Supporting Information


Near-Field Coupling Induced Less Chiral Responses in Chiral Metamaterials for Surface-Enhanced Vibrational Circular Dichroism

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Supporting Information for
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Supporting Note 1 to 7
Supporting Figures. S1 to S20

Supporting Note 1: Derivation of TCMT equations

The model is expressed as:

\[
\frac{d}{dt} \begin{pmatrix} P_x \\ P_y \end{pmatrix} = \begin{pmatrix} j\omega_x - \gamma_{rx} - \gamma_{ax} \\ j\omega_y - \gamma_{ry} - \gamma_{ay} \end{pmatrix} \begin{pmatrix} P_x \\ P_y \end{pmatrix} + \begin{pmatrix} \kappa_x & 0 \\ 0 & \kappa_y \end{pmatrix} \begin{pmatrix} s_x^+ \\ s_y^+ \end{pmatrix} \tag{S1} \]

The mode amplitude term \( \begin{pmatrix} P_x \\ P_y \end{pmatrix} \) can be expressed as:

\[
\begin{pmatrix} P_x \\ P_y \end{pmatrix} = \frac{\begin{pmatrix} j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax} \\ -j\xi_{xy} \end{pmatrix} \kappa_x + 0 \begin{pmatrix} s_x^+ \\ s_y^+ \end{pmatrix}}{\xi_{xy}s_x + j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax}[\omega_0 - \omega_x] + \gamma_{ry} + \gamma_{ay}] \begin{pmatrix} 0 \\ 0 \end{pmatrix}} \tag{S2} \]

Substituting the \( \begin{pmatrix} P_x \\ P_y \end{pmatrix} \), we could obtain the relationship between the input and output parameters:

\[
\begin{pmatrix} s_x^- \\ s_y^- \end{pmatrix} = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} s_x^+ \\ s_y^+ \end{pmatrix} + \frac{\begin{pmatrix} 2\gamma_{rx}[\omega_0 - \omega_x] + \gamma_{ry} + \gamma_{ax} \\ -2j\xi_{xy}\sqrt{\omega_{rx}\omega_{ry}} \end{pmatrix} \kappa_x + 0 \begin{pmatrix} s_x^+ \\ s_y^+ \end{pmatrix}}{\xi_{xy}s_x + j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax}[\omega_0 - \omega_x] + \gamma_{ry} + \gamma_{ay}] \begin{pmatrix} 0 \\ 0 \end{pmatrix}} \tag{S3} \]

the reflectivity matrix can be determined as:

\[
\hat{r} = \begin{pmatrix} s_x^- \\ s_y^- \end{pmatrix} = \frac{\begin{pmatrix} -\xi_{xy}s_x + j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax}[\omega_0 - \omega_x] - 2j\sqrt{\omega_{rx}\omega_{ry}} \end{pmatrix} \kappa_x + 0 \begin{pmatrix} s_x^+ \\ s_y^+ \end{pmatrix}}{\xi_{xy}s_x + j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax}[\omega_0 - \omega_x] + \gamma_{ry} + \gamma_{ay}] \begin{pmatrix} 0 \\ 0 \end{pmatrix}} \tag{S4} \]

The circular dichroism is defined by the subtraction between LCP and RCP light, where the electric field of LCP and RCP light is given by:
\[
\begin{pmatrix}
(s_x^{+})^{\text{LCP}} \\
(s_y^{+})^{\text{LCP}}
\end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ j \end{pmatrix}, \quad \begin{pmatrix}
(s_x^{+})^{\text{RCP}} \\
(s_y^{+})^{\text{RCP}}
\end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -j \end{pmatrix}
\]

And the \( R_{\text{CD}} \) is derived as:

\[
R_{\text{CD}} = r^{LCP}r^{LCP}_R - r^{RCP}r^{RCP}_R = 2(r^{LCP}_x + j r^{LCP}_y) \]

where \( r^* \) is the conjugate matrix of reflectivity, \( r^R \) and \( r^I \) represent the real and imaginary part of the reflectivity, respectively. The final expression of \( R_{\text{CD}} \) is:

\[
R_{\text{CD}} = 4(\xi_{xy} + \xi_{yx}) \frac{y_{ax}}{y_{ry}} - y_{rx} + y_{ax}[j(\omega_0 - \omega_x) + y_{ry} + y_{ay}] \] \( \xi_{xy} + [j(\omega_0 - \omega_x) + \gamma_{rx} + \gamma_{ax}]\[j(\omega_0 - \omega_y) + \gamma_{ry} + \gamma_{ay}] \] \( (\gamma_{rx} - \gamma_{ax})^2 + 4(\xi_{xy} + \xi_{yx})[\gamma_{rx} + \gamma_{ax}]\[j(\omega_0 - \omega_y) + \gamma_{ry} + \gamma_{ay}] \) \]

\[ (S7) \]

**Supporting Note 2: Parameter mapping**

Based on these calculation results, we use a parameter mapping method to study the influence of two near-field coupling coefficients \( \xi_{xy} \) and \( \xi_{yx} \). It should be noted that the parameter mapping results are not to show quantitative results of our TCMT model, as it can be affected by many parameters. Therefore, when changing the geometries or relative positions of the metamaterials, we only assume a perturbation of one single parameter to simplify the model and to study the trend of parameter changes. The parameter mapping results of \( R_{\text{CD}} \) are shown in Figure S8. As the optical chirality field in the near-field coupling regions shows only negative field enhancement for L-Ring structures and positive field enhancement for R-Ring structures, shown in Figure 5(c).

We assume that the near field coupling coefficients \( \xi_{xy} \) and \( \xi_{yx} \) have identical sign. Therefore, when these two factors are both negative, the represented case is L-Ring structure, while the both positive case is R-Ring structure. Besides, we also varied the loss ratio \( \frac{y_{ay}}{y_{ry}} - \frac{y_{ax}}{y_{rx}} \) into three cases. When \( \frac{y_{ay}}{y_{ry}} = \frac{y_{ax}}{y_{rx}} \), the \( R_{\text{CD}} \) achieves zero value when the \( \xi_{xy} \) and \( \xi_{yx} \) reaches identical value, as the white lines shown in Figure S8(c) and S8(d), which is the case of our quadruple resonator chiral metamaterials. Besides, when a larger difference between \( \xi_{xy} \) and \( \xi_{yx} \) is achieved, the \( R_{\text{CD}} \) will also reach its maximum or minimum value. When \( \frac{y_{ay}}{y_{ry}} - \frac{y_{ax}}{y_{rx}} \neq 0 \), the zero CD white line will shift, as shown in Figure S8(a), S8(b), S8(e), and S8(f), which indicating the possibility of creating non zero CD through symmetric near field coupling.
Supporting Note 3: Analysis of resonators with different thicknesses

In the manuscript, we use the quadruple resonators with identical geometrical parameters to minimize the influence of radiative loss and absorption losses, which fits the condition shown in Figure S8(d) and S8(d). To give a more comprehensive demonstration of this model, we further fabricate devices with thickness differences to investigate the case with different losses. The AFM testing results of the device with different thicknesses are shown in Figure S9(a). We fabricated the devices using a two-step e-beam lithography process, followed by deposition of gold of two different thicknesses. The x-oriented modes have a thickness of 80 nm, while the y-oriented modes are 115 nm. Leveraging such variation, we assume that the absorption loss of the x-oriented and the y-oriented modes are varied, which is because the Ohm loss of the plasmonic structures is varied\cite{1}. Similar variation was also reported using different materials\cite{2}, or different resonant modes\cite{3}. The mapping results of $R_{CD}$ with different near-field coupling coefficients are shown in Figure S9(b) and S9(c). For Figure S9(b), the losses are identical with $\frac{\gamma_{ay}}{\gamma_{ry}} - \frac{\gamma_{ax}}{\gamma_{rx}} = 0$, and the experimental results shown in Figure 3 agrees well with predicted results, where the asymmetric near field coupling induce an additional negative $R_{CD}$. When there is a thickness difference of 35 nm, the predicted results are shown in Figure S9(c). In this case, due to that $\frac{\gamma_{ay}}{\gamma_{ry}} - \frac{\gamma_{ax}}{\gamma_{rx}} > 0$, as the y-oriented modes have larger absorption loss, hence, the case where the four nanogaps are all exposed should have an initial $R_{CD}$. With more PMMA covering the nanogaps, the $R_{CD}$ will gradually vanish to zero. The experimental results shown in Figure S9(c), S9(d), and S9(e) also agree with the predicted result. Therefore, we again experimentally demonstrate the importance of the near-field coupling coefficient with different loss ratios, which can significantly influence the far-field $R_{CD}$. Besides, the variation of same thickness and same width is also shown in Figure S20. We choose 0.8 $\mu$m and 80-120 nm as the optimized geometries for our structure.
Supporting Note 4: Analysis of resonators with different nanogaps

Another method of demonstrating the influence of near-field coupling to the far-field CD is to design the structures with different nanogaps. With the reduced size of nanogaps, the confined near-field intensity becomes larger and the coupling coefficient is considered to be stronger. The near-field simulation results of the L-ring and R-ring structures are shown in Figure S10(a) and S10(d), respectively. It can be observed that different gap sizes bring different near-field intensity, inducing asymmetric near-field coupling in this case. For L-ring structures, if the vertical nanogaps are decreased, the near-field coupling coefficient become more negative, and thus will lead to a positive $R_{CD}$. The simulation results of the reduced vertical gaps are shown in Figure S10(b) and S10(c), where the decreased $g_1$ and $g_3$ could induce a positive $R_{CD}$ and agrees with the predicted mapping results. Similarly, for the R-ring structures, when the near-field coefficient become more positive when the gap reduced, the $R_{CD}$ will get a negative value, which also agrees with the simulated results shown in Figure S10(e) and S10(f).

Supporting Note 5: Analysis of glucose vibrational signal on thin film and thick film

The glucose enantiomers are also characterized using our custom-made optical setup. The results are shown in Figure S12. When forming the glucose film, there are two different thicknesses, as shown in the IR camera image in Figure S12(a). The different colors show reflection difference between the thick film and thin film glucose. The reflection spectrum is shown in Figure S12(b). Compared with the reflection signal on substrate, the thin film shows slightly smaller reflection signals, while the thick film shows much lower reflection signals, indicating a much larger absorption. We have also
measured the thicknesses from the thin film, as shown from Figure S12(c) and S12(d), which is around 43 nm. The thickness coated on metamaterials is also around this range, as shown in Figure S12(e) and S12(f). However, for the thick film layer, we could not measure the thickness using AFM, as the thickness is beyond the measuring scale of AFM (~500 nm). Therefore, we use Beer-Lambert law to roughly estimate the thickness of the thick film from the original spectrum. The absorbance is calculated by $A=\log(I_0/I)$, where $I_0$ is the reflection signal of substrate, and $I$ is the reflection signal from the glucose film. After obtaining the absorbance from both thin film and thick film, we apply $A=\varepsilon cl$, where we assume the concentration $c$ and molar absorptivity $\varepsilon$ is the same for glucose film. The calculated length $l$ of thick film layer is ranging from 1700 nm to 2500 nm (the variation is because of different absorbance at difference wavenumbers, thickness of thin film is set as 45 nm). Furthermore, we extracted the vibrational signal of D-glucose and L-glucose from the reflection spectrum, as shown in Figure S12(g). For this result, if we plot them at the same reflection or absorption spectrum, the details of vibrational peaks can be difficult to observe. Therefore, we use the absorption of thick film and reflection of thin film so that they can be plotted at the same scale with the name “vibrational signal”. We also tried to measure the VCD signal from the glucose films. However, the VCD signal on thin film is too weak to be detected using our optical setup. We only detected a flipped VCD signal from the glucose thick film, as shown in Figure S12(h). Although it is also not a perfectly mirror symmetry, the VCD signal shows similar distribution as the results in Figure 4(e) and 4(f). However, as we mentioned before, the thick film is not for calculation and characterization, as the thickness and concentration are not fixed. Therefore, this result is only to show the original VCD signal of L-glucose and D-glucose without enhancement, but we do not use it for quantitative analysis.

**Supporting Note 6: Analysis of concentration error between glucose solutions**
We notice that our measurement results from L-glucose and D-glucose do not present perfect mirror symmetry. As we also mentioned in the main text, conventional VCD instrument could provide larger SNR ratio and accuracy, especially when dealing with weak molecular VCD signal ($\sim 10^{-5}$), which can obtain molecular signals with complete mirror symmetry. To study the reason of this phenomenon, we use the results in Figure 5d and 5e for case study, which contains variation for both wavenumber and concentration. Given our definition of mirror symmetry as $CD$ (left-handed molecule) +$CD$ (right-handed molecule) =0, we sum the VCD signal of L-glucose and D-glucose with the same labelled concentration. The results are shown in Figure S15. It can be observed that: 1) When the concentration is lower, it becomes difficult for the summation signal to reach zero. This is because that lower concentration approaches the noise level, thus resulting in fluctuation of the signals. 2) The summation signal is more closely to zero from 1040 cm$^{-1}$ to 1080 cm$^{-1}$. This is because the noise level of different wavenumbers is also different (see Supporting figure S13(a)), where the noise level from 1040 cm$^{-1}$ to 1080 cm$^{-1}$ is lower than that from 1210 cm$^{-1}$ to 1250 cm$^{-1}$. As a result, the signals ranging from 1040 cm$^{-1}$ to 1080 cm$^{-1}$ behave more mirror symmetry. Besides, we also notice that the summation at same concentration sometimes can be far from zero. This is because of potential error between labelled concentration and the real concentration. However, this will not affect the VCD signal from a certain analyte. The VCD signal is obtained using a two-step subtraction, which is 1) Subtracting the signals between LCP and RCP light: $\Delta A = A_{LCP} - A_{RCP}$; 2) Subtracting the signals between with analyte and without analyte: $\Delta A_{w.glu.} - \Delta A_{w.o.glu.}$. We understand that potential signal variation may come from the second step, given that the IR signals between L-glucose and D-glucose are different. However, such difference can be subtracted at the first step when processing the CD signals, which is $\Delta A_{w.glu.} = A_{LCP(w.glu.)} - A_{RCP(w.glu.)}$. The absorption data is obtained from the measured reflection spectrum. We also show the original reflection spectra in Figure S16. Indeed, the vibrational signal can be different for L-glucose and D-glucose, but the subtraction between LCP and RCP signal is for single analyte, where the vibrational signal is the same and the signal difference reveals the glucose chirality. It can be observed that the reflection signal of L-glucose with L-
ring is higher when the incident light is RCP (\(\sim 1074 \, \text{cm}^{-1}\)), while the LCP signal is higher when D-glucose coated with R-ring at the same wavenumber.

**Supporting Note 7: Control experiment of racemic mixture and linear response**

We have measured the metamaterials with left, right and racemic analytes under front-side illumination, as shown in Figure S17 (a), the enhanced signal of L-glucose and D-glucose is stronger than the racemic glucose mixture. Besides, the signal from the bare device (without glucose) also does not provide any additional VCD signals. Therefore, the VCD signal is not considered from any artifacts on the metamaterial substrate. In Figure S17 (b), we also compare the VCD signals on gold substrate. However, for D-glucose, L-glucose and racemic mixtures, there are no observable signals in the VCD spectrum, where we consider the unenhanced signal is too weak to be detected by our optical setup. We also show the results of vibrational signals of these three different analytes from the thick film, as shown in Figure S17 (c). This figure is only to show the identical vibrational peaks, such as 1022 cm\(^{-1}\), 1076 cm\(^{-1}\), 1184 cm\(^{-1}\) and 1216 cm\(^{-1}\), indicating the existence of these analytes on the chip. The different signal intensities are mainly due to different concentrations of the thick film. The reason why we use thick film to characterize the vibrational signal is because the signal is more observable than thin film. From these two measured results, we confirm the signal is mainly from the glucose chirality, rather than any artifacts on the substrate or metamaterials.

We also implement another measurement to verify potential interference from linear dichroism effect by using a manual rotary stage (Thorlab, MSRP01). The optical setup is shown in Figure S18(a). The orientation of the linear polarization is fixed, while we rotate the sample plane to change the relative orientation between sample and light beam. We first measured the reflection spectra of R-ring metamaterials without analyte, as shown in Figure S18(b). The length of the metamaterials is 2.6 µm, matching the
resonant wavenumber around 1225 cm\(^{-1}\). When the rotary stage is changed to 45
degrees, 90 degrees, and 135 degrees, corresponding to the orientation change of linear
polarizations, the metamaterials behave insensitively. Our simulation results of the
near-field intensity are shown in Figure S18(c). When the orientation of the linear
polarization is different, the near-field distribution is also different. However, the
maximum intensities of the electric field of all polarizations are identical, indicating the
same enhancement provided to the molecules when coating onto the metamaterials.
Based on these insensitive responses to the linear polarizations, we coated D-glucose
onto the metamaterials and rotate the sample plane to measure the linear polarization
response of both metamaterials and the analyte, as shown in Figure S18(d). After
removing the signal of metamaterials, we extract the infrared absorption spectra of D-
glucose. For all four polarizations, the vibrational peaks have almost identical
absorption intensity. Although small fluctuations can be observed between each
polarization, this fluctuation approaches noise level. Therefore, we do not consider this
as a linear dichroism signal from D-glucose.
Fig. S1: Geometric parameters of MIM double resonators and quadruple resonators. The light is incident in normal direction, and the polarization is left-handed polarization (a) Parameters for quadruple resonators. (b) Parameters for double resonators. The thickness difference between two resonators is constructed by two-step e beam lithography process, followed by evaporation of gold layer with different thicknesses.
Fig. S2: Study of double resonators and quadruple resonators. (a) Reflection spectrum of double resonators and quadruple resonators with length of L=3.5 μm under LCP illumination. (b) Reflection spectrum of double resonators and quadruple resonators with length of L=3.6 μm under LCP illumination. (c) Reflection spectrum of double resonators and quadruple resonators with length of L=3.7 μm under LCP illumination. The detailed quality factor (Q factor) for each structure is shown in the table. (d) and (e) Comparison of simulation and experiment results of double resonators and quadruple resonators under LCP and RCP light, indicating double resonators have a larger circular dichroism than the quadruple resonators.

<table>
<thead>
<tr>
<th>Q factor</th>
<th>L=3.5 μm</th>
<th>L=3.6 μm</th>
<th>L=3.7 μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Double nanorods</td>
<td>18.76</td>
<td>18.92</td>
<td>13.26</td>
</tr>
<tr>
<td>Quadruple nanorods</td>
<td>17.45</td>
<td>17.02</td>
<td>17.16</td>
</tr>
</tbody>
</table>
Fig. S3: Fabrication process flow the quadruple resonators with selectively exposed PMMA. The dielectric layer is chosen as aluminum oxide (Al$_2$O$_3$), because Al$_2$O$_3$ is less absorptive and does not cover the resonant signal from the metamaterials.
Fig. S4: Atomic force microscope (AFM) image of the selectively exposed PMMA and their measured heights.
Fig. S5: Near-field illustration of the uncoupled parallel modes. (a) Near-field image of the x-polarized modes. (b) and (c) Cross-section data of the electric field intensity. The blue and green curves represent the cross-section lines shown in (a). The shaded areas indicate near-zero field intensity in the middle areas, illustrating no field overlapping and therefore, no near field coupling between the x-polarized modes. (d) Near-field image of the y-polarized modes. (e) and (f) Cross-section data of the electric field intensity. The black and red curves represent the cross-section lines shown in (d). The shaded areas indicate near-zero field intensity in the middle areas, illustrating no field overlapping and therefore, no near field coupling between the y-polarized modes.
Fig. S6: Optical microscope (OM) and scan electron microscope (SEM) images of the 0 gap exposed, 1 gap exposed, and 2 gaps exposed quadruple resonators.

Fig. S7: Simulated results of the 0 gap exposed, 1 gap exposed, and 2 gaps exposed quadruple resonators. (a)-(c) Reflection spectrum of the 0 gap exposed, 1 gap exposed, and 2 gaps exposed quadruple resonators with LCP and RCP illumination. (d)-(f) Reflected circular dichroism (RCD) spectrum of the 0 gap exposed, 1 gap exposed,
and 2 gaps exposed quadruple resonators. The 0 gap exposed resonators show no \( R_{CD} \) signals, while the 1 gap exposed, and 2 gaps exposed resonators show negative \( R_{CD} \) signals, which agrees with the experimental results shown in Fig. 3(e) to Fig. 3(g).

Fig. S8: Mapped \( R_{CD} \) with varied near-field coupling coefficient \( \xi_{xy} \) and \( \xi_{yx} \). (a), (b) Calculation of \( R_{CD} \) when \( \frac{\gamma_{ax}}{\gamma_{rx}} < \frac{\gamma_{ay}}{\gamma_{ry}} \). The range of \( \xi_{xy} \) and \( \xi_{yx} \) are both negative shown in (a) and both positive shown in (b). (c), (d) Calculation of \( R_{CD} \) when \( \frac{\gamma_{ax}}{\gamma_{rx}} = \frac{\gamma_{ay}}{\gamma_{ry}} \). The range of \( \xi_{xy} \) and \( \xi_{yx} \) are both negative shown in (c) and both positive shown in (d). (e), (f) Calculation of \( R_{CD} \) when \( \frac{\gamma_{ax}}{\gamma_{rx}} > \frac{\gamma_{ay}}{\gamma_{ry}} \). The range of \( \xi_{xy} \) and \( \xi_{yx} \) are both negative shown in (e) and both positive shown in (f).
Fig. S9: Analysis of R_{CD} with thickness-varied quadruple resonators. (a) AFM results of the resonator thicknesses. The characterized thickness difference (ΔH) is 35 nm. (b), (c) Mapped R_{CD} of same thickness and different thicknesses. The thicknesses difference is assumed to change the loss ratio difference. (d) to (f) Measured R_{CD} of 0 gap exposed, 1 gap exposed, and 2 gaps exposed structures with ΔH = 35 nm. The R_{CD} agrees with the calculated results in (c).
Fig. S10: Analysis of $R_{CD}$ with nanogap-varied quadruple resonators. (a) and (d) Simulated near-field electric intensity of different size of nanogaps. The 100 nm gap could induce a larger field enhancement than the 200 nm gaps, indicating asymmetric near-field coupling. (b), (c) Simulated $R_{CD}$ of same nanogaps and different nanogaps of L-Ring structure. The results agree with the blue and red dot in (a). (e), (f) Simulated $R_{CD}$ of same nanogaps and different nanogaps of R-Ring structure. The results agree with the blue and red dot in (d).
**Fig. S11: Control experiment of metamaterials.** (a) to (c) Measured reflection spectra of chiral metamaterials coated with different number of PMMA pillars. The resonant lengths of chiral metamaterials are design differently to achieve the same resonant wavelength after coating with PMMA. (d) to (f) Compared circular dichroism of the metamaterials structures when changing the length of each nanorod. (g) Measured enhanced VCD spectrum of R-ring with and without D-glucose (concentration: 2000 ng/μl). A negatively enhanced VCD signal is observed when coated with the R-ring structure. (h) Measured enhanced VCD spectrum of D-glucose on R-ring and L-glucose on L-ring structures. It can be observed that the strongest VCD peak is around 1216 cm\(^{-1}\). The difference of D-glucose signals between S11 (g) and S11 (f) is due to the concentration error between different devices.
Fig. S12: Measured glucose vibrational signal of both thin film and thick film. (a) IR camera image shown the reflection difference between thin film and thick film glucose. (b) Reflection spectrum of glucose thin film, thick film, and bare gold substrate. (c) and (d) Measured thickness of glucose thin film on substrate. (e) and (f) Measured thickness of glucose thin film on gold metamaterials. (g) Vibrational signal extracted from the reflection spectrum. For both D-glucose and L-glucose, the vibrational signal
for thick film is higher than thin film. However, once the thick film is formed, it will hinder the signal from the metamaterials due to enormous absorption. Therefore, we still use thin-film glucose for all the characterization of chiral metamaterials. (h) Measured VCD signal from the thick film glucose. This is only used to show the shape of glucose VCD signal, but not used for calculations.

![Detection limit of glucose at different absorption peaks.](image)

**Fig. S13: Detection limit of glucose at different absorption peaks.** (a) Noise characterization of the experimental setup from the reflection spectrum. The $3\sigma$ noise is around $2.388\times10^{-4}$ and $0.00191$ for the wavelength band around $1076$ cm$^{-1}$ and $1216$ cm$^{-1}$, respectively. (b) to (e) Calculated detection limit for D-glucose and L-glucose at these two wavelengths. The detection limit is around $\mu$M level.
Fig. S14: Experiment setup of the optical measurement (Spero® Chemical Imaging Microscope). The waveplate is fixed on an optical bench to ensure the normal direction of incident light.
Figure S15: Summation of the VCD signal from L-glucose and D-glucose. VCD signal of L-glucose, D-glucose, and their summation. (a) to (d) VCD signal ranging from 1040 cm\(^{-1}\) to 1080 cm\(^{-1}\). Concentration is varying from 1000 ng/μl to 125 ng/μl. (e) to (h) VCD signal ranging from 1210 cm\(^{-1}\) to 1250 cm\(^{-1}\). Concentration is varying from 1000 ng/μl to 125 ng/μl.

Figure S16: Comparison of the reflection signal difference between LCP and RCP light. (a) L-glucose coated on L-ring metamaterials (b) D-glucose coated on R-ring metamaterials.
Figure S17: Measured D-glucose, L-glucose, and racemic glucose signals on (a) metamaterials with glucose thin film, (b) gold substrate with glucose thin film and (c) gold substrate with glucose thick film. The racemic glucose mixture is made by mixing the solution of L-glucose and D-glucose. The volume and concentration of the L- and D-glucose solutions are the same (10ul, 1000ng/ul), and we consider it a racemic mixture.

Figure S18: Control experiment of linear response. (a) Optical setup of the rotary measurement. The orientation of the linear polarization is fixed, while the sample plane is rotating to change the relative orientation. (b) Measured reflection spectra of R-ring metamaterials under different rotation angles. (c) Simulated near-field distribution and...
intensity under different polarization angles. (d) Extracted absorption spectrum from D-glucose coated onto R-ring metamaterials under different linear polarizations.

Figure S19: Simulation of optical chiral field at different heights. (a) Simulation of the electric field intensity of cross-section plane. (b) to (g) Simulated chiral field distribution of the metamaterials from on different heights.
**Figure S20: Geometry study on width and height of the metamaterials.** (a) Parameter sweep of the width of each plasmonic rod. (b) Parameter sweep of the thickness of the metamaterials.

**References**

