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ADVANCED MATERIALS

Supporting Information

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High-Quality-Factor Mid-Infrared Toroidal Excitation in Folded 3D Metamaterials

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Supporting Information

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1. Fabrication strategy of the 3D folding metamaterial

In the fabrication process, the sample was reversed to be up-side-down before ion beam process, in order to protect the metal patterns during ion beam folding. As shown in Figure S1a, a conventional strategy is to scan the SiN_x film with metal patterns on the top of the film. However, as the SiN_x flake is folded towards vertical direction, the ion beam will etch part of the metal patterns due to its thickness. A solution is illustrated in Figure S1b, where the metal pattern is located on the backside of the SiN_x film, and it is obvious that the ion beam cannot damage the metal pattern as the inclined angle approaches 90°. Excessive ion dose will lead to damage on the top of the SiN_x flake but the metal pattern can be protected.



Figure S1 Two different ion beam scanning strategies. a) The metal patterns are on the top side of SiN_x film, of which the metal pattern can be easily damaged. b) The sample is put up-side-down to protect the metal patterns.

2. Extraction of the Q factors

The transmission spectrum of the SiN_x film was measured and shown in Figure S2a. It can be found that there is strong absorption around 25 THz, thus unfortunately, the baseline of the transmission curve in Figure 3a is not flat. In order to extract the Q factors of the two resonance modes accurately, the baseline of the experimental transmission curve was subtracted, as shown in Figure S2b. The Q factors of both resonances were calculated in the form of $Q_i = f_i / \Delta f_i$, where f_i is the center frequency of each resonance, and Δf_i is the peak width at half maximum.



Figure S2 a) Transmission spectrum of flat SiN_x film. b) Transmission curve of

toroidal metamaterial after baseline subtraction.

3. Excitation of LC resonance on SRR unit

When the SRR structures are perpendicular to the wave vector k, the electric and magnetic resonance can only be excited by electric field (E-field), because there is no magnetic field (H-field) component that passes through the ring.^[1] But when the SRRs are not perpendicular to k, both electric and magnetic field can excite resonant modes,^[2] which is exactly the case in this work. The angle between the incident E-field and bottom sides of SRRs is 45°, thus both

electric and magnetic field contribute to the resonance. However, SRRs with opposite openside directions have different ways of excitation. It can be easily concluded from right hand rule that, for "atom 1", the surface current that excited by E-field on the bottom side of SRR is the same direction with that excited by H-field through Lenz"s law (red and blue dotted arrow in Figure S3a). The red and black curves show the H-field in the center point of SiN_x flakes of both samples with and without SRR structure. The phase difference of H-field in "atom 1" and reference is about 150°, indicating that the excited H-field has almost opposite phase compared to incident light, which is in agreement with the Lenz's law. But for "atom 2", the surface currents excited by E-field and H-field are opposite (red and blue dotted arrow in Figure S3a). From the waveform curves, the phase of H-field in "atom 2" is only about 22° lagging behind the reference, which is not agree with the Lenz's law, indicating that the surface current is mainly excited by E-field. Since the responses of "atom 1" and "atom 2" to the incident field are contrary, the coupling when they are combined in toroidal metamaterial can be more complicated.



Figure S3 Waveform comparison of H-field in the center of SRR, and schematic of

light polarization and SRR units of "atom 1" a) and "atom 2" b).

4. Excitation of toroidal response

The H-field waveform in the center of "atom 1" and "atom 2" after they are combined in toroidal metamaterial was also simulated, and plotted in Figure S4. At ω_1 (toroidal resonance) in Figure S4a, "atom 1" has π phase difference while "atom 2" has the same phase compared to the reference sample (SiN_x flakes without SRR structure), which is the same with the SRR unit analyzed in Part 1. This means that the toroidal resonance is a natural feature of the toroidal metamaterial when combining four SRRs together, due to the oppositely excited H-field. However at ω_2 (magnetic resonance), the phases in the center of "atom 1" and "atom 2" have a 73° and 69° delay compared to the incident light. This means that the LC resonances were not solely excited by E-field or H-field, but the combination of both. At the phase of incident H-field maximum in the center of SRRs, the wave front of the E-field with a minimum intensity passes through the SRR units, as shown in Figure S4c. This means that since the SRRs are parallel to the wave vector *k*, there are phase differences when the incident light passes through and meet the bottom side of "atom 1" and "atom 2". Therefore, the surface current on the bottom sides of "atom 1" and "atom 2" are excited by electric field with opposite directions, leading to the "magnetic resonance" with a total magnetic dipole of **M**_{*y*}.



Figure S4 Comparison of H-field waveform in the center of SRR units at a) ω_1 and b) ω_2 . c)

The electric field is plotted in the plane that parallel to two of the SRRs at the phase of

maximum magnetic field in the center of SRR units.

5. Meta-molecule composed of single direction SRRs

The simulated transmission spectrum of meta-molecule composed of only "atom 1" is shown in Figure S5a. There is only one response at 29.8 THz, and the H-field distribution indicates that this is a magnetic response, because the H-field does not form a loop but has a vector resultant in x direction (blue arrows in Figure S5b).



Figure S5 a) Transmission spectrum of metamaterial composed of single direction SRR only;

b) The H-field distribution on the plane that cross the center of SRRs

6. Multipole moments calculation by surface current

In order to calculate the moments of different kinds of multipoles, the following expressions were used:^[3]

Electric dipole moment: $\mathbf{P} = \frac{1}{i\omega} \int \mathbf{j} d^3 r$

Magnetic dipole moment: $\mathbf{M} = \frac{1}{2c} \int (\mathbf{r} \times \mathbf{j}) d^3 r$

Toroidal dipole moment: $\mathbf{T} = \frac{1}{10c} \int [(\mathbf{r} \cdot \mathbf{j}] d^3 r]$

Electric quadrupole moment: $Q_{\alpha\beta} = \frac{1}{i\omega} \int \left[r_{\alpha} j_{\beta} + r_{\beta} j_{\alpha} - \frac{2}{3} (\mathbf{r} \cdot \mathbf{r}) \right]$

Magnetic quadrupole moment: $M_{\alpha\beta} = \frac{1}{3c} \int \left[\left(\mathbf{r} \times \mathbf{j} \right)_{\alpha} r_{\beta} + \left(\mathbf{r} \times \mathbf{j} \right)_{\beta} r_{\alpha} \right] d^{3}r$

where **j** is the current density extracted from simulation, *c* is the speed of light, ω is the angular frequency, and α , $\beta = x, y, z$.

7. Magnetic field and surface current distribution at an incident angle of 75°



Figure S6 a) Magnetic field and b) surface current distribution at ω_1 with an incident angle of

75°.

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