

Iron–Germanium Multilayer Neutron Polarizing Monochromators*

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A new type of thermal neutron polarizing monochromator, consisting of alternate thin layers of iron and germanium, has been constructed and tested. Bragg reflection from such artificial 'crystals' in the fully magnetized state yields a highly polarized beam with high reflecting efficiency (~ 0.84) for the reflected spin state. These multilayer monochromators have the additional advantage that higher orders in the reflected beam are almost completely suppressed. Since d spacings are typically large (~ 100 Å), they produce a broader wavelength distribution than conventional single-crystal polarizing monochromators. Nevertheless, there are many applications where wavelength resolution is of secondary importance and the large gain in intensity (~ 40 -fold) over conventional polarizing crystals can be a considerable advantage. Multilayers can also be used to advantage in combination with good monochromating crystals such as pyrolytic graphite or beryllium to produce polarized beams of high intensity and good wavelength resolution.

1. Introduction

Although polarization analysis substantially enhances the sensitivity and selectivity of neutron scattering experiments (Moon, Riste & Koehler, 1969), it is rarely attempted because of the low reflecting efficiencies of the commonly available neutron polarizers and analyzers. These are most often ferromagnetic single crystals in which neutrons of one spin state are selectively Bragg reflected by virtue of interference between the nuclear and magnetic contributions to the scattering. What occurs can be briefly described as follows. In a fully magnetized crystal the reflected intensity is proportional to (Marshall & Lovesey, 1971)

$$|F^\pm|^2 = \left| \sum_j [b_j \pm p_j(\tau)] \exp(i\tau \cdot \mathbf{r}_j) \right|^2 \quad (1)$$

where τ identifies the reciprocal lattice vector of the reflection under consideration, \mathbf{r}_j locates the position of the j th atom within the unit cell and the summation extends over all atoms in the unit cell. The other quantities are the nuclear scattering amplitude b_j , which is independent of the wave vector transfer $\mathbf{\kappa} = \mathbf{k}_I - \mathbf{k}_F$, and the magnetic scattering amplitude p_j defined by the expression

$$p_j(\mathbf{\kappa}) = -(\gamma e^2 / 2mc^2) \mu_j f(\mathbf{\kappa}) \quad (2)$$

It is evident that p_j depends on $\mathbf{\kappa}$ through the magnetic form factor $f(\mathbf{\kappa})$, which is the Fourier transform of the normalized magnetization distribution.

The other terms in the equation are $\gamma e^2 / 2mc^2$, a constant with the value -0.2695×10^{-12} cm μ_B^{-1} , and μ_j which represents the magnetic moment of the j th atom

in units of the Bohr magneton μ_B . Good polarizers are crystals which are relatively easy to magnetize and in which b and p closely match for low-index Bragg reflections. As is apparent from (1), when b and p are equal only one of the two neutron spin states \pm is reflected and the resulting beam is fully polarized.

Not many crystals satisfy these stringent requirements. Of those which do, the most commonly employed are $\text{Co}_{0.92}\text{Fe}_{0.08}$ and Cu_2MnAl (Heusler alloy). Although producing highly polarized beams, both types of crystals contain materials with relatively large absorption and incoherent scattering cross sections. Furthermore, the sizes and mosaic distributions of the available crystals are far from optimum. As a result, the reflected intensities fall well below those routinely obtained from good nonpolarizing monochromators such as pyrolytic graphite or beryllium. Crystals of $(^{57}\text{Fe}_{0.86}^{56}\text{Fe}_{0.14})_{0.96}\text{Si}_{0.04}$ alloy perform considerably better (Koehler, 1975), but their high cost and limited availability put them beyond the reach of most laboratories.

As an alternative to magnetic crystals, we have recently begun to explore the use of Fe–Ge multilayer films as polarizing monochromators. These are made by depositing alternate layers of Fe and Ge on glass substrates, producing a periodic variation in the index of refraction (for neutrons) in the direction normal to the film planes. Such 'artificial crystals' are known to provide intensities comparable to the best single-crystal neutron monochromators currently available (Schoenborn, Caspar & Kammerer, 1974). Like other multilayer systems tested, Fe–Ge multilayers are highly reflecting monochromators. In addition, when fully magnetized, they produce highly polarized neutron beams.

In the monochromators investigated thus far, the individual layers are relatively thick, with d spacings typically of the order of 100 Å. As a consequence, the Bragg angles tend to be small and the wavelength res-

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olution correspondingly poor unless the beam is highly collimated. Nevertheless, there are many applications in which wavelength resolution is less important than high intensity. In these cases, the large intensity gain of Fe-Ge multilayer monochromators over conventional magnetic crystals (approximately 40-fold) can be a considerable advantage.

2. Diffraction from multilayer films

Although, in general, Bragg reflection from multilayers is similar to Bragg reflection from conventional crystals there are differences which have important effects on the performance of multilayers as monochromators. To see what these differences are, let us consider neutron reflection from the array of Fig. 1(a) containing alternate layers of atoms u and v , with v being a magnetic species. These layers, of thickness $d/2$, are assumed to be uniform in composition and to have well-defined boundaries. In this case, the structure factor is of the form (Saxena & Schoenborn, 1975):

$$F = \sum_{j=1}^N \left[\int_{(j-1)d}^{(j-1/2)d} g_1 \exp(ik_z z) dz + \int_{(j-1/2)d}^{jd} g_2 \exp(ik_z z) dz \right]. \quad (3)$$

Here N is the total number of bilayers on the substrate, κ_z is the component of κ normal to the film plane (*i.e.* in the z direction in Fig. 1) while $g_1 = \rho_u b_u$ and $g_2 = \rho_v [b_v + p_v (\hat{s}_n \cdot \mathbf{q})]$ where ρ indicates the number of atoms per unit volume, \hat{s}_n is a unit vector in the direction of the neutron spin and $\mathbf{q} = (\hat{S} \cdot \hat{\kappa})\hat{\kappa} - \hat{S}$ where \hat{S} and $\hat{\kappa}$ are unit vectors in the direction of the magnetization and wave vector transfer respectively (Marshall & Lovesey, 1971).

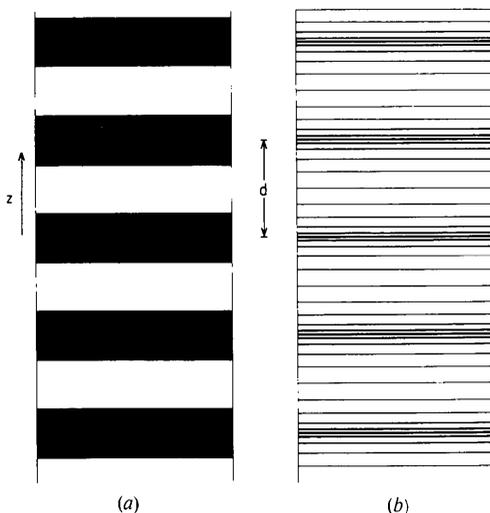


Fig. 1. Assumed distributions of the Fe and Ge layers in the direction normal to the plane of the film: (a) separate regions of Fe and Ge with distinct boundaries; (b) sinusoidal variation of the densities, with the two materials being 180° out of phase.

The reflected intensity is proportional to F^2 . From (3) we find

$$I \propto \left[\frac{2 \sin(\kappa_z d/4) \sin(\kappa_z N d/2)}{\kappa_z \sin(\kappa_z d/2)} \right]^2 \times [g_1^2 + g_2^2 + 2g_1 g_2 \cos(\kappa_z d/2)]. \quad (4)$$

Diffraction maxima will occur whenever

$$\kappa_z = 2m\pi/d \quad \text{where } m = 1, 3, 5, \dots,$$

i.e. when the Bragg condition is satisfied for odd values of m . At the Bragg position, the reflected intensity depends (in the extinction-free limit) on N, d, g_1 and g_2 through the relation

$$I \begin{cases} = (Nd/\pi m)^2 (g_1 - g_2)^2 & m = 1, 3, 5, \dots \\ = 0 & m = 2, 4, 6, \dots \end{cases} \quad (5)$$

Expressing g_1 and g_2 in terms of the appropriate number densities and scattering amplitudes we then obtain (for odd m)

$$I(\text{unmagnetized}) \propto (Nd/\pi m)^2 \times [(\rho_u b_u - \rho_v b_v) + \langle 1 - (\hat{\kappa} \cdot \hat{S}) \rangle (\rho_v p_v)^2] \quad (6a)$$

$$I^\pm(\text{magnetized}) \propto (Nd/\pi m)^2 [\rho_u b_u - \rho_v (b_v \pm p_v)]^2. \quad (6b)$$

The angular brackets in (6a) indicate an average over domain orientation in the magnetic layers. In (6b) the plus sign applies when the neutron spin is antiparallel to the magnetization (\hat{s}_n antiparallel to \hat{S}) and the minus sign to the parallel configuration. It is also assumed that κ is perpendicular to \hat{S} ; in other words, that the applied magnetic field is parallel to the film planes and perpendicular to the scattering plane.

During the deposition process (which takes place at room temperature) there is undoubtedly some diffusion of atoms between layers. Therefore the uniform composition and sharp boundaries assumed in the derivation of (4) are probably not very realistic representations of the actual density distributions in the films. Since we are dealing with thin layers, we might consider as an alternative and possibly more realistic hypothesis that the densities of the two components vary sinusoidally from layer to layer as shown in Fig. 1(b). In this case we would have

$$g'_1 = \frac{1}{2} \rho_u b_u (1 + \sin 2\pi z/d) = \frac{1}{2} g_1 (1 + \sin 2\pi z/d) \quad (7a)$$

$$g'_2 = \frac{1}{2} \rho_v [b_v + p_v (\hat{S}_n \cdot \mathbf{q})] (1 - \sin 2\pi z/d) = \frac{1}{2} g_2 (1 - \sin 2\pi z/d), \quad (7b)$$

which leads to an expression for the structure factor of the form

$$F = \sum_{j=1}^N \int_{(j-1)d}^{jd} (g'_1 + g'_2) \exp(ik_1 z) dz. \quad (8)$$

It then follows that the expression equivalent to (4) is

$$I \propto \sin(\kappa_z N d/2) \left\{ \frac{(g_1 + g_2)^2}{\kappa_z^2} + \frac{(2\pi/d)^2 (g_1 - g_2)^2}{[\kappa_z^2 + (2\pi/d)^2]^2} \right\}. \quad (9)$$

Equation (9) has a single diffraction maximum at $\kappa_z = 2\pi/d$. Only the first-order Bragg reflection is present and this with an intensity

$$I \propto (Nd/4)^2 (g_1 - g_2)^2, \quad (10)$$

a result similar to (5) but with the important difference that all higher orders are suppressed. Substituting for g_1 and g_2 in (10) will lead to expressions identical to (6a) and (6b) (with $m=1$) except for a slight change in the numerical coefficient which does not concern us here.

It is apparent from inspection of (6b) that only one of the two neutron spin states will be reflected if $q_u b_u$ matches either $q_v(b_v + p_v)$ or $q_v(b_v - p_v)$. Thus polarization is produced in multilayers not by matching the nuclear and magnetic scattering amplitudes but rather by matching the *indices of refraction* of adjacent layers for one of the two neutron spin states. Surprisingly, a number of combinations of magnetic and nonmagnetic materials have indices of refraction near enough to be potentially useful in multilayer polarizers. Our reasons for selecting Fe and Ge in preference to other possible combinations involved a number of factors which we will review in the following section. But before turning to these considerations, it ought to be noted that there is evidence of cross-diffusion between layers in all of the monochromators examined to date. Higher-order reflections are always noticeably suppressed and in some instances absent altogether. Therefore, for the most part (9) seems to give a better description of the actual behavior of multilayer monochromators than (4). This is of course a highly fortunate circumstance since it opens up the possibility of using multilayers as continuously tunable higher-order filters as well as monochromators.

3. Selection of components and preparation of monochromators

As we remarked earlier, a number of combinations of magnetic and nonmagnetic materials can be found which meet the matching condition discussed in the previous section. But aside from having indices of refraction which match extremely well, Fe and Ge were selected in preference to other possible combinations for the following reasons: (i) both have relatively low interdiffusion rates at room temperature and can be evaporated without particular difficulty, (ii) both have reasonably small absorption and incoherent scattering cross sections and rather large coherent scattering cross sections, (iii) Fe is easily saturated in small magnetic fields and (iv) Ge is resistant to corrosion and therefore can be used to protect the Fe layers from atmospheric exposure.

Preparation of the monochromators involved vacuum evaporation (Chopra, 1969) of Fe and Ge films on optically flat 1×6 inch glass substrates. The two materials were placed in tungsten boats and evaporated alternately by resistance heating. During the evapora-

tion process, layer thickness was monitored by means of a quartz crystal oscillator and controlled by a shutter which was opened and closed automatically at preset values. Since performance of the multilayers is degraded if the film thickness varies significantly from layer to layer, evaporation was done at the low rate of 2 \AA s^{-1} with one material held just below its melting point while the other was evaporated.

After preliminary tests of the method, two Fe-Ge multilayer plates were prepared. Both films were made with layers approximately 80 \AA thick, with Ge on the top surface to seal the underlying Fe layers from oxidation. One plate was covered with 32 bi-layers, the other with 28 giving a total film thickness of roughly 5000 \AA .

We have not noted any deterioration in the performance of the monochromators during a one-year period of testing. It should be emphasized, however, that the surfaces can be easily damaged if the plates are handled carelessly.

4. Experimental studies

(i) Quality of the reflected beam

To investigate the overall performance of Fe-Ge multilayers as monochromators, the 32-layer plate (in the unmagnetized condition) was first scanned in the θ - 2θ mode with a $\frac{1}{4}$ mm wide, unpolarized, monochromatic 4.2 \AA beam. The results, plotted on a logarithmic scale, are shown in Fig. 2. It is evident that there is a well defined first-order Bragg peak at $\theta = 0.72^\circ$. From this, we establish that $d = 167 \text{ \AA}$. Assuming uniform densities and sharp boundaries, the Fe and Ge layers can be considered to be nominally about 83 \AA thick. A broader peak centered at 0.3° is also

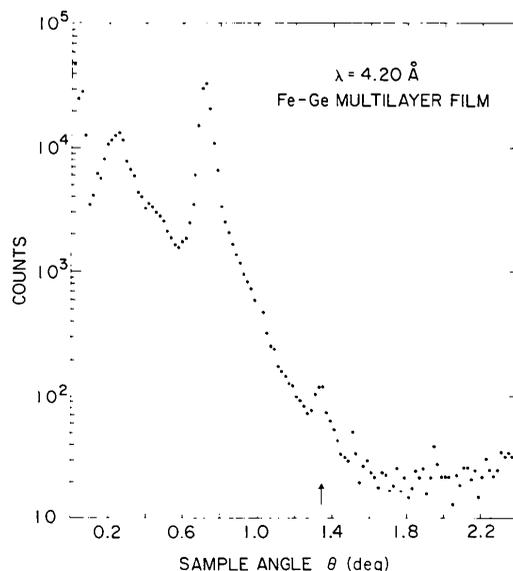


Fig. 2. The observed intensity of neutrons scattered from an unmagnetized Fe-Ge multilayer as measured in a θ - 2θ scan with an unpolarized monochromatic beam incident.

apparent in Fig. 2. This is due to mirror reflection of neutrons from the film and the glass substrate. No doubt a considerable part of the background underlying the Bragg reflection also comes from this source.

At the position noted by the arrow in Fig. 2 there is a small second-order peak. No third order could be detected. Most of the observed intensity in the second-order peak (which is lower by a factor of 500 than the first-order intensity) is probably due to inhomogeneities and aperiodicities in the Fe and Ge layers (Saxena & Schoenborn, 1975). We therefore infer that the atomic density distributions in this particular monochromator are close to the sinusoidal form of (7a) and (7b).

(ii) Reflectivity and wavelength resolution

A narrow, well collimated, broad-spectrum, cold neutron beam was used to measure the reflectivity and wavelength resolution of the Fe-Ge multilayer monochromator. This beam was directed on the monochromator surface at an angle of 0.80° and the wavelength distribution in the resulting Bragg reflected beam at $2\theta = 1.60^\circ$ was analyzed with a pyrolytic graphite crystal. Fig. 3 shows (for the unmagnetized monochromator) the wavelength dependence of the ratio of reflected to incident intensity. It is apparent from this figure that the reflectivity is 0.65 at the Bragg wavelength $\lambda_B = 4.66 \text{ \AA}$. Also, the full width at half maximum is 0.33 \AA indicating that the wavelength resolution $\Delta\lambda/\lambda_B$ is 0.07. It should be noted that corrections for the resolution of the analyzing crystal can be safely ignored here because of the comparatively high resolution of pyrolytic graphite in the wavelength range under investigation.

Repeating this measurement with a fully magnetized monochromator led to an apparent reflectivity of 0.42.

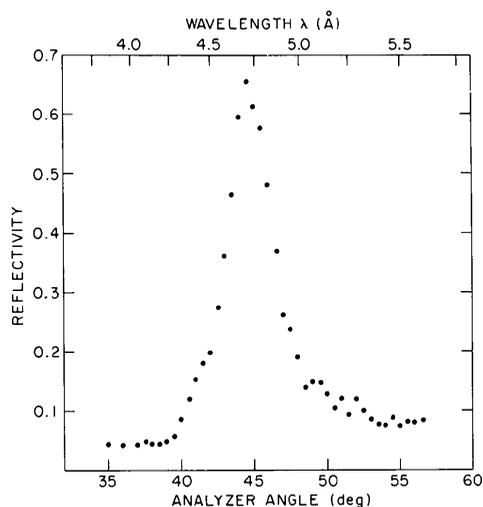


Fig. 3. The observed reflecting efficiency of the unmagnetized Fe-Ge multilayer measured with an unpolarized Be-filtered 'white' beam. The multilayer was set to reflect neutrons nominally of wavelength 4.7 \AA .

But keeping in mind that in the magnetized condition only one of the two neutron spin states is in effect reflected (as we will show in the next section), the actual reflectivity is very nearly $0.42/0.50 = 0.84$ – a considerable improvement over the unmagnetized case. According to (6), this would be expected as a consequence of the increase in the coherent scattering of Fe when the nuclear and magnetic parts of the scattering interfere constructively.

(iii) Polarizing efficiency

We used the 'shim ratio' method (Gurevich & Tarasov, 1968) to measure the polarizing efficiency of the Fe-Ge monochromators. This entails comparing the intensity of the beam doubly reflected from polarizer and analyzer crystals with the intensity which results when the same beam is depolarized by a thin shim of unmagnetized iron placed between the two crystals. The ratio of the observed intensities is related to the polarizing efficiencies of the crystals by an equation of the form

$$\frac{I(\text{without shim})}{I(\text{with shim})} = 1 \pm f_p f_a. \quad (11)$$

In this expression, the polarizing efficiency f is defined by the relation

$$f = \frac{I^+ - I^-}{I^+ + I^-} = \frac{R - 1}{R + 1}, \quad (12)$$

where I^+ and I^- represent the intensities of neutrons in the $+$ and $-$ spin states in the beam reflected by the polarizer and R is the so-called polarization ratio I^+/I^- . In (11), the plus sign is intended to apply when both polarizer and analyzer reflect neutrons of the same spin state and the minus sign when they reflect opposite spin states.

The measuring sequence employed was as follows: using a Heusler alloy crystal, we first produced a polarized, monochromatic 4.05 \AA beam. Then, with a second essentially identical crystal as analyzer, we used the shim ratio method to determine f_p , the polarizing efficiency of the monochromator, which was found to be 0.99. After this, the second crystal was replaced by the Fe-Ge multilayer plate which was mounted between the pole pieces of an electromagnet. Application of a magnetic field to the multilayers produced an immediate decrease in the Bragg reflected intensity. This we interpreted as indicating a 'crossed polarizer and analyzer' configuration which is expected because the Heusler alloy crystals are known to reflect neutrons in the $+$ spin state while Fe-Ge multilayers would be expected to reflect preferentially the $-$ spin state of the neutrons. Fields exceeding 100 Oe were found to extinguish completely the Bragg reflected intensity thus indicating magnetic saturation of the Fe layers.

We then proceeded to scan the multilayer (in the fully magnetized condition) and detector in a θ - 2θ mode both with and without a depolarizing shim be-

tween the (Heusler alloy) polarizer and the (Fe-Ge) analyzer. The results of these scans are plotted on the lower half of Fig. 4. Subtracting background from the two curves and taking their ratio yields the product $f_p f_a$ [see (11)]. Knowing f_p , we can then determine f_a which in this case represents the polarizing efficiency of our multilayer monochromator. This appears as the curve plotted at the top of the figure.

At the Bragg peak position $2\theta_B$, the polarizing efficiency is 0.98, only slightly less than the value predicted by substituting in (12) the polarization ratio $R = I^+ / I^-$ calculated from (6b) [or (10)]. It is also evident from the figure that neutrons reflected from the multilayer plates at angles less than the Bragg angle are substantially polarized. This is due to the fact that the magnetized Fe layers act in effect as neutron polarizing mirrors.

Similar measurements for incident wavelengths of 2.44 and 1.40 Å established that the polarization efficiency did not depend on the wavelength over the entire range $4.1 \geq \lambda \geq 1.4$ Å.

5. Discussion

From the foregoing, it is clear that Fe-Ge multilayers offer the advantages of high polarizing efficiencies and high reflectivities and produce beams relatively free of higher-order contamination. It is also clear that their major disadvantages stem from the large plane spacings inherent in multilayer films and the correspondingly small Bragg scattering angles. Thus multilayers have comparatively poor wavelength resolution and must, in addition, be awkwardly long to produce neutron beams of reasonable width. At 4 Å for example, a multilayer plate such as the one we have been investigating would have to be 70 cm long to reflect a beam 1 cm wide.

As we mentioned earlier, there are applications in which high polarization and high intensity are essential while wavelength resolution is not of itself particularly important. Among such applications are spin-echo spectroscopy (Mezei, 1972), studies of the circular polarization of capture γ -rays (Abrahams & Ratynski, 1969) and studies of ferromagnetic domain formation (Burgy, Hughes, Wallace, Heller & Woolf, 1950) to name but a few. All would presumably benefit from the use of Fe-Ge multilayers.

We can also see advantages to combining good monochromator crystals such as pyrolytic graphite with Fe-Ge multilayer polarizers for three-axis spectroscopy. Operated in series, pyrolytic graphite and Fe-Ge should provide good wavelength resolution, high polarization (when needed) and effective suppression of higher-order contamination – all over a continuous wavelength range and without prohibitive losses in intensity. In special cases (such as studies of magnetic spin dynamics or for the separation of isotopic from spin incoherent scattering) an Fe-Ge multilayer in combination with a good monochromating crystal

could be used as a polarizing monochromator and an Fe-Ge multilayer alone would be satisfactory as an analyzer. More generally, however, both monochromator and analyzer would have to be a combination of polarizing multilayer and crystal monochromators.

Another interesting application of multilayer monochromators is possible if their construction is modified in the following way. We choose the magnetic and nonmagnetic materials so that their *nuclear* indices of refraction are equal. In this case the term $q_u b_u - q_v b_v$ in (6a) will be identically zero. If the magnetization is perpendicular to the scattering vector, then $\hat{\mathbf{k}} \cdot \hat{\mathbf{S}}$ in (6a) is zero and $I \propto (q_r p_r)^2$ independent of the neutron spin direction. On the other hand, if the magnetization is rotated parallel to the scattering vector, then the term $1 - (\hat{\mathbf{k}} \cdot \hat{\mathbf{S}})^2$ in (6a) is zero and no reflection takes place. This would provide a convenient method of rapidly switching the incident (unpolarized) neutron beam on and off. Such a device might be particularly useful for correlation time-of-flight spectroscopy (Mook, Snodgrass & Bates, 1974).

At present, the most serious difficulty with all of these potential applications appears to be the length of the multilayer plates required. One obvious way to resolve this problem is to reduce the layer thickness. This would involve improving evaporating techniques. It is also conceivable that the length problem can be solved by evaporating the films on thin, nonmagnetic backings (such as stainless steel or titanium) and then mounting them in a parallel 'Soller' configuration to accept a wider beam. Both of these possibilities are now being investigated.

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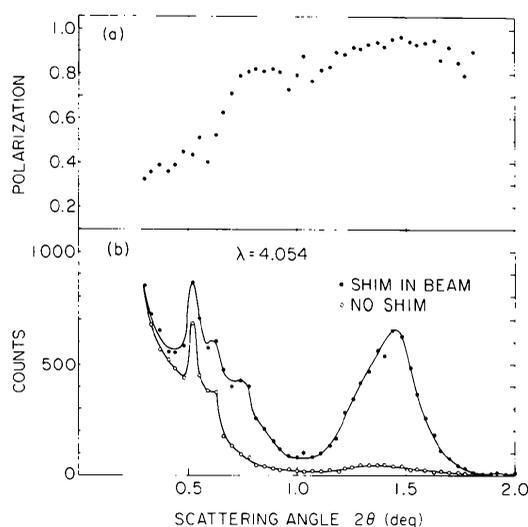


Fig. 4. θ - 2θ scan of the magnetized Fe-Ge multilayer with a polarized (no iron shim) and unpolarized (iron shim in beam) incident monochromatic neutron beam: (a) polarization dependence; (b) observed intensity.

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