

Flexible, Stretchable, Transparent Carbon Nanotube Thin Film Loudspeakers

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ABSTRACT

We found that very thin carbon nanotube films, once fed by sound frequency electric currents, could emit loud sounds. This phenomenon could be attributed to a thermoacoustic effect. The ultra small heat capacity per unit area of carbon nanotube thin films leads to a wide frequency response range and a high sound pressure level. On the basis of this finding, we made practical carbon nanotube thin film loudspeakers, which possess the merits of nanometer thickness and are transparent, flexible, stretchable, and magnet-free. Such a single-element thin film loudspeaker can be tailored into any shape and size, freestanding or on any insulating surfaces, which could open up new applications of and approaches to manufacturing loudspeakers and other acoustic devices.

Most of the loudspeakers used today consist at least of a cone, a voice coil attached to the apex of the cone, a permanent magnet fixed to the loudspeaker's frame, and an enclosure. By applying an audio current waveform to the voice coil, an audio frequency movement of the cone is produced due to the magnetic interaction between the voice coil and the magnet, thus reproducing the sound pressure waves. Recently we found that just a piece of carbon nanotube (CNT) thin film could be a practical magnet-free loudspeaker simply by applying an audio frequency current through it. This CNT thin film loudspeaker can generate sound with wide frequency range, high sound pressure level (SPL), and low total harmonic distortion (THD). The nanothickness CNT thin films we used are flexible, stretchable, and transparent and can be tailored into many shapes and sizes, freestanding or placed on a variety of rigid or flexible insulating surfaces. Furthermore, the CNT thin film loudspeaker has a very simple structure, without magnets and moving parts. Such a single-element thin film loudspeaker might open up new applications of and approaches to manufacturing loudspeakers and other acoustic devices.

The CNT thin film can be directly drawn out from CNT arrays.¹⁻⁶ Here we use superaligned CNT arrays on a 4 in. wafer to draw out continuous CNT thin films up to 10 cm

wide (Figure 1a).⁴⁻⁶ The CNT thin film is composed of CNTs around 10 nm in diameter, which are sparsely parallel-aligned in the drawing direction (Figure 1b). It is tens of nanometers thick, extremely lightweight (typical mass per unit area is 1.5 $\mu\text{g}/\text{cm}^2$), transparent (78% at 550 nm for a single-layer CNT thin film), and conductive (typical sheet resistance is around 1 k Ω per square for a single-layer CNT thin film). The ac impedance of a CNT thin film is pure resistance and shows no frequency dependence within 1 MHz. (See Supporting Information Figure S1.)

The as-drawn CNT thin film was directly put on two electrodes, forming a simple loudspeaker, as shown in Figure 1a. Several CNT thin films can be put together to make a large area loudspeaker. Figure 1c shows an A4 paper size loudspeaker. Furthermore, the CNT thin films can be tailored into some arbitrary shapes or put on some arbitrarily curved surfaces to make loudspeakers with special functions. Figure 1d shows the cylindrical cage-like CNT thin film loudspeaker, which can emit sound in all directions. Simply by applying a sinusoidal voltage across the two electrodes, one can hear clear and loud tones emitted from the CNT thin film.

Since our CNT thin film loudspeakers can produce sounds as loud as a commercial voice-coil loudspeaker, their audio performances can be directly tested by using a conventional audio analyzer. As shown in Figure 2a, we embedded the CNT thin film loudspeakers (dimensions 3 cm by 3 cm) in the central hole of a baffle plate. A sheet of sound absorption

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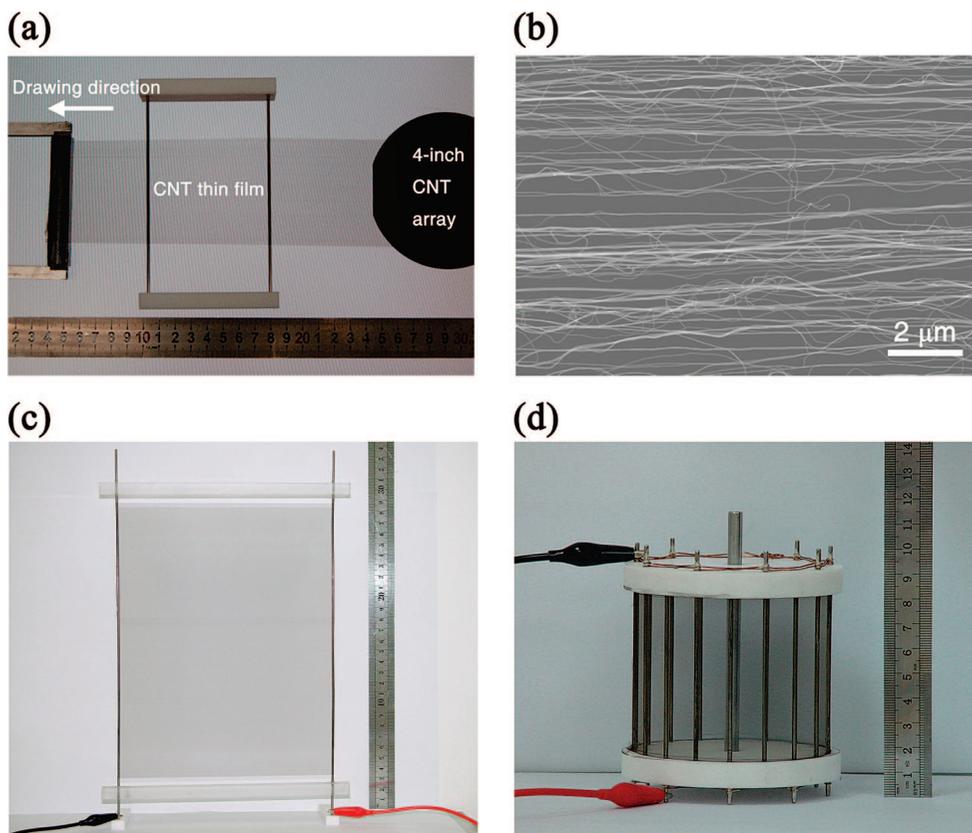


Figure 1. Carbon nanotube thin film loudspeakers. (a) The CNT thin film was pulled out from a superaligned CNT array grown on a 4 in. silicon wafer and put on two electrodes of a frame to make a loudspeaker. (b) SEM image of the CNT thin film showing that the CNTs are aligned in the drawing direction. (c) A4 paper size CNT thin film loudspeaker. (d) The cylindrical cage shape CNT thin film loudspeaker can emit sounds to all directions, diameter 9 cm, height 8.5 cm.

material was placed at the rear side of the loudspeaker to avoid reflection interference. A microphone (BK, model 4939) was put in front of the loudspeaker. The distance between the loudspeaker and microphone is 5 cm. The frequency response was measured by an audio analyzer (Audio Precision 2722).

We have made two kinds of CNT thin film loudspeakers for testing, i.e., one-layer and four-layer loudspeakers. The one-layer speaker was made by putting one CNT thin film on two electrodes. By stacking four layers of CNT thin films on the electrodes, we made the four-layer loudspeakers. It is easy to understand that the sheet resistance of the four-layer CNT loudspeaker is 4 times smaller than that of one-layer CNT loudspeaker.

Figure 2b shows the SPL versus frequency for one-layer (red) and four-layer (blue) CNT loudspeakers with the same input voltage (50 V_{rms}). Their input powers are 3 and 12 W, respectively. From Figure 2b, we can see that the SPL increases with increasing frequency. If we do linear fitting for the SPL curves, the result is $SPL \propto f^{0.7-0.8}$, in which f is the frequency. The total harmonic distortion is extremely small except for the low-frequency end, where the environment noise is severe. It is also found that the sound pressure is proportional to the input power, as is shown in Figure 2c. The more the power input, the higher the output sound pressure level. We have tested the maximum input power densities of our CNT loudspeakers, 5×10^4 W/m² for one-

layer and 1×10^5 W/m² for four-layer CNT loudspeakers. Since there is no limitation on their size, CNT loudspeakers with huge loudness can be practically realized.

Despite the excellent acoustic performance of the CNT loudspeaker, it has a drawback. As shown in Figure 2d, the output frequency doubles that of the input. The human voice and music sound strange when a commercial bipolar audio amplifier is used to drive the CNT thin film loudspeaker. A simple solution is to add a direct current bias I_0 to the alternating current for driving such CNT loudspeakers. To achieve this, we use a very simple single transistor amplifier to drive our CNT loudspeaker, which can reproduce the input sound wave signals faithfully. The schematic circuit is shown in Figure S2 of the Supporting Information. Note that the impedance of the CNT loudspeaker is pure resistance (see Supporting Information, Figure S1), and the design of the amplification circuit is much simpler than that for inductive voice-coil loudspeakers. Also the resistance can be tuned to any value to meet the requirement of driving circuits, simply by adjusting the position of the electrodes (e.g., the resistance of the loudspeaker shown in Figure 1c is 500 Ω, in Figure 1d it is 8 Ω).

With the self-made simple amplifier, the CNT thin film loudspeaker possesses all the functions of a voice-coil loudspeaker, as well as the merits of being magnet-free and without moving components. Besides these, there are

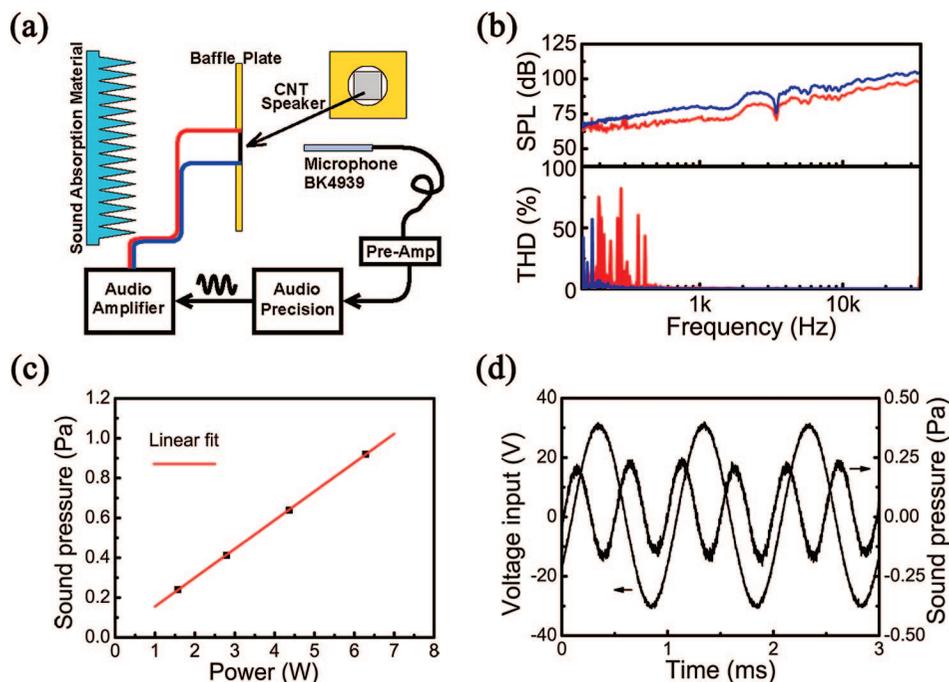


Figure 2. Testing of the acoustic performance of the CNT thin film loudspeaker. (a) Schematic illustration of the experimental setup. (b) Sound pressure level (in dB) and total harmonic distortion of the one-layer (red) and four-layer (blue) CNT loudspeaker for a distance of 5 cm between loudspeaker and microphone. The input power is 3 W and 12 W for one-layer and four-layer loudspeakers, respectively. (c) Sound pressure generated by the four-layer CNT loudspeaker versus the input power, showing a linear dependence. Black squares represent experimental results, and the red line is the fitting result. (d) Real time signals of the input voltage of the four-layer CNT thin film loudspeaker and the output sound pressure from the microphone, indicating that the frequency of the sound pressure doubles that of the input voltage. The phase shift between input voltage and output sound pressure signal is primarily due to the sound propagation from the CNT thin film to the microphone.

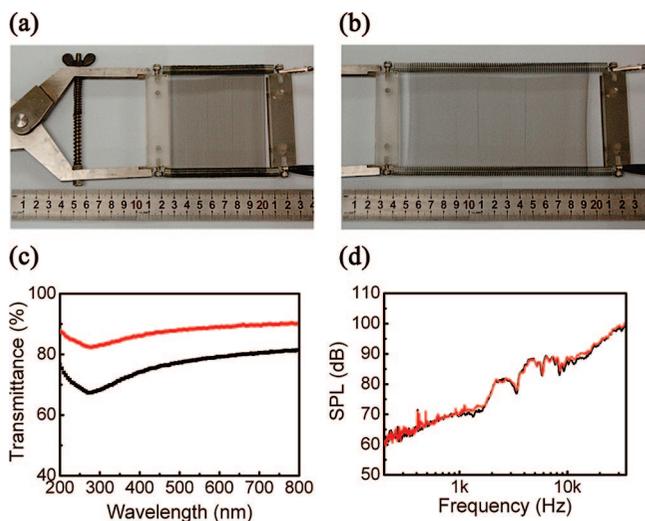


Figure 3. Stretchable CNT thin film loudspeaker. (a) A one-layer CNT thin film is put on two springs that serve also as electrodes. The CNTs are aligned perpendicular to the two spring electrodes. (b) The CNT thin film was stretched to 200% of its original width. (c) Transmittance spectra of the one-layer CNT thin film before (black) and after (red) being stretched to 200% of its original width, measured by using a UV-vis spectrophotometer (PGENERAL TU-1901). (d) Measured SPLs versus frequency for a one-layer CNT thin film before (black) and after (red) being stretched to 200% of its original width. Both input powers are 2.5 W.

some remarkable added values of the CNT thin film loudspeaker inherited from CNT materials.

The most striking property of the CNT thin film loudspeaker is that it is stretchable. Figure 3a shows that a CNT thin film is put on two springs that serve also as electrodes. The CNTs are aligned perpendicular to the two spring electrodes. When the springs are uniformly stretched, the CNT thin film is also uniformly stretched. Figure 3b shows that the CNT thin film loudspeaker was stretched to 200% of its original size and became more transparent than before stretching. From Figure 3c, we can see that the transmittances of the original (black curve) and 200% stretched (red curve) thin films are around 80% and 90%, respectively, for visible light. Movie S1 shows a CNT thin film loudspeaker being periodically stretched during singing. The sound intensity is almost unvaried during stretching. An audio analyzer also reports almost identical frequency responses (see Figure 3d) for the original (black curve) and 200% stretched (red curve) thin film loudspeakers with equal power inputs (2.5 W). The stretchable properties of CNT thin film loudspeakers might be used in future stretchable consumer electronics.⁷

The second unique property is that the CNT thin film loudspeaker is transparent. This property allows us to integrate the CNT thin film loudspeaker with an LCD module. Figure 4a shows a CNT thin film loudspeaker directly placed on the frame of a 17 in. LCD screen. The logo of Tsinghua University displayed on the LCD screen can be clearly seen when looking through the CNT thin film loudspeaker. Compared with the uncovered part, the part mounted with the loudspeaker is slightly darker. The

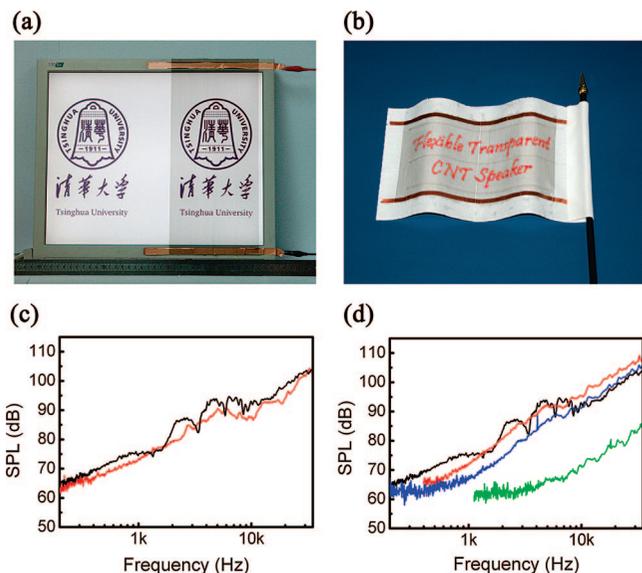


Figure 4. Transparent, flexible, CNT thin film loudspeaker. (a) A transparent CNT thin film loudspeaker is directly mounted on the frame of a 17 in. LCD screen. (b) The CNT thin film was put on a flag to make a flexible flag loudspeaker with dimensions of 8 cm by 14.5 cm. (c) Measured SPLs versus frequency for a one-layer CNT thin film that is either freestanding (black) or put on a piece of cloth (red). Both input powers are 4.5 W. (d) Measured SPLs versus frequency for a one-layer CNT thin film when it is freestanding (black), put on a ground glass (red), put on window glass (blue), or put on window glass with further ethanol treatment (green). The process of ethanol treatment is as follows: drop and spread some ethanol on the window glass covered by CNT thin films, then wait until the ethanol evaporates. All the input powers are 4.5 W.

corresponding transmittance is around 80% according to Figure 3c. Furthermore, by stretching and/or laser treatment, the transmittance can be improved to around 95%, which is almost equal to the transmittance of an uncoated optical glass.⁸ Movie S2 shows that a transparent CNT thin film loudspeaker is playing the songs of the movie played by the iPod beneath it. The CNT thin film can also be put on window glass, over paintings, etc., to make versatile transparent loudspeakers.

Another unique property is that the CNT thin film is flexible, and it can be tailored into many shapes and put onto a variety of rigid or flexible insulating surfaces. Figure 4b shows a single layer of CNT thin film put on a flag, which can act as a loudspeaker as it dances in the wind (see Movie S3). One can also imagine putting CNT thin films on clothes to make a singing and speaking jacket! The frequency response curves are shown in Figure 4c for a one-layer CNT thin film put on a piece of cloth (red) and freestanding (black), respectively. The SPL of the loudspeaker on a piece of cloth is slightly smaller than that of the freestanding loudspeaker. We have also made CNT thin film loudspeakers directly placed on window glass, ground glass, etc. The corresponding frequency response curves are shown in Figure 4d, which is smoother than that of a freestanding CNT loudspeaker (black). The sound generated by a CNT thin film on window glass (blue) is softer than that on ground glass (red). If some ethanol was dropped and spread on the

glass covered by CNT thin films, the CNTs will adhere to the glass tightly after the ethanol evaporates. But the ethanol-treated CNT thin film can generate only very weak sound. The corresponding frequency response curves are also shown in Figure 4d (green). The SPL is more than 20 dB less than that of untreated with the same power input (4.5 W).

Despite the excellent properties and versatile applications of the CNT thin film loudspeakers, we have to clarify the sound generation mechanism. The first idea that occurred to us is that the vibration of the thin film results in the sound generation. We have tried to use a laser vibrometer (Polytech PSV 300-F) to measure the vibration of the thin film, but failed to detect any vibration. Also we found that if our thin film was partially broken or put on some clothes, it can still generate proper sounds, which is impossible for a vibrating film. All these results indicate that the CNT thin film does not move during sound generation.

As is well known, sound is a pressure wave, and sound propagation is an adiabatic process. Thus pressure oscillation is always accompanied by temperature oscillation. On the other side, if we can excite a temperature oscillation, a pressure wave will also be excited. Such an acoustic effect induced by temperature oscillation is called “the thermoacoustic effect”, which has been studied for more than 200 years⁹ and led to the inventions of thermoacoustic engines and refrigerators in recent years.¹⁰

In case of the CNT thin film loudspeaker, the sound generation mechanism can be understood with the aid of a thermoacoustic picture. The alternating current periodically heated the CNT thin films, resulting in a temperature oscillation. The temperature oscillation of the thin film excites the pressure oscillation in the surrounding air, resulting in the sound generation (see the flash Movie S4). In this process, it is the thermal expansion and contraction of the air in the vicinity of the thin film that produces sound, not the mechanical movement of the thin film itself.

A direct consequence of this thermoacoustic mechanism is that the frequency of the output doubles that of the input. For an alternating current passing through, the CNT thin film will be heated during both positive and negative half-cycles, resulting in a double frequency temperature oscillation, as well as a double frequency sound pressure. Our experimental results shown in Figure 2d obviously agree well with this conclusion, thus providing support for the thermoacoustic mechanism.

Similar phenomena of thin metal wires or foils had been reported by Preece¹¹ and Braun et al.¹² at the end of 19th century, resulting in an invention called the “thermophone”.^{13–16} When an alternating current passes through the very thin metal foil, the thermophone will emit sound. But at that time, “the thermophone, listened to in the open air, sounds extremely weak”; no data of the acoustic performance of their thermophones in the open air were recorded.

However, Arnold and Crandall¹⁴ had proposed the correct physical picture for the thermophones: “When alternating current is passed through a thin conductor, periodic heating takes place in the conductor following the variations in the current strength. This periodic heating sets up temperature

waves which are propagated into the surrounding medium; the amplitude of the temperature wave falling off very rapidly as the distance from the conductor increases. On account of the rapid attenuation of these temperature waves, their net effect is to produce a periodic rise in temperature in a limited portion of the medium near the conductor, and thermal expansion and contraction of this layer of the medium determines the amplitude of the resulting sound waves.” On the basis of this picture, they derived the formula of the sound pressure produced by the thermophone:

$$p_{\text{rms}} = \frac{\sqrt{\alpha\rho_0}}{2\sqrt{\pi T_0}} \cdot \frac{1}{r} \cdot P_{\text{input}} \cdot \frac{\sqrt{f}}{C_s} \quad (1)$$

in which C_s is the heat capacity per unit area (HCPUA) of the thin film conductor, f is the frequency of sound, P_{input} is the input power, r is the distance between the thin film conductor and the microphone, ρ_0 , T_0 , and α are the density, temperature, and thermal diffusivity of the ambient gas, and p_{rms} is the root-mean-square sound pressure.

Equation 1 tells us that the sound pressure generated by the thermophone increases with increasing frequency and input power and decreasing HCPUA. In the case of frequency and input power, the agreement between Arnold and Crandall’s theory and our experiments is obvious if we look at the results shown in Figure 2b and c, respectively. For the HCPUA of the CNT thin film, it is the degree of the thermal contact with the substrate that determines its effective value. Better thermal contact means larger HCPUA. Thus the effective HCPUA of an ethanol-treated loudspeaker is the largest, and the HCPUA of the loudspeaker on ground glass is the least. According to eq 1, the loudspeaker on ground glass should produce the largest SPL, while the ethanol-treated loudspeaker should produce the smallest, which agrees qualitatively with our experimental results shown in Figure 4d.

To make a quantitative comparison, we have to know the HCPUA of our CNT thin film loudspeakers and that of the Pt thin film thermophone. The heat capacity of CVD grown CNT is 500 J/kg·K.¹⁷ One 4 in. wafer of superaligned CNTs (height of the CNT array is around 250 μm) weighs about $(6\text{--}7) \times 10^{-5}$ kg, which can be converted to a CNT thin film with an area around 500–600 times the area of the wafer. Thus the HCPUA of a one-layer CNT thin film is $(6.4\text{--}8.9) \times 10^{-3}$ J/m²·K. In the following calculations, we adopted a middle value of 7.7×10^{-3} J/m²·K for a one-layer CNT thin film speaker. For a four-layer CNT loudspeaker, it is easy to understand that the HCPUA is 4 times of that of a one-layer CNT loudspeaker. For Pt foil, we use $C_s = d\rho c_p$ to calculate the HCPUA. $\rho = 21450$ kg/m³, $c_p = 133$ J/kg·K,¹⁸ and d is the thickness of the Pt foil, which is 7×10^{-7} m according to ref 14. Then the HCPUA of the Pt foil is 2.0 J/m²·K, which is 260 times that of the one-layer CNT thin film. According to eq 1, the sound pressure generated by a one-layer CNT loudspeaker should be 260 times that generated by the 700 nm thick Pt film, corresponding to 48 dB difference in SPL. Now we can understand why the thermophones sound extremely weak,^{13–16} but our CNT loudspeakers are loud.

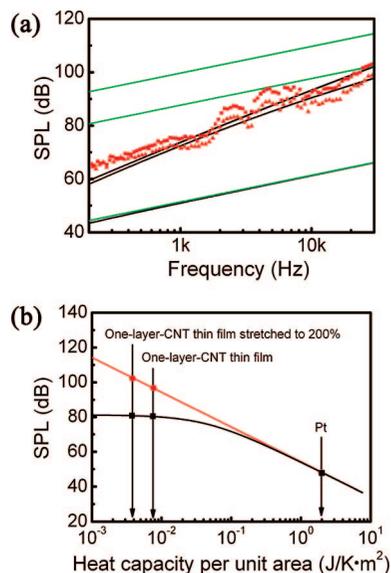


Figure 5. Theoretical and experimental data for the thermoacoustic thin film loudspeakers. (a) Theoretical and experimental results of the SPLs versus frequency of thermoacoustic thin film loudspeakers. The experimental data are represented by red solid squares and triangles for one-layer and four-layer CNT thin films, respectively. The green lines and black lines are SPLs calculated according to Arnold and Crandall’s theory (eq 1) and our theory (eq 2), respectively, for one-layer (upper) and four-layer (middle) CNT loudspeakers and a 700 nm thick Pt thermophone (lower). The input powers are all 4.5 W. (b) The dependence of SPLs (at 10 KHz with input power 1 W) on HCPUA C_s is calculated according to Arnold and Crandall’s theory (eq 1, red line) and our theory (eq 2, black line), respectively.

But there are discrepancies between Arnold and Crandall’s theory¹⁴ and our experimental results in the quantitative dependence of p_{rms} on f and C_s . The measured SPLs for one-layer and four-layer CNT thin film loudspeakers with input power of 4.5 W are shown in Figure 5a as red solid squares and triangles, respectively. The green lines are SPLs calculate according to eq 1 for one-layer (upper) and four-layer (middle) CNT loudspeakers and a 700 nm thick Pt thermophone (lower), respectively. The discrepancies between their theoretical and our experimental results lie in the absolute SPL value, the difference in the SPL value between one layer and four layers, and the slope of the SPL versus frequency. As shown in Figure 5a, the calculated SPLs for a one-layer CNT loudspeaker is 20–30 dB larger than the measured SPLs. One may argue that this discrepancy might due to the error in estimation of the HCPUA of the one-layer CNT thin film. But the difference in SPL value between one-layer and four-layer CNT thin films is $20 \log 4 \approx 12$ dB according to eq 1, which is independent of the absolute value of HCPUA. The experimental value is only around half of the theoretical prediction, as shown in Figure 5a. Also for a 200% stretched one-layer CNT thin film, their theory predicts a SPL value 6 dB larger than that of an unstretched thin film. But the experimental results shown in Figure 3d give almost identical SPLs with the same input power. Besides the aforementioned discrepancies, the slope of the theoretical SPL versus frequency is obviously smaller than that of the experimental one, as shown in Figure 5a. All these discrepancies imply

that some unknown factors have been omitted in Arnold and Crandall's theory.¹⁴

Since we believe that the thermoacoustic mechanism is correct, we adopted the same physical picture as Arnold and Crandall's to derive the theory. But in the derivation, we take into consideration the rate of heat loss per unit area of the thin film (due to conduction, convection, and radiation) per unit rise in temperature of the thin film above that of its surroundings, β_0 , and the instantaneous heat exchange per unit area between the thin film and its surrounding air due to thermal conduction, $Q_0 = -\kappa(\partial T(x,t))/(\partial x)_{x=0}$, in which κ is the thermal conductivity of ambient gas and $T(x,t)$ is the temperature distribution in the ambient gas. These two factors have both been omitted in Arnold and Crandall's derivation. The details of our derivation are included in the Supporting Information. The sound pressure generated by the thermoacoustic thin film loudspeaker should have the following form:

$$p_{\text{rms}} = \frac{\sqrt{\alpha}\rho_0}{2\sqrt{\pi}T_0} \cdot \frac{1}{r} \cdot P_{\text{input}} \cdot \frac{\sqrt{f}}{C_s} \cdot \frac{\frac{f}{f_2}}{\sqrt{\left(1 + \sqrt{\frac{f}{f_1}}\right)^2 + \left(\frac{f}{f_2} + \sqrt{\frac{f}{f_1}}\right)^2}} \quad (2)$$

in which $f_1 = (\alpha\beta_0^2)/(\pi\kappa^2)$ and $f_2 = (\beta_0)/(\pi C_s)$. Equation 2 is almost the same as eq 1, except for the last term. When $ff_2 \gg \sqrt{f/f_1}$, i.e., $f \gg (\kappa^2)/(\pi\alpha C_s^2)$, eq 2 turns into eq 1. For a 700 nm thick Pt foil, this condition is $f \gg 2.45$ Hz, while for a one-layer CNT thin film, $f \gg 1.65$ MHz. That is to say, within the audible range 20 Hz to 20 kHz, eq 1 is a good approximation of eq 2 for the 700 nm thick Pt foil. But for a one-layer CNT thin film, we have to use eq 2 to calculate the SPLs.

The black lines in Figure 5a represent our calculated SPLs according to eq 2 for one-layer (upper) and four-layer (middle) CNT thin films and a 700 nm thick Pt foil (lower), respectively. It is clear that calculations according to eqs 1 and 2 give almost identical SPLs for a 700 nm thick Pt foil, as we expected. But our calculations according to eq 2 for one-layer and four-layer CNTs thin films give rise to better agreement with experimental results, as compared to Arnold and Crandall's theoretical results (see Figure 5a). The dependence of SPLs (at 10 KHz with input power 1 W) on HCPUA C_s is calculated according to eqs 1 and 2 and plotted in Figure 5b as black and red lines, respectively. For large HCPUA C_s such as that of a 700 nm thick Pt foil, both theories give identical SPLs. But our theory predicts a plateau in small HCPUA C_s where the HCPUA of stretched and unstretched one-layer CNT thin films is located. This prediction agrees very well with our experimental results shown in Figure 3d. Thus eq 2 gives a more accurate description of the thermoacoustic thin film loudspeakers or thermophones than eq 1.

Now looking back to the 1920s, Arnold and Crandall have already proposed the criteria for a good thermophone: "the conductor be very thin; its heat capacity must be small, and it must be able to conduct at once to its surface the heat produced in its interior",¹⁴ which is just a vivid description

of our CNT thin films. Unfortunately they could not obtain such materials at that time. The only applications demonstrated are precision source of sound¹⁴ and minimum intensity for audition,¹⁶ which have been ignored for more than 80 years due to the very weak effect. With the help of our CNT thin films, the thermoacoustic mechanism can be used to build not only very small sound intensity instruments, but also loudspeaker with arbitrary loudness and many unique properties such as being flexible, stretchable, transparent, etc.

To summarize, we reported a kind of thermoacoustic loudspeaker made by CNT thin films, which has a very simple structure and a very easy fabrication process and are ready to be mass-produced. One superaligned CNT array grown on a 4 in. silicon wafer can be totally converted to a continuous CNT thin film up to 10 cm wide and 60 m long, which can be further made into approximately 500 loudspeakers with a size of 10 cm by 10 cm! These CNT thin film loudspeakers are transparent, flexible, and stretchable, which can be tailored into many shapes and mounted on a variety of insulating surfaces, such as room walls, ceilings, pillars, windows, flags, and clothes without area limitations. Furthermore, CNT thin films can also be made into small area devices, such as earphones and buzzers. There is no doubt that more and more applications will be developed as time goes on. This technique might open up new applications of and approaches to manufacturing loudspeakers and other acoustic devices.

In principle, other thin films of CNTs, such as solution-based random networks of CNTs,¹⁸ might also be a practical loudspeaker. The challenges might be how to make them nanometer thick, how to make them freestanding, and how to make them into any shape and any size, as compared to the simple method of dry drawing from superaligned CNT arrays we used here.

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Supporting Information Available: PDF file containing supporting online figures, derivation of the theory for thermoacoustic thin film loudspeakers, and tables containing the constants used in numerical calculations; a movie showing a singing CNT thin film loudspeaker that is being periodically stretched; a movie showing that a transparent CNT thin film loudspeaker is playing the songs of the movie played by the iPod beneath it; movie showing a single layer of CNT thin film put on a flag forming a flexible flag loudspeaker; an AVI animation illustrating the principle of thermoacoustic thin film loudspeakers. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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