each point of the two unknowns, the frequency of the particular elastic wave and the intensity of the Compton scattering. Given the complex temperature dependence of the first order thermal scattering, this solution for the unknowns is carried out most easily by a self-consistent method: at each point along this line in reciprocal space a value for the Compton scattering is assumed, which, in conjunction with the 300°K data, yields a frequency ν. From this frequency the intensity of the first-order scattering at 5°K can be calculated, which, taken with the 5°K data, gives a new value for the Compton scattering. The procedure is repeated, using the new value for the Compton scattering as the starting value, until initial and final values for this intensity are equal. It is assumed in this procedure that the thermal vibrations can be described as harmonic oscillators. This assumption is not rigorously satisfied, but the error in the Compton scattering arising from this does not appear important when compared with the errors in the original measurements.

RESULTS AND DISCUSSION

The results of this measurement of the Compton scattering are plotted as solid circles in Fig. 2. Errors in the experimental measurements and in the calculated corrections are such that the probable error of these values is about ±0.5 e.u. The measurements of Laval, shown as the solid squares, are in reasonable agreement with the present results where the comparison can be made. Curien[32] has developed a different method for measuring the Compton scattering, based on the use of a filter which has an absorption edge falling between the wavelengths of the modified and unmodified radiation. The values found by this method for the Compton scattering for aluminum are in accord with the present measurements, though showing considerably greater scatter.[33]

Two theoretical curves are also given in this figure. Curve *A* is obtained from the usual expression, Eq. (1), using values for $|f_{jj}|^2$ calculated by James and Brindley,[14] but with the Breit-Dirac correction appropriate

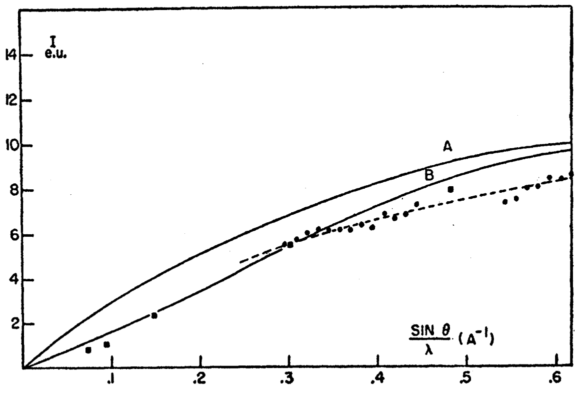

FIG. 2. Intensity of the Compton scattering from aluminum. Present measurements are given as the solid circles. The solid squares give the results obtained by Laval. Curve *A* is calculated from the Compton-Raman-Wentzel equation, and curve *B* is calculated from a modified Waller-Hartree expression.

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[28] See, for example, J. Laval, J. phys. radium **15**, 545 and 658 (1954), or R. W. James, *The Optical Principles of the Diffraction of X-Rays* (G. Bell and Sons, London, 1948), Chap. 5.

[29] C. B. Walker, Phys. Rev. **103**, 547 (1956) preceding paper.

[30] G. W. Brindley, Phil. Mag. **21**, 778 (1936), and G. W. Brindley and P. Ridley, Proc. Phys. Soc. (London) **50**, 96 (1938). A Hönl factor of +0.18 was used, as confirmed by these references.

[31] An x-ray Debye characteristic temperature Θ of 402°K was used, as checked by the temperature dependence of the integrated intensity of the (400) reflection.

[32] H. Curien, J. phys. radium **16**, 494 (1955).

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to counter detection. Curve B is obtained from a modified Waller-Hartree approximation: the contribution of the core electrons is calculated from Eq. (2), using the James and Brindley values for $|f_{jj}|^2$ for the core electrons of a free aluminum atom and the values of $\sum\sum_{j\neq k}|f_{jk}|^2$ calculated for a neon atom by Harvey, Williams, and Jauncey,[13] and to this is added the contribution of the three conduction electrons, calculated from Eq. (3). The difference between these two calculated curves for values of $(\sin\theta)/\lambda > 0.28$ is due exclusively to the exchange term in the Waller-Hartree expression. Its inclusion in this case reduces the intensity of the Compton scattering by as much as 20%, an amount which is certainly not negligible. Even greater differences appear at the lower angles.

The measured values for the Compton scattering are not fitted by either of the two calculated curves. The usual calculation, Curve A, is uniformly too high, the discrepancy being as much as 25%. The modified Waller-Hartree calculation shows much better agreement, but there are still significant differences at the higher values of $(\sin\theta)/\lambda$. While these differences may be due in part to the approximations employed in the calculation, such as using the exchange term calculated for neon, they may also arise in part from the band structure effect discussed above.

In summary, none of the present theoretical expressions for the intensity of the Compton scattering are reliable over a reasonable range of angles for the case of aluminum. The usual expression is seriously in error over the entire range of angles investigated, and the modified Waller-Hartree expression, while reasonably accurate at the low angles, shows significant errors at the higher angles. The source of these errors is probably to be found in the incomplete inclusion in the theory of the effects of the lattice periodicity on the electronic wave functions. Thus, if an accurate knowledge of the intensity of the Compton scattering is required, at present one must rely only on experimental measurements.

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Gyromagnetic Ratios of the Iron-Nickel Alloys

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Measurements by magnetomechanical experiments show that the gyromagnetic ratios for the alloys of the iron-nickel series undergo changes for low values of the induced magnetic intensity. This leads to a high- and a low-intensity value for g' for each metal. Two distinct curves of g' *vs* percent nickel are thus obtained. Nine different rods were used in which the content of nickel in iron varied from 0% Ni to 100% Ni.

INTRODUCTION

IN these experiments on the iron-nickel alloys, nine different rods were used[1] in which the content of nickel in iron varied from 0% Ni to 100% Ni. The results for the 100% Fe and 100% Ni rods have already been reported.[2] The same dependence of g' on the intensity of magnetization, noted for Fe and Ni in the above references, was observed also for the alloys of these metals.

Each of the iron-nickel rods, after casting, was inspected by x-ray to make certain of the absence of voids. The rods were then ground to size and annealed for two hours in dry hydrogen at 2000°F. They were then furnace cooled. These alloys contained impurities of a few tenths of one percent; mostly Mn, Al, or C. Each rod was wound with its own magnetizing winding and supported in the apparatus as a torsional pendulum as has been previously described.[2,3]

RESULTS

The following relationship was used to calculate the gyromagnetic ratio from the experimental data. Factors which are common to all of the rods are given values in the list of symbols. The other factors were determined independently for each rod. For the method of obtaining the data, see reference 2.

$$\rho\frac{e}{m}=\left(\frac{\pi Id}{4PXkm/e}-2i_e\sum A_e\right)\Big/(M_e-i_e\sum A_e),$$

where ρ = gyromagnetic ratio (g coul^{-1}), I = moment of inertia (g cm^2), d = (double) amplitude change per reversal of i_e (cm), P = period of torsional pendulum (sec), X = optical length = 1576.9 cm, k = phase angle constant = 0.99941, m/e = mass-charge ratio of electron $=5.6844\times10^{-9}$ g coul^{-1}, i_e = magnetizing current (amp), $\sum A_e$ = winding constant (cm^2), and M_e = magnetic moment (rod and winding) (amp cm^2).

Table I gives a condensation of the data taken for the 35% Ni 65% Fe rod. Each value of d given in this table

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[1] The Fe-Ni rods were cast by the metallurgical department of the General Motors Research Staff.

[2] G. G. Scott, Phys. Rev. **99**, 1241 and 1824 (1955).

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