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POLARIZED-NEUTRON SPECTROMETER DEVELOPMENT AND EXPERIMENTS AT BROOKHAVEN

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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 $S(\vec{Q})$ in amorphous ferromagnets[3] recently performed at Brookhaven using polarization analysis will be discussed.

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II. Heusler monochromator and analyser. - The performance of a conventional triple-axis spectrometer with a single Heusler crystal 30x30x5 mm in the transmission geometry as monochromator and two Heusler crystals of the same dimension in the reflection geometry (and tilted for vertical focussing) as analyser was tested. Intensities $\approx 30\times$ lower compared to unpolarized beams utilizing bent PG crystals were obtained at the detector. The (111) peak reflectivities of the Heusler crystals for one spin state were measured to be about 1/3 that of a good (flat) PG crystal while their mosaic spreads were found to be between 2/3 and one degree. This reflectivity together with the absence of vertical focussing at the monochromator position and the loss of one-half the incident beam upon polarization accounts for the observed net loss in intensity. These Heusler crystals are smaller and less efficient than those recently reported on by Zeyen[4]. It is hoped that a vertically bent, composite Heusler monochromator currently being constructed will significantly increase the overall intensity.

An important measure of the performance of a polarized beam spectrometer is the instrumental flipping ratio, i.e. the ratio of intensity for flipper off divided by that for flipper on (as obtained using a known, purely non-spin-flip scatterer, for example). In the process of improving the flipping ratio of the Heusler-Heusler combination described above, a striking variation of the flipping ratio as a function of neutron energy was observed as shown in Fig. 1. This effect is due to simultaneous or multiple scattering in the Heusler crystals. The variations are more pronounced at lower energies where the Ewald sphere is relatively small and the number of reciprocal lattice points within resolution distance is low. At higher energies the larger number of reflections near the surface of the Ewald sphere results in a smoother variation of the flipping ratio with energy. By judicious selection of neutron energy, flipping ratios greater than 100 have been obtained employing a modification of the Mezei flat coil flipper[2]. Inside the standard flat coil is a second flat coil whose interior magnetic field direction is perpendicular to that of the outer coil. The windings of the two coils are in physical contact with each other so that the coils have practically the same thickness. This compound coil is then placed in a vertical guide field of adjustable magnitude and is oriented so that the internal field of the outer coil is horizontal and perpendicular to the beam direction. The internal magnetic field of the inner coil is consequently vertical and is adjusted to cancel the externally applied vertical guide field in the common interior region of the flat coils. The magnetic fields of the coils and guide field are of sufficient magnitude that stray fields are negligible in comparison. Therefore, the magnetic field is almost exactly vertical outside and horizontal inside the flat coils, as is required for the optimum performance of the flipper. This flipper has proven to be highly efficient even when large horizontal magnetic fields are located at the sample position.

III. FeGe polarizing multilayers. - 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 $\lambda=1.5\text{Å}$, the Bragg angle θ (given by the usual relation, $\lambda = 2d \sin \theta$) 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[6], 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 $\vec{G}_0$; 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 $\vec{G}_0$ through the mean Bragg angle θ with a detector fixed at a scattering angle of 2θ . This is the well-known "rocking curve" scan which traverses the circular path of radius 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.

Fig. 2. Distribution of reciprocal lattice vector lengths and directions.

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 minutes of arc (full width at half maximum) in θ (or 8 min in 2θ) 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. For example, at 14.7 meV on a triple axis spectrometer with PG monochromator and analyzer and all 40 min collimators, a multilayer whose peak reflectivity for a given wavelength is 90% is found to reflect approximately 25% of one spin state incident at the sample position. (Even so, a multilayer such as this when used in conjunction with a bent PG crystal as a polarizing monochromator combination yields about 2X more intensity at the sample position than that obtained with our flat Heusler crystal in the transmission geometry).

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 $d = 40 \text{ \AA}$ can be successfully deposited: unfortunately, approximately 1000 bilayers of a single d are needed to obtain peak reflectivities $> 90\%$. Progress is being made in developing a technique where self-supporting multilayer films with a total thickness of $\sim 100,000 \text{ \AA}$ can be removed from the substrate and physically superimposed on other similar films until the desired properties are obtained. 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 $\cot\Delta\theta$ 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 θ 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_0 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 $v_0 = \gamma H_0 / \pi$ (where γ 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 monochromatic 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 $\Delta E/E \approx 1/M$ 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.

Fig. 3. Fundamental unit of the polarizing multilayer spectrometer.

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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 $\vec{Q}$, 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 $S(\vec{Q})$ and reported a large, 20 meV, magnon energy gap at room temperature. We have recently reexamined [3] these magnetic excitations near the maximum in $S(\vec{Q})$ 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 $\vec{Q}$. The direction of the magnetic guide field for the scattered beam was rotated smoothly from along $\vec{Q}$ 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, $\vec{Q}$, and the momentum of the excitation, $\vec{q}$, must be considered. The distinction is clear in crystalline systems where the two may differ by any reciprocal lattice vector $\vec{G}_{hkl} = \vec{Q} - \vec{q}$. 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 $\vec{q}$. These excitations then cause scattering about elastic diffraction maxima. The expressions S_{sw}^{+} and S_{sw}^{-} for the appropriate generalization of "umklapp" ferromagnetic spin wave scattering for a monatomic amorphous solid are derived in Ref. 3. The "umklapp" model was tested by convoluting $S_{sw}^{+} - S_{sw}^{-}$ 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 $\vec{Q}$ and frequency ω . 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 ω . The measured intensity is a convolution of $S^{+} - S^{-}$ with the

Fig. 4. A numerical calculation of $S_{\text{SW}} - S_{\text{SW}}^*$ 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 ω) 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 $\chi''(\vec{Q}, \omega)$ rather than the scattering function $S(\vec{Q}, \omega)$. The susceptibility $\chi''(\vec{Q}, \omega)$ is related to the scattering function by the Boltzmann factor: $\chi''(\vec{Q}, \omega) = [n(\omega) + 1]^{-1} S(\vec{Q}, \omega)$. Since for $\hbar\omega \ll kT$, $\chi''(\vec{Q}, \omega) \approx (\hbar\omega/kT) S(\vec{Q}, \omega)$ is odd in ω and shows a peak at finite ω even if $S(\vec{Q}, \omega)$ does not. A peak in $\chi''(\vec{Q}, \omega)$ 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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