REDUCTION IN RADIATION DAMAGE DUE TO CHANNELING OF 51-MeV IODINE IONS IN GOLD*

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Anomalous energy-loss rates for energetic light[1-3] and heavy[4,5] ions, as well as orientation-dependent yields for atomic[6] and nuclear reactions,[7,8] have shown that crystal orientation effects are important at high energies as well as at low[9-11] energies. The basic feature of channeling is the steering of ions by a correlated sequence of small-angle collisions with the lattice atoms, such that the ions follow paths characterized by a reduced probability of small-impact-parameter events. This steering has been demonstrated by the shift in position of the transmission patterns of protons penetrating Si.[12] Since channeling reduces the rate of elastic energy loss,[9,11] it is expected that the damage rate to the crystal would be less when channeling occurs than when it is absent.[9,13] Although it has been shown that radiation damage leads to a suppression of channeling,[10,14] to date no observations have been reported on the effect of channeling on damage rates. This note describes preliminary observations of the effect of channeling of 51-MeV ^{127}I ions in gold on the damage rate, as determined by electron microscope observation of the damage structures. These observations have indicated a reduction in the observed damage rate by more than an order of magnitude when channeling occurs.

Single-crystal gold films of (001) orientation, 2200 Å thick, were prepared by evaporation and epitaxial growth upon cleavage surfaces of NaCl, using the procedure described by Bassett and Pashley.[15] The films were detached from the substrate and mounted over 1-mm apertures in 3-mm disks to give specimens suitable for direct study by transmission electron microscopy. A special procedure to be described elsewhere was employed in mounting the films which minimizes wrinkling and retains the planar, single-orientation character of the films established by the epitaxial growth. A group of eight specimens was mounted in a holder, oriented by rotation in the plane of the films, such that with beam incidence at 45° systematic misalignments of beam direction away from [011] were obtained in the different specimens. This array was irradiated in the Oak Ridge tandem accelerator with a beam of 51-MeV ^{127}I ions of $\simeq 10^8$ ions/cm^2 sec over an area 1.5 cm in diameter. Four MoO_3 smoke specimens in the extreme positions of the array were employed to measure the irradiation dose. Track counts on electron micrographs from these monitor specimens indicated a gradient of $\simeq 40\%$ in the dose across the array, and this information was employed to normalize the statistics of each specimen to the average dose of 4.26×10^{10} ions/cm^2.

Following irradiation, the gold specimens were removed from the holder and examined in the electron microscope. A number of areas from each specimen, selected at random, were photographed under as nearly identical conditions as possible. Figure 1 shows examples of the micrographs obtained. In accordance with previous work on single-crystal films of gold[16,17] irradiated with fission fragments, the effect of irradiation on the microstructure is to generate spot structures which range in apparent size from several hundred angstroms down to less than 50 Å. It is apparent from this set of micrographs that the spot density varies greatly as the ion-beam direction is systematically varied from the [011] direction. Figure 2 shows statistics obtained from these observations in which the spot density is plotted against the measured deviation of the ion beam from [011]. These statistics indicate that the damage rate differs by a factor of about 14 for aligned and unaligned beams. Further, the angular width (width for damage density one-half the maximum density) is about 1°.

This marked decrease in the damage rate over an appreciable angular range is taken as evidence that channeling is the dominant process in this orientation-dependent effect. Furthermore, the observed reduction in damage is believed to be a direct consequence of the reduced nuclear collisional losses due to channeling, as it is generally accepted that radiation damage in metals is due to the nuclear component of the energy loss. That is, although the electronic component is by far the dominant

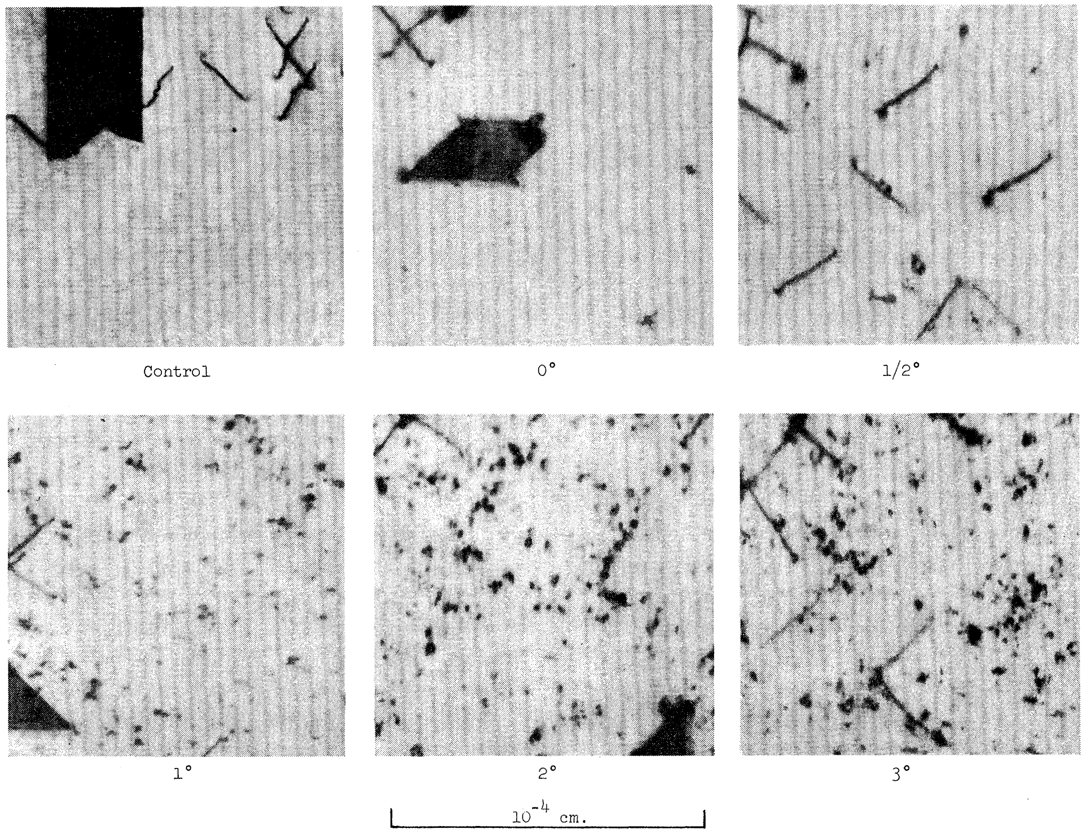

FIG. 1. Transmission electron micrographs of 2200-Å thick single-crystal gold films irradiated with 51-MeV ^{127}I ions. Captions under individual micrographs refer to the nominal misalignment of ion beam from [011]. Damage due to the irradiation appears as dark spots, variable in size and contrast, while large, dark, regular-shaped areas are microtwins, and dark lines are dislocations. Control micrograph is typical of unirradiated films. Magnification ~43 000×.

energy-loss mechanism (>95%) at these energies, it is the nuclear component which, although small (<5%), gives rise to the damage through the atomic recoils. This point is supported by earlier energy-loss measurements[4,5] in which channeling led to a reduction in the energy loss of less than a factor of two, whereas the present results show a damage reduction of more than an order of magnitude. From the above results it is concluded that the nuclear stopping component is much more sensitive to channeling than the electronic component. This result is in general agreement with the theory of Lindhard,[18] which predicts that the ratio of nuclear stopping to the electronic stopping is much smaller for a channeled beam than for a random beam. The damage reduction, however, does not vary with angle as rapidly as the nuclear stopping, which is predicted[18] to decrease as the fourth power of the channeling angle for angles smaller than the critical angle. The observed damage, on the

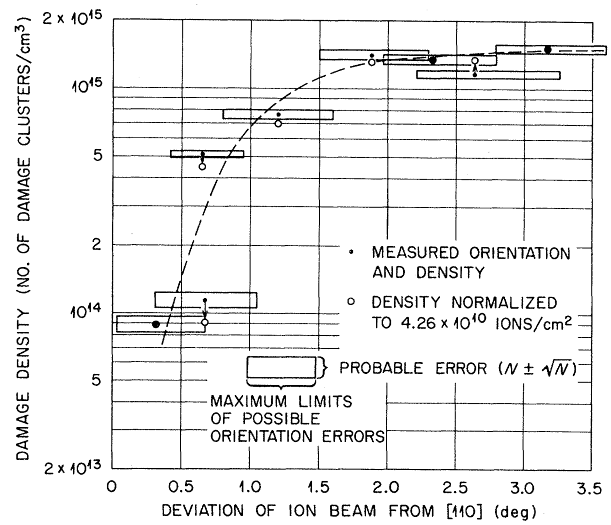

FIG. 2. Damage density versus misalignment angle of 51-MeV ^{127}I ion beam relative to the [011] direction for 2200-Å thick single-crystal gold films of (001) orientation. The maximum spot density of $\simeq 1.5\times 10^{14}$ spots/cm^3 corresponds to an average of $\frac{3}{4}$ of a spot per incident ion.

other hand, decreases approximately as the square of the angle. Although the reasons for this difference are not completely clear, it appears (as will be discussed later) that the damage "seen" by the electron microscope is associated only with the more energetic encounters between the projectile and the lattice atoms, so that a comparison of the damage with the total nuclear stopping is not completely appropriate.

A comparison of the present results with electron-microscope studies of fission-fragment damage in gold is of interest and suggests a possible more detailed interpretation. Merkle[16] concludes on the basis of charged-particle irradiation of gold that the spot damage observed in the electron microscope arises from those displacement cascades which are produced by primary knock-on events of energies greater than a threshold value of about 34-35 keV. From the present density data at maximum beam misalignment and by associating each spot with the displacement cascade of a single energetic recoil atom, it is calculated that each event corresponds to the scattering of the projectile through an angle of 1.5° or greater with an energy transfer to the recoiling lattice atom of 22 keV or greater. Here the scattering probability was taken to be that of a random solid of the same density, an assumption expected to be good for this case of maximum beam misalignment. The scattering potential used was the Molière approximation to the Thomas-Fermi potential, together with the impulse approximation of classical scattering theory. Although an energy threshold of 22 keV is somewhat lower than that of Merkle, this may be explained by the latter's use of an unscreened Coulomb potential which, if used with the present data, would give a threshold of 34 keV.

On the basis of the interpretation of the spot damage as the more or less direct observation of displacement cascades associated with nuclear encounters transferring energy in excess of about 22 keV, the orientation-dependent reduction in the damage rate represents the reduction in the frequency of high-energy collisions when channeling occurs. It is noted that a single deflection angle of 1.5°, corresponding to an energy transfer of 22 keV, is somewhat larger than the observed critical angle of 1°. This suggests that the projectile ion either becomes dechanneled while making a spot-producing collision or has already been dechanneled (by lattice imperfections such as thermal vibrations) prior to such an event. This conclusion is supported by noting that a scattering of 1.5° corresponds to an impact parameter of 0.037 Å, a distance considerably smaller than the rms transverse thermal displacement amplitude which is estimated to be 0.14 Å. It is expected that an ion moving nearly parallel to a principal crystal direction will be readily dechanneled if it has sufficient transverse kinetic energy to approach the atomic row within the rms displacement amplitude. In fact, this rms displacement may be regarded as a critical distance of approach that an ion may make to an atomic row and still remain unchanneled. Inserting this distance into the row potential given by Erginsoy[19] for Molière's approximation to the Thomas-Fermi potential gives a critical channeling angle of 1.2°, which agrees well with that observed here. On the other hand, Lindhard's[18] expression for the critical angle gives a value too large by a factor of about 2.

For angles slightly greater than the critical angle, it has also been predicted[18] that there should be an enhancement for processes involving close approach to the nucleus. The present work gives no indication of such enhancement. However, such enhancement is expected to be rather small and, therefore, could be masked by other effects.

The minimum damage observed is about a factor of 5 larger than would be expected from the minimum fraction of ions scattered into the random beam as a result of thermal vibrations and incidence at small impact parameters on the atom rows.[18] At least part of this difference may be due to the fact that the ion beam was slightly off (0.3°) of the [011] with the result that an absolute minimum was not observed.

Although the interpretation of the present experimental work is uncertain with respect to many details, it is clear that substantial changes in the damage rate occur which are in qualitative agreement with the expectations of channeling theory. Additional experimental work on different film thicknesses and orientations is in progress which should resolve some of the uncertainties in the interpretation of the present work. However, it is also apparent that a more detailed theoretical treatment, particularly with respect to the collision spectra, will be necessary before adequate interpretation of damage studies will be possible.

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EFFECT OF EXCHANGE SCATTERING ON NUCLEAR SPIN RELAXATION IN MAGNETIC MATERIALS*

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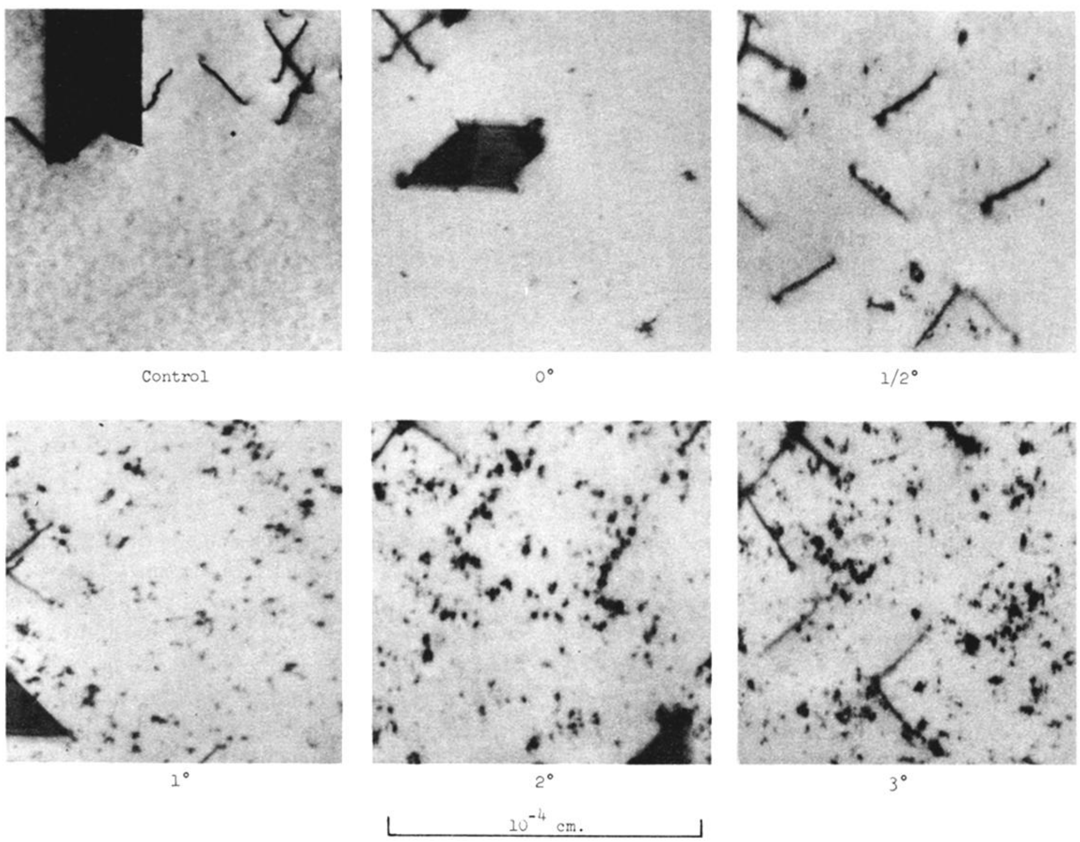
There has recently been a growing body of evidence that three-magnon processes play an important role in relaxing the nuclear magnetization in ordered magnetic insulators. In particular, (1) Fromhold and Narath[1] have measured the Cr^{53} T_1 in metamagnetic $CrCl_3$ and find temperature and field dependences characteristic of the three-magnon process but about an order of magnitude shorter than the theoretical prediction. (2) Welsh and Portis[2] have observed a T^5 dependence for the relaxation rate of Mn^{55} in antiferromagnetic $CsMnF_3$. This is the predicted[3] temperature dependence for the three-magnon process in antiferromagnets at temperatures high compared to the effective spin-wave gap temperature. (3) Kaplan and Jaccarino[4] have investigated T_1 of Mn^{55} in antiferromagnetic MnF_2 and find strong evidence for intrinsic spin-wave relaxation processes in the region $k_BT \gtrsim \Delta$ (Δ is energy gap in the spin-wave spectrum).

The necessity for considering the three-magnon process for nuclear relaxation relative to lower order spin-wave mechanisms can be summarized as follows: (1) The direct process, i.e., emission of single spin waves of arbitrarily well-defined energies, is not allowed by energy conservation because the nuclear resonance frequency is invariably much smaller than the minimum spin-wave energy (Δ). (2) The Raman process,[5] i.e., scattering of a thermal magnon with a simultaneous nuclear spin flip, has a vanishing matrix element in the situations of interest here where we have an isotropic hyperfine interaction, $A\vec{I}\cdot\vec{S}$, and the electronic spins and nuclear are quantized along the same axis. The selection rule against this process is a result of angular-momentum conservation. Thus the lowest order spin-wave process which can relax the



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