Limits on Dark Matter Using Ancient Mica

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The combination of the track etching method and atomic force microscopy allows us to search for weakly interacting massive particles (WIMPs) in our Galaxy. A survey of 80 720 μm^2 of 0.5 Gyr old muscovite mica found no evidence of WIMP-recoil tracks. This enables us to set limits on WIMPs which are about an order of magnitude weaker than the best spin-dependent WIMP limits. Unlike other detectors, however, the mica method is, at present, not background limited. We argue that a background may not appear until we have pushed our current limits down by several orders of magnitude.

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Much research is being devoted to the questions of the nature and detectability of the dark matter that comprises more than 90% of the mass of the Universe [1]. One of the most promising candidates is a weakly interacting massive particle (WIMP) which is being sought with instruments capable of detecting the ~keV/amu recoiling ions which would be produced in elastic collisions between WIMPs and nuclei [1]. The best limits on the mass and scattering cross section of WIMPs trapped in the Galactic halo result from the use of natural Ge, NaI, and CaF detectors [2]. These limits, however, fall short, by several orders of magnitude, of ruling out one of the favored WIMP candidates, the neutralino [3]. We show here that the natural mica crystals, with an integration time of $\sim 10^9$ yr, can record and store the tracks of recoil nuclei struck by WIMPs, and that these tracks can be measured with an atomic force microscope (AFM). Our approach is an extension of the etching method for studying ancient tracks in minerals [4,5]. With it, we report a new limit that is about an order of magnitude weaker than the best spin-dependent limits from NaI and CaF detectors, but show that we have the potential to push these limits down by several orders of magnitude.

As with the Ge, NaI, and CaF detectors, mica serves both as the target and as the detector. Muscovite mica is primarily composed of ^{1}H ($I = \frac{1}{2}$), ^{16}O ($I = 0$), ^{27}Al ($I = \frac{5}{2}$), ^{28}Si ($I = 0$), and ^{39}K ($I = \frac{3}{2}$). The range of one of these nuclei with a typical recoil energy of ~keV/amu is only a few hundred angstroms [6] and the etched depth is even smaller. Although such etched tracks cannot be studied with an optical microscope, we have shown that their dimensions can be accurately measured with an AFM [7]. As shown in Fig. 1, the technique is to cleave open a mica crystal, etch the freshly exposed surfaces, and use an AFM to scan and measure the tracks crossing the cleavage plane. For each area scanned (typically 40 μm × 40 μm) a 256 × 256 grid of heights is obtained and fitted line by line (to remove the effect of the piezo motion on the heights) with a fourth order polynomial using a robust fitting algorithm [8]. New, flattened heights are calculated from the difference between the old height and the fit. All contiguous pixels with depths below 20 Å are then grouped into clusters. Clusters with three or more pixels are then further analyzed. Clusters passing this 20 Å, 3 pixel cut are shown in Fig. 2 with the height of the deepest pixel in the cluster displayed alongside. The xy location of this pixel is taken to be the location of the recoiling ion and its depth is taken to be the depth of the etched pit.

An example of a scan of one surface of ancient mica etched for 1 h in room temperature 49% hydrofluoric acid

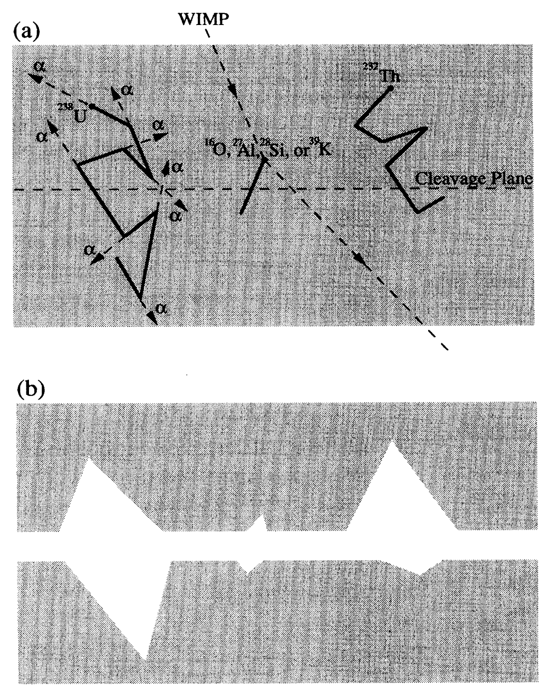

FIG. 1. An illustration of the etching technique. (a) If WIMPs exist they would cause the constituent atoms of muscovite mica, mainly ^{16}O, ^{27}Al, ^{28}Si, and ^{39}K, to recoil across a cleavage plane. (b) When both halves of the cleavage plane are etched matching pits will appear. The illustration also shows the development of α-recoil tracks and that these tracks will have longer summed depths than WIMP-recoil tracks.

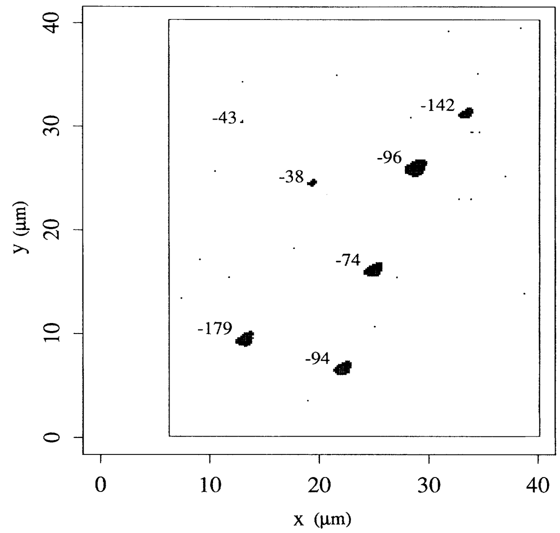

FIG. 2. A processed AFM image. The numbers are the heights in Å of the deepest pixels in clusters passing a 20 Å, 3 pixel cut.

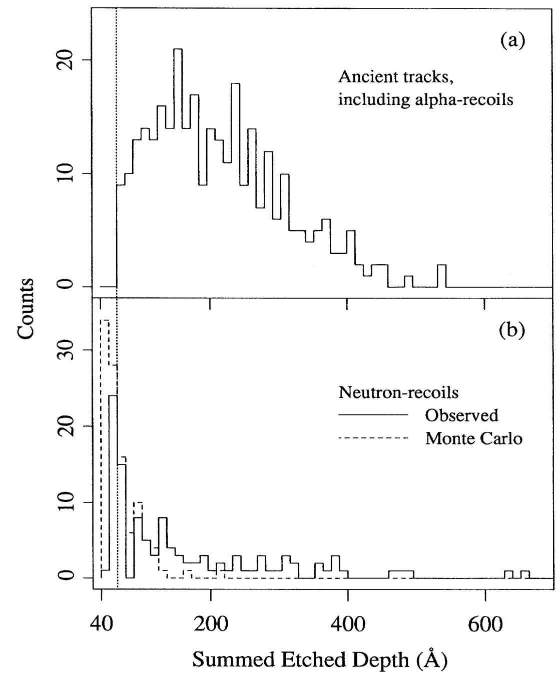

FIG. 3. (a) The summed etched depths of tracks recorded in a 80 720 μm^2 scan of 0.5 Gyr old muscovite mica. No events appear between our cutoff of 40 and 64 Å (shown with a dashed vertical line). (b) The solid line shows the summed depths of etched neutron-recoil tracks. The dashed line shows the results of a MC program of these data. In both the real and MC data a large fraction of the events appear in the 40–64 Å gap.

(RTHF) is shown in Fig. 2. Etched mica is completely insensitive to the β and γ rays which plague the Ge, NaI, and CaF experiments. There is, however, a background of tracks, shown in Fig. 2, due to the α decay of U and Th in the mica [9]. These etch pits are due to the recoiling daughter nuclei. The daughter nuclei are also radioactive and decay via a combination of β and α decays to Pb. The ^{238}U chain has eight α decays while the ^{232}Th chain has six, as shown in Fig. 1(a). These latent tracks are indistinguishable from WIMP-recoil tracks if only one surface, created by cleaving the mica open, is analyzed. To distinguish the etched pits created by α recoils from WIMP recoils we match etched pits across the cleavage plane. The summed depths of each pair of α-recoil etch pits should be larger because of their higher stopping power and longer range, as illustrated in Fig. 1(b).

For our search we selected, from $\sim$50 samples, a large, nearly perfect crystal of muscovite mica with low density of ancient fission tracks, low concentration of uranium, high fission track age, and low degree of thermal annealing—criteria previously applied to mica scanned for tracks of monopoles [5]. This mica was cleaved open and both freshly created surfaces were etched for 1 h in RTHF. Using an optical microscope attached to the AFM we were able to view matching fission tracks revealed by the etch. This allowed us to scan roughly complementary areas on both sides of the mica. α-recoil tracks on both sides then allowed us, in software, to match tracks to within a few tenths of a micron. We considered tracks to be matched if they passed a 20 Å, 3 pixel cut on both sides and fell within 0.75 μm of each other. After scanning 80 720 μm^2 we obtained the histogram of summed depths shown in Fig. 3(a). As expected there is a gap at small summed depths from our cutoff of 40 Å to the minimum observed summed depth of 64 Å.

To set limits on WIMPs we must be able to predict how many WIMP-recoil tracks will appear on this histogram. We begin with a model for the response of mica developed specifically for this purpose [10]. There are two parameters in this model, k_e and k_n, which characterize the effectiveness of the electronic and nuclear stopping at track formation. These must be calibrated for each mica, etching time, and cut (e.g., 20 Å, 3 pixel). We exposed our mica to 10, 100, and 400 keV ^{16}O and 20, 80, 180, and 400 keV ^{29}Si ions and then etched it for 1 h in RTHF. Using a laser interferometer [11] we measured a general etch rate of 160 ± 20 Å/h for our mica. We then placed a 20 Å, 3 pixel cut on the data and derived values of $k_e = (0.8 \pm 0.2) \times 10^{-5}$ (MeV cm^2/g)$^{-1}$ and $k_n = (1.25 \pm 0.25) \times 10^{-5}$ (MeV cm^2/g)$^{-1}$.

For a variety of reasons (charge state of the ion, sputtering, and other surface effects) one might question whether this model, based on ions which penetrate an already cleaved surface, can predict the etched depths of ion tracks created, and contained, within the mica. To test the model we exposed our mica to the fast neutrons inside a nuclear reactor. The recoil spectrum from these fast neutrons is very similar to the recoil spectra calculated for massive WIMPs. To eliminate slow neutron induced reactions the exposure canister was lined with cadmium. The flux of neutrons was measured during the mica exposure using nuclear activation foils. From these measurements we found a neutron energy spectrum very close to the Watt fission spectrum. Using a program [12] which models the transport of neutrons through matter we were able to obtain a better fit to the foil measurements. The integrated flux of fast neutrons was $(2.4 \pm 0.2) \times 10^{13}/\mathrm{cm}^2$. In an attempt to remove the α-recoil tracks (a background for this experiment) but retain the fission tracks (useful for matching) the mica was annealed at 475 °C for 1 h before the neutron exposure. Except for the annealing the neutron irradiated mica was etched and analyzed in exactly the same way as described earlier for the ancient mica. We found $(5.8 \pm 0.5) \times 10^5/\mathrm{cm}^2$ matched etch pits. After subtracting a measured background of residual α recoils, $(0.6 \pm 0.2) \times 10^5/\mathrm{cm}^2$, due to incomplete annealing, and of random matches, $(0.7 \pm 0.2) \times 10^5/\mathrm{cm}^2$, due to crowded fields, we were left with $(4.5 \pm 0.6) \times 10^5/\mathrm{cm}^2$ matched neutron-recoil tracks. Figure 3(b) shows the summed depth distribution. Roughly 25% of these events fall in the gap at low summed depths.

We then used our model to predict the surface density and distribution of matched neutron-recoil etched tracks as follows. From the known chemical composition of the major elements in muscovite mica, supplemented with measurements of the heavy elements ($Z \geq 13$) with an energy dispersive spectrometry system, we found the molar composition of elements heavy enough and abundant enough to produce etchable recoil tracks to be 58% O, 12% Al, 16% Si, and 5% K. For these elements we used the energy-dependent differential elastic neutron scattering cross section [13] along with the measured flux and energy spectrum of the neutrons to generate the surface density and energy spectra of recoils crossing a cleavage plane. The ranges of the various ions were computed with the TRIM92.3 program [6]. These ions were then used in a track etching Monte Carlo (MC) program based on our track etching model to determine the etched depths on both sides of the cleavage plane. With no free parameters this model predicts a density of $(4.4 \pm 0.4) \times 10^5/\mathrm{cm}^2$ matched tracks in excellent agreement ($2\% \pm 16\%$) with our experimental results. Figure 3(b) shows the predicted summed depth distribution. Unlike the real data, roughly 60% of the summed depths from the MC program fall in the 40–64 Å gap, indicating that, at least for massive WIMPs, we will overestimate our limits by a factor of ~ 2.4. As discussed in the track etching paper [10] this is probably due to the lack of a pixel cut in the model.

With our scan of ancient mica and our calibrated model in hand we now turn our attention to setting limits on WIMPs. Using the fission track dating method [4] we measured the age of our mica to be 0.5 ± 0.1 Gyr. To predict the number of WIMP recoils occurring in our scan of mica this old we need to know the density, velocity distribution, mass, and interaction cross section of WIMPs. Following [2] we assume a mass density of 0.3 GeV/cm^3 and a Maxwellian velocity distribution with $v_{\mathrm{rms}} = 261$ km/s cutoff at $v_c = 640$ km/s. Our motion through the Galaxy, $v_{\mathrm{sun}} = 220$ km/s, modifies this distribution as calculated by Spergel [14]. Using these parameters and a MC program identical to the one used for the neutron data we can calculate for any mass and cross section the number of matched tracks that will show up in Fig. 3(a). Before we present our limits we need to address one final question.

How do we know that WIMP-recoil tracks, created long ago, will survive until today? It is well known that latent tracks in mica can be erased by heating the mica, a process known as annealing. In connection with the search for monopoles in ancient mica, Price and Salamon [15] discovered a class of tracks in ancient mica, known as α-interaction tracks, caused by inelastic collisions of energetic α particles emitted in the decay chains of ^{238}U and ^{232}Th with ^{27}Al and ^{28}Si in the mica. They then calculated the ratio of the surface density of these tracks to the surface density of fission tracks to be 0.2. For our mica we find a ratio of 0.26 ± 0.08. We also find this ratio to decrease to 0.10 ± 0.02 for a 1 h anneal at 200 °C. 1 h anneals at lower temperatures have no effect. We therefore claim that our mica never experienced an annealing event greater in magnitude than that equivalent to a 1 h anneal at 200 °C. For this temperature and time we then ask what would have happened to WIMP-recoil tracks. When we anneal the neutron-recoil tracks at 200 °C for 1 h we find that the surface density of matched tracks decreases by a factor of 3.0 ± 0.5. To predict the annealing behavior of WIMP-recoil atoms we introduce a new parameter into our etching model, P_{ann}, which is the probability that an etching defect will be annealed after it is created by an ion. P_{ann} will depend on temperature and time of annealing. Using the annealing data for the neutron-irradiated samples as input we can fix this at $P_{\mathrm{ann}} = 0.35$ for a 1 h anneal at 200 °C. To obtain the limits shown in Fig. 4 we annealed the recoil tracks generated by our WIMP MC simulation and calculated how many tracks would appear with summed depths in the gap shown in Fig. 3(a). A from factor correction term [16] was included in this calculation. From this 90% confidence level limit curves can be calculated for each element. These curves are shown in Fig. 4. They are generous by a factor of ~ 2.4 because of our model and conservative by a factor of ~ 3 because of the annealing. These limits are, at present, about an order of magnitude weaker than the best spin-dependent limits from NaI and CaF detectors at high WIMP masses [2] but represent the first limits on WIMPs using ^{39}K. Like ^{19}F, ^{23}Na, and ^{127}I the spin of ^{39}K is mainly associated with an odd proton which leads to a large spin-dependent cross section [17].

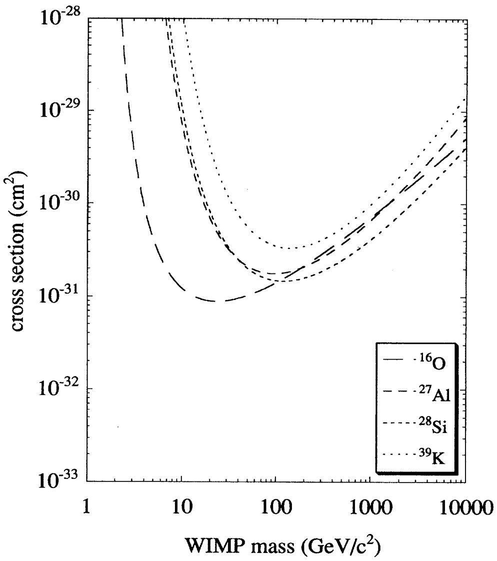

FIG. 4. Exclusion curves for each of the main constituent nuclei of mica. For a given mass, WIMPs with cross sections above these curves are ruled out at the 90% confidence level.

These limits are about an order of magnitude weaker than the best spin-independent limits from Ge detectors which are useful for ruling out heavy Dirac neutrinos.

With a variety of techniques, however, we expect to greatly improve our limits. First, we will simply analyze more mica. Second, so as not to run into an α-recoil background, we will need to etch the mica longer to widen the gap at low summed depths. Third, by selecting mica only from deep mines (a few hundred m) we can avoid muon-recoil tracks. For a thin mica deposit surrounded by rock containing 1 ppm uranium, typical of the Earth's crust, we would expect to see fast-neutron-recoil tracks due either to spontaneous fission of ^{238}U or to (α, n) reactions with light nuclei, somewhat below our current limits. However, for thick, self-shielded mica (dimensions >5 m are typical of pegmatitic mica) typically having <0.1 parts per 10^9 U, we expect the background of neutron recoils to be several orders of magnitude below our present limit.

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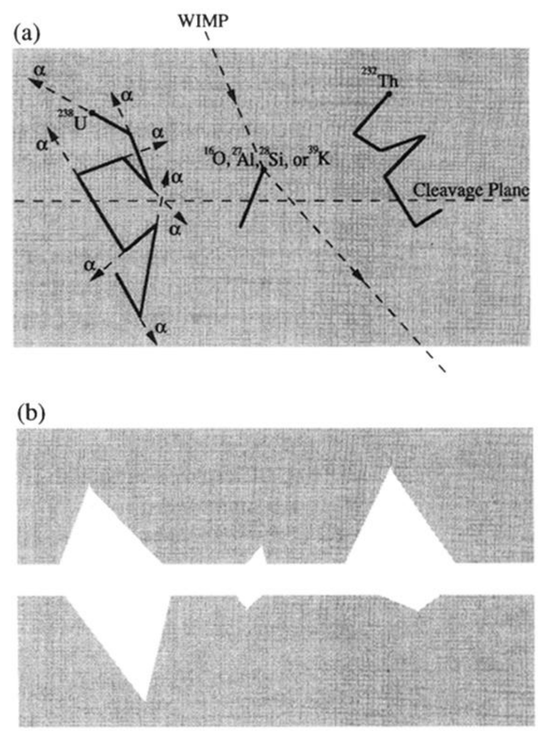

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