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Sound and Light from Fractures in Scintillators

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Prompted by intriguing events observed in certain particle-physics searches for rare events, we study light and acoustic emission simultaneously in some inorganic scintillators subject to mechanical stress. We observe mechanoluminescence in Bi₄Ge₃O₁₂, CdWO₄, and ZnWO₄, in various mechanical configurations at room temperature and ambient pressure. We analyze the temporal and amplitude correlations between the light emission and the acoustic emission during fracture. A novel application of the precise energy calibration of Bi₄Ge₃O₁₂ provided by radioactive sources allows us to deduce that the fraction of elastic energy converted to light is at least 3 × 10⁻⁵.

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Recent intriguing events observed by the CRESST II dark matter search using cryogenic scintillation-phonon CaWO₄ detectors [5], study mechanoluminescence as a form of photon-producing background in such devices. We also argue that photon emission can help characterize the rupture dynamics in a manner analogous to AE [13–15] and can provide complementary information about the fracture energy. To investigate these two points, we have carried out what we believe are the first experiments to correlate the acoustic and light emission from common scintillators under mechanical stress and to quantify the conversion of elastic energy into light.

Our studies involve the measurement of acoustic phonons and luminescence as a piece of scintillating crystal is stressed to the point of rupture. The inorganic scintillators studied here are widely used in particle detection [16,17]: bismuth germanate (Bi₄Ge₃O₁₂, also known as BGO), zinc tungstate (ZnWO₄), and cadmium tungstate (CdWO₄). Except where specified, all samples were kindly supplied by Crystal Manufacturing Lab, Ltd., Novosibirsk. We have tested two loading geometries, at room temperature and ambient pressure. Initially, 20 × 10 × 5 mm³ rectangular prisms were indented by a steel bead driven by a manual screw, causing the material to break in multiple fragments. In order to better control the rupture process, we subsequently adopted the double cleavage drilled compression (DCDC) geometry [18] (Fig. 1). These samples were
20 × 5 × 3 mm³ rectangular prisms, polished to optical quality, with a 1 mm diameter circular hole drilled perpendicularly in the middle of the 20 × 5 mm² face. Under compression along the long axis, cracks formed reproducibly on either side of the hole in the plane parallel to the 20 × 3 mm² face. Unlike BGO, tungstates have a cleavage plane [19]; orientation of the crystals was chosen so that this plane was parallel to the DCDC fracture direction. The DCDC samples were pressed against a backstop by a push rod that was driven by an actuator (controlled manually in early experiments and by computer in later ones). This geometry provides some control of crack velocity via the applied compressive stress, up to a critical length, at which point the sample cleaves abruptly [18]. A force gauge in the push rod measured the load imposed by displacements of the actuator. The acoustic activity was recorded by two piezoelectric transducers in contact with opposite sides of the crystal via ultrasound gel. Light emission was recorded by a standard bialkali photomultiplier (PM) covering the crystal. The output of the PM was integrated with a time constant ranging from 1.8 to 10 μs, depending on the sample, to produce a signal that could be digitized more slowly than the raw PM output. All four channels (force, 2 AE, PM) were digitized at a rate of 2 × 10⁶ samples per second in a continuous stream. The set point of the computer-controlled actuator was also recorded.

The fracture procedure involves advancing the actuator by small steps (down to 0.1 μm). Each step is followed by a waiting time of at least 30 s. The whole procedure lasts 1–2 h, during which we continuously monitor AE and PM activity. Figure 2 shows typical observations for the tungstate crystals. Spurts of activity on the AE and PM channels are correlated to drops in the force. These abrupt force relaxations are a direct consequence of crack growth in the samples. There is also a regime in which the force decreases slowly and during which other processes, such as rearrangements and dislocations, produce AE and PM signals [8]. Similar correlations have been observed for other materials, such as colored KBr [8], composites [20], and silica glass [21].

Indentation of BGO samples (from Fibercrst, France) also produces correlated AE and light signals. These correlations can be analyzed more quantitatively by looking at the statistical properties of individual acoustic and light events. In this analysis, individual events have been determined on each channel by requiring that the signal surpass a threshold and that it be separated from the previous event by at least a minimum time. Figure 3 shows that the distributions of waiting times between events are similar for AE and light over two decades. For light events, it is possible to obtain a much smaller minimum separation time (1.5 μs) than for AE (250 μs), since the former pulses are shorter than the latter.

To quantify the amount of light emitted and to investigate the emission mechanism, we next compared the spectra of light emitted by BGO (sample from Fibercrst) during fracture and during scintillation stimulated by x rays. To avoid afterglow from the high dose of x rays, the fracture spectrum was measured first, and then one of the fragments was measured while being irradiated by x rays. The measurements of the light spectra were carried out with a monochromator and a CCD camera. In the x-ray excitation configuration, a generator provided x rays with a broad spectrum up to a few tens of keV. Over the recorded range of wavelengths, both resulting spectra (Fig. 4) show a very similar shape (as is the case for many materials [22]), indicating a common underlying luminescence mechanism, at least for the last stages of light emission. The likeness of our spectra to previous work on BGO indicates this last stage involves ¹P₁ → ¹S₀ transitions.
electronic transitions of the Bi$^{3+}$ luminescence centers [23]. This measurement does not determine if the fractures excite the luminescence centers directly. It cannot be excluded that the crystal is indirectly excited by particles produced by fractoemission [9] interacting in the scintillator. For instance, if x rays or electrons are produced, the BGO sample would be quite efficient at detecting them itself (attenuation length = 20 µm for 10 keV x rays [24]), as opposed to needing an external x-ray detector [11]. Alternatively, any UV light (potentially from arcs in the atmosphere near the fracture) might be reabsorbed in the scintillator since BGO absorbs wavelengths below = 300 nm [25].

We then calibrated the light channel for a DCDC BGO sample using radioactive γ-ray sources as we would calibrate a regular scintillator used for particle detection. Calibration was carried out in the main DCDC setup and with the standard data acquisition system (Fig. 1). The analysis involves extracting individual events as described earlier and then building the distribution of the event integrals. The integral of an event, like its amplitude, is a proxy for the energy deposited by the γ ray, so we can identify the peaks in the distribution corresponding to the energies of the radioactive sources and of backscattering [16], as shown in Fig. 5. Over the energy range available with these sources, the crystal and readout behave in a linear manner with respect to the energy deposited in the scintillator, and we extrapolate this calibration to higher energies. Calibrations before and after the fracture are compatible to within 10%. From the standpoint of the scintillation mechanism, γ rays and x rays can interact in inorganic scintillators via the photoelectric effect or through Compton scattering, creating a primary electron-hole pair which eventually transfers a portion of the deposited energy to the luminescence centers (Bi$^{3+}$ for BGO). For a given light signal, calibrations provide the equivalent amount of energy deposited by a γ ray. This can be converted to the actual energy of the emitted light by knowing the light yield of the scintillator (8 photons/keV for γ rays in BGO [16]) and the energy of individual scintillation photons (2.6 eV; cf. Fig. 4).

The calibrated BGO crystal was then fractured in the main DCDC setup, using the protocol described earlier. We focus on the last drop in force, occurring when the crack splits the sample in two. Figure 6 shows that the overall behavior is similar to that observed for the tungstates. The photon channel displays a wide distribution of event amplitudes. They in fact reach up to the saturation level of the integrator, equivalent to γ-ray energy deposits of ≈ 85 MeV—a testimony to the amount of light emitted. There is also a hint of increased photonic activity before the main fracture occurs. The correlations are better quantified in Fig. 7, illustrating the cumulative number of events on the acoustic channels and those on the photon channel. Minimum separation time required between photon events was 10 µs, and 1 ms between AE events. On all three
The amount of emitted light is equivalent to that caused by calibrations with decay into heat), and light. For the emitted light, the (that are either measured as acoustic emission or BGO. Elastic energy is converted into broken bonds, photons, or heat. The standard room-temperature technique to study brittle fracture only involves measurement of acoustic phonons using acoustic emission. We are not aware of any work quantifying the fraction of energy going into acoustic phonons or calibrating the energy scale of acoustic phonons. The harder-to-implement technique of cryogenic calorimetry would allow measurement of all the phonons and would therefore provide more insight into the energy budget.

We have evidenced correlated mechanical-stress-induced emission of photons and phonons in several common inorganic scintillators used for particle detection (Bi$_4$Ge$_3$O$_{12}$, ZnWO$_4$, and CdWO$_4$), at room temperature and in a normal atmosphere. At least for BGO, both emissions share similar distributions of waiting times over several decades. Also for BGO, the two energy distributions are similar, and the wavelength of fractoluminescence is the same as that of scintillation. In a mechanically stressed scintillation-only detector, the power-law distribution of mechanoluminescence energies means that spurious low-energy events may mimic dark matter ones. It seems reasonable to assume our results apply to other tungstates, and it would be interesting to study the light-phonon correlations of individual fracture events in CRESST II–like conditions of vacuum and low temperature (other background-based explanations of the intriguing CRESST II events have been proposed [31]). In addition, the reabsorption of products of fractoemission could affect other types of solid-state particle detectors (e.g., ionization or ionization-phonon ones [1,16]). From the standpoint of fracture physics, in contrast to previous
studies by acoustic emission or by spectrally resolved luminescence (e.g. Ref. [32]), our use of the light channel offers a precisely calibrated energy scale. This allows us to quantify the amount of elastic energy converted into light and is a step toward a better fundamental understanding of fracture mechanisms.

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