Quantum Theory of Triboelectricity

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(Received 16 May 2019; accepted 18 September 2020; published 30 October 2020)

We propose a microphysical theory of the triboelectric effect by which mechanical rubbing separates charges across the interface between two materials. Surface electrons are treated as an open system coupled to two baths, corresponding to the bulks. Extending Zel'dovich's theory of bosonic superradiance, we show that motion-induced population inversion can generate an electromotive force. We argue that this is consistent with the basic phenomenology of triboelectrification and triboluminescence as irreversible processes, and we suggest how to carry out more precise experimental tests.

DOI: 10.1103/PhysRevLett.125.186101

Introduction.—The word *electricity* comes from the ancient Greek ἤλεκτρον for amber, a solid material that charges when rubbed with silk or fur. In the sixth century BCE, pre-Socratic philosopher Thales of Miletus pointed to magnets and amber as evidence of "a soul or life even to inanimate objects" [1,2]. The microphysics of dry friction remains poorly understood and there is still no widely accepted theory of triboelectrification, the separation of charges by rubbing. The Bohr–van Leeuwen theorem establishes that classical physics cannot explain the properties of magnetic materials [3], but it is less widely appreciated that classical electrodynamics is insufficient to account for triboelectricity.

Consider the triboelectric generator shown schematically in Fig. 1. The inner cylinder of material A rotates about its axis with angular velocity Ω. For the right choice of material B in the outer, hollow cylinder, a voltage is established between A and B, which can sustain a current I through an external circuit. The classical electromotive force (emf) $\mathcal{E}$ vanishes by the Maxwell-Faraday law:

$$\mathcal{E} \equiv \oint \mathbf{E} \cdot d\mathbf{s} = -\frac{d}{dt} \int \mathbf{B} \cdot d\mathbf{a} = 0, \qquad (1)$$

as there is no significant variation of the net magnetic flux through the plane of the circuit. Thus, at the interface between the two materials A and B, electrons are being transported *against* the average electric field by a nonconservative force (the emf), effectively acting as a negative resistance. The power for this evidently comes from the motor that spins A. But how mechanical energy is converted into the electrical work done by the emf calls for explanation.

Note that the generation of an emf by the relative motion of A and B must be irreversible, since the direction of the emf cannot depend on the sign of Ω. On the emf as an active nonconservative force, and on the impossibility of accounting for it using potentials, see [4]. Recently, the irreversible dynamics of work extraction by a quantum system coupled to an external disequilibrium has become a subject of theoretical and practical interest in quantum thermodynamics [5].

In 1971, Zel'dovich described a process, later dubbed "superradiance" by Misner, by which the kinetic energy of a moving dielectric can be partially converted into coherent radiation [6,7]. This result played a key role in the development of black-hole thermodynamics and it provides a useful guide to a broad class of active, irreversible processes [8,9]. As in a laser, superradiance depends on population inversion, which in the case of rotational superradiance results from the disequilibrium associated with the dielectric's macroscopic motion. Work may then be extracted from the population-inverted states through stimulated emission while generating entropy in the rotating dielectric, which we may treat as a moving heat bath [10].

The exclusion principle prevents stimulated emission of fermions, and therefore their superradiance. However, we will show here that the motion-induced population inversion of fermions can sustain a macroscopic current between

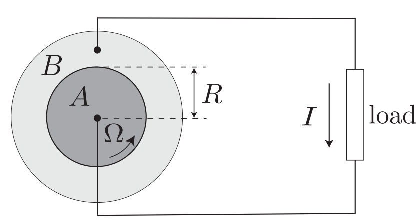

FIG. 1. The triboelectric generator sketched maintains a current I along the circuit if an external motor spins the cylinder of material A and radius R at a sufficient angular velocity Ω with respect to another material B.

two baths coupled to those fermion states. Such a process has not, to our knowledge, been considered before, although the authors of [11] noted the presence of Fermi surfaces of singularities in the Green's functions of fermions in the background of a charged black hole. Here, we argue that this offers a plausible theory of triboelectricity, including such remarkable phenomena as the generation of x rays by peeling ordinary adhesive tape [12,13].

Experimentalists have stressed that triboelectrification and associated effects depend strongly on the relative velocity of the materials in contact and are therefore essentially off equilibrium [14]. The process that we describe here is irreversible and velocity dependent. As such, it is qualitatively different from the reversible processes, describable in terms of Hamiltonians, considered in recently proposed theories of the triboelectric effect [15] and the related phenomenon of contact electrification [16]. More details on the theoretical and experimental motivations for our non-Hamiltonian, open-system theory of triboelectricity are provided in the Supplemental Material [17].

Open system.—Consider surface electrons as an open quantum system, weakly coupled to two baths corresponding to bulk materials A and B. In accordance with the setup of Fig. 1, we assume cylindrical symmetry so that each electron mode, both in the surface and in the bulk, is labeled by the common magnetic quantum number m (our final results will not, however, depend on this cylindrical symmetry). Any remaining quantum numbers are labeled by σ and κ.

The second-quantization formalism and notation are similar to those applied to rotational superradiance in [10]. Annihilation and creation operators are, respectively, denoted by $c_{\cdot}(\cdot,\cdot)$ and $c_{\cdot}^{\dagger}(\cdot,\cdot)$, while the corresponding energies are denoted by $\omega_{\cdot}(\cdot,\cdot)$. The subsystem is indicated by the index, while the quantum numbers of the mode are given as arguments. We work in $\hbar = 1$ units.

At rest, the system Hamiltonian is the sum of terms

$$H_0^x = \sum_{\sigma,m} \omega_x(\sigma,m) c_x^{\dagger}(\sigma,m) c_x(\sigma,m) \tag{2}$$

for $x = a, b$, with a corresponding to the surface attached to material A, and b corresponding to the surface attached to material B. Meanwhile, the Hamiltonians for the baths are

$$H_0^X = \sum_{\kappa,m} \omega_X(\kappa,m) c_X^{\dagger}(\kappa,m) c_X(\kappa,m) \tag{3}$$

for $X = A, B$.

If the material A rotates with an angular velocity Ω small enough that its internal states are not excited by the rotation, then we have effective Hamiltonians

$$H_\Omega^a = \sum_{\sigma,m} [\omega_a(\sigma,m) - m\Omega] c_a^{\dagger}(\sigma,m) c_a(\sigma,m) \tag{4}$$

and

$$H_\Omega^A = \sum_{\kappa,m} [\omega_A(\kappa,m) - m\Omega] c_A^{\dagger}(\kappa,m) c_A(\kappa,m). \tag{5}$$

The sign of Ω in Eqs. (4) and (5) is arbitrary and has been chosen for later convenience. The shift from the H_0's to the H_Ω's may be interpreted as a Doppler shift.

The experimental evidence is now clear that triboelectrification of solids is dominated by electron tunneling processes [18]. We therefore consider a weak interaction between the surface electrons and each of the two baths,

$$H_X^x = \sum_{\kappa,\sigma,m} g_X^x(\kappa,\sigma,m) c_X^{\dagger}(\kappa,m) c_x(\sigma,m) + \text{H.c.}, \tag{6}$$

where the g_X^x's correspond to direct transition amplitudes, to which the Coulomb interaction probably contributes significantly.

We expect the surface states a and b to be localized along the transport direction (i.e., perpendicular to the surface), so that their mutual interaction plays no role in transport. We therefore neglect ab interactions, which would give only a hybridization absorbable into modified wave functions. Moreover, since the ab interaction is not needed to obtain a triboelectric effect, it is reasonable to neglect it for the sake of simplicity since our present goal is to formulate a qualitatively new model rather than a detailed one. We therefore take the full Hamiltonian to be

$$H_{\text{full}} = H_\Omega^a + H_0^b + H_\Omega^A + H_0^B + H_A^a + H_B^a + H_A^b + H_B^b. \tag{7}$$

Kinetic equations.—The occupation numbers for the surface electron states are

$$n_x(\sigma,m) = \langle c_x^{\dagger}(\sigma,m) c_x(\sigma,m) \rangle. \tag{8}$$

In the limit of weak coupling between the system and the baths, we may compute the decay rates $\gamma_{\downarrow}^{xX}$ using Fermi's golden rule [19,20]. The pumping rates $\gamma_{\uparrow}^{xX}$ are related to the decay rates by the Kubo-Martin-Schwinger (KMS) condition. Omitting the quantum numbers, the corresponding kinetic equation may be written as

$$\dot{n}_x = \gamma_{\uparrow}^{xA} + \gamma_{\uparrow}^{xB} - (\gamma_{\downarrow}^{xA} + \gamma_{\downarrow}^{xB} + \gamma_{\uparrow}^{xA} + \gamma_{\uparrow}^{xB}) n_x. \tag{9}$$

Let us define

$$n_X(y) \equiv \frac{1}{e^{\beta(y-\mu_X)} + 1}, \tag{10}$$

where μ_X is the chemical potential of the corresponding bulk material in equilibrium.

By Fermi's golden rule, the rate of decay of the a surface electrons into the bath A is

$$\gamma_{\downarrow}^{aA}(\sigma,m) = 2\pi\{1-n_A[\omega_a(\sigma,m)]\}\overline{g_A^a}^2(\sigma,m), \quad (11)$$

where

$$\overline{g_A^a}^2(\sigma,m) \equiv \sum_{\kappa}|g_A^a(\kappa,\sigma,m)|^2\delta[\omega_a(\sigma,m)-\omega_A(\kappa,m)]. \quad (12)$$

For the pumping rate we have, by the KMS condition,

$$\begin{aligned}\gamma_{\uparrow}^{aA}(\sigma,m) &= 2\pi n_A[\omega_a(\sigma,m)]\overline{g_A^a}^2(\sigma,m)\\ &= e^{-\beta[\omega_a(\sigma,m)-\mu_A]}\gamma_{\downarrow}^{aA}(\sigma,m).\end{aligned} \quad (13)$$

Because of the shift of the energies in Eq. (4), for the rate of decay of a surface electrons into the bath B we have

$$\gamma_{\downarrow}^{aB}(\sigma,m) = 2\pi\{1-n_B[\omega_a(\sigma,m)-m\Omega]\}\overline{g_B^a}^2(\sigma,m;\Omega), \quad (14)$$

where

$$\begin{aligned}\overline{g_B^a}^2(\sigma,m;\Omega) \equiv \sum_{\kappa'}&|g_B^a(\kappa',\sigma,m)|^2\\ &\times\delta[\omega_a(\sigma,m)-m\Omega-\omega_B(\kappa',m)].\end{aligned} \quad (15)$$

The pumping rate is given by the modified KMS relation

$$\gamma_{\uparrow}^{aB}(\sigma,m) = e^{-\beta[\omega_a(\sigma,m)-m\Omega-\mu_B]}\gamma_{\downarrow}^{aB}(\sigma,m). \quad (16)$$

Thus, when

$$m\Omega > \omega_a(\sigma,m)-\mu_B \quad (17)$$

the corresponding state exhibits population inversion ($\gamma_{\uparrow}^{aB} > \gamma_{\downarrow}^{aB}$), making it possible to extract electrical work from it. A similar analysis gives us $\gamma_{\downarrow}^{bX}$ and $\gamma_{\uparrow}^{bX}$. Equation (17) corresponds to the "anomalous Doppler shift" of the Ginzburg-Frank theory of radiation by uniformly moving sources [21,22].

Work may be extracted by superradiance from a single moving bath because the pumping of the population-inverted bosonic state leads to stimulated emission [10]. In the case of fermions, on the other hand, a second bath is needed to remove the pumped fermion from its population-inverted state, before another fermion becomes available to sustain an active current. Whereas superradiance and other forms of bosonic radiation by uniformly moving charges may be described classically [8,21], the fermionic case (which we propose here as the microphysical basis of the triboelectric effect) requires a quantum treatment.

Tribocurrents.—In the steady state ($\dot{n}_a = 0$), Eq. (9) implies that

$$n_a = \bar{n}_a \equiv (\gamma_{\uparrow}^{aA}+\gamma_{\uparrow}^{aB})/\Gamma^a, \quad (18)$$

where

$$\Gamma^a \equiv \gamma_{\uparrow}^{aA}+\gamma_{\downarrow}^{aA}+\gamma_{\uparrow}^{aB}+\gamma_{\downarrow}^{aB}. \quad (19)$$

For each channel (σ,m), the number of electrons per unit time that flow from A to a is

$$j_a = \gamma_{\uparrow}^{aA}-(\gamma_{\downarrow}^{aA}+\gamma_{\uparrow}^{aA})\bar{n}_a. \quad (20)$$

By Eqs. (13) and (16), this can be reexpressed as

$$j_a = \gamma_{\uparrow}^{aA}\gamma_{\downarrow}^{aB}[1-e^{\beta(m\Omega+\mu_B-\mu_A)}]/\Gamma^a. \quad (21)$$

In the steady state this is also the current the flows from B to a (see Fig. 2).

Similarly, $\dot{n}_b = 0$ implies that

$$n_b = \bar{n}_b \equiv (\gamma_{\uparrow}^{bA}+\gamma_{\uparrow}^{bB})/\Gamma^b, \quad (22)$$

where

$$\Gamma^b \equiv \gamma_{\uparrow}^{bA}+\gamma_{\downarrow}^{bA}+\gamma_{\uparrow}^{bB}+\gamma_{\downarrow}^{bB}. \quad (23)$$

The current that flows from B to b (which in the steady state equals the current from b to A) is then

$$\begin{aligned}j_b &= \gamma_{\uparrow}^{bB}-(\gamma_{\downarrow}^{bB}+\gamma_{\uparrow}^{bB})\bar{n}_b\\ &= \gamma_{\downarrow}^{bA}\gamma_{\uparrow}^{bB}[1-e^{-\beta(m\Omega+\mu_B-\mu_A)}]/\Gamma^b.\end{aligned} \quad (24)$$

As illustrated in Fig. 2, the total electric current from A to B is

$$J = -e\left[\sum_{\sigma,m}j_a(\sigma,m)-\sum_{\sigma',m}j_b(\sigma',m)\right]. \quad (25)$$

By Eqs. (13) and (14) we have that

$$\gamma_{\uparrow}^{aA}\gamma_{\downarrow}^{aB} \sim n_A[\omega_a(\sigma,m)]\{1-n_B[\omega_a(\sigma,m)-m\Omega]\}. \quad (26)$$

As the ratio μ/k_BT for ambient temperature is $\simeq 10^2$, we replace the Fermi-Dirac distributions by step functions $n_X(y) \simeq H(\mu_X - y)$ giving

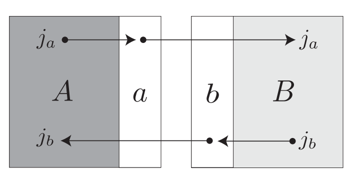

FIG. 2. Sketch of the currents j_a of Eq. (20) and j_b of Eq. (24), for the open system a, b in a steady state.

$$\gamma_{\uparrow}^{aA}\gamma_{\downarrow}^{aB} \sim \chi_{[\mu_B+m\Omega,\mu_A]}[\omega_a(\sigma,m)], \tag{27}$$

where χ_E is the indicator function of the set E. Thus, only surface modes of electrons satisfying

$$m\Omega < \mu_A - \mu_B, \tag{28}$$

contribute to the tribocurrent j_a in Eq. (21), so that $j_a > 0$. By a similar reasoning we find that only modes satisfying

$$m\Omega > \mu_A - \mu_B \tag{29}$$

contribute to j_b in Eq. (24) and therefore $j_b > 0$.

Phenomenology.—The j_x currents depend on surface-to-bulk tunneling rates that are exponentially sensitive to potential barrier heights and widths. The sums in Eq. (25) also depend on the density of surface electron states. But even without detailed characterization of this complex landscape we can show that our theory is nontrivially consistent with key observations.

The sign of J in Eq. (25) depends on the relative magnitudes of $\gamma_{\uparrow}^{aA}\gamma_{\downarrow}^{aB}/\Gamma^a$ and $\gamma_{\downarrow}^{bA}\gamma_{\uparrow}^{bB}/\Gamma^b$, controlled by the couplings between bulks and surfaces. For two materials in rubbing contact, the sign of J can therefore vary with the surface's geometry, corrugation, stress, etc. This agrees with the observation of patches of positive and negative charge, with sizes at the roughness scale $\simeq 1\ \mu$m [23].

According to Eqs. (28) and (29), as $|\mu_A - \mu_B|$ increases under net charging, fewer modes contribute to the j_x in Fig. 2 giving the charging, while more modes contribute to the opposing current. This may explain why significant triboelectrification is usually seen only when two materials well separated in the "triboelectric series" are rubbed against each other [24]. It may also explain why the net current between the rubber belt and the metal brush is opposite at the two terminals of a Van de Graaff generator, where the brushes are identical except for their respective voltages [25].

A larger work function for material A implies a higher barrier for a to B tunneling, thus suppressing $\gamma_{\downarrow}^{aB}$ in Eq. (21), whereas a larger work function for material B suppresses $\gamma_{\downarrow}^{bA}$ in Eq. (24). We therefore expect net J (for zero initial voltage) to tend to point from the material with greater work function to the one with smaller work function, as reported in [26]. Work functions do not, however, determine triboelectric properties entirely. The details of the interface barrier can play an important role, especially for insulators [27].

Let (k_z, k_m) be the cylindrical components of the wave vector and let k_F be the maximum value of $\sqrt{k_z^2 + k_m^2}$, corresponding to the Fermi wave vector for the surface electrons. In terms of the linear speed $V_s = |\Omega R|$ with which the surface of material A slides against the surface of material B in Fig. 1,

$$|m\Omega| = |k_m V_s| \le k_F V_s. \tag{30}$$

From Eqs. (28) and (29) we conclude that

$$e\phi_{\rm oc} = |\mu_A - \mu_B|_{\text{at zero current}} \lesssim \hbar k_F V_s, \tag{31}$$

where $\phi_{\rm oc}$ is the tribovoltage (note that we have reintroduced $\hbar$). The bound of Eq. (31) is saturated if and only if j_a is negligible compared to j_b, or vice versa.

Taking $k_F \simeq 1\ \text{Å}^{-1}$ and $V_s \simeq 1$ m/s in Eq. (31), we obtain $\phi_{\rm oc} \lesssim 10^{-5}$ V. Rapid mechanical separation of the charged surfaces increases the voltage accordingly [16]. If the distance between the charged surfaces grows from angstrom to meter scale, the resulting voltage will be $\lesssim 10^5$ V, as in a Van de Graaff generator [25]. If the distance goes from interatomic to $\simeq 10\ \mu$m scale, the energy of the electrons can be in the visible range ($\simeq 1$ eV). On triboluminescence, see [28] and references therein.

The surface charge density generated by peeling adhesive tape increases strongly with the peel rate [14]. The surface charge density $\simeq 10^{10}\ e/\text{cm}^2$ reported in [13] may be consistent with our theory, supposing that the maximum velocity of slippage between the dissimilar materials in contact is larger, by a couple of orders of magnitude, than the average peel rate $\simeq 1$ cm/s. The x-ray bursts produced by the peeling are preceded by a further hundredfold increase in the charge density, in a process connected with macroscopic stick-slip oscillations [13]. Such acoustic oscillations can enhance the effective $m\Omega$ in the exponential of Eq. (16), pumping the $\phi_{\rm oc}$ by another 2 or 3 orders of magnitude.

Recent experiments, in which various materials are charged using a uniform technique, find triboelectric charge densities σ lying on an approximately symmetric interval $[-\sigma_{\max}, \sigma_{\max}]$; see Fig. 3 in [26]. Since the maximum and minimum values of σ correspond to entirely different materials, this symmetry has no obvious explanation in potential models. On the other hand, it agrees with our Eq. (31), according to which $\sigma_{\max}$ (proportional to the upper bound on $\phi_{\rm oc}$) should be determined by the technique used. More detailed comparison to data will require a better understanding of how the effective V_s depends on the various experimental setups.

Discussion.—Ginzburg stressed that "radiation during the uniform motion of various sources is a universal phenomenon rather than an eccentricity" [22], with counterparts "in any field theory" [21]. Considering bosonic superradiance in terms of open quantum systems clarifies the respective roles of macroscopic motion, dissipation, and stimulated emission [10]. Here, we have extended that analysis to fermions, allowing us to propose a microphysical explanation of the persistent conversion of macroscopic motion into an emf, something that cannot be obtained from density functional theory or other equilibrium descriptions [28].

In our treatment, the emf results from motion-induced enhancement of pumping over decay (i.e., population inversion) in the modified KMS relation of Eq. (16). This allows us to obtain active currents from the kinetic equations for the populations of the surface electron states coupled to the two bulk materials. This theory has other key features qualitatively different from what one might expect in a potential description: Rubbing produces opposing currents j_a and j_b (see Fig. 2), and the upper bound on charging of Eq. (31) (approached when the two materials are very far from each other on the triboelectric series) depends only on the Fermi wave vector of the surface electrons and on sliding velocity. We have argued that these and other aspects of our theory are compatible with reported observations. New experiments with precise control of the sliding velocity (possibly based on setups closer to Fig. 1) could test our predictions more directly.

Some authors have interpreted triboelectrification as resulting from phonon production by mechanical rubbing [29]. The irreversible consumption of mechanical power by dry friction may result from the generation of phonons that then thermalize in the bulk [30]. Such phonons may contribute to the tribocurrent by assisting electron tunneling, enhancing the effective g_X^x's in Eq. (6). On the other hand, the direct j_x's consume power even when dry friction is not accompanied by significant net charging. The roles of phonons and j_x currents in both dry friction and triboelectrification therefore call for further investigation. In the Supplemental Material [17] we sketch an argument for why we expect the contribution of phonon-assisted tunneling to triboelectrification to be relatively small.

We thank Carlos Díaz, José Gracia-Bondía, Gian Guzmán-Verri, Itamar Kimchi, Lok Lew Yan Voon, John McGreevy, and Niclas Westerberg for discussions. R. A. was supported by the International Research Agendas Programme (IRAP) of the Foundation for Polish Science (FNP), with structural funds from the European Union (EU). A. J. was supported by the University of Costa Rica's Vice-rectorate for Research (Project No. 112-B6-509), by the EU's Horizon 2020 research and innovation program under the Marie Skłodowska-Curie Grant No. 690575, and by Narodowa Agencja Wymiany Akademickiej (NAWA's) Ulam Programme (Project No. PPN/ULM/2019/1/00284, "Energy conversion by open quantum systems: Theory and applications").

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[17] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.125.186101 for a more detailed discussion of how this work relates to previous theoretical treatments of triboelectrification, and for further insight into why our description is physically plausible and consistent with available data.

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