Triboluminescence dominated by crystallographic orientation

Kuifang Wang¹, Liran Ma², Xuefeng Xu², Shizhu Wen¹ & Jianbin Luo¹

Triboluminescence (TL) is an optical phenomenon that has a long and varied history with broad applications, such as damage detection, X-ray source, and mass health monitoring sensor. So far, the properties and mechanisms of TL remain not completely understood. The TL properties emitted during the sliding contact between Al₂O₃ and SiO₂ surfaces were studied along different crystallographic orientations. In this study, the TL intensity of Al₂O₃ was significantly enhanced as Al₂O₃ surface was along a particular crystallographic orientation, which is an unconventional phenomenon. TL enhancement of Al₂O₃ was not affected by air atmosphere and atomic stacking mode of Al₂O₃. The enhancement mechanism of Al₂O₃ may be influenced by the surface state of Al₂O₃. This work provides a new method to control the intensity of TL and novel ideas to elucidate the TL mechanism.

Triboluminescence (TL) is a luminescence phenomenon by solid materials when they are stressed or fractured¹,². TL is also regarded as fracture, piezoelectric, and mechanical luminescence. Since the 20th century, this phenomenon has gained increasing attention because of its broad application³. TL has been successfully used in mass health monitoring sensor⁴, X-ray source, and damage detector⁵, among others. The triboluminescent properties of crystal materials are mostly investigated because many common crystal materials exhibit TL performance⁶.

TL properties of crystal are often affected by external factors, such as gas atmosphere, gas pressure, temperature, and speed. The photon emission intensity of Al₂O₃, ZrO₂, and Si₃N₄ with a diamond stylus decreases with increasing number of carbon atoms in the hydrocarbon molecules⁷, whereas the emission intensity enhances to a maximum value at a particular n-butane gas pressure⁸. The TL intensities of NaCl and LiF doped with Br, Sr, Ca, and Pb decrease with temperature and disappear completely at 105 ± 5 and 180 ± 10 °C, respectively⁹. Hollerman et al.¹⁰ reported that the TL emission of Zn:Mn appears to be a function of speed for collision up to 6 Kms⁻¹. The doped impurities can change the TL properties of crystal materials; the TL emission intensity of ZnS:Mn is extremely strong, but TL properties are absent in ZnS¹⁰,¹¹.

The physical properties of crystal structure also greatly influence TL. The discussion about TL properties of crystal with different space groups indicated that a non-centrosymmetric crystal structure is necessary but not sufficient for TL in crystal materials¹²,¹³. Hird¹⁴ reported that the intensity of TL emission during diamond polishing in ‘hard’ direction is greater than ‘soft’ direction. The crystallographic orientation of Al₂O₃ influences the atomic and electronic structures of alumina surfaces¹⁵. Brewer et al.¹⁶ investigated the fluorescence band at 3.0 eV, which was produced by photoexcitation in high-purity Al₂O₃ crystal. The results showed that the emitted light is plane polarized with the maximum intensity that occurs when the electric vector is perpendicular to the c axis of the crystal (E⊥c) and with the minimum intensity that occurs when the electric vector is parallel to the c axis (E∥c)¹⁶. Kurita et al.¹⁷ explored detailed atomic structures and electron states of stable and metastable surfaces of three important planes of Al₂O₃, namely, C plane (the (0001) surface), R plane (the (1102) surface), and A plane (the (1120) surface). They found that the stoichiometric surfaces of the C plane have the lowest surface energy, followed by the stoichiometric surfaces of the R plane and then the A plane. In other studies, Cs-corrected high-resolution electron microscopy that combines first-principle calculations and image simulations was used to observe and investigate the quantitative and qualitative structures of (1120) and (0001) surfaces¹⁸. Al₂O₃ possesses good TL properties, but is not clearly investigated. SiO₂ is a common crystal material, and the investigation of SiO₂ of TL properties is very less.

The TL properties of crystal materials in different crystallographic orientations are rarely reported. We explored the TL properties of Al₂O₃ along different crystallographic orientations by measuring TL emission during sliding with SiO₂. We discovered an unconventional phenomenon that the TL intensity was enhanced several tens of times as Al₂O₃ plane was in a particular crystallographic orientation. This work may provide a novel method to control the intensity of TL.

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**Results**

**TL in ambient air.** Schematic diagram of light emission is shown in Fig. 1(a), the intensity of light emission of Al₂O₃ is greatly enhanced due to crystallographic orientation changed from (1120) to (0001). The images of photon emitted in ambient air during the sliding contact between Al₂O₃ (0001), (1120), (1010), and (1102) surfaces and SiO₂ (110) under normal force (F) of 10 N and relatively shear velocity (V) of 33 mm/s are shown in Fig. 1(b–e). Images of (c) and (d) are much brighter than those of (b) and (e). The mean intensities of I(b), I(c), I(d), and I(e) were 249, 1605, 4512, and 222, respectively. The mean intensities of (b) and (e) are much weaker compared with those of (c) and (d). I(c) is about 10 and 7 times higher than I(b) and I(e), and I(d) is about 22 and 14 times higher than I(b) and I(e). The images of photons emitted during sliding between Al₂O₃ (0001), (1120), (1010), and (1102) surfaces and SiO₂ (003) are also shown in Fig. 1. In this condition, the intensity of TL has the same appearance although crystallographic orientation of SiO₂ is changed. Thus, the TL intensities of Al₂O₃ (1120) and (1010) surfaces are significantly enhanced compared with the TL intensities of Al₂O₃ (0001) and (1102) surfaces.

The spectra of photons emitted in ambient air during the sliding contact between Al₂O₃ (0001), (1120), (1010), and (1102) surfaces and SiO₂ (110) under F = 10 N and V = 33 mm/s are shown in Fig. 2(a). Many sharp peaks in regions 300–450 nm and 600–900 nm are on spectra, but the spectra intensities of diverse Al₂O₃ surfaces are quite different. The maximum spectra intensities of Al₂O₃ (1120) and (1010) surfaces are much higher than those of Al₂O₃ (0001) and (1102) surfaces. The maximum spectrum intensity of Al₂O₃ (1120) is nearly five times of spectra intensity of Al₂O₃ (0001) and (1102) surfaces, in quantitative terms, while the maximum spectrum intensity of Al₂O₃ (1010) is more than 10 times than those of Al₂O₃ (0001) and (1102) surfaces. As the crystallographic orien-
tation of SiO₂ is changed from (110) to (003), the spectra intensities of Al₂O₃ (1120) and (1010) surfaces of Al₂O₃ are much stronger. Thus, the intensity of emission is enhanced, when Al₂O₃ (1010) and (1120) surfaces are sliding with SiO₂.

We conducted other sliding experiments to study further the enhancement mechanism of TL intensity of Al₂O₃. The images and spectra of photons emitted in ambient air during the sliding contact between Al₂O₃ (0001), (1120), (1010), and (1102) surfaces and SiO₂ (110), (b) during the sliding contact between Al₂O₃ (0001), (1120), (1010), and (1102) surfaces and SiO₂ (003).

Figure 2. The spectra of photons emitted under F = 10 N and V = 33 mm/s in air: (a) during the sliding between (0001), (1120), (1010), and (1102) surfaces of Al₂O₃ and SiO₂ (110), (b) during the sliding contact between (0001), (1120), (1010), and (1102) surfaces of Al₂O₃ and SiO₂ (003).
(1010) surfaces are higher than those of Al2O3 (0001) and (1102) surfaces. The maximum intensity of images of 
Al2O3 (1010) is nearly three times stronger than those of Al2O3 (0001) and (1102) surfaces. In this condition, the photons emitted in ambient air are extremely weaker through eliminating the effect of external electric potential difference. The spectra of photons of Al2O3 (1120) and (1010) surfaces have sharp peaks as shown in Fig. 3(e), whereas those of Al2O3 (0001) and (1102) surfaces have no peaks. Thus, the TL intensities of Al2O3 (1010) and 
(1120) surfaces are much higher.

TL in vacuum. To explore better the TL properties of Al2O3, we conducted subsequent experiments in vac-
umum instead of ambient air. Figure 4 shows the images and spectra of photons emitted during the sliding contact between Al2O3 (0001), (1120), (1010), and (1102) surfaces and SiO2 (110) in vacuum under F = 10 N and 
V = 33 mm/s. The spectra of Al2O3 in vacuum which are composed of instrument noise, have no characteristic peaks due to very lower photons intensity. The mean intensities of photon images of Al2O3 (1120) and (1010) surfaces are higher than those of Al2O3 (0001) and (1102) surfaces. The mean intensity of Al2O3 (1120) is nearly two times stronger than those of Al2O3 (1120) and (1010) surfaces.

The spectra of photons emitted in vacuum have no peaks because of extremely low light intensity as shown in Fig. 4(e). In this condition, the TL properties of Al2O3 are also affected by crystallographic orientation, and the TL intensities of Al2O3 (1120) and (1102) surfaces are enhanced.

Friction and abrasion. The friction coefficients of the sliding contact between Al2O3 (0001), (1120), (1010), and (1102) surfaces and SiO2 (110) are measured using a Universal Micro-Tribotester (UMT-3; Bruker, America), where the values of friction coefficients are 0.179, 0.258, 0.261, and 0.329. The friction coefficient of Al2O3 (0001) surface is lowest and that of Al2O3 (1102) surface is highest. The friction coefficients of Al2O3 (1120) and (1010)
surfaces are much closer. The results prove that the TL intensity has no evident linear relationship with friction coefficient as the TL intensities of Al2O3 (1120) and (1010) surfaces are much higher.

The depth and width of grinding cracks on SiO2 (110) surfaces, which are sliding with Al2O3 (0001), (1120), (1010), and (1102) surfaces, are measured using a TalySurf (5P-120; Taylor Hobson, England). Figure 5 is the mean depth and width of the grinding crack of SiO2 (110) by sliding with Al2O3 (0001), (1120), (1010), and (1102) surfaces. The values of mean depth of four groups are close to 0.02 mm, whereas those of SiO2 (110) surfaces have no distinct differences. The results stated that abrasions of SiO2 (110) surfaces sliding with Al2O3 (0001), (1120), (1010), and (1102) surfaces have no apparent variations. Thus, the TL intensity of Al2O3 is not influenced by wear of SiO2, and enhancement mechanism of TL properties of Al2O3 is dominated by crystallographic orientation.

Discussion
The Al2O3 crystal is a hexagonal crystal, and the side views of atom arrangement of Al2O3 (0001), (1120), (1010), and (1102) surfaces are shown in Fig. 6(a–d), respectively. In the hexagonal unit cell, the atoms are stacked along the [0001] direction in a sequence of an oxygen layer and Al double layers: -AlAlO3-AlAlO3-R (Fig. 6(a)). The atoms along the [1120] direction are a layer unit that consists of five atomic layers: an O layer constructed of one O atom in the 1×1 unit, an O layer constructed of two O atoms, an Al layer constructed of four Al atoms, and an O layer constructed of one O atom. This -O-O2-Al4-O2-O- repeating layer unit has no dipole moment along the [1120] direction. The atoms are stacked along the [1010] direction in a sequence of an O layer and an Al layer: -O-Al-O-Al-R (Fig. 5(f)). The 1×1 lateral unit of the (1102) surface is a layer unit in a sequence of an Al layer constructed of two Al atoms and an O layer constructed of two O atoms. The -Al2-O2-Al2-O2- repeating layer unit has no dipole moment along the direction perpendicular to the (1102) surface. The atomic stacking mode of

Figure 4. Photons under F = 10 N and V = 33 mm/s in vacuum during the sliding between SiO2 (110) and Al2O3 (0001) (a) images with I = 35 and (e) spectra, Al2O3 (1120) (b) images I = 114 and (f) spectra, Al2O3 (1010) (c) images I = 119 and (g) spectra, Al2O3 (1102) (d) I = 81 and (h) spectra.
Al$_2$O$_3$ is extremely complicated along different crystallographic orientations, where (1010) and (1120) Al$_2$O$_3$ surfaces have no similar stacking mode. No regular discipline exists to indicate that the enhancement of TL intensities of (1010) and (1120) Al$_2$O$_3$ surfaces is related to atomic stacking mode.
The results of rough measurements of surface charge of Al₂O₃ and SiO₂ by using faraday cup showed that Al₂O₃ surface was negatively charged and SiO₂ was positively charged. Tribocharging mechanism is that electrons transferred from a surface with a low work function to a mating surface with a high work function. In surface state theory, charge is exchanged between surface states in proportion to the difference between the effective or surface work functions of the two materials. Surface potential difference is the fermi level difference between...

Figure 7. (a) Single-crystal X-ray diffractometer results of Al₂O₃ surfaces with miller indices (0001), (11̅20), (1010), (1102), and SiO₂ surfaces with miller indices (110) and (003). (b) Al₂O₃ planes in hexagonal unit cell.

Figure 8. Schematic diagram of the sliding experiment setup for observation of the images and spectra of photons.
original surfaces as well as the work function difference. In equation (1), \( V_c \) is the surface potential difference, \( \phi_1, \phi_2 \) are the work functions of two surfaces respectively.

\[
V_c = \frac{\phi_1 - \phi_2}{e}
\]

Silicon oxide (SiO\(_2\)) surface has a lower work function than aluminum oxide (Al\(_2\)O\(_3\)) surface. Thus, electrons are transferred from SiO\(_2\) to Al\(_2\)O\(_3\), resulting in the former being positive and the latter being negative, then electric field between SiO\(_2\) and Al\(_2\)O\(_3\) contacting surfaces is formed. Electrons of ambient air molecules in electric field will be excited from ground level to the exited levels, then fall down to the lower or ground level, photons are emitted. These sharp peaks of spectra are caused by the electrical breakdown of ambient gas. The spectra peaks of photons in the region 300–450 nm are assigned to C\(_2^+\)−→B\(_3^+\) electron transitions of N\(_2\). Other sharp peaks of spectra are mainly due to the B\(_3^+\)−→A\(_2^+\) electron transitions of N\(_2\) and the b\(_3^+\)−→X\(_3^+\) electron transition in O\(_2\). The pressure of vacuum chamber is between 1 to 10 Pa during the sliding experiment. Air molecules are much smaller under this condition. Thus, photons emitted during sliding between SiO\(_2\) and Al\(_2\)O\(_3\) in vacuum is very few, the spectra of Al\(_2\)O\(_3\) in vacuum have no peaks.

As shown in Fig. 6(e), the band gap of SiO\(_2\) is 9 eV, and the conduction band (Ec) is −9.6 eV. The surface states (S\(_s\)) of Al\(_2\)O\(_3\) (0001) and Al\(_2\)O\(_3\) (1120) are 9 and 12 eV, respectively. The bottom of Fermi level of Al\(_2\)O\(_3\) (1120) is 0.8 eV. Al\(_2\)O\(_3\) (1120) has two surface-state bands at 12 and 15 eV. The bottom of the Fermi level of Al\(_2\)O\(_3\) (0001) is −6.8 eV, which is much lower than that of (1120). Al\(_2\)O\(_3\) (0001) has two surface-state bands at 9 and 12.5 eV. The surface states of Al\(_2\)O\(_3\) (1120) are much higher than those of Al\(_2\)O\(_3\) (0001). Thus, enhancement mechanism of Al\(_2\)O\(_3\) may be related to the energy levels of Al\(_2\)O\(_3\), while much higher energy levels of Al\(_2\)O\(_3\) (1120) surface result in increasing TL emission.

The enhancement mechanism of TL properties of Al\(_2\)O\(_3\) may be influenced by the surface state of Al\(_2\)O\(_3\). The enhancement mechanisms of Al\(_2\)O\(_3\) still need further exploration, and our work may provide a novel method to control the TL intensity.

Methods

Materials. Silicon oxide (SiO\(_2\)) and aluminum oxide (Al\(_2\)O\(_3\)) crystals with trigonal and hexagonal crystal structures, respectively, were used in the sliding experiment. The two types of crystal planes of SiO\(_2\) were (110) surface by X cut and (003) surface by Z cut. The results of single-crystal X-ray diffractometer of SiO\(_2\) surfaces are shown in Fig. 7(a). Four surface planes of Al\(_2\)O\(_3\) crystal, including C plane (0001), A plane (1120), M plane (1010), and R plane (1010) were used as shown in Fig. 7(b). (1120) and (1010) planes are parallel to axis, and (0001) plane is perpendicular to axis. (1010) plane is crossed with C axis. The dielectric constant of sapphire at 298 K in 10\(^3\)–10\(^9\) Hz interval is \( \epsilon = 11.5 \), \( \perp \) C = 9.3. Dielectric constant of four planes of Al\(_2\)O\(_3\) have little difference. The SiO\(_2\) with width of 3 mm and thickness of 2 mm and the Al\(_2\)O\(_3\) with diameter of 30 mm and thickness of 2 mm were purchased from Shanghai Daheng Optics & Fine Mechanics Co. Ltd. The Vickers hardness of (110) and (003) surfaces of SiO\(_2\) are 1257 and 1167, respectively, and those of Al\(_2\)O\(_3\) (0001), (1120), (1010), and (1102) are 2060, 2119, 2076, and 2329, respectively. The thickness of (110) and (003) surfaces of SiO\(_2\) are 1.6 and 1.44 nm, respectively, and those of Al\(_2\)O\(_3\) (0001), (1120), (1010), and (1102) surfaces are 5.43, 5.21, 5.18, and 5.51 nm. In our experiments, Al\(_2\)O\(_3\) (0001), (1120), (1010), and (1102) surfaces would be slid with SiO\(_2\) (110) and (003) surfaces, respectively.

Experimental setup. The schematic of the experiment setup used to observe the images and spectra of photons during sliding between SiO\(_2\) and Al\(_2\)O\(_3\) is shown in Fig. 8. Optical fiber was used to gather light and then transmitted the light to a spectrograph (SP2500; Princeton Instruments, America). The images and spectra of photons were obtained with the spectrograph and CCD. The spectra of photons ranged from 300 nm to 900 nm, and the image of photons reflected the overall intensity of light.

Experimental condition. Silicon oxide (SiO\(_2\)) was adhered to the rotating platform along with the motor, and the Al\(_2\)O\(_3\) wafer was fixed on a holder under a normal force of 10 N, as shown in Fig. 8. The bottom surface of Al\(_2\)O\(_3\) wafer was sliding over the top surface of SiO\(_2\) wafer. The integration time (T) of CCD camera was 10 min, and the relative shear velocity (V) between Al\(_2\)O\(_3\) and SiO\(_2\) wafer was 33 mm/s. The sliding experiment was performed in ambient air and vacuum. The vacuum pressure was between 1 and 10 Pa, and air humidity was nearly 10%.

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Author Contributions
L.M., X.X. and S.W. conceived the experiments. K.W. conducted the experiments and processed the experimental data. L.M., X.X. and K.W. analysed the results. All authors reviewed the manuscript.

Additional Information
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