Optical Čerenkov radiation from microscopic $BaSO_4$ grains

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In the analysis of an experiment to determine the response of scintillators to relativistic heavy ions, we have discovered that the $BaSO_4$ grains (radius ~ 0.5 μm), which constitute the highly reflective interior surface of our light-integration box, emit Čerenkov radiation with an index of refraction intermediate to that of crystalline $BaSO_4$ and to that of the air-crystal volume-averaged reflective coating. The small amount of radiation emitted below threshold ($\sim 2\%$ of the $\beta = 1$ signal) is consistent with the expected level due to Čerenkov emitting delta rays and/or to diffraction radiation with no residual scintillation.

Over the past twenty years there has been considerable interest in the nature of radiation-induced luminescence, or scintillation.[1] We have recently performed an experiment at the Lawrence Berkeley Laboratory Bevalac in which we exposed 25 samples of liquid, plastic, and inorganic scintillators to ^{20}Ne, ^{40}Ar, and ^{56}Fe heavy-ion beams with energies between 0 and 600 MeV/amu. This extends an experiment completed at an earlier date[2] as part of a cosmic-ray telescope calibration. To facillitate comparison of the response of the various samples we utilized a light-integration box similar to that described in Ref. 3. The samples are placed sequentially in a well and are thus exposed to the interior of the box. Scintillation light injected into the box is randomized by highly reflectant $BaSO_4$ paint[4,5] and is collected by a single EMI9817QAM photomultiplier tube. Analysis of the scintillator response data as a function of charge, velocity, and sample type is currently underway and will be reported at a future date. In this paper we will report on the background emission, i.e., the emission of light by heavy ions with no sample in the well. This emission is found to consist of two components: (i) unsaturated scintillation by the 51-cm air column of the box interior and (ii) Čerenkov emission by the $BaSO_4$ paint with an effective (i.e., properly weighted by light collection and quantum efficiencies and by the Čerenkov emission spectrum) index of refraction equal to 1.61 ± 0.02. The former component is consistent with observed α-particle-induced scintillations in air[1] where excellent linearity is indeed observed. The latter component represents the first observed polarization-induced radiation from grains comparable in size to the wavelength of the emitted radiation. It is this grain size and small ultraviolet absorption down to 150 nm which renders powders such as $BaSO_4$ such good diffuse reflectors.

It is well known from the theory of the scattering of light[6,7] that formidable theoretical difficulties are encountered when the size of the scattering object is comparable to the wavelength of the light which is incident upon it. Similarly, the familiar classical Čerenkov formula[8,9] is derived in the approximation that the wavelength of the emitted radiation is much smaller than the dimensions of the medium through which the charged particle passes. Pogorzelski and Yeh[10] have considered the general problem of the emission of radiation by a charged particle moving through a dielectric sphere. Although they do not reproduce the Čerenkov formula in the limit of a large sphere, they do show that the usual threshold velocity given by $v = \beta c = c/n$, where n is the refractive index of the sphere, appears in this limit. For spheres with diameters comparable to the emitted wavelength, Pogorzelski and Yeh show that there is no sharp threshold velocity due to the presence of transition and diffraction radiation which is required to satisfy the boundary conditions at the surface of the sphere. Calculation of the scattered intensity in this regime is made difficult by the need to include all higher-order multipoles and by the rapidly oscillating expansion coefficients with respect to both multipole order and sphere radius. This latter dependence on sphere radius reduces the reliability of these results for application to nonspherical grains.

In Fig. 1 we plot the data obtained for ^{40}Ar. The ordinate is the mean number of photoelectrons $\langle \Delta L \rangle_{av}$ (corrected for the asymmetry of the Poisson distribution) which arise from light emitted by the air and paint surface. The front door of the light-integration box, 1.27-cm black polyvinyl toluene, was covered by black photographic tape to eliminate the small amount of residual scintillation emanating from this surface. The energy of the beam

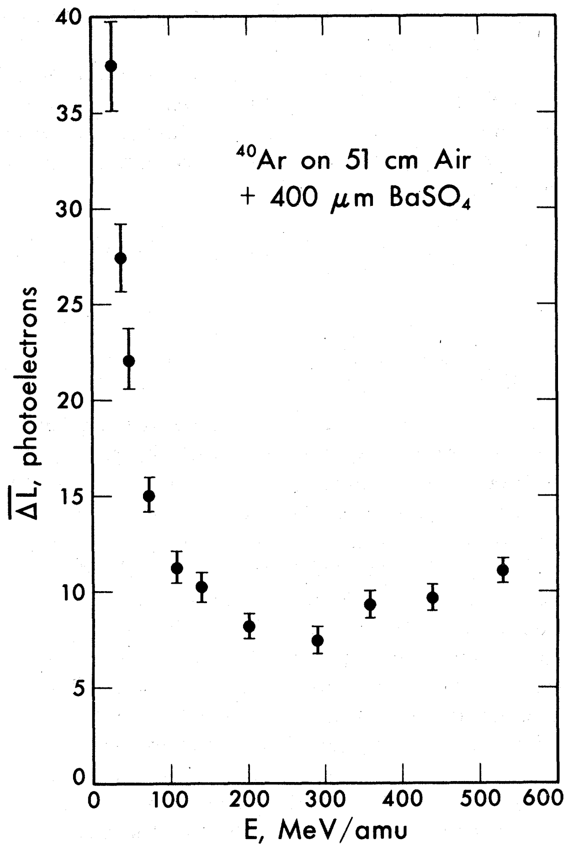

FIG. 1. Light output for ^{40}Ar incident on test box with no sample.

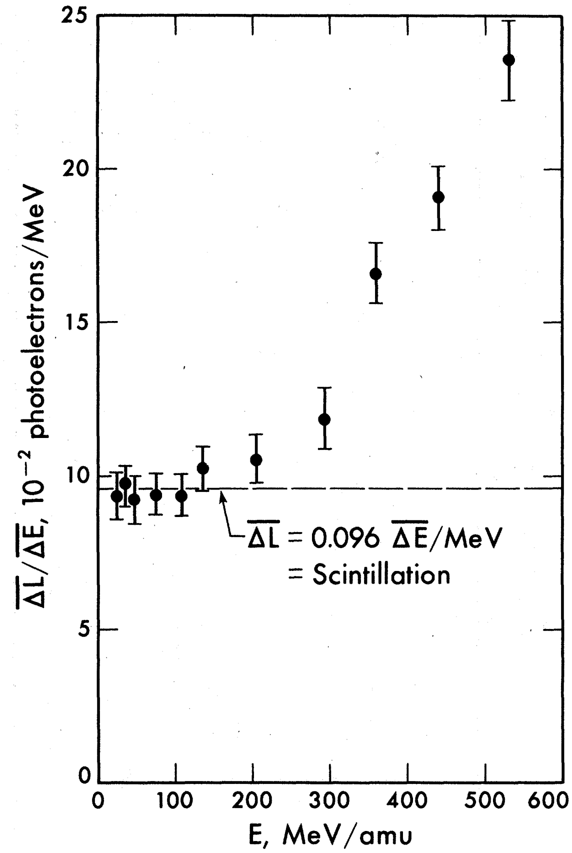

FIG. 2. Conversion efficiency for air plus powder surface.

was varied with a high-precision automated lead absorber consisting of eight slabs (the three thinnest of which were made of aluminum) each being twice as thick as its predecessor. Thus 256 switch-selectable energies were available. Discrimination against charge-changing nuclear interactions was accomplished by gating the pulse height analysis of the PMT by a sufficiently large signal from a 299-μm Si solid-state detector which was immediately in front of the light-integration box. This detector also limited analysis to particles which passed within 1.2 cm of the center of the light-integration box. Uncertainties in the incident beam energy (of the order of 0.5%) are sufficiently great to prevent accurate calculation of the beam energy after it passes through the automated absorber. We have calculated these energies by going backwards, demanding that the "cusp point," i.e., the point of maximum energy deposition, in our plastic scintillators corresponds to the energy which just barely penetrates the entire thickness of the scintillators. This procedure gave an energy of 565.7 MeV/amu incident on the automated absorber for our Pilot F scintillator. A similar technique was applied to the air scintillation of the box to eliminate positive and negative saturation. In this case we obtained an energy of 565.9 MeV/amu, which is consistent with the Pilot F value in view of the drift in beam energy which is possible. We have used an incident energy of 566.0 ± 0.3 MeV/amu for the data analysis. In Figs. 1, 2, and 3 we have used this uncertainty to estimate the errors of E, β, and $\langle \Delta L \rangle_{av}/\langle \Delta E \rangle_{av}$ (E is the energy of the beam per amu incident on the air column, βc is the velocity of beam after penetration of the air column and incident on the paint, and $\langle \Delta E \rangle_{av}$ is the mean energy loss in the air column). The error in energy or $1/\beta^2$ is always less than the size of the dot. In most cases the error has been dominated by the difficulty in determining the most probable channel for the pulse height distributions. Fluctuations due to energy straggling and photoelectron statistics prevented us from evaluating this channel to much better than 5%. This error is much greater than any due to asymmetric energy straggling or escaping δ rays. Phototube drift

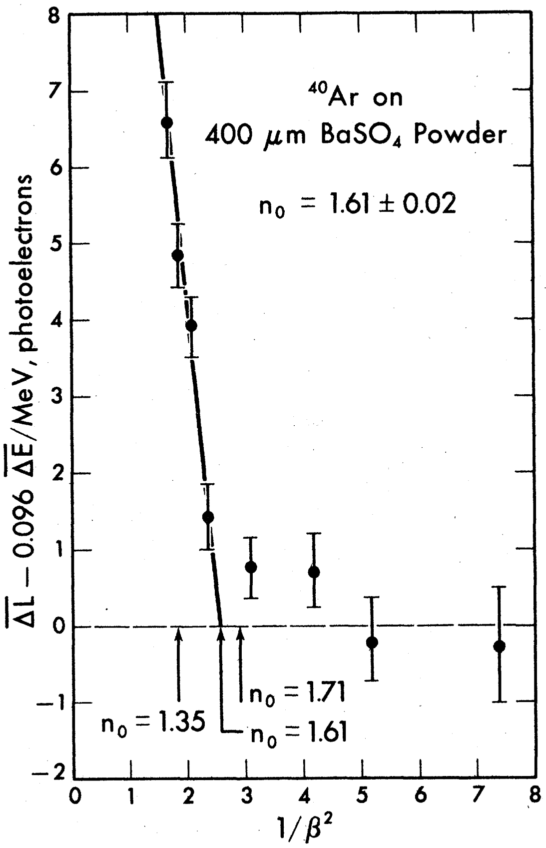

FIG. 3. Pure Čerenkov yield obtained by subtracting scintillation from total light output. The intercept of the $1/\beta^2$ axis indicates an index of refraction of 1.61±0.02. The intercept corresponding to the index of crystalline $BaSO_4$ (1.71) is shown for comparison. We also show a conservative upper limit for the intercept of a powder-air averaged mixture corresponding to a density of 2.2 g/cm^3. This represents a 95% confidence level upper limit of the actual paint density.

corrections have been applied through the use of an ^{241}Am doped NaI crystal with light emission uniform to better than 1% over the course of the experiment.

In Fig. 2 we plot the scintillation efficiency $\langle \Delta L\rangle_{av}/\langle\Delta E\rangle_{av}$ and see that this is constant for energies ≤ 150 MeV/amu, indicating the unsaturated scintillation coming from the air. By using the six lowest energy points we obtain the mean value of 0.096 ±0.001 photoelectrons per MeV of deposited energy. By comparing this result with that for the exposure of NE110 (which has an energy conversion efficiency of 3%) to atmospheric muons in the same light-integration box, we find that 93g photons/MeV are emitted by the air where g is the fraction of scintillation light which escapes from the NE110 scintillator. If $g=0.38$, a value which is consistent with the geometry of the well, we obtain the standard result for air (35 photons/MeV) obtained with α and β particles.[1]

In Fig. 3 we plot the pure Čerenkov yield by subtracting the scintillation component. This is plotted as a function of $1/\beta^2$ so that the zero intercept gives the square of the index of refraction, as required by the Čerenkov law[11]:

$$\frac{dL}{dx}=\frac{Z^2e^2}{\hbar c^2}\int_{n(\omega)>1/\beta} f(\omega)g(\omega)q(\omega)\left(1-\frac{1}{\beta^2 n^2(\omega)}\right)d\omega, \tag{1}$$

$$\frac{dL}{dx}\equiv \eta Z^2\left(1-\frac{1}{\beta^2 n_0^2}\right), \tag{2}$$

where ω is the circular frequency of the radiation, $f(\omega)$ is the light collection efficiency, $g(\omega)$ is the efficiency for escape from the Čerenkov radiator, $q(\omega)$ is the quantum efficiency of the phototube, and $n(\omega)$ is the index of refraction of the Čerenkov radiator. The projectile charge is Ze and η and n_0 are the system figure of merit and effective index of refraction, respectively. The units of dL/dx are photoelectrons/cm. Values for the index of refraction of crystalline $BaSO_4$ for several wavelengths are to be found in Ref. 12. $BaSO_4$ is biaxial and so there are three relevant indices. However, these are very close (1.636, 1.637, and 1.648 at the sodium D line) and we have simply averaged them (to obtain 1.640 for the sodium D-line value). We have fitted the tabulated values to Sellmeier's equation[13] for extrapolation into the ultraviolet. If we assume that (a) $g=1$ for $\lambda>150$ nm and $g=0$ for $\lambda<150$ nm (see Ref. 4); (b) $q(\omega)$ is as given by the results of tests performed by EMI Gencom (New York) at our request; and (c) f is determined at ~350 nm by the requirement that the air scintillation is given by the standard efficiency of 35 photons/MeV and is determined at smaller wavelengths by scaling to the results of Refs. 3 and 4 we get that

$$\frac{dL}{dx}=4.2Z^2\left(1-\frac{1}{(1.71\beta)^2}\right)\text{ photoelectrons/cm.} \tag{3}$$

The effective index $n_0=1.71$ is considerably larger than for the sodium D line due to the dispersion and to the wide spectral range of $BaSO_4$ paint and our quartz window on the phototube.

The best-fit straight line through the four high-velocity points of Fig. 3 yields $n_0=1.61\pm0.02$ where the error in index is calculated as the standard deviation of the square root of the ratio of the intercept to slope of the best-fit straight line assuming uncorrelated fluctuations of the Čerenkov signals. This index is significantly less than that expected for the pure crystal. Linney *et al.*[14] have shown that fine grain (~70 Å) silica powder has a variable Čerenkov index of refraction which is given by

$$n_0 = 1 + (0.21 \pm 0.02)\rho \text{ cm}^3/\text{g},$$

where ρ is the powder density. This is simply the volume-averaged index of the air + silica powder mixture and is what one might expect when the grain size is much smaller than the wavelength of light. Since the density of our paint is 1.7 ± 0.3 g/cm^3 and that of crystalline $BaSO_4$ is 4.5 g/cm^3, one would expect an average index of 1.27 ± 0.05 if the same prescription held for micron-size grains. This is clearly inconsistent with the data. To eliminate the possibility that large grains could be predominantly responsible for the relatively large index observed, we examined the paint under a high-power optical microscope. The bulk of the grains at the edges of paint flakes, which were most easily observed, had diameters ≤ 1 μm. Also, since the fraction of dissolved $BaSO_4$ in the wet paint is 10^{-8}, it is unlikely that any significant recrystallization occurred resulting in grains larger than initial diameters.

It can be seen from Fig. 3 that the level of light emitted below threshold is quite low, being ~2% of the signal at $\beta = 1$ (this number was obtained by averaging the four low-velocity values in Fig. 3). This level is consistent with what one would expect from Čerenkov emitting δ rays,[11] although the δ-ray transport problem is more complex here than that considered in Ref. 11. It is difficult to evaluate the level of diffraction radiation to be expected due to the above mentioned difficulties. From the graphs presented in Ref. 10 it seems safe to conclude that for grain radii of 0.3 μm and wavelengths of the same size, the level of diffraction radiation for $3 < 1/\beta^2 < 4$ is $\leq 2\%$ of the $\beta = 1$ signal. In view of the results of Ref. 14 and those presented here, it does not appear that diffraction radiation is ever a serious competitor with Čerenkov radiation.

Finally, we calculate the effective thickness of the paint surface by equating Eq. (2) (with $\eta = 4.2$, $n_0 = 1.61$) to the observed level. This yields a thickness of ~140 μm, which is significantly less than the paint thickness of ~400 μm. This is roughly what one might expect if the Čerenkov emission were restricted to the actual grain volume. Indeed, by using the measured paint density, one calculates that a 400 μm thick layer of paint has a mean total grain thickness of 150 ± 30 μm. Thus it seems possible that a substantial thickness of the paint surface is contributing to the Čerenkov signal. However, it should be emphasized that the paint layer is opaque when examined under the microscope which means that self-absorption will prevent the Čerenkov signal from increasing for increased thicknesses. This also means that the underlying primer layer and aluminum surface do not enter in any way into considerations of the experiment described here.

CONCLUSIONS

We have found that the index of refraction of the $BaSO_4$ surface is 1.61 ± 0.02. The index of refraction of crystalline $BaSO_4$ is 1.71. If the density of a very fine powder of $BaSO_4$ (i.e., with grain size much smaller than the wavelength of light) is taken to be that of the paint, then one would expect an index of refraction of the powder to be 1.27 ± 0.05. Hence, the observed value is intermediate to these values. This conclusion could be altered only if the dry-paint density is 3.9 instead of 1.7, a discrepancy well outside of any possible experimental error. We interpret this result as being due to the Čerenkov radiation emitted by micron-sized grains in response to the passage of the charged projectile. This conclusion is justified by the following observations:

(i) Microscopic examination of paint chip edges shows that the grain sizes are distributed about a mean diameter of ~1 μm, with a variation of the order $\frac{1}{2}$ μm.

(ii) Consideration of the extremely small solubility product for $BaSO_4$ implies that the opaque region of the paint chips must similarly be composed of micron-sized grains since recrystallization is impossible (the use of K_2SO_4 in the preparation of the paint strengthens this argument due to the common ion effect).

(iii) The measured optical depth of the paint surface is consistent with that expected if the grains alone are responsible for the Čerenkov radiation and not a grain-air averaged conglomerate.

(iv) The work of Pogorzelski and Yeh[10] shows that it is not unlikely for the index of refraction (in the sense of characterizing a threshold velocity) to be dependent on grain size, going over to the optical index in the limit of grain size large compared to the wavelength of light.

Finally, we have found that although diffraction radiation is in principle capable of substantially contributing to subthreshold light emission, in practice it is limited to less than 2% of the maximum possible Čerenkov emission.

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