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POLARIZED NEUTRON SPECTROMETER DEVELOPMENT AND EXPERIMENTS AT BROOKHAVEN

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Résumé - A Brookhaven un travail récent centré sur le développement de spectromètres à neutron polarisé reflète un renouveau d'intérêt pour l'analyse de polarisations. Des cristaux de Heusler et des multicouches de FeGe utilisés conjointement avec des cristaux de graphite pyrolytique ont été testés comme monochromateurs sur un spectromètre trois-axes. Le but est d'abord d'obtenir des faisceaux polarisés d'intensité raisonnable et d'évaluer ensuite, en utilisant les propriétés de faisceaux polarisés, les différentes améliorations instrumentales qui contrôlent la résolution du spectromètre. Des progrès obtenus, dans cette voie, seront reportés. Une étude récente des excitations magnétiques dans des ferromagnétiques amorphes, utilisant ces possibilités nouvelles de l'analyse de polarisations, sera aussi discutée.

Abstract. - Recent work at Brookhaven has focussed on the development of polarized neutron spectrometers and reflects the resurgence of interest in polarization analysis. Both Heusler crystals and FeGe multilayers used in conjunction with pyrolytic graphite crystals have been tested as polarizing monochromators on a triple-axis instrument. The goal is to first obtain polarized beams of adequate intensity and then to evaluate various instrumental innovations which affect control of spectrometer resolution by utilizing the properties of polarized beams. Progress in these regards is reported. A recent study of the magnetic excitations in amorphous ferromagnets employing the instrumentation for polarization analysis developed thus far is also discussed.

I. Introduction. - Possessing a magnetic moment, the neutron is a unique and often indispensable probe of many condensed matter systems in which the constituent atoms have spin-dependent cross sections. To take full advantage of the neutron moment usually requires both a polarized incident beam and polarization analysis of the scattered neutrons as described in the classic paper by Moon, Riste, and Koehler[1]. However, because of the relatively low reflecting efficiencies of available polarizing monochromators, this powerful technique has in practice been limited primarily to either elastic scattering or experiments for which the neutron spin echo technique introduced by Mezei[2] is applicable.

Recent work at Brookhaven has focussed on the development of polarized beam spectrometers. The first goal is to obtain on a triple-axis machine a highly polarized beam of intensity comparable to that of an unpolarized beam. Two polarizing schemes have been tried thus far: 1) Heusler crystal monochromator and analyser; and 2) polarizing FeGe multilayers used in conjunction with bent pyrolytic graphite(PG) crystals. The performance of these two configurations are described in the following sections. Secondly, once polarized beams of adequate intensity have been obtained, various instrumental innovations affecting the control of spectrometer resolution by utilizing the properties of the polarized beams are possible. One particular idea will be presented in some detail below. Finally, a study of the magnetic excitations near the maximum of the scattering function S(Q) in amorphous ferromagnets[3] recently performed at Brookhaven using polarization analysis will be discussed.
Several years ago Lynn et al. [5] demonstrated that alternating thin films of Fe and Ge (with the Fe layers magnetized to saturation) can act as an efficient polarizing monochromator with high reflectivity. However, these multilayers were made by evaporation onto small glass substrates and had relatively large d-spacings, of the order of 150 Å. Thus for neutrons of wavelength λ=1.5Å, the Bragg angle θ (given by the usual relation, λ = 2d sin θ) is approximately 0.3° which in order to reflect a beam 1 cm wide requires a multilayer 2 m in length.

Fig. 1. Flipping ratio of Heusler-Heusler combination for (111) reflections: vertical axes are not identified.
Using a radio frequency sputtering apparatus designed specifically for the purpose, FeGe multilayers with d-spacings as small as 40 Å (20 Å thick Fe layers and 20 Å thick Ge layers) have been deposited uniformly on float glass substrates approximately 5 cm x 50 cm (although substrates as large as 10 cm x 90 cm can be accommodated). For \( d = 40 \) Å, a multilayer 50 cm long can reflect a beam almost 1 cm wide at a wavelength of 1.5 Å. Peak reflectivities greater than 90% with polarizing efficiencies of 98% have been obtained. Nonetheless, it is important to consider the reflectivity of a multilayer in some detail.

A mosaic crystal can be described in terms of an angular distribution of perfect microcrystallites which reflect a given wavelength over a finite range of incident angles. The angular acceptance of a multilayer (with parallel layers) depends, for a particular wavelength, not on an angular mosaic but on a distribution of d-spacings. Fig. 2 shows a plane in reciprocal space in which lie two distributions (taken to be Gaussian for the purpose of illustration) of reciprocal lattice vectors: one of the directions of the vector \( \mathbf{G}_0 \); the other of reciprocal lattice vector magnitudes along a single direction. The former characterizes a mosaic crystal while the latter corresponds to a multilayer with a distribution of d-spacings. Position along the vertical axis labeled \( N \) is proportional to the number of reciprocal lattice vectors. Ideally, to measure directly the peak reflectivity and angular acceptance first requires a perfectly parallel and monochromatic incident beam. To make such a measurement on a mosaic crystal then requires that it be rotated about an axis perpendicular to \( \mathbf{G}_0 \) through the mean Bragg angle \( \theta \) with a detector fixed at a scattering angle of \( 2\theta \). This is the well-known "rocking curve" scan which traverses the circular path of radius \( \mathbf{G}_0 \) in Fig. 2. For multilayers, on the other hand, a \( \theta:2\theta \) scan tracing a path along the \( y^* \) axis is appropriate.

In practice, the peak reflectivity and angular acceptance of a multilayer (for a given wavelength) is measured on a spectrometer with an energy resolution \( \Delta E/E < 1\% \) and with 1 min and 2 min non-reflecting, single slit collimators before and after the multilayer, respectively. Angular acceptances of about 4 min of arc (full width at half maximum) in \( \theta \) (or 8 min in 2\( \theta \)) for a \( \theta:2\theta \) scan were measured for multilayers with a nominal d-spacing of 40 Å at 5 meV. The observed width is greater than both the instrumental width and that calculated for an ideal multilayer having a perfectly uniform d-spacing (with the same number of bilayers). This inherent width is presumably due to a spread in d-spacings corresponding to deposition rate fluctuations in the sputtering system. Rocking curve widths, on the other hand, were only 1 to 2 min and of the order of the instrumental resolution. This is consistent with a picture in which the bilayers are almost perfectly parallel but have a finite distribution of d-spacings. For very fine collimation, the multilayers have been measured to be more efficient reflectors than PG (for one neutron spin state). However, at coarser collimation, the overall reflectivity of the multilayer can be less than that of PG. This is due to the relatively narrow angular acceptance of the multilayer. For example, the triple-axis spectrometer referred to above with 40 minutes collimation yields intensities at the detector \( \approx 30\times \) lower than that which would be obtained with PG monochromator and analyser. If the Heusler monochromator is replaced with a PG and FeGe multilayer monochromator-polarizer combination, the loss factor is reduced to \( \approx 15\times \) at 40 min collimation and \( \approx 8\times \) at 10 min collimation.
In order to enhance the overall reflectivity of a multilayer for beams of greater angular divergence requires, in principle, an increase in the angular acceptance for each wavelength component in the beam. As discussed above, this can be accomplished with a broader distribution of d-spacings. Numerical calculations of the dynamical structure factor were performed in order to determine the interference effects to be expected for a particular sequence of different d-spacings. These calculations are analogous to those encountered in the solution of thin-film optical interference problems[7]. Various sequences have been tried and found to perform as predicted. The current limitation, however, is the total number of bilayers which can be deposited before rising stresses in the film eventually break it apart. At present, 2000 to 3000 bilayers with \( d = 40 \) Å can be successfully deposited: unfortunately, approximately 1000 bilayers of a single d are needed to obtain peak reflectivities > 90%. The difference between the multilayer monochromators described here and the Mezei supermirror should be mentioned. The supermirror employs a continuously incremented sequence of d-spacings to extend the critical angle for specular reflection while the multilayer monochromator exhibits a distinct Bragg diffraction separated from the region of specular reflection. Thus far the practical advantage of the multilayer monochromator over the supermirror is the higher angle of reflection, especially for shorter neutron wavelengths. A comprehensive discussion of the technical problems encountered in the actual fabrication of the multilayers will be published in a separate paper.

In concluding this section we mention several other pertinent properties of the FeGe multilayers: 1) 98% polarizing efficiencies are obtained in magnetic fields as low as 100 Oe; 2) reflectivities for higher-order wavelengths are typically one percent or less; this in most cases eliminates the need for a higher-order filter, thereby yielding an increase in the intensity of the primary wavelength and removing the constraint of working within filter "windows"; and 3) the effect of simultaneous reflections on polarizing efficiency, as observed in Heusler crystals, for example, is not a problem with multilayer polarizers.

IV. FeGe multilayers as spectrometer elements. - In the Bragg reflection process for a perfect crystal with a single d-spacing, the limiting wavelength resolution \( \Delta \lambda / \lambda \) is given by \( \cot \theta \Delta \theta \) where \( \Delta \theta \) represents the angular divergence of a polychromatic incident beam. \( \Delta \lambda / \lambda \) is therefore relatively poor for a typical multilayer since the angle \( \theta \) is small. This is one of the reasons for using the multilayer in conjunction with a PG crystal. If the beam is polarized, however, the wavelength or energy resolution can also be significantly improved by using a wavelength dependent flipper such as the one developed by Drabkin[8,9]. The fundamental unit of the polarized-neutron spectrometer we propose to build consists of a pair of polarizing multilayers between which a corrugated, current carrying Al foil is inserted as shown in Fig. 3. The foil produces a small, spatially oscillating magnetic field perpendicular to the beam direction. A larger, uniform magnetic field \( H_o \) is superimposed perpendicular to both the oscillatory field and the beam direction. The resultant magnetic field acts as a velocity selective resonance flipper; i.e., only those neutrons with velocities in the neighborhood of \( v_o = \gamma H_o / \hbar \) (where \( \gamma \) is the gyromagnetic ratio) undergo a spin flip. Neutrons with other velocities are unaffected and are subsequently not reflected by the second multilayer in which the magnetization direction is opposite to that of the first. Together, the pair of multilayers and flipper produce polarized beams with an energy or wavelength spread which can be varied by simply changing the length of the current carrying section of the corrugated foil. The energy resolution \( \Delta E / E = 1 / M \) where \( M \) is the number of reversals in the oscillatory magnetic field direction. It has been calculated that for \( M=400 \), a \( \Delta E / E \) of about 1% can be obtained (independent of \( E \)). The fundamental unit described above would function collectively as polarizing monochromator with a second identical unit serving as the analyser of the spectrometer. Because the angular divergence of the neutron beam, which is a couple of degrees at most, has a negligible effect on the energy resolution of this flipping device (for resolutions of the order cited above), this spectrometer would have the remarkable property that energy and momentum resolution are essentially decoupled. Energy and momentum resolution could then be selected independently to best suit a particular experiment.
V. Magnetic excitations in amorphous ferromagnets. — Magnetic excitations can be studied using a polarized incident beam without polarization analysis of the scattered beam. If a ferromagnet is placed at the sample position in a horizontal magnetic field which is parallel to the scattering vector \( \mathbf{Q} \), then the difference of spin-up (+) and spin-down (−) cross sections selects out only the inelastic spin-wave component. Using a time-of-flight polarized beam spectrometer, Mook and Tsuei [10] performed a detailed study of the magnetic excitations in the amorphous ferromagnetic alloy \( \text{Fe}_{75}\text{P}_{15}\text{Co}_{10} \) in this way. They observed a minimum in magnetic excitation energy near the first maximum of \( S(\mathbf{Q}) \) and reported a large, 20 meV, magnon energy gap at room temperature. We have recently reexamined [3] these magnetic excitations near the maximum in \( S(\mathbf{Q}) \) for small energy transfers (up to 15 meV) in the same sample studied by Mook and Tsuei and in two other Fe-based alloy compositions. Our measurements were made on a triple-axis spectrometer at Brookhaven where the sample was exposed to an unpolarized beam from a PG monochromator. A horizontal magnetic field of 7.5 kOe was applied along the scattering vector \( \mathbf{Q} \). The direction of the magnetic guide field for the scattered beam was rotated smoothly from along \( \mathbf{Q} \) at the sample to the vertical at the flipper which was located just before a Heusler (111) analyser. This geometry is the inverse of the conventional arrangement in which the incident beam is polarized but polarization analysis of the scattered neutrons is not done. The two configurations are in principle equivalent insofar as the measurements are concerned.

For each of the three samples examined, our results are consistent with a mechanism involving "umklapp" scattered, long wavelength spin waves with no energy gap. This is in contrast to the large energy gap reported by Mook and Tsuei. For inelastic magnetic scattering measurements at large momentum transfer, the possible distinction between the momentum transfer of the scattering event, \( \mathbf{Q} \), and the momentum of the excitation, \( \mathbf{q} \), must be considered. The distinction is clear in crystalline systems where the two may differ by any reciprocal lattice vector \( \mathbf{G}_{hk\ell} = \mathbf{Q} - \mathbf{q} \). In this "umklapp" scattering the deficit momentum is taken up by the lattice as a whole. Essentially identical processes occur in amorphous solids as well [11,12]. Neutron scattering experiments have established the existence of well-defined spin-wave excitations at small wavevectors \( \mathbf{q} \). These excitations then cause scattering about elastic diffraction maxima. The expressions \( S_{\text{w}}^+ \) and \( S_{\text{w}}^- \) for the appropriate generalization of "umklapp" ferromagnetic spin wave scattering for a monotonic amorphous solid are derived in Ref. 3. The "umklapp" model was tested by convoluting \( S_{\text{w}}^- - S_{\text{w}}^+ \) with the appropriate instrumental resolution function and then comparing directly with the data. A typical example is shown in Fig. 4. The measured intensity differences display apparent gaps which can be shown to be caused by an instrumental resolution effect. The measurements consist of taking the difference of (+) and (−) intensities at a given \( \mathbf{Q} \) and frequency \( \omega \). Neglecting magnon lifetime effects, \( S^+ \neq 0 \) and \( S^- = 0 \) for \( \omega > 0 \) while \( S^- = 0 \) and \( S^+ \neq 0 \) for \( \omega < 0 \). In the limit \( \hbar \omega \ll kT \), \( S^- (\omega > 0) = S^+ (\omega < 0) \) so that \( S^+ - S^- \) is an odd function of \( \omega \). The measured intensity is a convolution of \( S^+ - S^- \) with the

![Diagram of polarizing multilayer spectrometer](image-url)
A numerical calculation of $g g' - 45$ according to the amorphous model of Ref. 3 is convoluted with the appropriate spectrometer resolution function and compared directly to the experimental data.

Appropriate instrumental resolution (which is even in $\omega$) and therefore passes through zero at $\omega = 0$. This method will thus produce a peak at a finite energy even for a gapless excitation spectrum. The better the resolution, the smaller the size of the apparent gap observed. However, the cross sections near $\Delta E = 0$, where the difference between flipper off and on should be zero, can in some cases be significantly distorted by a transmission polarization effect. If (+) neutrons are more efficiently scattered than (−) neutrons by the sample, then the sample on average is irradiated by a larger number of (−) neutrons. This is a relatively minor effect of the order of a few percent at most and is inconsequential in most scattering experiments. However, this can become a substantial correction when the difference of two large numbers is taken near $\Delta E = 0$.

The energy gap reported by Mook and Tsuei might be due to the effect of finite instrumental resolution discussed above. Furthermore, they chose to represent the susceptibility $\chi''(Q, \omega)$ rather than the scattering function $S(Q, \omega)$. The susceptibility $\chi''(Q, \omega)$ is related to the scattering function by the Boltzmann factor: $\chi''(Q, \omega) = \frac{1}{n(\omega) - 1} S(Q, \omega)$. Since for $\hbar \omega \ll kT$, $\chi''(Q, \omega) = (\hbar \omega/kT) S(Q, \omega)$ is odd in $\omega$ and shows a peak at finite $\omega$ even if $S(Q, \omega)$ does not. A peak in $\chi''(Q, \omega)$ does not necessarily imply a gap. The interested reader is referred to the original papers of Mook and Tsuei [10] and Shirane et al. [3] for more detailed accounts.

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