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X RAYS FROM SYNCHROTRON:
NEW CHALLENGE FOR NEUTRON SCATTERING*

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Abstract

A brief review is given of current developments in x-ray scattering techniques at synchrotron radiation facilities. Highly collimated, intense white radiations open up new areas of research in condensed matter physics and challenge the traditional domains of neutron scattering. These include energy dispersive scattering, the use of anomalous dispersion, magnetic diffraction and direct energy analysis by backscattering. The relative merits of x-ray and neutron scattering techniques will be discussed. The unique advantage of neutron scattering is the capability of performing polarization analysis. We will discuss in some detail the current developments at Brookhaven in inelastic scattering of polarized neutrons. In addition, we will also discuss special technical problems associated with the search for phasons utilizing a high resolution triple axis spectrometer.

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I. Introduction

When I selected the title of my talk I had hoped that the Brookhaven National Synchrotron Light Source would be operating by now and I believed, optimistically, that I could present new and exciting results of our own. As it turned out, delay of construction of the Brookhaven X-ray ring has been such that I have not yet participated in any synchrotron x-ray scattering experiments. Meanwhile, I have been involved heavily with the development of polarized neutron spectrometers at Brookhaven. My talk today is therefore shifted more to the technical problems encountered with neutron scattering.

We will first discuss the characteristics of synchrotron x-ray radiation¹ with emphasis on comparison with the steady state and pulsed neutron sources. New research tools, such as synchrotron x-ray radiation and the pulsed neutron source, always open up new areas of scientific research. Then the more established research tools, like steady state research reactors, have to reevaluate their relative merits and shift emphasis accordingly.

There is no need to emphasize the importance of neutron research for the participants of this conference. I would like to discuss two specific technical aspects of neutron research, namely, polarization analysis and high (energy) resolution measurement, which represent the unique advantage of neutron scattering.

II. X-rays from Synchrotron

Several synchrotron sources have already been in operation and many more are now under construction.¹ One of the most important characteristics of x-rays emerging from a synchrotron is their narrow vertical divergence (≈ 40 seconds of arc). Combined with the small source size (< 1 mm), this gives an extraordinarily bright x-ray source throughout a continuous energy spectrum. The improvement over the conventional x-ray machine is of the order of a thousandfold, and with wigglers and undulators, this improvement factor may be considerably higher. With this type of source one can easily attain a very high momentum resolution, $\Delta q \approx 10^{-4} \text{ \AA}^{-1}$.

Typical examples are given in Fig. 1 and Fig. 2; these were taken recently at Stanford Synchrotron Radiation Laboratory by the spectrometer developed by D. E. Moncton and G. S. Brown.² X-ray beams are tailored for a particular experimental need by a combination of mirrors and monochromators; these types of spectrometers will be available soon in several laboratories in the United States, Europe and Japan. As you can see in Fig. 1, the synchrotron experiments vastly improve the equivalent data at a rotating anode, which in turn is usually superior than a neutron spectrometer in q resolution. High q resolution studies will clearly be the future research area which can be expected to be dominated by synchrotron x-ray facilities. We neutron scatterers should keep

in mind that one can extend the range of $S(Q)$ into smaller q ($Q = G+q$) by utilizing synchrotron x-rays. However, if one is interested in the dynamical aspects of condensed matter physics, namely the study of $S(Q, \omega)$, then the neutron scattering technique will remain the most versatile tool.

In regard to the high q resolution of a synchrotron x-ray spectrometer, it is interesting to consider the possibility of x-ray scattering from magnetic crystals. Magnetic x-ray diffraction has been demonstrated³ already but it appears so much more difficult to measure compared with the "natural" neutron diffraction. With the built-in high q resolution, however, this aspect of x-ray scattering may offer an interesting area of research and even challenge the neutron domain.⁴

Another important new aspect of synchrotron x-rays are their continuous spectrum; the neutron monopoly until now. One can immediately see new applications such as the use of anomalous dispersion and energy dispersive scattering. Then in these areas, x-ray and neutron must now compete with other factors such as relative cross sections of constituent atoms and radiation damages during the exposure.

The final topic I would like to discuss is the direct energy analysis of $S(Q, \omega)$ by high resolution x-ray scattering. With the use of higher order reflections of perfect crystals, it was already demonstrated that an energy resolu-

tion of 10 meV may be obtained.⁵ In order to get sufficient intensities from excitations in solids, one has to relax the angular resolution, without sacrificing the energy resolution. A back scattering spectrometer for this purpose has been thought about at Brookhaven and some progress has been made.⁶ If successful, it may offer a very fascinating tool to study excitations from extremely small single crystals.

I would like to switch my talk now to the neutron scattering problems with which I have recently been involved. As I have just outlined, the synchrotron sources offer plenty of challenges to neutron scattering. However, as we all agree, the neutron technique will remain as one of the central research tools in condensed matter physics. Two aspects I will discuss today are closely related to the special characteristics of the neutron scattering techniques: polarization analysis and high-energy resolution (meV to μ eV). These are the areas where x-rays cannot possibly challenge neutrons.

III. Polarized Beam Development

Recently we have been engaged in the development of polarized beam techniques. Almost twenty years ago, with C. Shull and R. Nathans, we had a very active double axis polarized beam program for the study of spin density distributions in solids. The last ten years have seen a dramatic comeback of the utilization of polarization, with particular interest

in polarization analysis, which was described in the classic paper⁷ by Moon, Riste and Koehler in 1969. A special impetus was given to the use of polarized beams by the development of Mezei's neutron spin echo⁸ in 1972.

Last year we started the joint US-Japan project on neutron scattering and an advanced polarized beam spectrometer is now under construction. Our goal is first to obtain a highly polarized beam of reasonable intensity on a triple axis machine and then to evaluate various beam modulation techniques. We have thus far tried two polarizing schemes, (1) composite Heuslers and (2) a polarizing FeGe multilayer following a bent pyrolytic graphite crystal. These give intensities at the sample position from 4 to 6 times lower than bent pyrolytic graphite at 14 meV. Our composite Heusler consists of two 30x30x5 mm pieces arranged for vertical focussing. It is smaller and somewhat less efficient than the ILL Heusler composite recently reported by Zeyen.¹⁰

The properties of polarizing multilayers were recently reported by Majkrzak, Passell and Saxena.⁹ We have arranged a multilayer polarizer in the exit collimator after a bent graphite as shown in Fig. 3. Conceptually this is an unbalanced double monochromator scheme. Preliminary results are very promising¹¹; high polarization and high reflectivity. At present, efforts are made to improve the reflectivity at wider collimators; this requires a larger d-spacing variation.

During the process¹¹ of improving the flipping ratios¹² of the Heusler-Heusler combination, we have discovered an interesting variation of the flipping ratio as a function of neutron energy (Fig. 4). This is due to a simultaneous scattering in the Heusler crystals. The flipping ratio is mainly controlled by smaller additional components $\delta(\pm)$

$$F = \frac{(F_N + F_M)^2 + \delta_{(+)}^2}{(F_N - F_M)^2 + \delta_{(-)}^2} .$$

Simultaneous scattering is very common in neutron scattering because of the relatively large size of the resolution function in ω -Q space. We have utilized the energy scan extensively to elucidate spurious peaks. It is sometimes very difficult to remove the simultaneous scattering as demonstrated in Fig. 5. In the case¹³ of SrTiO_3 , it requires a tight collimation and lower energy to reach clear regions. Thus judicious selection of neutron energy can give a very high flipping ratio at lower energies.

We have carried out several polarized beam experiments using our new set up. A study of the magnetic excitations near the maximum of $S(Q)$ in amorphous ferromagnets¹⁴ and the magnetic excitations in O_2 .¹⁵ We also plan to investigate possible spin density waves in several organic conductors, by utilizing the polarization analysis after scattering.

IV. High Resolution Study Near a Bragg Peak

In recent years extensive triple axis measurements have been carried out very near the Bragg peaks. The motive of these studies is to observe second magnons in antiferromagnets or phasons in incommensurate crystals. Inelastic neutron studies are usually the principal method to probe these interesting regions. Recently, we came across one important resolution problem, unique to the study of excitations at small q and ω . As schematically shown in Fig. 6(a) the transverse Bragg tails in (b) are well known. Another mechanism of spurious peaks are depicted in Fig. 6(c) as L or M. On these lines, the crystal satisfies Bragg conditions either at E_i or E_f ; as described in detail by Currat and Axe,¹⁶ the incoherent (or thermal diffuse) cross section of monochromator (or analyzer) produce the false peaks. The regular phonons are Bragg-phonon-Bragg in sequence of the scattering, the Currat-Axe peaks are phonon-Bragg-Bragg or Bragg-Bragg-phonon.

Looking at the diagram Fig. 6(c), it appears safe to scan along exactly the X direction ($\Delta y = 0$), then the spurious peaks should not appear. However, we have noticed, for more than ten years,¹⁷ the mysterious peaks appear at very small q_x with the energy slope of about $4k$, in meV-Å. Very recently, similar peaks were observed again in Cr¹⁸ around the magnetic satellite position $(1-\epsilon, 0, 0)$. (See Fig. 6(d)).

We felt it was now important enough to clarify the origin of these spurious peaks. Uemura, Grier and Shirane¹⁹ first repeated the Cr measurements of ILL. Then we characterized the shape of resolution function at $k = 1.33 \text{ \AA}^{-1}$ and all 20' collimators using perfect Ge(111) reflection. The careful test measurement produced only three spurious branches L, T and M in Fig. 6(c) but not along the x-axis. It became apparent that Q and R branches are actually L and M, namely Currat-Axe peaks.

The very important feature which escaped our attention until now is the effect of the sample mosaic spread. At higher neutron energy, the mosaic spread effectively increase the Δy dimension of the resolution function. When we use smaller and smaller neutron energies, the mosaic effect becomes more important compared with the size of resolution function. For example, Δy spread of 0.5° mosaic crystal is $0.02 \text{ \AA}^{-1}$ at $Q = 2 \text{ \AA}^{-1}$; this is bigger than the Δy span of the resolution function at 3 meV. Thus the L and M branches appear at the nominal $\Delta y = 0$ scan because of the mosaic. This effect alone explains all of the longitudinal Bragg tails we have observed in MnF_2 , RbMnF_3 and Cr. Additional confirmation of this explanation was given by the fact that only the Q branch of Fig. 6(d) is observed with a spectrometer with double monochromators, not R. The consecutive scattering essentially eliminate the incoherent portion of the cross section in the monochromator which is responsible for R.

In the light of this new discovery of the longitudinal Bragg tails, it is important to reexamine all of the recent neutron scattering studies which reported sharp phason modes at small energy transfer.

This review is benefited by many stimulating discussions with J. D. Axe, S. Burke, B. Grier, C. Majkrzak, D. E. Moncton, W. Stirling, and Y. Uemura.

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Figure Captions

- Fig. 1: Comparison of q resolutions of x-ray scattering with rotating anode and synchrotron sources. After R. M. Fleming, D. E. Moncton, J. D. Axe and G. S. Brown to be published.
- Fig. 2: High resolution scattering data of xenon absorbed on graphite taken at Stanford Synchrotron Radiation Laboratory by P. A. Heiney, R. J. Birgeneau, G. S. Brown, P. M. Horn, D. E. Moncton, and P. W. Stephens (Phys. Rev. Letters 48 (1982) 104).
- Fig. 3: Polarizing monochromator scheme: bent graphite followed by FeGe multilayer with d -spacing of 50 Å.
- Fig. 4: Flipping ratio of Heusler-Heusler combination with (111) reflections; vertical axes are not identified. After Majkrzak and Shirane (Ref. 11).
- Fig. 5: Example of very persisting simultaneous scattering. These data demonstrate that the $1/2$ (111) reflection is missing in SrTiO_3 below 110 K transition (Ref. 13).
- Fig. 6: Resolution problems near Bragg reflections. (b) well known Bragg tail, T , for transverse scan. (c) Currat-Axe peaks, L and M (see text). (d) Q and R branches are actually L and M produced by finite mosaic spread of crystal.

Figure 1

XENON ADSORBED
ON GRAPHITE (001) SURFACE

Figure 2

Figure 3

Figure 4

Figure 5

(a)

(b)

(c)

(d)

Figure 6