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Reversible and Continuously Tunable Control of Charge of Close Surfaces

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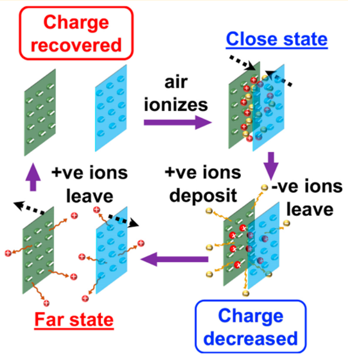
Supporting Information

ABSTRACT: Surfaces of almost all types of materials are often charged easily by contact electrification or deposition of ions; hence, surface charge is ubiquitous and has a vast range of influences in our lives and in industry. Since the 19th century, scientists have been measuring the charge of multiple materials collectively. The common expectation is that the total charge of multiple materials is equal to the sum of the charges of the individual materials. This study describes a previously unreported phenomenon in which the total charge of two insulating surfaces decreases when the surfaces are brought close to each other. The charge varies continuously and reversibly depending on the distance of separation between the surfaces. Experimental results derived from analyzing the movement of charge suggest that the changes are due to a rapid exchange of charge between the surfaces and their surrounding air. This change can be used to control the surface charge of the materials flexibly and reversibly.



Surfaces with a net charge are ubiquitous: almost all types of materials (e.g., metals, semiconductors, inorganic materials, and polymers) can be charged significantly and easily by different ways (e.g., contact electrification).[1] Charged surfaces have a wide range of applications and profound influences in many aspects of our lives. Traditionally, they have been used in applications such as electrophotography,[1] electrostatic filtration/precipitator, and electrostatic coating.[2] More recently, they have been used in many interesting applications, including energy generation (e.g., triboelectric nanogenerators), sensors,[3−5] manipulation of multiphase fluids in microfluidic systems,[6] electrical conductance modulation in heterogeneous nanostructured media,[7−9] actuators of nanomaterials,[10] spontaneous immobilization of liposomes on surfaces,[11] and water disinfection.[12] Besides useful applications, charged surfaces can have many different types of undesirable consequences. In industry, they can cause a reduction in the efficiency of many manufacturing processes.[2,13,14] In addition, excessive accumulation of charge on surfaces can result in electrostatic discharge. This discharge can cause damage to electronic components and costs the industry billions of dollars per year.[15] More severely, electrostatic discharge can cause explosion of flammable materials.[13] The interaction of charged surfaces of particles is also important for various natural phenomena such as lightning and dust or sand storms.[13,16−18] Because of these wide-ranging consequences, it is important to measure accurately and control the amount of charge on charged surfaces.

On the other hand, the investigation of the behaviors of charge on insulating surfaces can be challenging. For contact electrification (i.e., the generation of charge by the contact and separation of two solid surfaces), for example, the mechanism by which charge is generated on insulating surfaces is still incompletely understood. Different research groups have proposed that the charge species may be an electron,[1,19,20] an ion,[1,21] or a small quantity (e.g., nanoscopic) of charge material that transferred from one surface to another.[22,23] Another complication is that it is important to consider the dynamic interactions of multiple charge surfaces in proximity. Charging by contact electrification, for example, involves the contact between surfaces; hence, the charged surfaces are usually found to be close to one another during the charging process (e.g., charging by interaction of granular particles).

This study describes a previously unreported natural phenomenon in which charge on insulating surfaces can be changed continuously and reversibly depending on the distance of separation between the charged surfaces. In our experiments, we charged the surfaces either by depositing ions onto them (i.e., via corona discharge generated by applying stress on a piezoelectric material)[24] or contact electrification; these are common ways in which surfaces become charged. After charging, we used a Faraday cup to measure the charge of the materials. Charge on insulating surfaces is generally observed to be immobile; thus, it is possible to pattern charge on surfaces at the nano-scale.[25,26] As such, charge can be measured easily using many methods (e.g., a Faraday cup).

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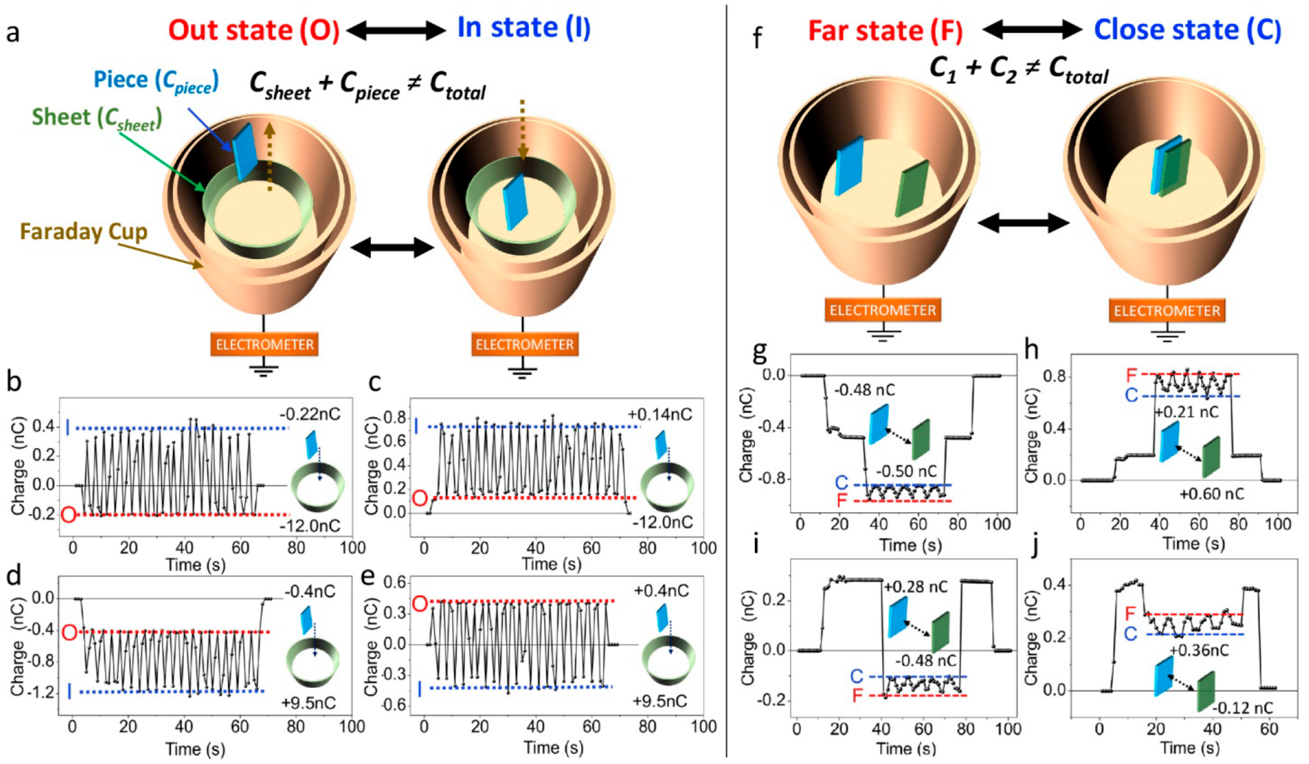

Figure 1. The total charge of two pieces of materials close to each other is different from the sum of their individual charges. (a) Scheme illustrating the experiment in which a small piece of charged material is inserted into a Faraday cup with a relatively much larger sheet of charged material at the bottom of the cup. The position of the small piece of material was varied repeatedly from above the large sheet at the "out" state (O) to within the large sheet at the "in" state (I). (b−e) Real-time measurements of the charge. Different polarities of a small piece of silicone rubber (blue) and a large sheet of Teflon (green) were used, as indicated by the insets. (f) Scheme illustrating the experiment in which the distance of separation between the two pieces of materials of the same size was changed repeatedly from the "far" state (F) to the "close" state (C). (g−j) Real-time measurements of the charge. Different polarities of the pieces of silicone rubber were used, as indicated by the insets.

Since the 19th century, scientists have routinely been measuring the total charge of multiple materials collectively.[2,27,28] The rationale of these collective measurements is the expectation that the total charge of multiple materials is equal to the sum of the charges of the individual materials.[29] We found, however, that this expectation is not always true: the charge of the insulating materials can change depending on the distance of separation between them.

In our experiments, we used a small piece of silicone rubber (1.7 × 1.2 cm and 1.0 mm thick) and a much larger hollow cylindrical sheet of polytetrafluoroethylene (Teflon; 4.0 × 15.5 cm and 1.0 mm thick) to demonstrate that their charges can change when they are brought close to each other. We first charged the materials (e.g., by either spraying ions from a Zerostat gun onto their surfaces or contact electrification) and measured their charges by a Faraday cup (see Experimental Methods in the Supporting Information and Figure S1a,b) connected to an electrometer. When a single piece of material was inserted into the Faraday cup, the charge of the material was approximately constant from a depth of around 2 cm or more; hence, this constant value represented the charge of the material (Figure S1h,i).

In order to investigate the charge of multiple materials, we first placed the hollow cylindrical sheet of Teflon at the bottom of the Faraday cup (Experimental Methods, Supporting Information and Figure 1a). We then set the reading of the electrometer to zero and measured the charge in real time. The silicone rubber was subsequently inserted at mainly two specific positions: at a depth of 2.5 cm (above the sheet of Teflon; "out" state) and 7.5 cm (surrounded by the sheet of Teflon; "in" state) inside of the Faraday cup.

In our first demonstration, we charged both the sheet of Teflon and the small piece of silicone rubber negatively (i.e., measured separately as −12.0 nC for Teflon and −0.22 nC for silicone rubber). At the "out" state, the charge was measured to be the charge of the piece of silicone rubber (−0.22 nC), as expected (Figure 1b). However, when we lowered it farther to the "in" state, the charge increased in the positive direction instead. This change in charge was significant: the change in charge in the positive direction was around 0.6 nC, which was much more than the charge of the silicone rubber. The process was reversible. Changing the position of the silicone rubber repeatedly between the "out" and "in" states resulted in a reversible change in charge. Similar changes in charge were observed for Teflon and silicone rubber charged to different polarities (Figure 1c−e). The charge measured at the "out" state was always approximately the charge of the silicone rubber; however, the charge at the "in" state was always significantly different than that of the "out" state. In general, the direction of change in charge from the "out" to the "in" state was always opposite to the polarity of the sheet of Teflon (e.g., in Figure 1b, charge increased in the *positive* direction from the "out" to "in" state while the polarity of Teflon was *negative*). Hence, one possible explanation is that the sheet of Teflon discharged from the "out" to "in" state. Because the sheet of Teflon had a much higher charge than the small piece of silicone rubber, the change in charge of Teflon dominated. The same phenomenon was observed for different types and

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combinations of materials, including inorganic materials (glass, zirconium dioxide, and alumina), polymers (low-density polyethylene and polytetrafluoroethylene), and another rubber (natural rubber) (Figures S2–S5). Different methods of handling the materials yielded similar results (Figure S6). On the other hand, numerical calculations showed that the charge should be approximately constant from the "out" to the "in" state according to conventional expectation (Figure S1c–e,j).

The same phenomenon can be observed when two pieces of charged materials of the same size are used (Figure 1f). We inserted two pieces of charged silicone rubber of the same size into the Faraday cup and varied the distance of separation between them from 3 cm ("far" state) to 2 mm ("close" state). Similarly, the results showed that the total charge was approximately equal to the sum of the charges of the two individual materials at the "far" state; however, the charges were different at the "close" state (Figure 1g–j). The total charge changed continuously with the distance of separation between the materials (see Figure S7 for a more detailed plot of the variation of charge with respect to the distance of separation). When both materials were of the same polarity, the magnitude of the total charge decreased from the "far" to "close" state, hence the pieces of silicone rubber discharged in the "close" state (Figure 1g,h). When the materials were of opposite polarity, the direction of change in charge from the "far" to "close" state seemed to correspond to the decrease in charge of the piece that had a higher amount of charge (Figure 1i,j). This phenomenon was also observed when other insulating materials were used (Figure S8) or when the experiment was conducted in different atmospheres (e.g., nitrogen or sulfur hexafluoride; see Figure S9). The change was reversible and could be repeated many times (Figure S10). Similarly, numerical calculations showed that the charge should be constant from the "far" to the "close" state (Figure S1f,g,k).

By the law of conservation of charge, if the total charge of the two materials changed, the difference in charge must have transferred elsewhere. One possibility is that charge might have moved in and out of the Faraday cup through the solid materials used to manipulate the charged materials (e.g., pairs of tweezers). However, the results were similar regardless of the type of material used (e.g., including highly insulating materials such as Teflon; see Figure S11).[30] In addition, the current through the solid materials was measured to be negligible (Figure S12).

Another possibility is air. In order to investigate if air is involved, we covered the Faraday cup partially with a piece of aluminum (i.e., a hole was made in the cover so that the charged materials can be handled from outside; see Experimental Methods, Supporting Information). In the first experiment, we inserted a small piece of Teflon into the partially closed Faraday cup with a large sheet of Teflon at its bottom. We then varied the position of the small piece repeatedly between the "out" and "in" states (Figure 2a). In the second experiment, we used two small pieces of Teflon and varied their distance of separation repeatedly between the "far" and "close" states (Figure 2b). Results showed that the difference in charge between the two states decreased significantly for both experiments compared to the case when the Faraday cup was fully opened. Hence, the amount of charged air molecules that moved in and out of the cup may have reduced by partially covering the Faraday cup.

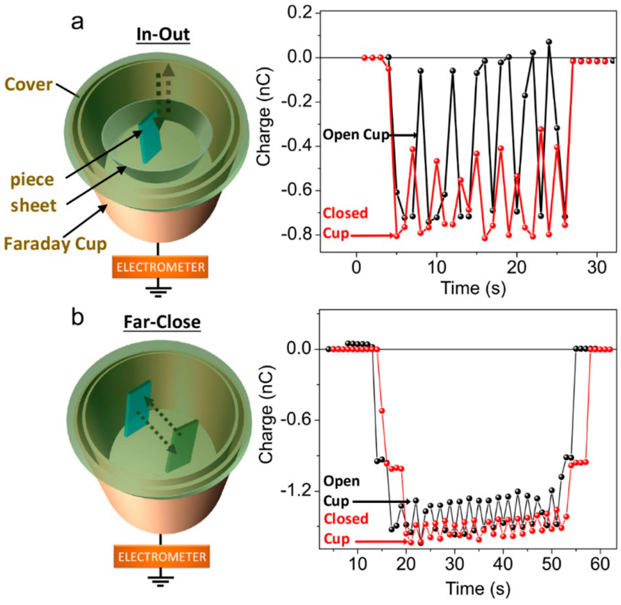

Figure 2. Investigating the involvement of air by covering the top of the Faraday cup. The total charge of the materials was measured when the top of the Faraday cup was either fully open (black) or partially closed (red). Changes in charge when the position of a small piece of Teflon was changed repeatedly (a) between the "out" and "in" states and (b) between the "far" and "close" states.

To understand the involvement of air, we calculated the electric field that surrounds the charged materials numerically (Figure 3a–c). Results showed that when the two materials are close to each other (i.e., separated by 2 mm), the electric field around various regions of the charged materials is higher than when only one material is present. This increase in electric field can lead to the ionization of gas molecules.[31] For example, when a piece of material is charged, it usually discharges naturally with time via the ionization of gas molecules.[32−35] In addition, a larger amount of charge of the material can lead to a higher electric field and a larger amount of discharge. This effect was observed experimentally: for two materials of the same polarity, a larger amount of charge resulted in a larger change (Figure 3d,e). For two materials of opposite polarity, however, they may discharge to similar extents; thus, the net charge did not change as much.

In general, we propose a mechanism for the change in total charge as follows. First, when the two materials are brought close to each other, the electric field surrounding the materials increases. This increase in electric field ionizes the air molecules and generates both positive and negative ions in air.[36] The gaseous ions with a polarity that is opposite to the charged surface are attracted to, and deposit onto, the surface; this deposition lowers the charge of the material. On the other hand, the gaseous ions with a polarity that is the same as the charged surface are repelled and move out of the Faraday cup. Once ions move out of the Faraday cup, the measurement of charge changes (Figure 4).

Subsequently, when the distance of separation between the materials is increased, the electric field around the materials decreases. This decrease in electric field may allow the gaseous ions that are deposited onto the surface to desorb; hence, the materials revert back to their original amounts of charge. We further showed experimentally that ions from air deposited onto the surface of a material can desorb rapidly in ∼1 s (Figure S13).

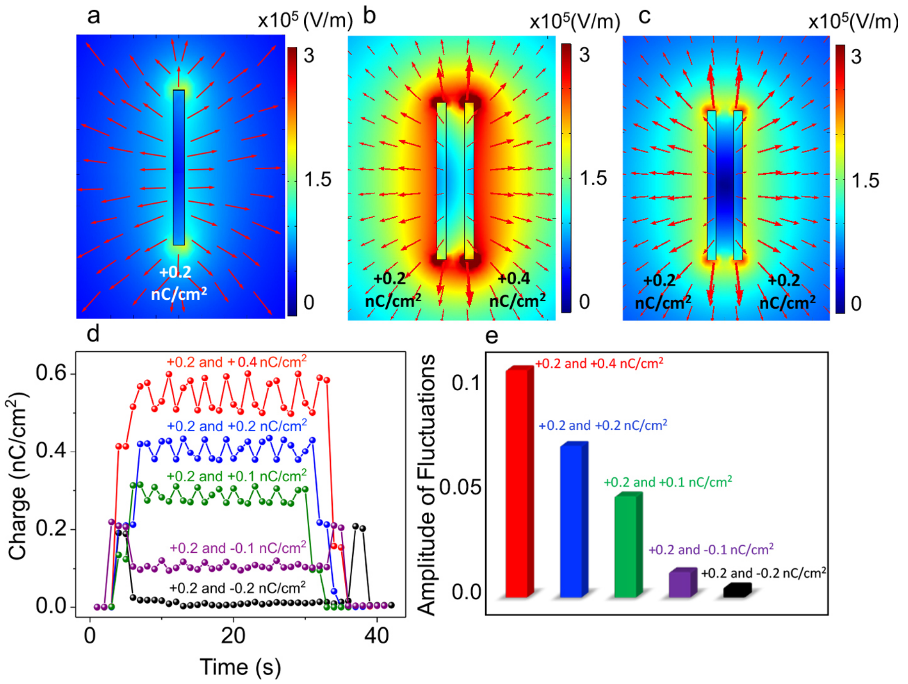

Figure 3. Electric field increased around the charged materials when the materials are close to each other. Numerical calculations of the electric fields for one (a) or two charged materials close to each other (b,c) of different amounts of charge. (d) Experimental results showing the changes in charge when two small pieces of materials of different amounts and polarities of charge were changed repeatedly between the "far" and "close" states. The amplitude of the changes (i.e., the average difference in charge between the "far" and "close" states) are plotted in (e).

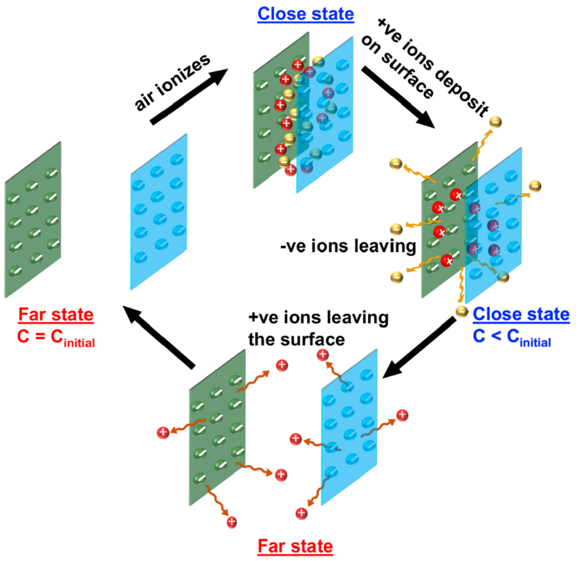

Figure 4. Scheme illustrating the proposed mechanism for the reversible change in charge of two initially negatively charged surfaces when the distance of separation between them is varied.

Because the total charge changes continuously with the distance of separation between the charged materials, we can control the charges of the materials flexibly. Controlling charge is important for the wide range of applications related to surface charge or for reducing the charge of materials when the presence of surface charge is undesirable.[1,15,37,38] For example, the adhesion of charged materials and particles onto surfaces due to attractive electrostatic forces is problematic for many manufacturing processes and in our daily lives (e.g., particles sticking onto screens of computers). As a demonstration, we charged a bead (polypropylene; 3.2 mm in diameter) positively and allowed it to stick onto the edge of a negatively charged piece of Teflon by their attractive electrostatic force (see the scheme in Figure 5a). Subsequently, we brought another two pieces of negatively charged Teflon gradually toward the piece of Teflon with the bead from two opposite sides (Experimental Methods, Supporting Information). Due to the proximity of the materials, the charge and the attractive electrostatic force decreased. When the pieces of Teflon were sufficiently close (~1.5 cm), the bead fell vertically downward (images in Figure 5a and Movie S1, Supporting Information). On the other hand, for the control experiment in which we brought two pieces of Teflon that were not charged toward the piece of Teflon with the bead, the bead remained stuck to the Teflon and did not fall.

An important consequence of surface charge is the electrostatic force: electrostatic force is necessary for many applications that rely on the surface charge of materials (e.g., in electrophotography, electrostatic precipitators, electrostatic spray painting, and manipulation of multiphase fluids in microfluidic systems).[6,13,39−41] We showed that because the charge decreases when the charged materials are close to each other, the electrostatic force is correspondingly smaller. First, we charged two pieces of the same material (i.e., silicone rubber, Teflon, natural rubber, or silicone foam) to the same polarity. We determined the amount of repulsive electrostatic force, F_{expt}, between them when they were close together (i.e., separated at a close distance of d_{sep}) using a weighing balance (Figure 5b and Experimental Methods, Supporting Information). We then measured the charges of the same materials in the Faraday cup at the "far" and "close" (i.e., at the same close distance of d_{sep}) states. Subsequently, we calculated numerically the repulsive force between the two materials when they were separated at d_{sep}. When we used the lower amount of charge measured experimentally at the "close" state, we found that the

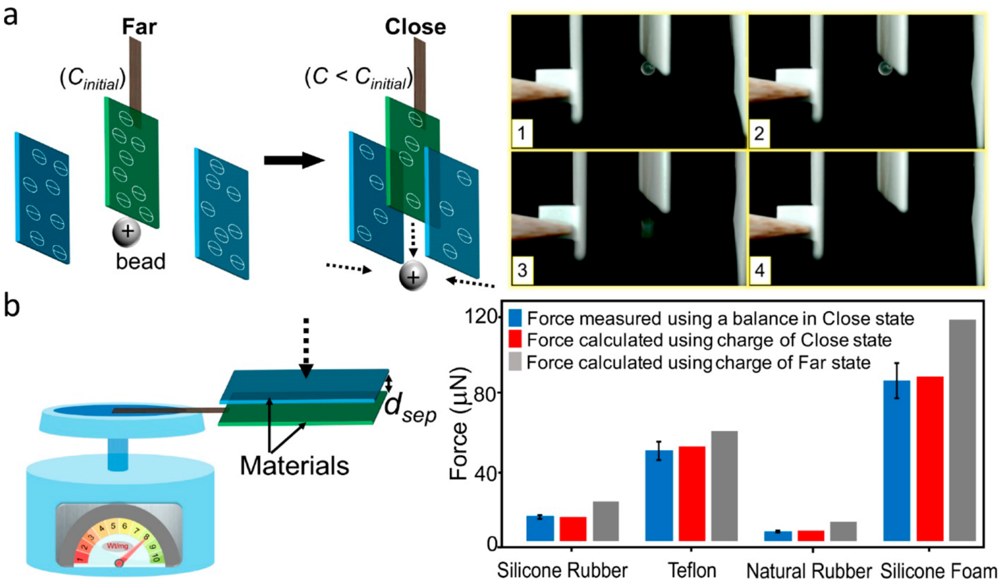

Figure 5. Consequences of the change in charge when the surfaces are close to each other. (a) Controlling the adhesion of a bead on a solid surface due to their attractive electrostatic force. (b) Controlling the electrostatic force between two charged surfaces due to the change in charge.

calculated force was approximately the same as F_{expt} (Figure 5b, plot on the right). When we used the higher amount of charge measured at the "far" state (i.e., the sum of charges of the two individual pieces), the calculated force was significantly larger than F_{expt}. Hence, the electrostatic force is smaller than commonly expected when the two pieces of charged materials are close to each other.

Since the 19th century, the expectation is that the total charge of multiple materials is equal to the sum of the charges of the individual materials. Therefore, the discovery that surface charge can change depending on the distance of separation between the surfaces is surprising. Our experimental results suggest that the change is due to a rapid exchange of charge between the charged surfaces and their surrounding atmosphere. The change in charge is reversible; the continuous range of "charge states" is readily accessible. This phenomenon can be used to control the charge of materials flexibly and reversibly. The change in charge affects the amount of electrostatic force between them. It is important to determine the amount of electrostatic force accurately for applications in which the electrostatic force is required. In terms of measurements, the materials need to be separated far apart in order to measure the charge of the individual material accurately.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jpclett.7b02763.

Experimental methods, measuring charge using a Faraday cup, in−out experiment in a Faraday cup experiment with several different materials, changes in charge when different combinations and polarities of the materials were used with the NR sheet, changes in charge when different combinations and polarities of the materials were used with the SR sheet, changes in charge when different combinations and polarities of the materials were used with the Teflon sheet, different methods of handling the materials between the "in" and "out" states, charge vs distance plot when two charged silicone rubber pieces moved toward each other from the far state to close state, far−close experiment in a Faraday cup with several different materials, changes in charge observed when the experiments were carried out in SF_6, N_2, and air atmospheres, changes in charge observed for many cycles of the far−close experiment, changes in charge observed when the charged materials were handled using different types of solid insulating materials, measuring the current through the solid materials used to handle the charged pieces, and experimental study showing ions desorbing rapidly from the charged materials into air (PDF)

Video showing that when the pieces of charged Teflon were sufficiently close the bead electrostatically attached to one Teflon fell vertically downward (MOV)

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Notes

The authors declare no competing financial interest.

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