Polarization of the $L\alpha_1$ X Rays of Mercury†

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(Received 12 May 1970)

The polarization of the $L\alpha_1$ characteristic x rays of mercury has been measured with respect to the emission plane. A mercury-vapor-target x-ray tube was used with a jet density such that most electrons encountered only a single exciting collision. A quartz crystal $(23\bar{5}4)$ plane which diffracted the $L\alpha_1$ line (1.241 20 Å*) at an angle $2\theta = 104°$ was used as the analyzer. The observed polarization was $(-14 \pm 4)\%$.

INTRODUCTION

Several polarization studies on the K characteristic x rays emitted by solid targets have shown no measurable effect.[1-4] However, if the K radiation were partially polarized, its observation with solid targets would probably be impossible because the scattering of the primary electrons is so great that approximately 30% emerge from the target in a backward direction. This problem was eliminated in the present experiments by the use of a gas-target x-ray tube.

Recently Mehlhorn[5] has shown that there should usually be partial polarization of the characteristic x radiation following the removal of an inner electron $(n, l > 0, j > \frac{1}{2})$ by electron or proton impact. This is due to the alignment of the ionized atoms with respect to the direction of the electron or proton beam. Mehlhorn's quantum condition is not satisfied for K radiation $(n = 1, l = 0)$, which could explain the failure of some earlier experiments. He calculated the polarizations for some lines in the L spectra of krypton and found them significant. It is interesting to note that the $L\alpha_2$ line should be polarized in a direction opposite to that of the $L\alpha_1$.

Bonse, Cleff, and Mehlhorn[6] carried out experiments on the L_{III} lines from solid targets of gold and tungsten, but observed no polarizations. These negative results are probably due to electron scattering, as discussed above, and suggest the need for employing a gas target. In fact, a reexamination of Koch's[7] data, obtained with a gas target, reveals clear indications of a small polarization in the unresolved $L\alpha_{1,2}$ doublet of mercury.

EXPERIMENT

The same mercury-vapor-target (jet) x-ray tube[8] was used as the source of x rays in the present work. Voltage and current were stabilized and a liquid-nitrogen condenser[8] was used to improve the vacuum and stability of operation. The x rays were emitted from the tube in a horizontal direction perpendicular to that of the electron beam. This determines the x-ray emission plane.

The polarization of the $L\alpha_1$ line (1.241 20 Å*) was determined by diffracting the x rays from a quartz $(23\bar{5}4)$ crystallographic plane ($d = 0.7891$ Å and $2\theta = 103° 42'$). The schematic diagram of the experimental arrangement is shown in Fig. 1. Measurements were taken in two different positions, viz., (a) crystal rotation axis parallel to the electron beam and detector in position D (with observed intensity $\mathcal{I}_{\parallel}$ and rotation angle β) (b) crystal rotation axis perpendicular to the emission plane and detector in position D′ (with observed intensity $\mathcal{I}_{\perp}$ and rotation angle γ).

The experiment required that the horizontal and vertical divergences of the x-ray beam be small enough to permit good resolution of the $L\alpha_1$ and $L\alpha_2$ lines. On the other hand, because of the low

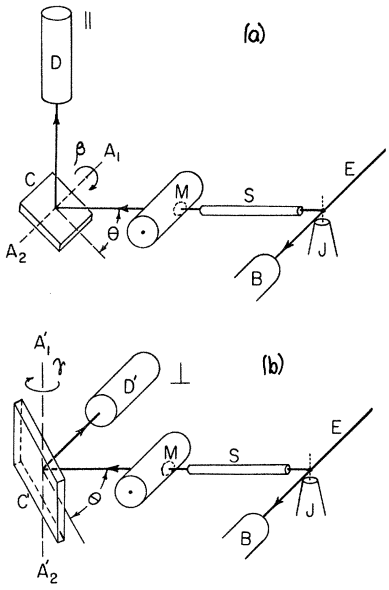

FIG. 1. (a) Schematic diagram of the experimental arrangement to record $\mathcal{I}_{\parallel}$, consisting predominantly of radiation polarized parallel to the emission plane. B is the beam stop, C is the crystal, D is the x-ray detector, E is the electron beam, J is the mercury jet, M is the monitoring counter, and S is the slit system. During a run, the crystal is rotated about axis $A_1 A_2$ through a small range of angles designated by β. (b) Arrangement to record $\mathcal{I}_{\perp}$, consisting predominantly of radiation polarized perpendicular to the emission plane. Crystal is now in position C′ and rotates about axis $A'_1 A'_2$, with rotation angle γ, while detector has been moved to D′.

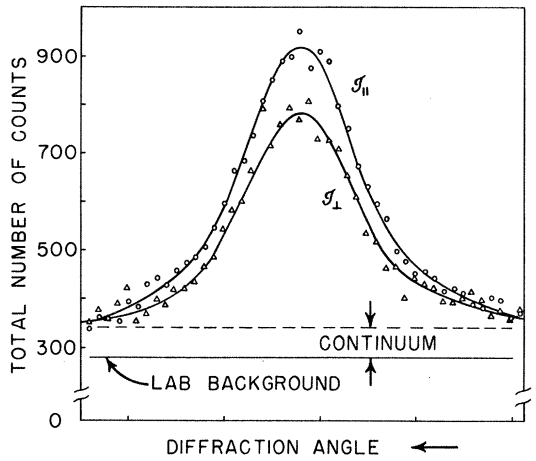

FIG. 2. Observed curves of Hg $L\alpha_1$ line and continuous background diffracted from $(23\bar{5}4)$ crystallographic plane of quartz. $\mathcal{I}_{\parallel}$ and $\mathcal{I}_{\perp}$ represent intensities recorded in positions shown by Fig. 1(a) and 1(b), respectively. The dashed line represents total background and the solid line that portion due to radioactivity (x-ray voltage off).

intensity of the x-ray source, it was vital that the observed intensity should not be unduly limited by slits. This problem was solved by the use of a slit system composed of 19 parallel thin-walled metal pipes (hypodermic tubing) with an inner diameter of 0.5 mm and 20 cm in length. This kind of slit provides about 20 min vertical and horizontal divergence and covers a relatively large part of the target volume.

Since the angle between the crystal surface and the crystallographic planes was negligibly small (about 15 sec), an optical adjustment procedure was used to align the x-ray beam, crystal, and detector. For this purpose the optical light source was temporarily located between the x-ray tube and the slit.

Diffracted intensity was registered by a scintillation (NaI) counter and conventional electronics. Twenty sets of diffracted intensity curves were taken with the crystal axis of rotation being alternated between the parallel and perpendicular positions. The purpose of this procedure was to minimize any possible effects due to the changes in the x-ray intensity. As a further precaution, incident intensity on the crystal was recorded by the monitor counter M (Fig. 1) throughout the experiment.

The counts observed in each angular position for each orientation of the polarimeter were totaled for all runs and plotted in Fig. 2. The upper curve shows $\mathcal{I}_{\parallel}$ as a function of the crystal rotation angle β; the lower one gives $\mathcal{I}_{\perp}$ as a function of γ. Before any further computation, the values were normalized to constant incident intensity by use of the monitor count.

Since the Bragg angle of the crystal differed from 45°, the diffracted x rays were not completely polarized, and a correction factor is required. Furthermore, in order to minimize the statistical error, it is desirable to make full use of all data in the observed curve and to avoid any need for unfolding. The desired expression is obtained as follows:

It will be assumed that the polarimeter axis (i.e., the axis about which the crystal is turned from $\parallel$ to $\perp$ position) is precisely normal to the crystal axis [A_1A_2 and $A_1'A_2'$ in Figs. 1(a) and 1(b), respectively]. Horizontal divergence α and vertical divergence φ will be defined with respect to this polarimeter axis; the angular distribution of the incident radiation will be denoted by $G(\alpha, \varphi)$. If the slit system S is symmetric and properly aligned along the polarimeter axis and the source intensity is uniform, then $G(\alpha, \varphi)$ will be a symmetric function; however, no such assumptions are required.

By a simple modification of a result given by Compton and Allison[9] the expressions for $\mathcal{I}_{\parallel}(\beta)$ and $\mathcal{I}_{\perp}(\gamma)$ may now be written

$$\mathcal{I}_{\parallel}(\beta) = \int\int\int I_{\parallel}(\lambda)G(\alpha,\varphi)C\{\varphi - \beta - (\tfrac{1}{2}\alpha^2)\tan\theta - [(\lambda-\lambda_0)/\lambda_0]\tan\theta\}d\alpha\, d\varphi\, d\lambda$$
$$+\int\int\int I_{\perp}(\lambda)G(\alpha,\varphi)\bar{C}\{\varphi - \beta - (\tfrac{1}{2}\alpha^2)\tan\theta - [(\lambda-\lambda_0)/\lambda_0]\tan\theta\}\, d\alpha\, d\varphi\, d\lambda, \quad (1a)$$

$$\mathcal{I}_{\perp}(\gamma) = \int\int\int I_{\perp}(\lambda)G(\alpha,\varphi)C\{\alpha - \gamma - (\tfrac{1}{2}\alpha^2)\tan\theta - [(\lambda-\lambda_0)/\lambda_0]\tan\theta\}d\alpha\, d\varphi\, d\lambda$$
$$+\int\int\int I_{\parallel}(\lambda)G(\alpha,\varphi)\bar{C}\{\alpha - \gamma - (\tfrac{1}{2}\varphi^2)\tan\theta - [(\lambda-\lambda_0)/\lambda_0]\tan\theta\}d\alpha\, d\varphi\, d\lambda. \quad (1b)$$

$I_{\parallel}$ and $I_{\perp}$ represent the wavelength distributions of the two polarization components incident on the crystal, i.e., the quantities needed to calculate polarization. C and $\bar{C}$ are, respectively, the Darwin-Prins crystal functions for components polarized perpendicular to and parallel to the *plane of incidence on the crystal.* The crystal rotation angles β and γ are defined to represent de-

viations from the Bragg angle for a ray of wavelength λ_0 with zero divergence. λ_0 denotes the wavelength of the line peak and θ the corresponding Bragg angle (51° 51′). The wavelength integration includes the entire range where $I_{\parallel}(\lambda)$ or $I_{\perp}(\lambda)$ is appreciable and the angular integration covers the range where $G(\alpha, \varphi)$ is nonzero.

From Eqs. (1) it easily follows that

$$\int \vartheta_{\parallel}(\beta)\, d\beta = S_G S_C \int I_{\parallel}(\lambda)\, d\lambda + S_G S_{\overline{C}} \int I_{\perp}(\lambda)\, d\lambda, \quad (2a)$$

$$\int \vartheta_{\perp}(\gamma)\, d\gamma = S_G S_C \int I_{\perp}(\lambda)\, d\lambda + S_G S_{\overline{C}} \int I_{\parallel}(\lambda)\, d\lambda, \quad (2b)$$

where

$$S_G = \int\int G(\alpha, \varphi)\, d\alpha\, d\varphi,$$
$$S_C = \int C(x)\, dx, \; S_{\overline{C}} = \int \overline{C}(x)\, dx. \quad (3)$$

By the Darwin-Prins theory,[10] it can be shown that $S_{\overline{C}} = S_C |\cos 2\theta|$. It will now be assumed that the two polarization components are proportional, i.e., $I_{\perp}(\lambda) = kI_{\parallel}(\lambda)$, which is consistent with experimental results.

With these substitutions the polarization fraction becomes

$$P = [\int I_{\perp}(\lambda)\, d\lambda - \int I_{\parallel}(\lambda)\, d\lambda]/[\int I_{\perp}(\lambda)\, d\lambda + \int I_{\parallel}(\lambda)\, d\lambda]$$
$$= \{[\int \vartheta_{\perp}(\gamma)\, d\gamma - \int \vartheta_{\parallel}(\beta)\, d\beta]/[\int \vartheta_{\perp}(\gamma)\, d\gamma + \int \vartheta_{\parallel}(\beta)\, d\beta]\}$$
$$\times [(1 + |\cos 2\theta|)/(1 - |\cos 2\theta)] \quad (4)$$

with the range of integration covering the entire region of appreciable intensity. Note that this expression is independent of the function $G(\alpha, \varphi)$ and hence involves no assumptions on symmetry of the angular distribution. It may be shown that the above result still holds when background intensity is significant, provided that background correction for each component is made in the usual way.

DISCUSSION AND RESULT

The background in the observed curves of Fig. 2 is due primarily to radioactive contamination of the laboratory, but also includes the partially polarized continuous x-ray spectrum. The dashed line at 340 counts represents the total background (as recorded in the $\vartheta_{\parallel}$ position well-removed from the $L\alpha_1$ line peak) and the solid line at 280 counts denotes the radioactive background with the x-ray voltage off, leaving a continuous background of 60 counts. From the results of Koch[7] for a 30 kV accelerating voltage and the inclusion of the correction factor in $|\cos 2\theta|$ from Eq. (4), it follows that the continuous x-ray background in the $\vartheta_{\perp}$ position should be roughly 60% of that for the $\vartheta_{\parallel}$ position.

The curves in Fig. 2 for $\vartheta_{\parallel}$ and $\vartheta_{\perp}$ do not show any clear evidence of the expected difference in background. This discrepancy may be due to three causes: (i) The continuous spectrum may be a smaller fraction of the background than indicated here; (ii) the approximate value derived from Koch's results may overestimate the polarization of the continuous background; and (iii) statistical errors in the two curves can account for part, but probably not all, of the discrepancy.

Thus there is considerable uncertainty as to how best to correct for polarization of the continuous background. One might assume a background of 60 counts for $\vartheta_{\parallel}$ and 36 counts (60% of 60) for $\vartheta_{\perp}$; in this case the application of Eq. (4) to the data shown in Fig. 2 (after correction for background and monitor count) yields a polarization $P = -14.2\%$. On the other hand, 60 counts might be assumed as more nearly an average value for the continuous background with 70 counts for $\vartheta_{\parallel}$ and 42 counts for $\vartheta_{\perp}$; this hypothesis gives a value $P = -12.9\%$. If one assumes the same continuous background of 60 counts for both curves, which is contrary to both theory and experiment, the polarization would increase to -26.9%. (If the monitor count correction were eliminated, all of these figures would be reduced in magnitude and would be -10.8, -9.6, and -21.8%, respectively.) It is clear that the uncertainties involved here dominate the statistical error of the data, which give a standard deviation of about 11% of P[e.g., the first value above would become $(-14.2 \pm 1.6)\%$].

If the crystal is not quite perfect, the factor in $|\cos 2\theta|$ in Eq. (4) should be decreased, but cannot fall below unity in any case. However, this effect, if any, is probably small.

From the above discussion it appears that the best value for P lies between -12.9 and -14.2%. The final result may be stated as $P = (-14 \pm 4)\%$, where the error is a rough estimate of the standard deviation (70% confidence interval).

In any event this experiment seems to establish unambiguously that the characteristic $L\alpha_1$ x-ray line of mercury is definitely polarized. The result is in fair agreement with Mehlhorn's calculated value for krypton when one considers the difference in atomic number. Thus the existence of polarized characteristic radiation has been established both theoretically and experimentally; further quantitative work is obviously desirable.

ACKNOWLEDGMENT

The authors are indebted to Dr. J. S. Thomsen for helpful discussion and his critical comments on the manuscript.

†This work was performed while one of the authors (J. H.) was on an exchange program sponsored by the Czechoslovak Academy of Sciences and the U. S. National Academy of Sciences.

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[1]J. A. Bearden, Proc. Natl. Acad. Sci. U. S. 14, 539 (1928). References to earlier experiments are listed in this paper.

[2]E. O. Wollan, Proc. Natl. Acad. Sci. U. S. 14, 864 (1928).

[3]A. H. Compton, Proc. Natl. Acad. Sci. U. S. 14, 549 (1928).

[4]H. Mark and K. Wolf, Z. Physik 52, 1 (1928).

[5]W. Mehlhorn, Phys. Letters 26A, 166 (1968).

[6]U. Bonse, B. Cleff, and W. Mehlhorn (private communication).

[7]R. J. Koch, dissertation, The Johns Hopkins University, 1969 (unpublished).

[8]J. A. Bearden, F. N. Huffman, and J. J. Spijkerman, Rev. Sci. Instr. 35, 1681 (1964).

[9]A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (Van Nostrand, New York, 1935), p. 716. This result is obtained from Eq. (9.50a) by including the angle of rotation of the crystal, integrating over divergence angles and wavelengths, and treating the two polarization components independently.

[10]B. W. Batterman and H. Cole, Rev. Mod. Phys. 36, 681 (1964).

PHYSICAL REVIEW A VOLUME 2, NUMBER 5 NOVEMBER 1970

Nonperturbative Approach to the Theory of Transition Probabilities

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(Received 18 February 1970)

A nonperturbative approach to the theory of transition probabilities is presented. The results of this theory are employed to investigate the validity of the assumptions inherent in the perturbative approach. For this purpose, the simple examples of absorption of radiation (first-order process) and Raman scattering (second-order process) are used. The method proposed, which is based upon finite-difference techniques, represents an accurate solution of the time-dependent Schrödinger equation in a given representation and has many advantages over methods based upon the perturbation series.

INTRODUCTION

In several recent papers[1,2] attention has been drawn to the conditions which are necessary to define the concept of "transition probability per unit time" within the framework of time-dependent perturbation theory. Since the most important formula of perturbation theory is the widely used "golden rule" for the transition probability per unit time, the validity of the assumptions employed to derive the rule is a matter of some importance. Briefly, the necessary conditions[3-5] are that the time should be sufficiently long that energy conservation can be assured and yet sufficiently short that the total probability of transition is small. At first sight, these conditions appear to be somewhat contradictory, but, in the absence of an accurate expression for the transition probability, there appears to be little possibility of ascertaining the correctness of the assumptions.

Brooks and Scarfone[1] have recently investigated this problem by a modified form of perturbation theory which still employs the idea of an infinite-series expansion, but in which it is comparatively easy to guarantee such necessary properties as the unitarity of transition amplitudes.

In this paper, we shall describe an accurate method by means of which transition amplitudes may be calculated and, therefore, by means of which the aforementioned difficulties may be resolved. Before doing so, however, it will be useful to survey the advantages and disadvantages of the perturbation-series approach to the calculation of transition probabilities.

This approach has an advantage which the alternative we shall propose does not possess: that a formal expression can be written for the transition amplitude. This formal expression, as an infinite series, cannot be employed in practice, it being necessary in general to terminate it at the first relevant nonvanishing term. For certain simple processes this technique appears to be fairly satisfactory, but for many others, such as the amplitude for a low-energy collision, it is certainly inadequate. Furthermore, this approach, since it is an "initial-rate" theory, cannot adequately describe important cases which involve single and multiple resonances. In addition, the calculated transition amplitudes are not ordinarily unitary.

The alternative which we shall propose is not based upon perturbation theory and has none of the disadvantages inherent in such methods. On the