Is there optical activity in the X-ray region?

by

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Abstract

In this lecture a Polarimeter for investigating polarisation effects in X-rays is described. Experiments performed to detect optical activity in quartz and cinnabar are presented. There is evidence (although quite tenuous) that optical activity exists in the X-ray region. It is less than 2% to 3% of the rotation in the visible region but if it does exist in these substances, the rotation is definitely in the same direction as that of the optical activity in the visible region. By replacing the analyser of this polarimeter with experimental crystals it hoped to study the mosaicity of crystals. The break-down of Friedel's Law has been observed in cinnabar. The possibilities of using two wavelengths to solve the phase problem are mentioned. It is also hoped to look for the optical analogue of the Borrmann effect in iridiscent potassium chlorate.

1. Introduction

When G.N. Ramachandran left the Indian Institute of Science to occupy the Chair of Physics at the Madras University I was asked to take over the X-ray section which was being planned at Bangalore. I was to start research in X-ray crystallography and to "look after" the students working in this field. I, therefore, greatly welcome this opportunity to speak here today. In this brief 15 minutes, I shall present my future plans so as to elicit comments from the senior scientists of the Academy present here today. I also will

Text of talk delivered at the Academy, 1952

present, what I consider to be, some extremely interesting results.

Till now I have been interested in optical phenomena like birefringence, optical activity, magneto-optic rotation etc. To begin with, therefore, I am planning the following programme which are really extensions of my earlier interests - to the X-ray region.

1) Set up an X-ray polarimeter.
2) Attempt to detect optical and magneto-optic rotation, in the X-ray region.
3) Study the absorption phenomenon in X-rays and see if it could be used for determining the phases of X-ray reflections.
4) See if well known X-ray effects like the Borrmann effect are observable in the optical region.

Slide I illustrates the polarimeter we have recently set up. X is the X-ray tube to which B a Brass box (lined with lead) is attached and which contains P the polarising KCl crystal with a ground face parallel to the (110) plane is mounted on a horizontal axis. T is the tangent screw by which P can be rotated and C_1 is the first collimator. Sl is the slide with adjusting screws by which the experimental crystals can be introduced into position in the polarised X-ray beam and normal to it. C is the second collimator, S is the spectrometer on whose vertical axis a goniometer G is mounted. A is the analyser, again a KCl crystal with a face ground parallel to (110). E is an extension moveable arm of the spectrometer with a post Po to which is attached the Geiger Muller counter GM. R is a rod which can couple the analyser crystal A and the GM counter (so that the crystal and the counter can be

The 440 reflection ($2\theta = 88$) from the first crystal P consists a monochromatic X-ray beam polarised to within 1% which goes down the vertical axis parallel to the axis of rotation of the spectrometer. It falls on the analyser crystal whose azimuth can be rotated. The (440) Bragg reflection is chosen for analysis. When the analyser (after proper adjustment) is rotated the intensity as measured by the counter should vary as

$$I_\phi = I\,(\cos^2\phi + \sin^2\phi \cos^2 2\theta)$$

where θ is the Bragg angle of the 440 reflection and ϕ is the angle between plane of polarisation and that of the analyser. Since $2\theta = 90^\circ$ the formula almost reduced to

$$I_\phi = I \cos^2\phi$$

The introduction of an experimental quartz crystal in the polarized beam decreases the intensity considerably. P and A are crossed - (apart from the background to the cosmic ray and other causes there is a residual intensity), since 2θ is not exactly 90°). When the crystal is introduced this intensity drops considerably. By changing the azimuth of the analyser it was difficult to see whether the position of the minimum is changed or not.

The most sensitive position for the detection of any change in intensity with azimuth is when $\phi = 45^\circ$. To get over the absorption problem the following technique was tried. 4 quartz plates were made, two right rotating and two left rotating. They were ground and polished perpendicular to the optic axis so that magnitude of these rotations in the optical region was the same to within two minutes of arc i.e. the thicknesses were same to about 0.005mm. This was also verified with a dial gauge. ϕ is set at 45° and the doublet system $R_1 + R_2$ (which was on one part of the slide)was introduced. The X-ray

intensity as detected by the GM counter, dropped by about two orders of magnitude. The number of counts was measured for a time T. The ($L_1 + L_2$) doublet was then introduced by pushing in the slide. The X-ray absorption (in theory) should be the same. Any change in intensity should be due to change in the state of polarisation of the merging X-ray beam. Although the counting statistics was poor and the high voltage was not stabilised there appeared to be an extremely small change in the number of counts - but <u>always</u> in the same direction. All permutation and combinations of the 4 plates taken two at a time to eliminate thickness effects were made and one could conclude (if one were prejudiced enough in favour of optical activity in the X-ray region!) that there was a minute rotation about two orders of magnitude less than the rotation in the optical region.

Since the optical activity of cinnabar is almost two orders of magnitude larger than that in quartz, a similar experiment would have shown whether optical activity existed in the X-ray region or not. There were some excellent cinnabar plates in Prof. Sir C.V. Raman's collection. Unfortunately, most of them were right rotating so the "doublet" experiment could not be tried. Secondly, the absorption was also very large in HgS for CuKα. But in this case one could detect (by crossing) a very small change in ϕ. The rotation (if it exists at all) is less than 2% to 3% of the optical rotation in the visible region.

The conclusion one arrives at is that there is weak evidence to show that optical activity in the X-ray region <u>may</u> exist. The optical activity is less than 2% to 3% of that in the optical region but if it does exist it is in the same direction as in the optical case for quartz and cinnabar.

The formula for the variation in intensity with azimuth for a perfect and mosaic crystals are different.

$I_{\phi}(\text{mosaic}) = I\,(\text{Cos}^2\phi + \text{Sin}^2\phi\ \text{Cos}^2\ 2\theta)$

$I_{\phi}(\text{perfect}) = I\,(\text{Cos}^2\phi + \text{Sin}^2\phi\ \text{Cos}\ 2\theta)$

one can use the polarimeter arrangement with minor modification for researches in estimating the mosaicity of crystals. The experimental crystal must in that case replace the analyser crystal and ~~it~~ tilted. the GM counter raised on the post and fitted.

Some experiments on absorption have also been started. Since HgS is noncentro symmetric one wished to know whether one could observe the violation of the Friedel's law (as Coster Knol and Prins (1930) did in ZnS). It was not difficult to observe the violation of Friedel's law. It seems, therefore, one can use the idea suggested by Bijvoet that the difference in intensity between I(hkl) and I ($\overline{hkl}$) can be used for phasing X-ray reflections.

By changing the wavelength to the other side of the absorption edge one can in theory get rid of the imaginary part of the atomic form factor and reduce the absorption. However, there should still be a change (a decrease) in the real part of the atomic form factor (Honl 1933). It is my intention to see whether we could exploit this change together with the breakdown of the Friedel's law to solve the vexed phase problem in X-ray crystallography.

(Lecture delivered at the 18th Annual Meeting of the Indian Academy (1952))