LETTERS

Correlation between nanosecond X-ray flashes and stick–slip friction in peeling tape

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Relative motion between two contacting surfaces can produce visible light, called triboluminescence[1]. This concentration of diffuse mechanical energy into electromagnetic radiation has previously been observed to extend even to X-ray energies[2]. Here we report that peeling common adhesive tape in a moderate vacuum produces radio and visible emission[3,4], along with nanosecond, 100-mW X-ray pulses that are correlated with stick–slip peeling events. For the observed 15-keV peak in X-ray energy, various models[5,6] give a competing picture of the discharge process, with the length of the gap between the separating faces of the tape being 30 or 300 μm at the moment of emission. The intensity of X-ray triboluminescence allowed us to use it as a source for X-ray imaging. The limits on energies and flash widths that can be achieved are beyond current theories of tribology.

When a continuous medium is driven far from equilibrium, nonlinear processes can lead to strong concentrations in the energy density. Sonoluminescence[7] provides an example in which acoustic energy concentrates by 12 orders of magnitude to generate subnanosecond flashes of ultraviolet radiation. Charge separation at contacting surfaces[8,9] is another example of a process that funnels diffuse mechanical energy into high-energy emission. Lightning[10], for instance, has been shown to generate X-rays with energies of more than 10 keV (ref. 11). Although triboelectrification is important in many natural and industrial processes, its physical explanation is still debated[10,12].

By peeling pressure-sensitive adhesive tape one realizes an everyday example of tribocharging and triboluminescence[1]: the emission of visible light. Tape provides a particularly interesting example of these phenomena because it has been claimed that the fundamental energy that holds tape to a surface is provided by the van der Waals interaction[13]. This energy—the weakest in chemistry—is almost 100-fold smaller than the energy required for generating a visible photon, yet, as demonstrated in 1939 (ref. 3), light emission from peeling tape can be seen with the unaided eye. That even more energetic processes were at play had already been suggested in 1930 (ref. 14); it was observed that when mica is split under vacuum "the glass of the vessel fluoresces like an X-ray bulb". This insight led to the discovery in 1953 (ref. 2) that peeling tape is a source of X-rays. The simultaneous emission of visible and X-ray photons from peeling tape is shown in Fig. 1a, in which the blue glow is due to a scintillator responsive to X-ray energies and the red patch near the peel point is neon-enhanced triboluminescence[3]. Figure 1b shows that when the vacuum pressure is 10^{-3} torr the high-energy emission is so strong that the photo is illuminated entirely with scintillations.

Motivated by these photos, we interpret triboluminescence[1], a phenomenon known for centuries, as being part of an energy-density-focusing process that can extend four orders of magnitude beyond visible light to X-ray photons. To learn about the processes occurring in peeling tape, we employed efficient high-speed X-ray detection equipment. Our measurements indicated that the scintillations in

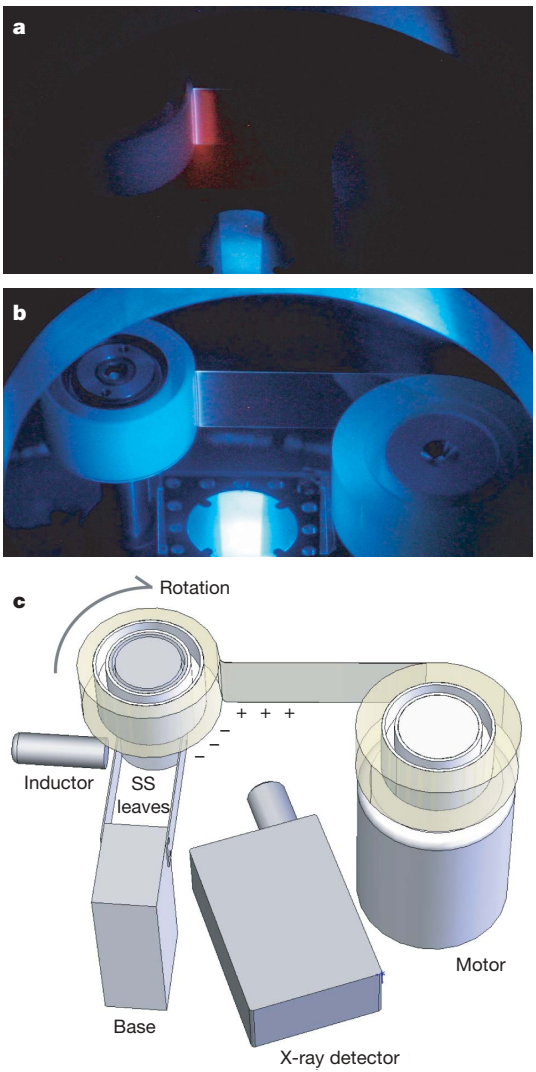

Figure 1 | Apparatus for studying high-energy emission from peeling tape. **a**, Photograph of the simultaneous emission of triboluminescence (red line) and scintillations of a phosphor screen sensitive to electron impacts with energies in excess of 500 eV (under neon at a pressure of 150 mtorr). **b**, Photograph of the apparatus (under a pressure of 10^{-3} torr) illuminated entirely by scintillations. **c**, Diagram of the apparatus used to measure peeling force; SS, spring steel (Methods).

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Fig. 1b contain nanosecond X-ray pulses whose emission is correlated with radiofrequency (r.f.) pulses and slips in the force required to peel the pressure-sensitive adhesive tape. Furthermore, the short duration of these X-ray pulses indicated that the emission originates from a submillimetre region near the vertex of peeling, with a transient charge density ($\sim 10^{12}$ electrons cm^{-2}); that is, more than an order of magnitude greater than is measured in typical tribocharging systems.

The correlation between X-ray emission and peeling force in a 10^{-3} torr vacuum is displayed in Fig. 2a. As the force (black trace) increases above its value under an applied pressure of 1 atm (ref. 15) (dashed green trace), emissions with X-ray energies are recorded (blue trace). No X-ray emission has been observed at 1 atm. The slips are also correlated with a signal detected by an r.f. antenna[16] (red trace). Figure 2b shows subnanosecond-resolved data used to correlate r.f. emission from peeling tape with liquid-scintillator signals (blue trace). The solid red and dashed red traces are the response of the antenna to signals generated, respectively, by peeling tape and by the relative motion of mercury and glass, in which r.f. discharges due to tribocharging are known to occur[16].

The data in Fig. 2a were acquired with tantalum foil shielding the window of a solid-state X-ray detector. This attenuates X-rays with energies below about 20 keV in favour of larger events synchronized to the slips. The spectrum[17] of all X-ray photons emitted from the peeling tape as recorded by an unshielded solid-state detector is shown in Fig. 3 and in Supplementary Fig. 1. To minimize the pile-up of photons the detector was placed 69 cm from the peeling vertex of the tape, so the plotted data have a solid angle correction of 120,000 relative to the raw data (see Methods). The total energy in the bursts that accompany the slips was obtained from events that were three-way coincident between a solid-state detector, the liquid scintillator and the characteristic r.f. pulse (Fig. 2b). The inset to Fig. 3 shows the spectrum of X-ray burst energies that accompany slip events out to 10 GeV. These pulses occur at a rate in excess of 1 Hz and their time traces fall within the 5-ns resolution of the liquid-scintillator detectors. The spectrum did not change significantly during ten rewindings of a given roll of tape.

According to studies of controlled vacuum discharges[18], the rise time of the current is the width of the X-ray flash. From the red trace in Fig. 2b this implies that the width of the coincident X-ray pulses is $\sim$1–2 ns. Thus a typical 2-ns burst with an energy of 2 GeV has a peak power of more than 100 mW. These bursts, which occur more than once per second, contain more than 50% of the total energy radiated as X-ray photons above 10 keV. This includes X-ray photons synchronized to slip events as well as 'precursor' X-rays emitted between slips. According to Fig. 3 the total emission is 1.2×10^{10} eV s^{-1} or 2 nW average X-ray power.

On the basis of the long-standing phenomenology of tribocharging[8,12], we propose the following sequence of events: as the tape peels, the sticky acrylic adhesive becomes positively charged and the polyethylene roll becomes negatively charged, so that electric fields build up to values that trigger discharges. At a reduced pressure, the discharges accelerate electrons to energies that generate Bremsstrahlung X-rays when they strike the positive side of the tape. To determine the current of high-energy electrons that drive this process we compared Fig. 3 with published scattering data[19]. A strand of adhesive tape is thick in comparison with an electron absorption length (the Kramers limit[19]) but not so thick as to absorb all the X-rays. Given that the difference is not significant[19], here we take the thick target limit. The peak near 15 keV with 3×10^5 X-rays per second is therefore due to electrons with energies of about 30 keV, which then create an integrated Bremsstrahlung X-ray spectrum with an efficiency of 10^{-4}. Only 5% of these X-rays are above 15 keV. These factors imply a discharge current of 6×10^{10} electrons per second, which corresponds to an average electric power of 0.2 mW; this is five orders of magnitude higher than the integrated X-ray spectra displayed in Fig. 3. As the 2-cm-wide tape peels at 3 cm s^{-1}, the average density of charge separated and discharged is 10^{10} electrons cm^{-2}, which is consistent with known tribocharging processes[12].

The X-ray bursts require charge densities that are substantially larger than those that characterize the average tribocharging discussed above. For a Townsend discharge[6], the bottleneck is the time it takes an ion to cross a gap of length l times the number of round trips (about ten) needed to build up an avalanche. For a hydrogen ion

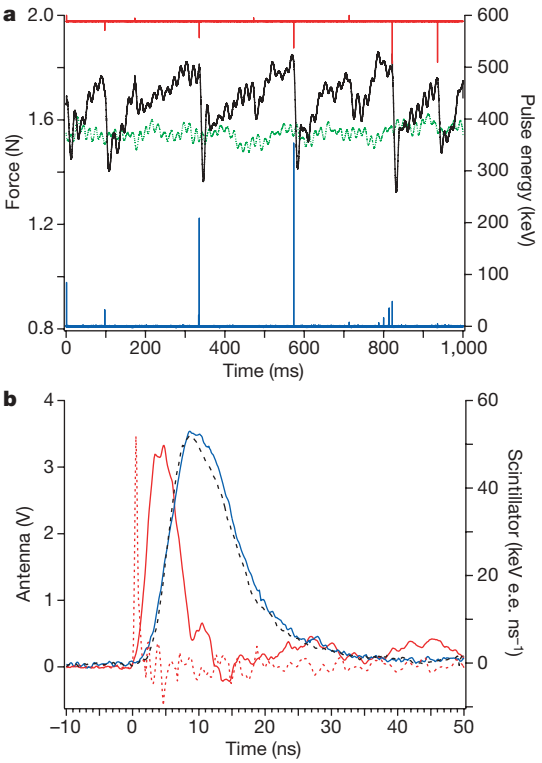

Figure 2 | Correlation between X-rays, force and radio frequency. **a**, The left axis shows the force for peeling tape at 3 cm s^{-1} in a 10^{-3} torr vacuum (black line) and at 1 atm (dashed green line). The right axis shows the X-ray signal (blue trace) from an Amptek detector with tantalum foil shield. The red upper trace is the r.f. antenna signal. **b**, Correlation of liquid scintillator (blue line) with radio frequency (red line) from peeling tape. The rise time of the scintillator is about 5 ns for the tape signal (blue line) and cosmic-ray calibration (dashed blue line). The dashed red line is an antenna calibration signal (Methods); keV e.e. ns^{-1} is the electron equivalent energy per

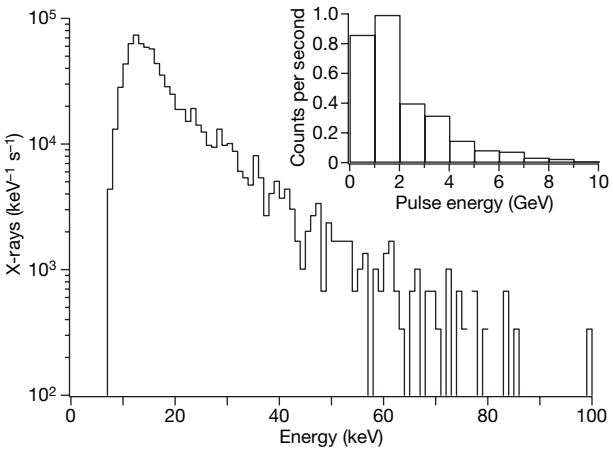

Figure 3 | Spectrum of X-ray energies from peeling one roll of tape. The peel speed was between 3 and 3.6 cm s^{-1} at 10^{-3} torr of air. Data were acquired with the Amptek CdTe detector. Inset: energies for nanosecond pulses out to 10 GeV for the same run taken with the Amptek 3-Stack detector (Methods).

moving with a velocity $v = \sqrt{(2eV/m)}$ in a potential $V = 30$ kV, a pulse width $\Delta t = 10l/v$ of ~1 ns implies a characteristic length l of ~300 μm, which in turn implies an accelerating field $E \sim V/l$ of $\sim 10^6$ V cm^{-1} and a charge density $\sigma \approx \varepsilon_0 E$ of 7×10^{11} electrons cm^{-2} (ref. 20). According to an alternative theory, the discharge consists of an explosive plasma emission[5]. The characteristic time for the current to flow is determined by the time it takes the plasma moving at 2×10^6 cm s^{-1} to expand across the gap[5,18]. It has been established experimentally that the duration of the pulse increases linearly with the gap size with proportionality factor of 5 ns/100 μm (refs 5, 18). This implies a gap l of the order of tens of micrometres, and the corresponding field of 10^7 V cm^{-1} requires a charge density of 7×10^{12} electrons cm^{-2}. An image of the X-ray emission region could distinguish between the various theories.

When the tape is peeled, part of the energy supplied is converted to elastic deformation of the tape[21], cavitation[22] and filamentation[23] of the adhesive, acoustic emission[24], visible light[3,25] and high-energy electron emission[2]. According to Fig. 2 the power required to peel the tape at a speed of 3 cm s^{-1} is 50 mW under ambient conditions of 1 atm. Under vacuum, an additional power of 3 mW must be supplied to overcome the observed stick–slip friction. Of this 3 mW at least 0.2 mW goes into accelerating electrons to 30 keV so as to generate an average X-ray power of 2 nW. The power going into visible triboluminescence is 10 nW, as shown by the spectrum (Fig. 4).

Although tribocharging has substantial technological applications[12], its physical origin is still in dispute. In one view, tribocharging of insulators involves the statistical mechanical transfer of mobile ions between surfaces as they are separated adiabatically[8]. A competing theory[9] proposes that a charged double layer is formed by electron transfer across the interface of dissimilar surfaces in contact. When these surfaces are suddenly pulled apart, the net charge of each layer is exposed. We have observed two timescales in dynamic tribocharging. One is the long timescale over which average charge densities of about 10^{10} electrons cm^{-2} are maintained on the tape. In addition, there exists a process that concentrates charge on a transient timescale of the order of 1 ns to reach densities that are about 100-fold larger than the average value. The physical process whereby such a large concentration of charge is attained involves the surface conductivity of the tape. This conductivity could be provided by mobile ions[12] or perhaps by means of precursor discharges stirring up the surface of the peeling tape. We propose that X-ray emission should yield insight into this and other fundamental aspects of tribology.

The intensity of emission is sufficiently strong (see Supplementary Fig. 2) to make peeling tape useful as a source for X-ray photography.

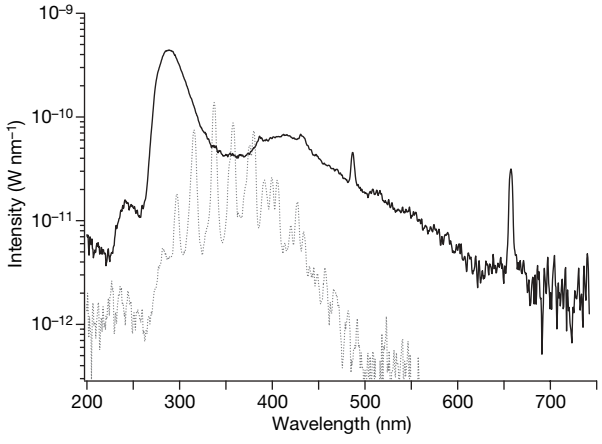

Figure 4 | Light spectra from peeling tape. The black trace was taken at 10^{-4} torr of air, and the grey dashed trace was taken at 1 atm. The nitrogen lines, which are prominent in air at 1 atm, are indicative of a gas discharge, which is typical of other processes such as fractoluminescence[27] and lightning. At low pressure the nitrogen lines are overshadowed by a process that leads to broadband emission with hydrogen lines.

Examples of X-ray photos are provided in Supplementary Fig. 3 and Supplementary Fig. 4. The correlation displayed in Fig. 2 has a resemblance to the geophysical effect called earthquake lights[26], in which the liberation of stress-induced charge during earthquakes generates electromagnetic radiation. The macroscopic physical processes that spontaneously organize an off-equilibrium throughput of low-energy density into X-ray emission suggest that it will be worth while to look for this phenomenon in systems that show fractoluminescence[27], stick–slip friction[16,28], triboluminescence[1] and gecko adhesion[29]. The charge density realized in these experiments is about the same as the effective charge that accumulates on the surface of pyroelectric crystals used to generate table-top nuclear fusion[30].

METHODS SUMMARY

All experiments were performed with off-the-shelf rolls of Photo Safe 3M Scotch Tape (19 mm × 25.4 m) that were secured to a precision ball-bearing mounted on a stage supported by two very stiff steel spring leaves (with spring constant $6.6 \times 10^3 \pm 3 \times 10^2$ N m^{-1}; Fig. 1c). The displacement of the leaves from their equilibrium position was measured with a commercial inductor position detector (Baumer Electric) with a resolution of 505 mm V^{-1}. A free portion of the tape was stuck to a cylinder connected to a rotating motor, and the whole setup was placed in a vacuum chamber. All X-ray data were acquired at a pressure of $\sim 10^{-3}$ torr and at a peel speed of ~3 cm s^{-1}. X-ray energy emissions were recorded with Amptek (XR-100T 3-stack and XR-100 CdTe) X-ray detectors and with Bicron 501A liquid scintillators 12.7 cm in diameter and 12.7 cm long, coupled to Hamamatsu 12.7-cm photomultiplier tubes (R1250)[30]. Radiofrequency signals were recorded with antennas made of the exposed inside conductor of BNC cables placed within millimetres of the peeling point. All data were digitized and saved to disk for offline analysis as detailed in the Methods section. The spectrum of visible photons (Fig. 4) was taken with a grating spectrometer (300i; Acton Research) coupled to an intensified camera (Princeton Instruments) and is corrected for the response function of the instrument.

Full Methods and any associated references are available in the online version of the paper at www.nature.com/nature.

Received 30 December 2007; accepted 27 August 2008.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

Acknowledgements We thank R. Lofstedt for bringing to our attention the importance of ref. 14; C. Regan for advice; B. Naranjo for many insights and for use of his uniquely designed liquid scintillator detection system; B. Kappus and S. Khalid for experimental assistance; J. Cambier for valuable discussions, E. Adams for archival assistance; and A. Bass for an independent translation of ref. 2. We thank Hamamatsu Corporation for lending us X-ray cameras. Various stages of this research were supported by the Office of Naval Research and the Defense Advanced Research Projects Agency (Microsystems Technologies Office and Defense Sciences Office) . J.E. thanks the Fulbright–Garcia Robles Scholarship Program and UC-MEXUS-CONACYT for support.

Author Contributions J.H. was instrumental in motivating this research. J.E. pinned down the correlation between the force and X-ray emission. C.C. obtained the GeV pulses, and S.P. is the principal investigator.

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METHODS

Figure 1a and Fig. 1b are 15-s exposures on a Canon EOS 10D camera. The electron scintillator visible in the forefront of these images is a Kimball Physics C5X5-R1000. The data shown in Fig. 2a were obtained with a National Instruments PXI-5122 14-bit digitizer at ten points μs^{-1}. The ~80-Hz oscillations on the force measurement correspond to the resonance frequency of the loaded spring. We note that although our data clearly show stick–slip motion, our peel speed of 3 cm s^{-1} is much lower than that referred to in the literature as the stick–slip regime for peeling pressure-sensitive adhesive tape[31]. The r.f. emission was recorded with a BNC chassis mount placed about 1 cm from the peel line terminated with 500 Ω (red upper trace) displayed in arbitrary units. For this figure the Amptek X-ray detector (XR-100T 3-stack) was placed about 5 cm from the peeling interface and its beryllium window was shielded with tantalum foil 25 mm thick, to prevent saturation. This detector has a background of about one count every 3 s from 5 to 400 keV (Supplementary Fig. 1), and pileup cannot be discriminated for events shorter than 600 ns. The possibility of pileup affecting spectral data also challenges efforts to resolve X-ray energy emission from lightning bolts in which similar energy scales are detected[11]. The X-ray data in Fig. 2b were acquired with a Hamamatsu 5-inch photomultiplier (R1250) looking at Bicron 501A liquid scintillator (5 inches in diameter by 5 inches long)[30] and recorded with an Infinium oscilloscope at 8×10^9 samples s^{-1} (1.5 GHz bandwidth). The units in the scintillator axis are kiloelectronvolt electron equivalent per nanosecond, and are with reference to the calibration performed with several Compton edges from different radioactive sources[30]. The centre of the scintillator was placed 15 cm from the peeling tape outside the vacuum chamber looking through a 2-cm quartz window. In this figure the antenna is 5 mm of exposed inside wire of a BNC cable terminated with 50 Ω. The relative timing of the signal has been corrected for the 54-ns transit time of the photomultiplier and the 3-ns length of the antenna. The characteristic rise time of the scintillator–photomultiplier arrangement can be determined by capturing a high-energy cosmic ray (dashed blue trace) and is seen to be about 5 ns, the same as for the X-ray pulse. The subnanosecond pulse (dashed red line) used to calibrate the antenna is generated by charge transfer between mercury and glass in relative motion[16]. Further studies of the timescales for discharge[18,32] could yield insight into the mechanisms involved.

The X-ray spectrum shown in Fig. 3 was obtained by unwinding an entire roll of tape at between 3 and 3.6 cm s^{-1}, which took about 700 s. The data were acquired with a solid-state X-ray detector (100-XR CdTe; Amptek), unshielded, placed outside the vacuum chamber at 69 cm from the peeling tape and looking through a ¼-inch plastic window. This detector has an active area of 25 mm^2, is 100% efficient from 10 to 50 keV, and has a background count rate of ~1 count per 100 s. The data were digitized with a National Instruments PXI-5122 board at a rate of 1 s every 1.9 s for a total of 364 s. The inset in Fig. 3 is the frequency of emission of nanosecond-long X-ray pulses as a function of the total pulse energy generated during the same unwinding. An X-ray pulse was deemed valid if a coincidence within 10 ns was recorded between the r.f. antenna and the liquid scintillator (501A; Bicron), and within 2 μs of a signal on an unshielded solid-state detector (XR-100 3-stack; Amptek) with more than 10 keV. All the Amptek coincidences are, however, found within a 400-ns window, which we believe is the limit of the internal electronics of the device. The antenna was 5 mm of exposed inside conductor of a standard BNC cable terminated with 50 Ω, placed 5 mm from the peel line. The X-ray detectors were placed outside the chamber looking through a ¼-inch plastic window, the 3-stack solid-state detector at 40 cm from the tape and the scintillator at 76 cm. Coincidence data were digitized at 10^9 samples s^{-1} with an Acqiris DC270 board[30] triggered on the antenna signal. The dead time of these acquisitions was less than 20 s for the 700-s run, and the background coincidences were found to be 0 for a 1,000-s wait.

The visible spectrum at 1 atm in Fig. 4 shows lines that are indicative of gas discharge, also observed in fractoluminescence[27] and lighting[33]. At low pressure the nitrogen lines are overshadowed by a process that leads to broadband emission with hydrogen lines.

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