We report a negative resistance, namely, a voltage drop along the opposite direction of a current flow, in the superconducting gap of NbSe\(_2\) thin films under the irradiation of surface acoustic waves (SAWs). The amplitude of the negative resistance becomes larger by increasing the SAW power and decreasing temperature. As one possible scenario, we propose that soliton-antisoliton pairs in the charge density wave of NbSe\(_2\) modulated by the SAW serve as a time-dependent capacitance in the superconducting state, leading to the dc negative resistance. The present experimental result would provide a previously unexplored way to examine nonequilibrium manipulation of the superconductivity.

**RESULTS**

\(2H\)-NbSe\(_2\) is a typical transition metal dichalcogenide showing the superconductivity below \(T_C = 7.2\) K. By using the mechanical...
exfoliation technique, NbSe₂ thin films were fabricated on a LiNbO₃ substrate, which is a strong piezoelectric material. The NbSe₂ film selected in the present work is about 30 nm in thickness, which corresponds to 25 unit cells. To induce SAWs along the x direction shown in Fig. 1A and to apply them to the NbSe₂ thin film on the LiNbO₃ substrate, we placed two opposite interdigital transducers (IDTs) so that the selected thin film was at the center of the two IDTs, as shown in Fig. 1B. The electrodes and the IDTs were prepared at the same time using electron beam lithography and depositions of Ti (60 nm) and Au (40 nm) in a vacuum chamber. Each IDT consists of 10 pairs of comb-shaped electrodes. The wavelength and the resonance frequency (f₀) for the designed IDTs are 1.0 μm and 3.25 GHz, respectively. The latter has been determined from the scattering parameter measurement with a network analyzer. The amplitude of the induced SAW at f₀ can be estimated at about 0.3 Å when the radio frequency (rf) power (or SAW power) applied to the IDTs is 30 μW (23). As reference samples, we also prepared 30-nm-thick 2H-NbSe₂ and Nb thin films on other LiNbO₃ substrates.

Figure 1C shows the temperature dependence of resistance, measured with the standard ac lock-in technique, for a typical NbSe₂ thin film on the LiNbO₃ substrate. A broad kink due to the CDW transition and a sharp superconducting transition can be seen at 33 and 7.2 K, respectively, as well established for bulk NbSe₂ (24, 25). A similar NbSe₂ thin-film device also shows a superconducting transition at 4 K (see Fig. 1C). In contrast, it has no CDW phase (25), which is supported by the observed featureless temperature dependence of resistance.

In Figs. 2 (A and B), we show dc current-voltage (I-V) properties of the NbSe₂ film exposed to the SAW sent from the left to right IDTs (see Fig. 1B) with different rf powers. The standard I-V curve is obtained except for in the vicinity of zero current. The critical current I_c for this device is about 0.8 mA, which is almost independent of the rf power (up to 30 μW). This shows that the superconducting state of the NbSe₂ device is robust for the irradiation of SAW. The shape of the differential resistance dV/dI just above I_c slightly changes by increasing the current, as depicted in Fig. 2C. This would be caused by the heating effect of the device.

An unexpected property emerges in the I-V curve within ±50 μA (see Fig. 2B). When the rf power is as small as 1 μW, zero voltage is detected within the critical current. Unexpectedly, however, a finite voltage appears at ±15 μA, and the voltage amplitude increases by increasing the rf power. Because the negative (positive) voltage is induced at + (−)15 μA, the slope, namely, resistance at I = 0, is negative. This fact indicates that the induced dc voltage drop is opposite to the current direction. To see the negative resistance more clearly, the differential resistance dV/dI numerically obtained from the dc I-V curve is plotted in Fig. 2D as a function of bias current measured at 1.6 K. (B) Close-up of the I-V curve shown in (A) near zero bias current. (C) Differential resistance obtained by numerically differentiating the I-V curve shown in (A). (D) Close-up of the dV/dI curve shown in (C) near zero bias current. (E) dc voltage of the NbSe₂ thin film exposed to the SAW with different rf powers as a function of bias current measured at 1.6 K. (F) Close-up of the I-V curve shown in (E) near zero bias current. (G) Differential resistance obtained by numerically differentiating the I-V curve shown in (E). (H) Close-up of the dV/dI curve shown in (G) near zero bias current.

DISCUSSION
What is the origin of the negative resistance in the superconducting gap observed only in the NbSe₂ devices? The negative resistance takes place without the external magnetic field. Thus, we can exclude microwave-induced resistance oscillations (3–8) mentioned
in Introduction. Recently, a negative local resistance has been reported in graphene, where the high viscosity of electronic states is essential (26), but this high viscosity cannot be expected in the NbSe$_2$ devices because the electron density in NbSe$_2$ is much higher than that in graphene. Although it is known that a very large contact resistance may induce a seeming negative resistance (27), this possibility can be ruled out safely from the following two reasons: first, the negative resistance is observed only when the SAW is applied to the NbSe$_2$ device, and is never observed for the NbS$_2$ device that has the same contact resistance (~100 ohms) as the NbSe$_2$ one; second, the I-V characteristic above $T_C$ is perfectly ohmic. We can also exclude the possibility that the sufficiently inhomogeneous current distribution (most probably due to the CDW phase as detailed in the next paragraph) in the NbSe$_2$ devices might induce a negative resistance. This is because the negative resistance does not appear just below $T_C$ where the coexistence of the CDW and the superconductivity is already realized but appears only far below $T_C$ (see Fig. 3A). This fact provides assurance that the current density is relatively homogeneous in the NbSe$_2$ devices. The emergence of a similar negative resistance was already reported in Josephson junctions (28–30). In (28) and (29), excess quasiparticles excited outside the superconducting gap tunnel opposite to the current bias direction due to the asymmetric profile of the density of states, leading to a negative resistance. In that case, however, the zero resistance state is not recovered again at finite bias regions, which is different from the present case (see Fig. 2 and fig. S5D). In (30), a negative resistance state was obtained only when the amplitude of rf current was much larger than the dc critical current of the Josephson junction. This situation is also different from the present setup, where the SAW power is small enough not to affect $I_C$, $T_C$, and $H_C$ of NbSe$_2$ thin films, as demonstrated in Figs. 2 and 3.

Therefore, it is reasonable to attribute the present negative resistance to cooperative interactions between the superconducting state and the CDW modulated by the SAW. At the moment, we do not have a conclusive model to fully explain our experimental results, but one possible scenario is that soliton-antisoliton pairs in the CDW phase (31–33) are generated by irradiating the SAW and they form local capacitances in the superconducting domains. In the CDW phase of NbSe$_2$ (below 33 K), selenium atoms have a periodic modulation that is three times the lattice constant for selenium atoms (24, 34, 35). When the SAW is irradiated to the NbSe$_2$ device, all the selenium atoms would be shifted from the commensurate position of the CDW, and thus, the phase shift of the CDW order parameter would be modulated over the wavelength of the SAW (=1 µm), as shown in the upper panel of Fig. 4A. This displacement increases the electrostatic energy and the elastic energy. Thus, it is energetically more favorable to nucleate soliton-antisoliton pairs and to induce a phase difference $\Delta \varphi$ of $2\pi$ in between the soliton-antisoliton pairs (31–33, 36), as illustrated in the lower panel of Fig. 4A. These soliton-antisoliton pairs have been intensively studied in quasi-1D CDW systems (31, 32) and also discussed even in 2D CDW compounds (33). Very recently, it has been revealed that there are several types of domains with a size of about 10 nm in the equilibrium NbSe$_2$ (35). This is an additional supportive observation for the soliton-antisoliton pairs in the NbSe$_2$ film. In addition to the complex CDW order, the superconductivity starts to develop below 7.2 K. Here, we assume the following two situations: (i) a superconducting domain described by a macroscopic wave function grows on each CDW domain with a size of ~10 nm [see (35)], and (ii) boundaries of the superconducting domains randomly distributed in the NbSe$_2$ film serve as weak junctions.

While the CDW opens an energy gap at the Fermi energy, it is not a full-gap state for NbSe$_2$ (37), and thus, the charge accumulation due to the soliton-antisoliton pair creation is immediately dissolved. On the other hand, the superconducting gap fully opens at the Fermi energy below $T_C$. Therefore, the charge accumulation is expected to survive longer in the superconducting state so that it can be regarded as a temporal and local capacitance. As for the superconducting part, the shape of the I-V curve near $I_C$ is typical of an overdamped Josephson junction because there is no hysteresis near $I_C$ (9). This fact indicates the existence of weakly coupled superconducting domains in the NbSe$_2$ thin film, which is also consistent with the above assumption that the superconducting parts developed on different CDW domains (35) are weakly coupled.

On the basis of the above experimental facts and ideas, we have developed a capacitively coupled Josephson junction model (see the Supplementary Materials for more details) and calculated an I-V property of the circuit shown in the upper panel of Fig. 4B. In this calculation, we assume that the time ($t$)-dependent local capacitance $C(t)$ between the soliton-antisoliton pairs is modulated by the same frequency as the SAW to have a sawtooth wave as shown in the lower panel of Fig. 4B. A typical I-V curve based on this model is displayed in Fig. 4C (see the Supplementary Materials for derivation). We qualitatively reproduce a negative resistance in the superconducting gap; the negative slope appears at zero current in the superconducting gap.

On the other hand, there is a difference between the experimental result and the theoretical calculation; in most cases (see Fig. 2B and
capacitively coupled Josephson junction under the SAW irradiation, we have qualitatively reproduced the experimentally measured negative resistance. Such a negative resistance state could be a promising stage to demonstrate Floquet engineering (39) in the superconducting state where quantum systems can be driven by the SAW.

MATERIALS AND METHODS

2H-NbSe$_2$ and 2H-NbS$_2$ thin films have been obtained using the mechanical exfoliation technique using Scotch tape. Because these thin films are sensitive to ambient air, the exfoliation process has been performed in a glove box filled with Ar gas of purity 99.9999%. Some of the exfoliated NbSe$_2$ (or NbS$_2$) flakes were transferred from the Scotch tape to a LiNbO$_3$ substrate. We then spin-coated polymethyl methacrylate resist onto the substrate for electron beam lithography and also for protecting the thin films from degradation. The substrate was taken out from the glove box, and electrode patterns including comb-shaped electrodes for irradiation of SAWs were deposited using electron beam deposition in a vacuum chamber next to the glove box. The NbSe$_2$ and NbS$_2$ thin-film SAW devices can be obtained after the lift-off process in acetone. For a reference sample, we also prepared Nb thin-film SAW devices using magnetron sputtering.

Low-temperature transport measurements of the SAW devices were performed using a cryogen-free superconducting magnet system (Cryomagnetics, C-Mag Vari-9) down to $T = 1.6$ K and up to $B = 9$ T. In dc measurements, the bias current was generated by using a voltage source (Yokogawa, 7651) and a large resistance (~1 megohm). The obtained dc voltage was measured with a digital multimeter (Keithley, 2000). We also performed ac measurements with a frequency of 173 Hz using a lock-in amplifier (Stanford Research System, SR830) to obtain a differential resistance. The fundamental properties of the IDTs were confirmed by measuring $S$-parameters using a network analyzer (Keysight, E5071C). The SAWs were induced by a microwave generated from a signal generator (Agilent, N5171B).

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/34/eaba1377/DC1

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