Can track-etch detector CR-39 record low-velocity GUT magnetic monopoles?

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A set of new data on the response of the CR-39 detector to low-energy heavy ions, collected with a recently developed technique based on atomic force microscopy, was used to evaluate the effectiveness of nuclear stopping in producing etchable tracks relative to electronic stopping. The results for two types of CR-39 are found to be significantly different from those obtained previously with other methods. I discuss the implications of the use of CR-39 for detecting particles with low ionization rates such as supermassive magnetic monopoles in cosmic rays as cosmological relics. [S0556-2821(98)06005-6]

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Among solid-state nuclear track detectors, CR-39, a polymer of allyl diglycol carbonate with composition $H_{18}C_{12}O_7$, is probably most frequently used. The principle of solid-state nuclear track detectors has been summarized in Ref. [1]. The wide spectrum of applications includes nuclear physics, cosmic ray astrophysics, and radiation dosimetry [2]. At high energies (velocity/c $\equiv \beta > 10^{-2}$), CR-39 is sensitive to a certain portion of the total ionization rate such as restricted energy loss caused by electronic stopping, as demonstrated in many relativistic heavy-ion experiments [3]. To place an upper limit on the flux of highly ionizing particles, such as magnetic monopoles, in galactic cosmic rays, Price [4] discussed the detectability of CR-39 in the low-energy regime ($\beta \simeq 10^{-4}$–10^{-2}) where nuclear stopping becomes important in comparison with electronic stopping. Using a scanning electron microscope (SEM) to measure the depths of etched tracks due to 200–400-keV ^{9}Be and 150–300-keV ^{28}Si ions, Snowden-Ifft and Price [5] found that for a particular type of CR-39 (made by American Acrylics) nuclear stopping appeared to be only $\simeq$20% as effective as electronic stopping in track formation. The threshold in total effective energy loss responsible for low-energy ion tracks is then extrapolated to be $(dE/dx)_{\rm th} \simeq 1$ GeV g^{-1} cm^2, implying that CR-39 will not record a low-velocity monopole. However, a set of significantly different data was recently reported by a group from Bologna [6] for another type of CR-39 (made by the Intercast Europe Co. of Parma, Italy) using the same technique with the same ions. Their work raises the need for further study of the response of CR-39 to low-energy ions, preferably with different methods, as it is important to determine the effectiveness of nuclear stopping in producing etchable tracks. These studies also have profound impacts on ongoing experiments of large scale designed to search for supermassive magnetic monopoles in galactic cosmic rays [7,8].

Hancox and I recently developed a novel technique for etched track measurements on short-distance scales using an atomic force microscope (AFM) in the region $dE/dx \sim 0.1$–4 GeV g^{-1} cm^2. This technique can be used to explore a new velocity regime in which nuclear stopping is comparable to or dominant over electronic stopping. The methodology, together with some data including depth distributions, was reported in a recent paper [9]. In this paper, I focus on the evaluation of the contribution from nuclear stopping to the detector signal based on the set of published data and an additional set of new data. This information should then put us in a position to infer more realistically whether a low-velocity magnetic monopole could be detectable in the CR-39 plastic.

We used TASTRAK CR-39 (DOS) manufactured by Track Analysis System Ltd. in Bristol, England and CR-39 (DOP) manufactured by American Acrylics in Wilmingtom, Delaware. Heavy ions studied so far include ^{2}D, ^{20}Ne, and ^{29}Si at kinetic energies ranging from 5 to 400 keV. The present analysis uses the set of data from ^{29}Si ions with energies 50, 100, 200, and 400 keV. Figure 1 shows the energy loss of ^{29}Si ions in CR-39, calculated using the 1992 version of the TRIM code [10], as a function of initial energy. The energy loss is decomposed into two components: one due to electronic stopping and one due to nuclear stopping. These particular four energies were chosen because at the

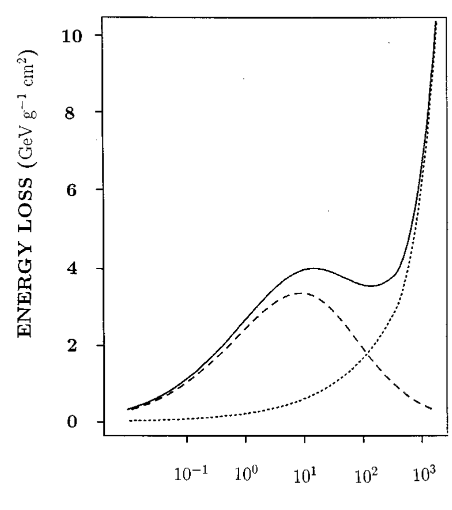

FIG. 1. The total-energy loss (solid line) as a function of kinetic energy for ^{29}Si ions in the CR-39 plastic, calculated using the TRIM code. The electronic stopping (dotted line) dominates at high energies and decreases with decreasing energy at low energies, while nuclear stopping (dash line) becomes dominant when the kinetic energy is less than 100 keV.

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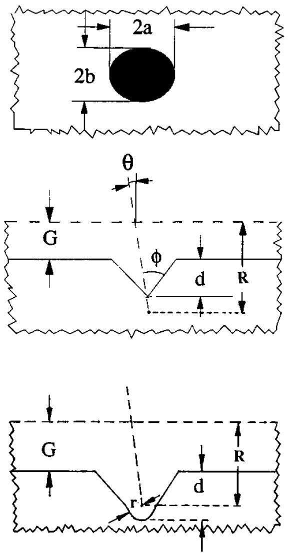

FIG. 2. Geometry of an etched nuclear track in a plate of a CR-39 detector at an incident angle θ with respect to the normal. The upper panel is the top view. The middle panel shows an etch cone when $G+d<R$. The lower panel shows an etch cone when $G+d>R$. One measures the depth d, the minor-axis b, or the half-opening angle ϕ to determine the detector response.

low-energy end energy loss is dominated by nuclear stopping and at the high-energy end energy loss is dominated by electronic stopping, and the sum is $\sim$3–4 GeV g^{-1} cm^2 with a minimum at $\sim$120 keV if the nuclear and electronic stopping have the same efficiency in producing etchable radiation damage.

The study of detector response requires an accurate measurement of the geometry of etched tracks. As shown in Fig. 2, an etch cone is characterized by its depth d, the minor-axis of an elliptical fit to its mouth at the surface b, and its half-opening angle ϕ. The thickness of material removed from one side during etch is G. If etching occurs beyond the end of range R, a bulb with radius r is formed at the bottom portion of the etch cone, as also shown in Fig. 2. The detector signal s, defined as the ratio of track etch rate to general etch rate, is a sensitive function of the average ionization rate over the sampling distance L_s.

The commonly used method of inferring the detector signal from track geometry utilizes measurement of b, with standard deviation σ_b by $s=(\cos\theta)^{-1}[(G^2+b^2)/(G^2-b^2)]$, with standard deviation $\sigma_s/s=[(s^2-1)/s](\sigma_b/b)$, and in its sampling distance $L_s=(1+s\cos\theta)^{-1}G$; or measurement of d with standard deviation σ_d and r with standard deviation σ_r by

$$s=(\cos\theta)^{-1}[(d+G-r)/(G-r)], \tag{1}$$

with standard deviation

$$\frac{\sigma_s}{s}=\frac{s-1}{s}\sqrt{\left(\frac{\sigma_d}{d}\right)^2+(s-1)^2\left(\frac{\sigma_r}{r}\right)^2}, \tag{2}$$

and in its sampling distance,

$$L_s=s(G-r). \tag{3}$$

In Eqs. (1)–(3), r and $\sigma_r \Rightarrow 0$ when etching does not occur beyond the end of range, i.e., $G+d \leqslant R$. If the half-opening angle ϕ is measured with standard deviation σ_ϕ, then s can be determined by

$$s=1/\sin\phi, \tag{4}$$

with standard deviation

$$\sigma_s/s=[\sqrt{s^2-1}\ \sin^{-1}(1/s)](\sigma_\phi/\phi). \tag{5}$$

The dispersion in the detector signal for a given energy, apart from measurement errors, represents the straggling in the energy loss. In this experiment, various efforts were made to ensure that an AFM scan yields adequate resolution and accuracy for etch cone measurement. The cone depth distribution, measured with an AFM, for each energy can be fitted by a Gaussian (see Figs. 12 and 13 in Ref. [9] for depth distributions of 50-, 100-, and 200-keV ^{29}Si ions in the two CR-39 samples). For the TASTRAK CR-39, the mean depths of etched cones due to 50-, 100-, 200-, and 400-keV ^{29}Si were measured to be $\langle d\rangle=30.7\pm1.1$, 22.6 ± 1.0, 39.4 ± 0.9, and 43.0 ± 0.7 nm with standard deviations $\sigma_d=6.6\pm1.1$, 5.6 ± 1.0, 5.4 ± 0.9, and 4.8 ± 0.7 nm. For the Acrylics CR-39, $\langle d\rangle=29.4\pm1.2$, 21.8 ± 1.1, 25.2 ± 0.9, and 28.6 ± 0.8 nm, and $\sigma_d=10.1\pm0.8$, 7.6 ± 0.7, 5.9 ± 0.6, and 4.2 ± 0.5 nm. A laser interferometer was employed to measure G at very short time scales [11].

To evaluate the contribution from nuclear stopping in track formation for CR-39, Snowden-Ifft and Price [5] adopted the following procedure. Assuming that the responsible energy loss for etchable tracks is $(dE/dx)_{\rm tot}=(dE/dx)_{\rm elec}+f(dE/dx)_{\rm nucl}$, they fitted measured s as a function of $(dE/dx)_{\rm tot}$ by a second-order polynomial where f was allowed to vary. The procedure yielded $f=0.2\pm0.1$. The Bologna group [6] used a similar procedure, but obtained $f\simeq1$. The significant difference between their results calls for the need of alternative approaches. The above procedure requires an accurate determination of s via d, b, r, and G. Systematic uncertainties involved in the measurement such as instrument calibrations at such a short scale are not easily estimated and corrected for.

To minimize the effect of unknown systematic errors in a determination of the effectiveness of nuclear stopping in producing etchable tracks, I developed a different approach. The procedure exploits one useful feature in Fig. 3, where the total effective energy loss averaged over L_s, $\langle dE/dx\rangle_{L_s}$, is plotted as a function of energy for a set of hypothetical values of f. One sees that $\langle dE/dx\rangle_{L_s}$ is a monotonic increasing function of energy for $f=0$ up to $\sim$0.7. As f increases further, a minimum develops in the dependence which occurs at $\simeq$100 keV when $f\simeq0.8$, and shifts to $\simeq$200 keV when $f\geqslant1$. Because the detector signal is a function of $(dE/dx)_{\rm tot}$, the trend in Fig. 3 can be compared directly with the experimentally determined mean value of the detector signal induced by ^{29}Si ions at 50, 100, 200, and 400 keV. For CR-39 samples made by both Track Analysis and American Acrylics, the monotonic increasing of the mean signal with energy is not observed, leading to a solid constraint $f\geqslant0.7$. The minimum signal is found to occur at 100 keV for both cases. For CR-39 made by Track Analysis, the maximum occurs at 400 keV, and the mean signal for 50-keV Si ions is smaller

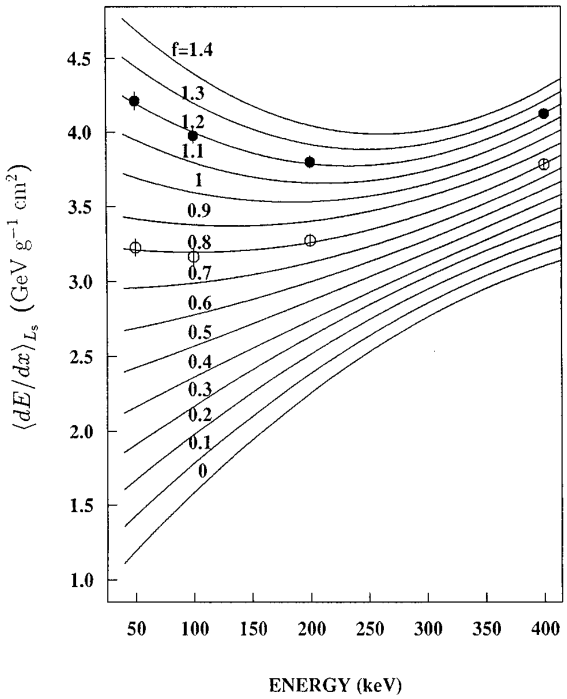

FIG. 3. The effective energy loss of ^{29}Si ions averaged over the sampling distance in the CR-39 plastic as a function of kinetic energy assuming the contribution from nuclear stopping is from 0% to 140% as effective as electronic stopping. Data are shown as open circles for the TASTRAK CR-39, and as solid circles for the Acrylics CR-39.

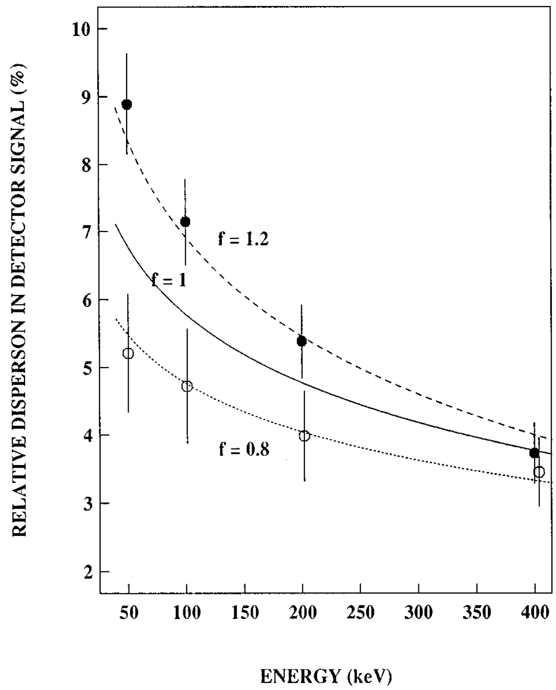

FIG. 4. The relative disperson in the measured signal of ^{29}Si ions at 50, 100, 200, and 400 keV in two types of CR-39 samples (open circles for the Track Analysis sample and solid circles for the American Acrylics sample). As a comparison, it also shows the predicted disperson in the total effective energy loss if nuclear stopping is 80% (dotted line), 100% (solid line), or 120% (dash line) effective in producing etchable tracks in the detector relative to electronic stopping.

than that for 200 keV. For CR-39 made by American Acrylics, the mean signal for 50-keV ^{29}Si is larger than that for 200 keV, and even slightly larger than that for 400 keV. To match the trend in the data, one can place a constraint $f \simeq 0.8$ for the TASTRAK CR-39 and $f \simeq 1.2$ for the Acrylics CR-39.

The energy-loss disperson can provide an additional stringent constraint on f. It is well known that nuclear stopping is associated with a larger fluctuation than electronic stopping. The measurement errors (~ 1–2 nm) are much smaller than the fluctuation induced by dE/dx disperson (which leads to range straggling). Therefore, the dispersion in the detector response represents the disperson in the effective energy loss that is responsible for etchable tracks. Consequently, the contribution of nuclear stopping to the detector signal can be evaluated based on the measured fluctuations. Figure 4 presents the relative disperson σ_s/s, together with the predicted relative fluctuation due to the energy-loss disperson in $(dE/dx)_{\rm tot}$ for hypothetical values of f up to a scaling factor. If f were unity, the solid line would describe both sets of data. The departure from the $f=1$ curve at lower energies indicates that nuclear stopping contributes less (more) than electronic stopping to detector response for the TASTRAK (Acrylics) CR-39. If we allow f to vary, the data require $f \simeq 0.8$ for the TASTRAK CR-39 and $f \simeq 1.2$ for the Acrylics CR-39, as shown in Fig. 4. These results are remarkably consistent with the ones derived from Fig. 3.

It is obvious from Eq. (4) that if one can measure ϕ at various points along the particle path, one is able to obtain a series of instantaneous values of the detector response. This information allows one to establish a relation between detector response and dE/dx using ions with only a single initial energy. This turns out to be possible with the excellent nm resolution of AFM for the TASTRAK CR-39, where the etched tracks can be identified against background on an event-by-event basis (for the Acrylics CR-39, the etched tracks could only be distinguished statistically from background but not on an event-by-event basis; see Figs. 11–13 in Ref. [9] for details). The fine structure of the walls of the etch cones was studied in detail at different energies for the TASTRAK CR-39. Examples from 200-keV ^{29}Si ions were shown in Fig. 7 of Ref. [9]. A remarkable phenomenon observed was that these track walls were found to be better described by a curve than a straight line, indicating that as an ion goes into the plastic, the signal becomes smaller and smaller along its path. This is understandable because dE/dx decreases gradually with energy near the end of a range. It was also observed that the disperson in the diameter of the track walls increases with depth, indicating that more and more nuclear stopping contributes to track formation near the end of a range. A detailed Monte Carlo simulation shows quantitatively that an etching model with a value of $f \simeq 0.8$ reproduces the observed wall structures as well as measured dispersons for the TASTRAK CR-39.

Three types of CR-39 (TASTRAK in the present work, Acrylics in Ref. [5] and in the present work, and Intercast in Ref. [6]) have been studied so far using two techniques (SEM in Refs. [5] and [6], and AFM in the present work). With the same technique (SEM), f for the Acrylics CR-39 [5] was found to be substantially different from that for the Intercast CR-39 [6] ($f=0.2\pm0.1$ vs $f\simeq1$), indicating possible intrinsic differences in the track-etch characteristics for different types of CR-39. This calls for the necessity of careful calibrations for each type of CR-39 when it is used in low-energy experiments. However, for the same type of CR-39 (Acrylics), the present result differs from that of Snowden-Ifft and Price [5] ($f \simeq 1.2$ vs $f = 0.2 \pm 0.1$). This implies some possible instrumental biases associated with techniques on which I comment below.

Because the ranges of low-energy ions are so short (R approximately equal to a few hundred nm) and their track dimensions are so small (d approximately equal to a few tens nm) one must use a microscope with nm spatial resolution to measure the detailed geometry of etched tracks on CR-39 surfaces. With the SEM technique, the surface of the sample to be viewed must first be coated with a thin conductive film in order to prevent the buildup of static charge. SEM observations can then be made either of the coated surface of the etched sample or of a coated replica of the etched surface that is cast from the original sample surface. In the first case, charging by the electron beam around the mouth of etch cones deflects the beam enough so that the measurements of depth are unreliable, as Snowden-Ifft observed [12]. In the second case, the influence of the sputtering process on the structure of the original surface to be viewed is likely. For instance, Snowden-Ifft [12] noted that the sputtering process appears to distort the wall angles of etch cones, and hence a good measurement of wall angles is impossible.

With the AFM technique, all these concerns become invalid, since AFM directly views the etched surface with ion tracks as it does for all other nonconducting materials. Moreover, the AFM can provide more measurements of the detailed characteristics of etched tracks than the SEM, even though both the SEM and AFM are equally good for observation of the existence of etched tracks. However, it should be pointed out that possible effects of AFM tips on the measurement need to be considered, especially when the tip is blunt and the cone angle is small. In our AFM measurement, various efforts were made, including the use of special AFM tips to ensure that the tip was able to reach the bottom portion of etch cones.

The effectiveness of nuclear stopping in comparison with electronic stopping is essential for a number of experiments that use CR-39 as primary detectors. This information sets the detector threshold. One important example is the search for magnetic monopoles in galactic cosmic rays. Supermassive magnetic monopoles have been predicted by grand unification theories (GUT's), and it has been suggested that they played an important role in the evolution of the early universe. GUT monopoles are expected to have $\beta \simeq 10^{-3}$ if they are either gravitationally trapped within our galaxy or passing through it, or $\beta \simeq 4 \times 10^{-5}$–$1.5 \times 10^{-4}$ if they are gravitationally trapped in semistable solar orbits. An upper limit on the flux of GUT monopoles in galactic cosmic rays using CR-39 was set by Price [4] under the assumptions that the response of CR-39 to a given value of dE/dx is the same at low energies as at high energies, and that nuclear stopping has the same effectiveness as electronic stopping. Among experiments underway to search for monopoles in galactic cosmic rays, experiments in Japan [8] and in the Monopole, Astrophysics, and Cosmic Ray Observatory (MACRO) at the Gran Sasso Underground Laboratory in Italy [7] have deployed arrays of CR-39 plastic detectors on the order of several thousand square meters. In the Japanese experiment, the CR-39 was the only detector used to record monopole candidates. In MACRO, the CR-39 was used to confirm the existence of a monopole when the scintillation counters and streamer tubes used in conjunction with the CR-39 array indicate a candidate.

Detection of a bare monopole in such a low-velocity regime requires that the detector be sensitive not only to electronic stopping but also to nuclear stopping to such an extent that $(dE/dx)_{\mathrm{tot}} \geqslant 0.1$ GeV g^{-1} cm^2. If nuclear stopping is only 20% as effective as electronic stopping at affecting the response of the CR-39, the threshold in the effective dE/dx is extrapolated to be ~ 1 GeV g^{-1} cm^2. Consequently, this type of CR-39 would be insensitive to a low-velocity monopole. Cecchini *et al.* [6] recently calibrated the CR-39 used in MACRO experiment using the SEM technique. They concluded that for their CR-39 the response to low-energy ions is the same as to high-energy ions for the equivalent value of dE/dx with the same efficiency for nuclear stopping and electronic stopping. The present work suggests that careful calibrations for each type of CR-39 are needed to determine the effectiveness of nuclear stopping relative to electronic stopping.

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[1] R. L. Fleischer, P. B. Price, and R. M. Walker, *Nuclear Tracks in Solids: Principles and Applications* (University of California Press, Berkeley, 1975).
[2] Y. D. He and P. B. Price, Nucl. Tracks Radiat. Meas. **20**, 491 (1992).
[3] Y. D. He, P. B. Price, and W. T. Williams, Phys. Lett. B **252**, 331 (1990).
[4] P. B. Price, Phys. Lett. **140B**, 112 (1984).
[5] D. P. Snowden-Ifft and P. B. Price, Phys. Lett. B **288**, 250 (1992).
[6] S. Cecchini *et al.*, Nuovo Cimento A **109**, 1119 (1996).
[7] G. Giacomelli *et al.*, Nucl. Tracks Radiat. Meas. **12**, 465 (1986): MACRO Collaboration, S. P. Ahlen *et al.*, *ibid.* **19**, 641 (1991).
[8] H. Ichinose *et al.*, Nucl. Instrum. Methods Phys. Res. A **286**, 327 (1990); S. Orito *et al.*, Phys. Rev. Lett. **66**, 1951 (1991).
[9] Y. D. He and C. I. Hancox, Rev. Sci. Instrum. **66**, 4575 (1995).
[10] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, Oxford, 1985).
[11] Y. D. He, C. I. Hancox, and M. Solarz, Nucl. Instrum. Methods Phys. Res. B **132**, 109 (1997).
[12] D. P. Snowden-Ifft, Ph.D. thesis, University of California at Berkeley, 1991.