## **Supporting Information**

## Acoustic Metamaterials for Remote Manipulation of Large Objects in

### Water

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#### Note S1: Acoustic properties measurement and calculation

Metal-resin composite samples (4 cm × 4 cm ×1 cm) were fabricated for measuring the acoustic properties. The measurement system is illustrated in Fig. S2. Two used frequencies: 1 MHz and 3 MHz, are measured. A sample is placed in between a transmitter (V303-SU from Olympus Inc. for 1 MHz measurement; TQ20-1030 from Siansonic Inc. for 3 MHz measurement) and a receiver (V303-SU, Olympus Inc.). A pulsed signal with a center frequency of 1 MHz is generated by the function generator. An example of the transmitted signal is shown in Fig. S3(b). The received signals are analyzed via oscilloscope and MATLAB.

The acoustic speed of samples with different mass ratios,  $c_{\text{sample}}$ , are calculated from the signal

arriving times with  $(t_1)$  and without  $(t_0)$  the sample:  $c_{\text{sample}} = \frac{w}{w/c_{water} - (t_0 - t_1)}$ . Here, w = 0.01 m, which is the thickness of the sample. Acoustic impedance, Z, is obtained by the product of the density  $\rho$  and the acoustic speed c:  $Z = \rho c$ . The reflection coefficient R is calculated by:  $p = \frac{\rho_2 c_2 - \rho_1 c_1}{\rho_2 c_2 - \rho_1 c_1}$ 

 $R = \frac{\rho_2 c_2 - \rho_1 c_1}{\rho_2 c_2 + \rho_1 c_1},$  where and  $c_1$  are the density and acoustic wave speed of transmission media and  $\rho_2$ ,  $c_2$  is the density and acoustic wave speed of reflecting media. In the calculation, the density of water  $\rho_{\text{water}}$  is set as 997 kg·m<sup>-3</sup> and acoustic speed  $c_{\text{water}}$  is set as 1500 m·s<sup>-1</sup>. The calculated acoustic properties are shown in Fig. 1(b, c).

#### Note S2: The z-direction acoustic radiation force measurement

As illustrated in Fig. S5(a), a metamaterial patch is attached to the bottom of a 3D printed hollow cylinder. The object floats on the surface of the water at a position with 30 mm distance to the transducer. Its *xy*-plane movement is limited by a ring-shaped structure (Fig. S4). When the acoustic beam in turned on, the acoustic radiation force will push the object upward. By measuring the z-direction displacement  $\Delta z$ , magnitude of the z-direction radiation force can be calculated according to  $F_{rz} = \rho_{water} \Delta V_z \cdot g$ , where  $g = 9.8 \text{ N} \cdot \text{kg}^{-1}$ ,  $\rho_{water} = 10^3$ kg·m<sup>-3</sup>, and  $\Delta V_z$  is the immersion volume change  $\pi R_s^2 \Delta z$ , where  $R_s$  is the radius of the cylindrical object. For the results shown in Fig. 3(a) and Fig. S5(b),  $R_s$  is 12.5 mm and the total weight of a metamaterial-attached cylinder is around 8 g.

We measured the acoustic radiation force generated by a metamaterial patch with  $\alpha = 0^{\circ}$  when the input power of the transducer varies. At each input power, the measurement was repeated by 20 times. It can be viewed from Figure S5b that the *z*-direction acoustic radiation force is linearly related to the input power of the transducer.

#### Note S3: Simulation of acoustic field distribution

COMSOL Multiphysics 6.0 is used to simulate the acoustic field distribution after reflection of from metamaterial patches with the pushing function. The simulation is performed in 2D in frequency domain. As shown in Fig. S7, the metamaterial patch's width in the *x*-direction is set as 15 mm (about 10 times and 30 times of the wavelengths at 1 MHz and 3 MHz, respectively). The surface of metamaterial is set as a Sound Hard Boundary and Perfect Matching Layers are placed at the boundaries of the 80 mm × 120 mm simulation area. An acoustic wave source is placed at a distance z = 37.5 mm away from the metamaterial patch, transmitting a *z*-direction acoustic field. The acoustic pressure and acoustic velocity along the black circle around the metamaterial patch are then exported from COMSOL for the acoustic radiation force calculation using equation (8). More details of acoustic radiation force calculation are in Note S4.

## Note S4: Acoustic radiation force calculation using simulated acoustic pressure and acoustic velocity

The perturbation theory is used to calculate the generated radiation force by placing an object into an acoustic field: <sup>1,2</sup>

 $p = p_0 + p_1 + p_2$  (S1a)  $\rho = \rho_0 + \rho_1 + \rho_2$  (S1b)  $u = \vec{0} + \vec{u_1} + \vec{u_2}$  (S1c)

Here  $p_0$  and  $\rho_0$  are the static pressure and density;  $p_1$ ,  $\rho_1$ , and  $\vec{u_1}$  are the acoustic pressure, pressure induced density change, and acoustic velocity;  $p_2$ ,  $\rho_2$ , and  $\vec{u_2}$  are the second order non-linear components. We also have  $p_1=c^2\rho_1$ .

The pressure *p*, density  $\rho$ , and velocity  $\vec{u}$  follows the following equations:  $p = p(\rho)$  (S2a)  $\partial_t \rho = -\nabla \cdot (\rho \vec{u})$  (S2b)  $\rho \partial_t \vec{u} = -\nabla p - \rho(\vec{u} \cdot \nabla)\vec{u} + \eta \nabla^2 \vec{u} + \beta \eta \nabla (\nabla \cdot \vec{u})$  (S2c)

The (S2a) is the thermodynamic equation, (S2b) is the kinematic continuity equation, and (S2c) is the dynamic Navier–Stokes equation. In (S2c),  $\eta$  is the dynamic viscosity of the fluid and  $\beta$  the viscosity ratio.

The first order perturbation of (S2b) and (S2c) is:  $\partial_t \rho_1 = -\rho_0 \nabla \vec{u_1}$  (S3a)  $\rho_0 \partial_t \vec{u_1} = -c^2 \nabla \rho_1 + \eta \nabla^2 \vec{u_1} + \beta \eta \nabla (\nabla \cdot \vec{u_1})$  (S3b)

If the viscosity of the liquid is small and can be neglected, (S3b) becomes  $\rho_0 \partial_t \vec{u_1} = -c^2 \nabla \rho_1$ 

The second order perturbation of (S2b) and (S2c) (neglecting the viscosity terms) is:  $\partial_t \rho_2 = -\rho_0 \nabla \vec{u_2} - \nabla \cdot (\rho_1 \vec{u_1})$  (S4a)  $\rho_0 \partial_t \vec{u_2} = \nabla p_2 - \rho_1 \partial_t \vec{u_1} - \rho_0 (\vec{u_1} \cdot \nabla) \vec{u_1}$  (S4b)

The acoustic radiation force is a time-average effect that does not resolve the oscillatory details in the acoustic field. A time average over a full oscillation period  $\tau$  can be defined as:

$$\langle X \rangle \equiv \frac{1}{\tau} \int_{0}^{t} dt X(t)$$
 (S5)

It is noted that the physical, real-valued time average  $\langle f g \rangle$  of two harmonically varying fields

*f* and *g* is given by the real-part rule  $\langle f g \rangle = \frac{1}{2} Re[f g^*]$ , where the asterisk denotes complex conjugation. In addition,  $\langle \rho_0 \partial_t \vec{u_2} \rangle = 0$ .

The time average of (S4b) can be expressed as:

$$-\nabla \langle p_2 \rangle = \langle \rho_1 \partial_t \vec{u_1} \rangle + \rho_0 \langle (\vec{u} \cdot \nabla) \vec{u_1} \rangle \tag{S6}$$

From  $\rho_0 \partial_t \vec{u_1} = -c^2 \nabla \rho_1$  (S3b), we have  $\rho_1 \partial_t \vec{u_1} = \rho_1 (-\frac{c^2}{\rho_0}) \nabla \rho_1$  (S7)

Inserting (S7) into (S6) gives:

$$-\nabla \langle p_2 \rangle = \left\langle \rho_1 (-\frac{c^2}{\rho_0}) \nabla \rho_1 \right\rangle + \rho_0 \langle (\vec{u} \cdot \nabla) \vec{u_1} \rangle \tag{S8}$$

Inserting  $p_1 = c^2 \rho_1$  into (S8), we have:

$$-\nabla\langle p_2 \rangle = \left\langle -\frac{1}{c^2 \rho_0} p_1 \nabla p_1 \right\rangle + \rho_0 \langle (\vec{u_1} \cdot \nabla) \vec{u_1} \rangle \tag{S9}$$

Further mathematical simplification of (S9) gives:

$$\langle p_2 \rangle = \frac{1}{2c^2 \rho_0} \langle p_1^2 \rangle - \frac{1}{2} \rho_0 \langle \vec{u_1}^2 \rangle$$
 (S10)

The force on the object is the integration of the time-averaged pressure  $\langle p_2 \rangle$  and the momentum flux tensor  $\rho_0 \langle \vec{u_1} \, \vec{u_1} \rangle$  over a boundary enclosing the object:

$$\vec{F}_{r} = -\oint \{\langle p_{2} \rangle \vec{n} + \rho_{0} \langle (\vec{n} \cdot \vec{u_{1}}) \vec{u_{1}} \rangle \} da = -\oint \{ \left[ \frac{1}{2} \kappa_{0} \langle p_{1}^{2} \rangle - \frac{1}{2} \rho_{0} \langle \vec{u_{1}}^{2} \rangle \right] \vec{n} + \rho_{0} \langle (\vec{n} \cdot \vec{u_{1}}) \vec{u_{1}} \rangle \} da$$
(S11)

Here,  $\kappa_0$  is the compressibility  $\kappa_0 = 1/(\rho_0 c^2)$ ,  $\rho_0$  is the density of liquid, and *c* is the acoustic wave speed in the liquid. The acoustic pressure  $p_1$  and acoustic velocity  $\vec{u_1}$  are given by the COMSOL simulation described in Note S3.

#### References

- 1 M. Settnes and H. Bruus, *Phys. Rev. E*, 2012, **85**, 016327.
- 2H. Bruus, Lab. Chip, 2012, 12, 1014.

# **Table S1.** The details of metamaterial patches, objects, and transducer input power in theexperiments in Movie S1-S3.

Video No.	Experiment name	Metamaterial pattern	Patch size (mm)/shape	Object material (density (kg/m <sup>3</sup> ))	Object size (mm)/shape	Object weight	Total weight	Transducer input power
S1	Pushing	pushing $(\alpha = 30^{\circ})$	3×30×2, square	Wood (~800)	65×65×3.175, cuboid	~10g	~15g	10 W
S1	Rotating	rotating $(RN = 3, -2)$	30×30×2, circle	Wax (~860)	35×35×18, cylinder	~19g	~23g	10 W
S1	Following	following $(\theta = 30^{\circ})$	30×30×2, circle	3D-printed resin (~1150)	50×50×14, hollow cylinder	~26g	~30g	10 W
S2	Pushing rotating	pushing_rotating $(\alpha = 45^\circ, RN = 0.5)$	30×30×2, square	Wood (~800)	64×64×22, cuboid	~35g	~40g	10 W
S2	Following rotating	following_rotating $(\theta = 45^\circ, RN = 0.3)$	30×30×2, circle	3D-printed resin (~1150)	50×50×14, hollow cylinder	~26g	~30g	10 W
S2	Multifrequency pushing	multiplexing_pushing $(\alpha_1 = -20^\circ, \alpha_3 = 40^\circ)$	30×30×2, square	Wood (~800)	65×65×3.175, cuboid	~10g	~15g	10 W
S2	Multifrequency rotating	multiplexing_rotating $(RN_1 = 0.4, \theta_1 = 30^\circ, RN_3 = -3)$	30×30×2, circle	Plastic foam (~30)	40×40×10, cuboid	~0.5g	~4.5g	3 W
S3	Multi-object manipulation	following×1 ( $\theta = 30^{\circ}$ ); following_rotating×1 ( $\theta = 45^{\circ}$ , $RN = 0.3$ )	30×30×2, circle	Plastic foam (~30)	40×40×10, Cuboid; 110×75×10, L-shape	~0.5g; ~1g	~4.5g; ~9g	3 W
S3	Multi-path manipulation	pushing_rotating×1 ( $\alpha = 45^{\circ}$ , $RN = 0.5$ ); multiplexing_pushing ×1( $\alpha_1 = -20^{\circ}$ , $\alpha_3 = 40^{\circ}$ )	30×30×2, square	Plastic foam (~30)	65×65×3.175, cuboid	~20g	~30g	10 W
S3	Manipulation in tube	pushing×1 ( $\alpha = 60^\circ$ )	6×10×2, rectangle	PDMS (~1500)	6×10×4, cuboid	~0.35g	~0.71g	3 W
S3	Underwater 3D manipulation	following×3 ( $\theta = 30^{\circ}$ )	30×30×2, circle	3D-printed resin (~1150)	26×26×20, hollow cylinder	~10g	~20.37g	1 W-4 W



Fig. S1. Pictures of metamaterial patches, corresponding surface patterns, and objects used in experiments. (a) "Pushing" metamaterial with  $\alpha$  ranging from 0° to 60°. (b) "Rotating" metamaterial with the different *RN* numbers. (c) "Following" metamaterial with  $\theta$  ranging from 0° to 60°. (d) Metamaterial patches attached on different objects.



Fig. S2. Illustration of the setup for acoustic property measurement.



**Fig. S3. Illustration of the incident acoustic beam characterization experiment.** (a) The measurement setup. (b) An example of the transmitted and received signals.



Fig. S4. 3D printed ring-shaped structures for blocking the objects' x-y plane movement.



Fig. S5. (a) Illustration of the z-direction acoustic radiation force  $(F_{rz})$  measurement method. (b) Measured  $F_{rz}$  as a function of transducer input power for a flat metamaterial patch ( $\alpha = 0^{\circ}$ ). The minimum, first quartile, median, third quartile, and maximum are shown, with the mean as a small square. (c) Illustration of the  $\Delta v_{max}$  measurement for the "following" function. (d) Experimentally measured  $\Delta v_{max}$  with different total weight of metamaterial and object. The used "following" metamaterial has  $\theta$  of 30° and input power of 10 W.



Fig. S6. (a) The needed acoustic radiation force  $F_{rz}$  (calculated as  $F_{rz}=F_b-F_g=V_t\rho_{water}g-m_tg$ ) for an object with total mass  $m_t$  and total volume  $V_t$  to reach the force balance in the z- direction.

(b) The additional acoustic radiation force needed,  $\Delta F_{rz}$  (calculated as  $\Delta F_{rz} = \pi (D_m/2)^2 h \rho_{\text{metamaterail}}g$ ), as a function of the metamaterial patch dimeter  $D_m$ , assuming that the patch is circular and have an average thickness of 2 mm, and the metamaterial density is  $\rho_{\text{metamaterial}} = 6340 \text{ kg} \cdot \text{m}^{-3}$ .



Fig. S7. Acoustic field simulation and acoustic radiation force calculation. (a-d) Simulation of the acoustic field distribution for a "pushing" metamaterial patch with: (a)  $\alpha = 0^{\circ}$ ; (b)  $\alpha = 20^{\circ}$ ; (c)  $\alpha = 40^{\circ}$ ; (d)  $\alpha = 60^{\circ}$ . (e) The calculated angle between the acoustic radiation force and the +z direction (arctan( $F_{rx}/F_{rz}$ )) as a function of the wave reflection angle  $\alpha$  based on simulated acoustic field distribution.



**Fig. S8. Simulation of acoustic reflection from the "multiplexing\_pushing" metamaterial.** (a) Surface pattern of the "multiplexing\_pushing" metamaterial. (b, c) Simulated acoustic pressure distributions for 1 MHz (b) and 3 MHz (c) incident waves.



**Fig. S9. Details of the experimental setups in Fig. 5 and Movie S3.** (a) Multi-object manipulation. (b) Multi-path manipulation. (c) Non-invasive manipulation in a tube. (d) 3D underwater manipulation.

Supporting Movie S1. Demonstration of acoustic manipulation functions: pushing, rotating, following.

Supporting Movie S2. Demonstration of multifunctional design of a single metamaterial patch.

Supporting Movie S3. Demonstration of underwater object manipulation with acosutic metamaterial.