Anomalous Yield Enhancement for Highly Collimated 180° Scattering of He^+ in Amorphous and Polycrystalline Materials

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A new and unusual yield enhancement [(20–30)% above Rutherford] has been observed in the energy spectra for He^+ backscattering to $180° \pm 0.25°$ in the energy range $0.2 \text{ MeV} \leq E \leq 2.5$ MeV for a variety of amorphous and polycrystalline targets. The depth and angular dependence of the yield enhancement has been established for a variety of materials and experimental conditions. Several possible explanations are discussed.

Experiments originally designed to study uniaxial double alignment in single crystals have resulted, because of their demanding collimation requirements,[1] in the observation of an unusual and previously unreported near-surface yield enhancement for narrow–acceptance-angle 180° scattering from amorphous and polycrystalline materials. Although the effect can also be demonstrated for single crystals continuously rotated to simulate a disordered lattice by averaging over all incidence angles, it is *not* an ion-channeling effect. Modification and reduction of the effect can, however, be achieved through partial or complete beam alignment with a good single-crystal channeling direction. Likewise the effect is not seen under small–acceptance-angle collimation at 90° on polycrystalline or disordered lattices, nor is it observed in wide–acceptance-angle geometries at 180°. These results lead to the conclusion that the effect is uniquely associated with small-impact-parameter, 180° scattering events.

Helium and proton beams from a 2.5-MV Van de Graaff accelerator were directed onto a variety of samples through a pair of collimators resulting in maximum incident half-angle beam divergence of 0.008°. Two in-line annular detectors (cooled to $-22°C$) could be positioned such that the one farthest from the target had an acceptance half-angle of $\theta_{1/2} = 0.2°$ and the closer one had a half-angle acceptance of $\theta_{1/2} = 4.5°$. Both detectors recorded scattering about the 180° direction. Two movable planar detectors were also used. One was at 90° (cooled to liquid-nitrogen temperature), and the other (at room temperature) could be scanned through a range of angles $0.11° \leq \pi - \theta \leq 0.66°$ about the 180° direction. Scattered-energy spectra could be collected simultaneously or separately from these detectors.

The nature of this anomalous yield enhancement is illustrated by Fig. 1 which contrasts the yields of 0.8-MeV He^+ ions backscattered to 180° from fine-grained polycrystalline Cu as measured in the wide- and narrow–acceptance-angle annular detectors. An energy-to-depth conversion was made for these spectra in the usual fashion with assumed stopping powers of 630 keV/μm for the inward path of the ions and 611 keV/μm for the outward path. These spectra have been normalized such that their yield distributions coincide at depths > 0.08 μm. The wide–acceptance-angle data show a typical thick-target Rutherford-backscattering yield distribution. The spectrum from the narrow–acceptance-angle detector contains a substantial and distinctive enhancement in yield observable as a broad peak in the near-surface region. The percentage difference in yield between the two spectra, over the first 700 Å, is 20%. The enhancement reaches a maximum within 150–200 Å of the surface and, within the limits of counting statistics, shows a regular decrease with depth, terminating at about 700 Å. Our experimental depth resolutions were ~ 100–200 Å.

It was initially speculated that crystalline texture effects might produce the above result in

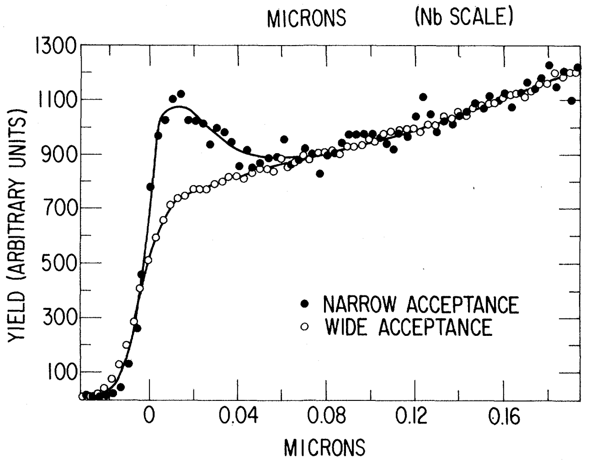

FIG. 1. Comparison of 180° scattering yields vs depth in annular detectors with narrow– and wide–acceptance-angle geometries for 800-keV He^+ on Cu.

polycrystalline materials through some collective, near-channeling compensation yields observed in the high-collimation geometry. To eliminate this possibility we performed similar experiments on amorphous germanium and also on the glassy metal alloy $Nb_{40}Ni_{60}$. The germanium crystal was rendered amorphous over the first 1800 Å by graded-energy phosphorous-ion implantation. Spectra were recorded in both wide– and narrow–acceptance-angle geometries for both ⟨100⟩-substrate channeling and a 5°-tilted–rotating-specimen mode. The wide–acceptance-angle measurements showed no yield enhancement. The narrow–acceptance-angle data showed pronounced surface peaks for the truly amorphous Ge layer in the aligned case and the rotating mode.

To further test the noncrystalline nature of the enhancement effect, measurements were made on a specimen of amorphous $Nb_{40}Ni_{60}$. These alloys, prepared by levitation melting and splat cooling, are known to retain their amorphous condition at and beyond second-nearest neighbors for temperatures below 600°C. It has been demonstrated that ion bombardment does not alter the amorphous condition, and in fact will turn the crystalline state amorphous after sufficient damage. The specimens used in this experiment were from a batch that was observed to be in the glassy state by transmission electron microscopy.[2] Since the alloy target is a binary system, the thick-target scattering spectra have two surface edges corresponding to scattering from Nb and Ni atoms. This is observed in Fig. 2 where the wide– and narrow–acceptance-angle spectra are superimposed for 0.8-MeV He^+ scattered to 180°. A weighted linear combination of stopping powers was used to establish the depth scale in Fig. 2 for scattering from Ni and Nb. It is seen in this figure that the wide-angle 180° spectrum shows a typical Rutherford yield whereas the narrow-angle spectrum again contains the near-surface enhancements for scattering from both the niobium and nickel. It must be concluded from these data, and those obtained from amorphous germanium, that the near-surface narrow-angle 180° scattering enhancement is not associated with crystallographic effects. An explanation must therefore be sought in terms of some other nuclear, electronic, or solid-state property.

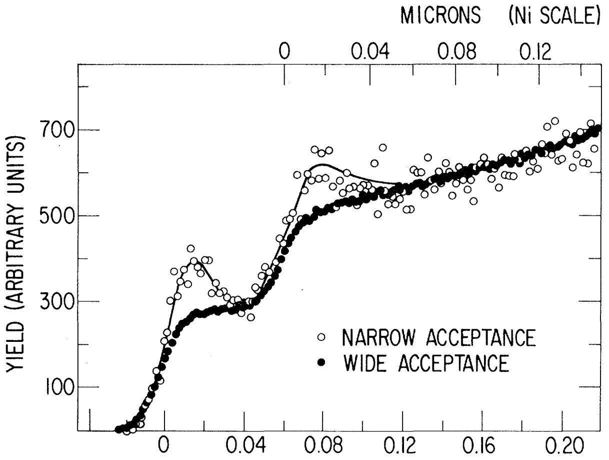

FIG. 2. Comparison of 180° scattering yields vs depth in annular detectors with narrow– and wide-acceptance-angle geometries for 800-keV He^+ on $Nb_{40}Ni_{60}$ (amorphous state).

In order to determine the angular extent of the enhancement we scanned the small (7-mm^2) 180° planar detector which had an angular resolution of 0.1° from 0.11° to 0.55° around the beam direction. At 0.11° from the 180° direction the energy spectrum enhancement was very similar to that seen in the annular detector. However, as the small planar detector was moved away from the incident-beam direction there was a reduction in the enhancement with complete disappearance occurring at about 0.5°. A comparison of the maximum yield enhancement at each angle compared to that at the 0.5° position is shown in Fig. 3 from

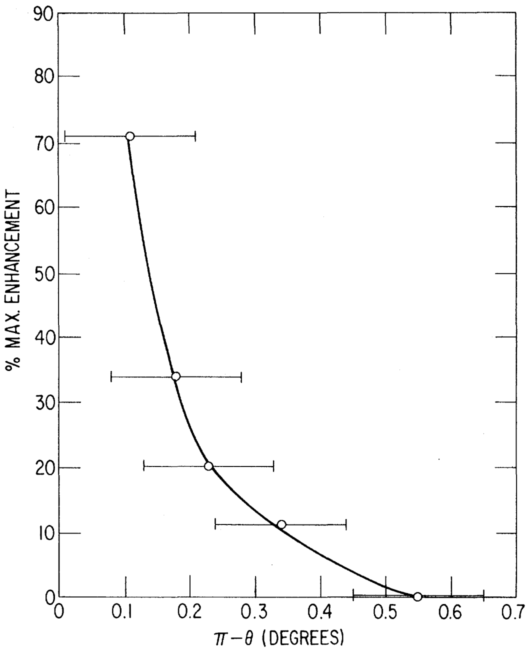

FIG. 3. Percent increase in the maximum yield enhancement of 1.0-MeV He^+ ions scattered from Pt near 180°. The reference yield spectrum was at 179.45°.

which a half-width of $\lesssim 0.2°$ can be estimated for the enhanced 180° scattering yield. It is also reassuring that the effect is observable when planar as well as annular solid-state detectors are used.

We have now established to our satisfaction the existence, and some systematic behavior, for this yield enhancement using several experimental arrangements, detectors, ions, and materials. However, we are not certain of its origin. It is difficult to believe that an elastic or inelastic nuclear resonance could account for this effect, since it has been observed for H, ^{4}He, and ^{3}He ions over a wide range of incident energies for many materials, and it is manifest in only the near-surface region.

One possible explanation[3] is that the details of some ion trajectories preceding the backscattering event will lead to a favorable set of atomic positions backwards along the beam direction. This tendancy may well be strong enough to produce an appreciably enhanced flux of backscattered ions in a small angular range near 180° since the favorable set of positions will persist for a brief period of time. The interplay between the conditions governing the reversibility of the trajectory, multiple scattering, and the ion transit times compared to thermal vibration times could explain the demise of the effect with depth and the observed angular dependence. This effect does not depend on any type of atomic displacement so that it may operate for single, polycrystalline, or amorphous targets. Although qualitatively plausible, this mechanism has not been verified by detailed calculations as yet.

Another possible explanation is that the positive ion, upon abrupt reversal of direction in nearly exact 180° scattering, interacts with its own polarization wake[3-6] set up in the electron gas of the solid during penetration, and that this interaction results in preferential alignment within $\simeq \pm 0.25°$ of the 180° direction. For the ions under consideration here with velocities $v_1 > v_0$ (v_0 = Bohr velocity $= 2.18\times 10^8$ cm/sec), electron density fluctuations which are set up are behind but stationary with respect to the moving ion. These electron-gas fluctuations typically extend < 100 Å, are established in times the order $\tau \sim \omega_p^{-1} \sim 10^{-16}$–$10^{-17}$ sec (ω_p = plasma frequency), and have relaxation times of several τ.[4-6] Thus, there is sufficient time to establish such wakes in the near-surface region, and the observed decrease in yield enhancement with depth could result from multiple scattering of the affected ions on their outward path. It is known, in fact, from molecular-cluster–breakup experiments that the wake field of a leading ion in a molecular cluster can exert an aligning influence on the trailing ion in transmission through 100-Å films with angular effects comparable to our acceptance angles.[6] The central question, however, is whether the aligning force exerted on backscattered ions by their own wake can produce the yield enhancements observed here. This process is obviously a very complicated problem but preliminary estimates suggest that the self-wake aligning force may be too small to account for the observed effect.[7] Ionic–charge-state effects on the stopping powers were considered a possibility. Ions backscattered to 180° near the polarization wake could have modified charge-state distributions and thus altered stopping powers, and this effect might redistribute stopping powers near the surface region over depths comparable to the mean free path for charge equilibration. However, measurements utilizing ^{4}He$^+$ and ^{4}He^{++} beams showed no significant differences in the yield enhancement or energy spectra. Additional work is clearly required to resolve these points. It is our hope that reporting these anomalous results will stimulate other experimental and theoretical investigations.

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[1] B. R. Appleton and L. C. Feldman, in *Atomic Collisions Phenomena in Solids*, edited by D. W. Palmer, M. W. Thompson, and P. D. Townsend (North-Holland,

Amsterdam, 1970), pp. 417–433.

[2] M. D. Rechtin, J. Vander Sande, and P. M. Baldo, Scr. Met. 12, 639 (1978).

[3] J. H. Barrett, private communication.

[4] J. Neufeld and R. H. Ritchie, Phys. Rev. 98, 1632 (1955), and Phys. Rev. 99, 1125 (1955).

[5] V. N. Neelavathi, R. H. Ritchie, and W. Brandt, Phys. Rev. Lett. 33, 302 (1974).

[6] D. S. Gemmell, J. Remillieux, J.-C. Poizat, M. J. Gaillard, R. E. Holland, and Z. Vager, Phys. Rev. Lett. 34, 1420 (1975).

[7] R. H. Ritchie, private communication.

Anisotropic Spin-Glass Behavior in Fe_2TiO_5

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It is shown that the insulating oxide Fe_2TiO_5 exhibits anisotropic (uniaxial) spin-glass behavior below 55 K. Extensive experimental results supporting this conclusion, including principal magnetic susceptibility, neutron scattering, ultrasonic, specific heat, and oriented single-crystal Mössbauer measurements, are described.

We report here on a comprehensive series of investigations of the structural and magnetic properties of the insulating oxide Fe_2TiO_5. Our results lead us to conclude that this undiluted compound containing only one type of magnetic ion exhibits *anisotropic* spin-glass behavior below 55 K. We believe this to be the first observation of such behavior and, since large crystals of Fe_2TiO_5 can be readily grown, we expect that this compound will be an ideal candidate for such studies as spin dynamics and critical behavior in spin-glasses. In particular, we note that there is the possibility of studying the angular dependence of spin-glass behavior.

Fe_2TiO_5 has been reported[1] to have an orthorhombic structure (*Cmcm*) with Fe^{3+} and Ti^{4+} ions filling (8*f*) and (4*c*) sites, respectively. Powder Mössbauer spectra[2] at 4 and 50 K exhibited hyperfine splitting, with rather broad linewidths. As we succeeded in growing single crystals of this compound, we were able to carry out a series of comprehensive studies on both single-crystal and powder specimens.

In Fig. 1 we present the results of low-field principal magnetic susceptibility measurements. These were obtained with use of a magnetic susceptometer in an alternating magnetic field of 30 Oe at 1100 Hz. Other measurements, carried out in fields of 20–100 Oe at 300–2000 Hz, yielded similar results. The temperature was varied at about 3 K/min. No hysteresis or relaxation ef-

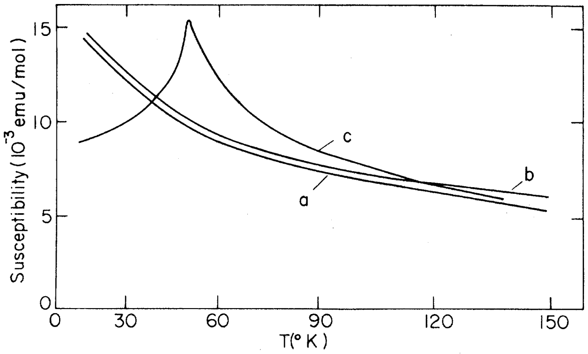

FIG. 1. Low-field principal magnetic susceptibilities parallel to *a*, *b*, and *c* crystallographic axes.