安徽理工大學
ANHUI UNIVERSITY OF SCIENCE & TECHNOLOGY

中国科学技术大学
University of Science and Technology of China

理论物理前沿进展研讨会

基于里德堡原子偶极相互作用的光场传输调控

郑迪迪

2024. 1. 25　安徽 合肥

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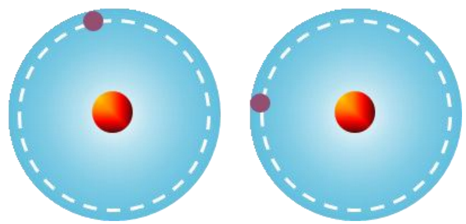
背景：里德堡原子

瑞典物理学家
Johannes Robert Rydberg
1854-1919

一般原子

里德堡原子



里德堡公式：

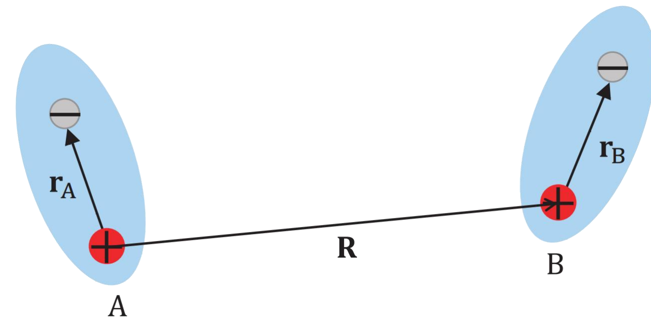
$$\frac{1}{\lambda} = R_{Ryd}\left(\frac{1}{n_1^2} - \frac{1}{n_2^2}\right)$$

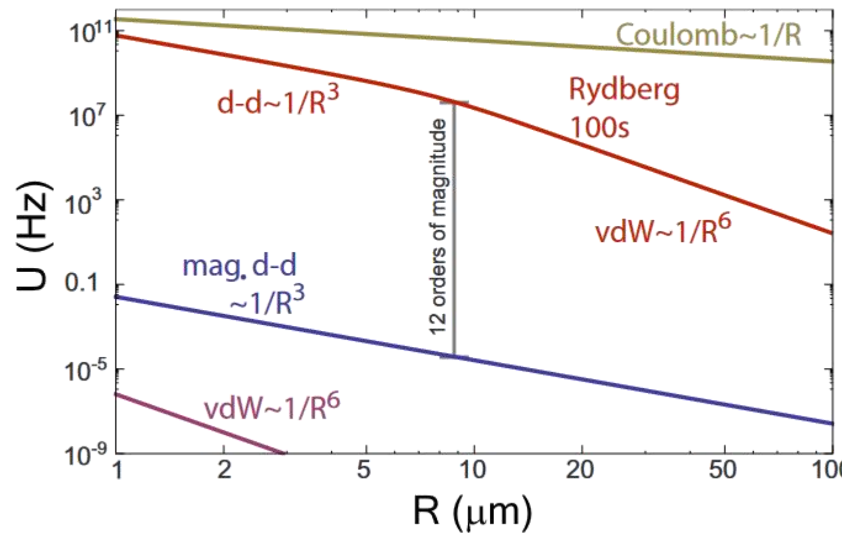
里德堡常数：

$$R_{ryd} = \frac{1}{(4\pi\varepsilon_0)^2}\frac{Z^2 m_e e^4}{2\hbar^2}$$

- 轨道半径：$\propto n^2$
- 极化强度：$\propto n^7$
- 能级寿命：$\propto n^3$

里德堡原子间强偶极-偶极相互作用

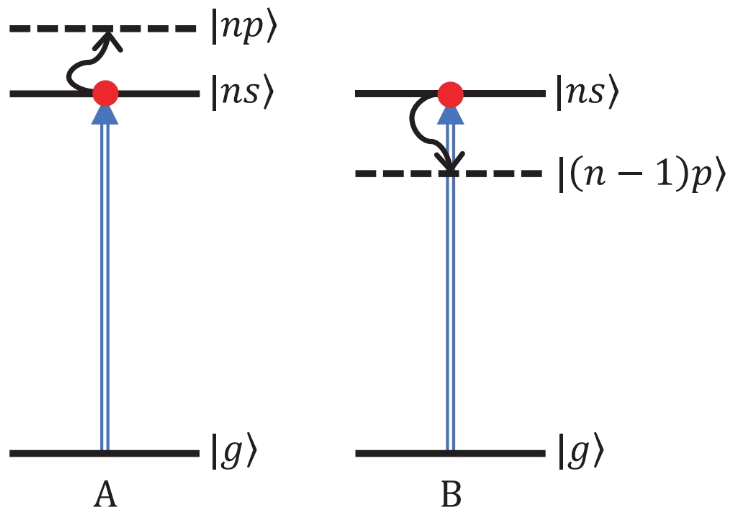




$$\frac{U_{\text{Rydberg}}}{U_{\text{ground}}} = 10^{12}$$

Rev. Mod. Phys. 82, 2313 (2010)

背景：里德堡原子



$$\hat{V}_{dd}(R) = \frac{1}{4\pi\epsilon_0 R^3}\left[\hat{\mu}_\mathrm{A}\cdot\hat{\mu}_\mathrm{B} - 3\left(\hat{\mu}_\mathrm{A}\cdot\frac{\mathbf{R}}{R}\right)\left(\hat{\mu}_\mathrm{B}\cdot\frac{\mathbf{R}}{R}\right)\right]$$

偶极耦合对态的能量差：

$$\Delta = E_{r'} + E_{r''} - 2E_r$$

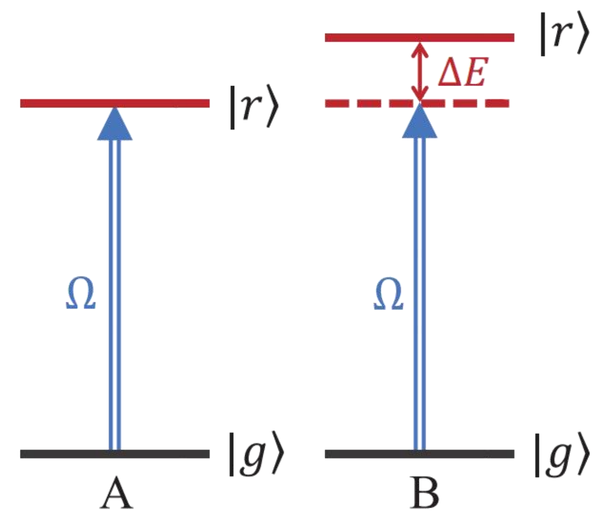
☐ 范德瓦尔斯机制：

$V_6 = \hbar(C_6/R^6)|r_A\rangle\langle r_A| \otimes |r_B\rangle\langle r_B|$ （ $C_6 \propto n^{11}$ ）

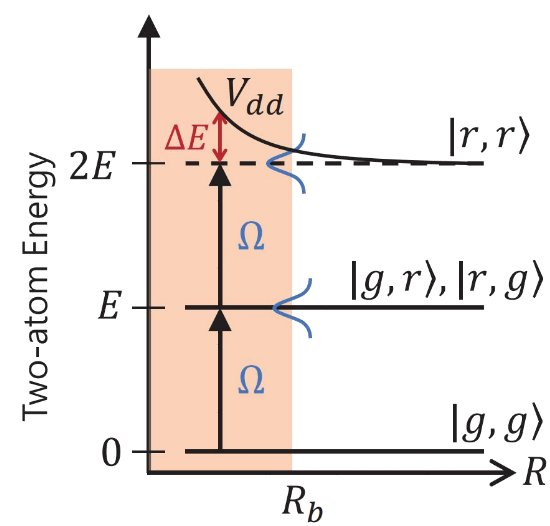
☐ 共振偶极–偶极机制：

$V_3 = \hbar(C_3/R^3)|r_A\rangle\langle r_A| \otimes |r_B\rangle\langle r_B|$ （ $C_3 \propto n^4$ ）

里德堡相互作用引起的频移：

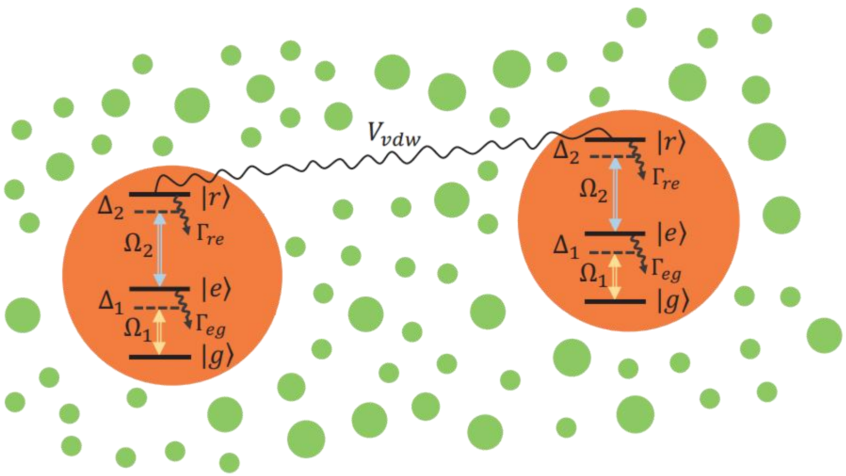


里德堡阻塞与反阻塞效应：

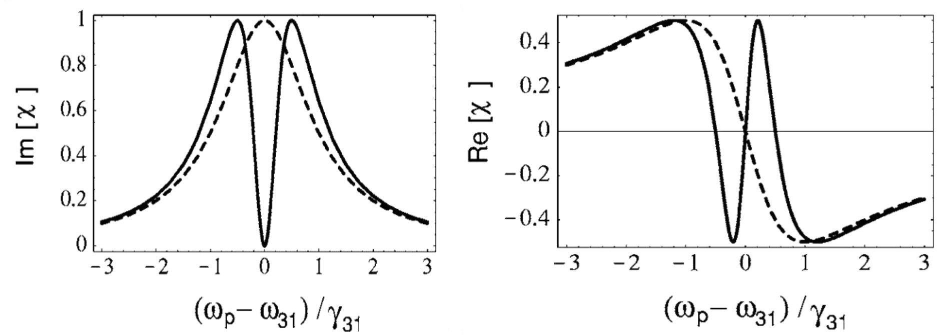


背景：里德堡原子

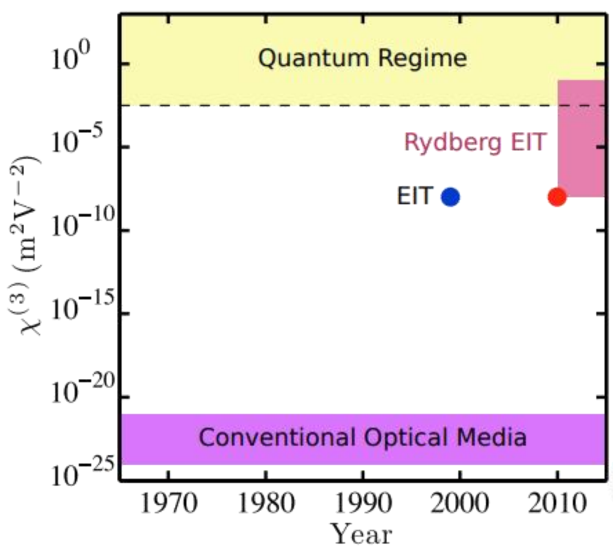
里德堡原子关联相互作用+电磁诱导透明(EIT)➞显著增强的光学非线性响应



电磁诱导透明（线性响应）：

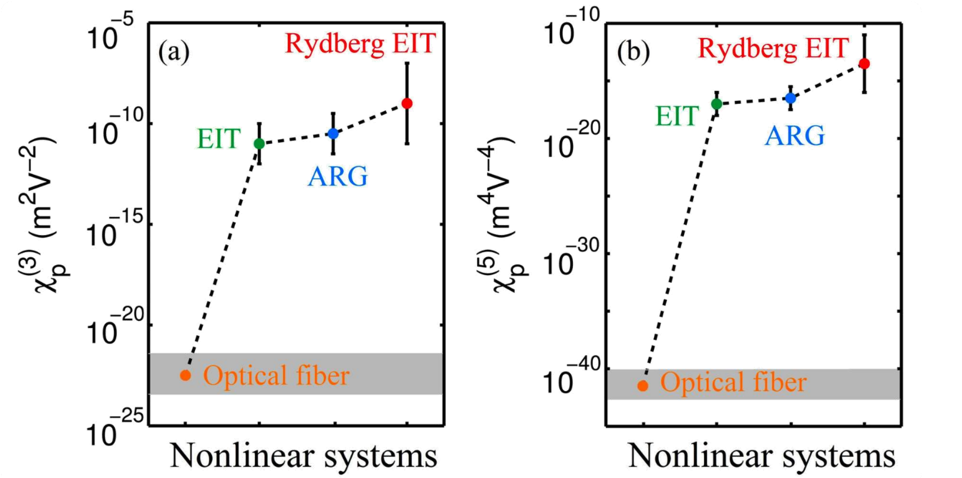


Rev. Mod. Phys. 77, 633-673 (2005)



Phys. Rev. Lett. 105, 193603 (2010)

非线性响应：$N = 1.0 \times 10^9 \sim 6.0 \times 10^{10}\ cm^{-3}$



Opt. Express 24, 4442 (2016)

背景：空间Kramers-Kronig关系

频率域：

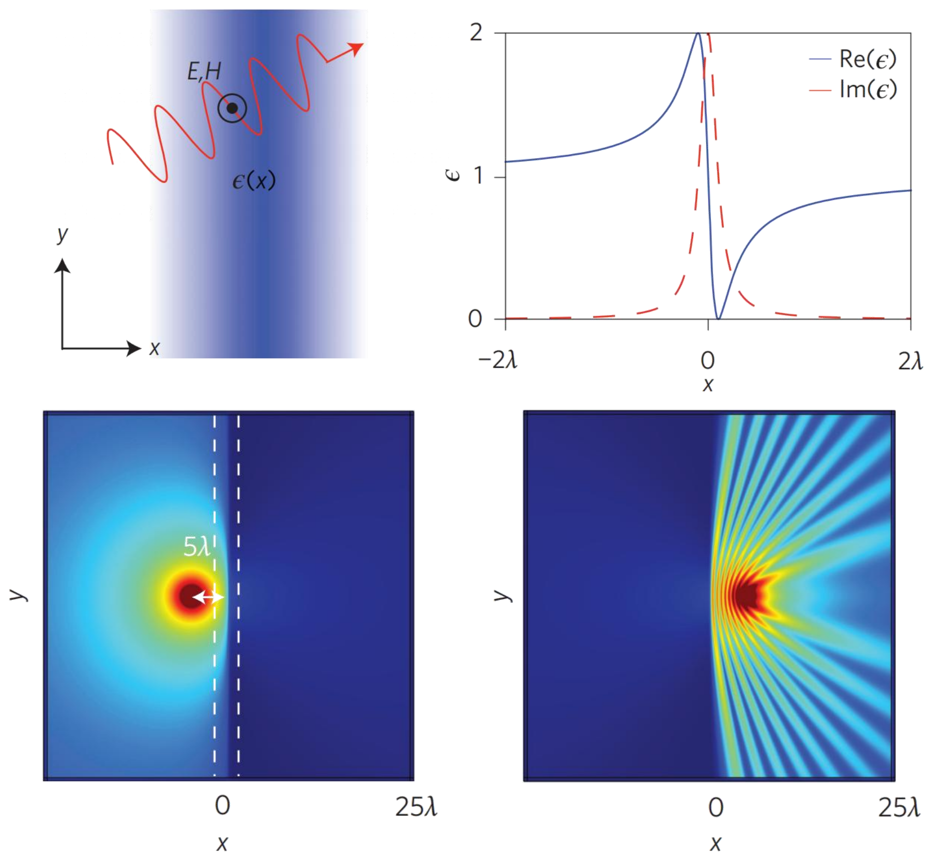
$$\mathrm{Re}[\chi(\omega)] = \frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\left[\chi\left(\omega'\right)\right]}{\omega'-\omega}d\omega'$$

$$\mathrm{Im}[\chi(\omega)] = -\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\left[\chi\left(\omega'\right)\right]}{\omega'-\omega}d\omega'$$

空间域：

$$\mathrm{Re}[\chi(x)] = \frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\left[\chi\left(x'\right)\right]}{x'-x}dx'$$

$$\mathrm{Im}[\chi(x)] = -\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\left[\chi\left(x'\right)\right]}{x'-x}dx'$$



- 实部和虚部不同步（out of phase）

✓ 不依赖增益/损耗区域

✓ 不需要磁性、各向异性

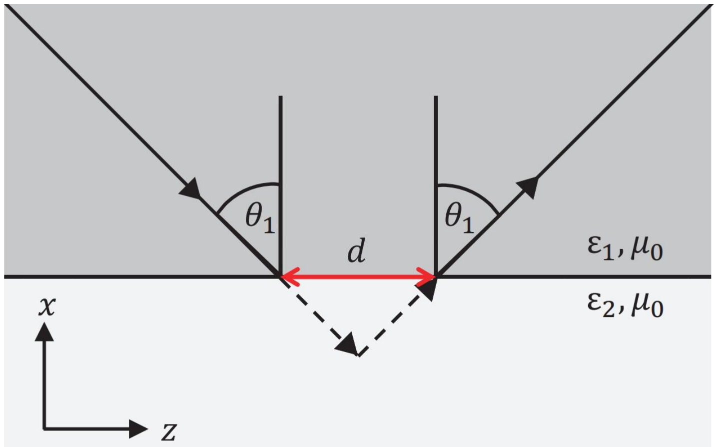
× 动态调控

- 设计光隔离器、芯片集成光学器件和隐形斗篷等器件

Laser Photonics Rev. 11, 1600253 (2016)

Nat Commun 8, 51 (2017)

背景：Goos-Hänchen横向位移



Ann. Phys. 436, 333 (1947)

静态相位理论计算GH位移：

k：波矢

θ：入射角

ϕ：反射系数的相位

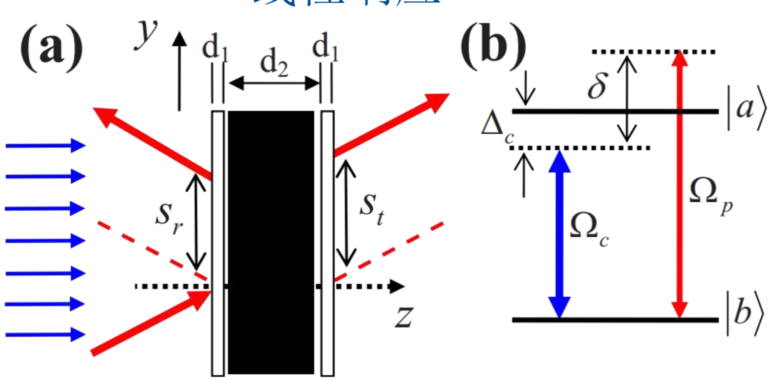
$$S = -\frac{1}{k}\frac{\partial \phi}{\partial \theta}$$

Ann. Phys. 437, 87 (1948)

- 开发光学开关、光学传感器和光束转向器等器件

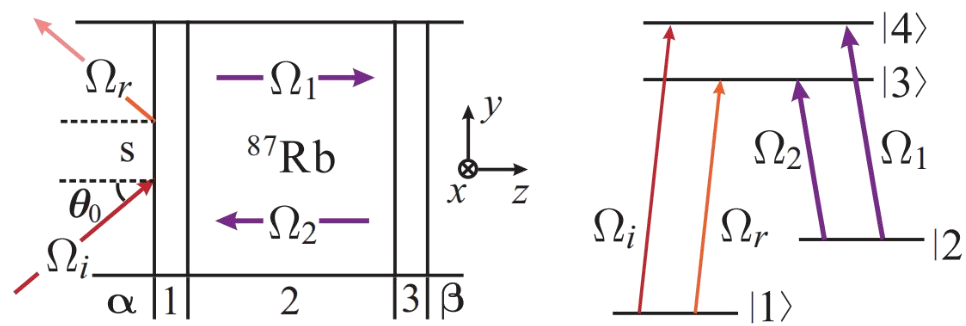
J. Opt. 18, 025612 (2016)
Physical Review A 101, 023837 (2020)
Nanophotonics 11, 4531-4536 (2022)
Opt. Lett. 48, 1710 (2023)

线性响应



Phys. Rev. A 77, 023811 (2008)

非线性效应（四波混频）

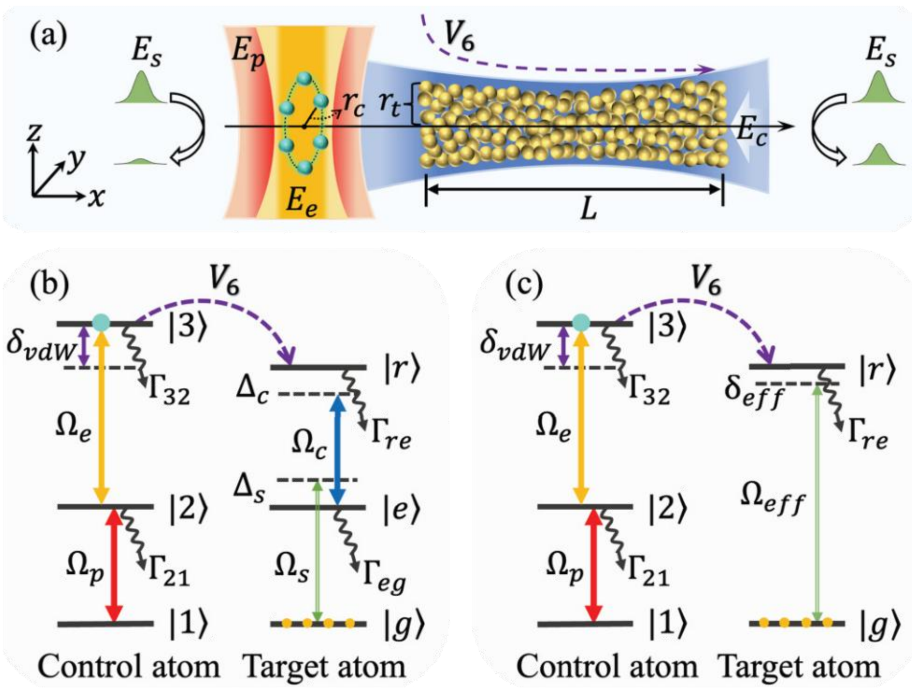


Phys. Rev. A 91, 033831 (2015)

✓ 在非线性介质表面，GH位移具有更加有趣的物理特性和更多的操控方式

× 大多数光学材料中场的非线性响应太弱

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射



$$\mathcal{V}_6 = \sum_{k=1}^{6} \frac{C_6}{[(x-x_0)^2 + |\vec{r}-\vec{r}_k|^2]^3}$$

控制原子和目标原子的哈密顿量：

$$H_c/\hbar = -\Delta_p\sigma_{22} - (\Delta_p + \Delta_e + \delta_{vdW})\sigma_{33} - \Omega_p\sigma_{21} - \Omega_e\sigma_{32} - \Omega_p^*\sigma_{12} - \Omega_e^*\sigma_{23}$$

$$H_t/\hbar = -\Delta_s\sigma_{ee} - (\Delta_s + \Delta_c)\sigma_{rr} - \Omega_s\sigma_{eg} - \Omega_c\sigma_{re} - \Omega_s^*\sigma_{ge} - \Omega_c^*\sigma_{er} + \mathcal{V}_6\sigma_{33}\sigma_{rr}$$

主方程： $\partial_t\rho = -i[H/\hbar, \rho] + \mathcal{L}(\rho)$

$$\mathcal{L}(\rho) = \sum \Gamma_{\mu\nu}[\sigma_{\nu\mu}\rho\sigma_{\mu\nu} - \frac{1}{2}(\rho\sigma_{\mu\nu}\sigma_{\nu\mu} + \sigma_{\mu\nu}\sigma_{\nu\mu}\rho)]$$

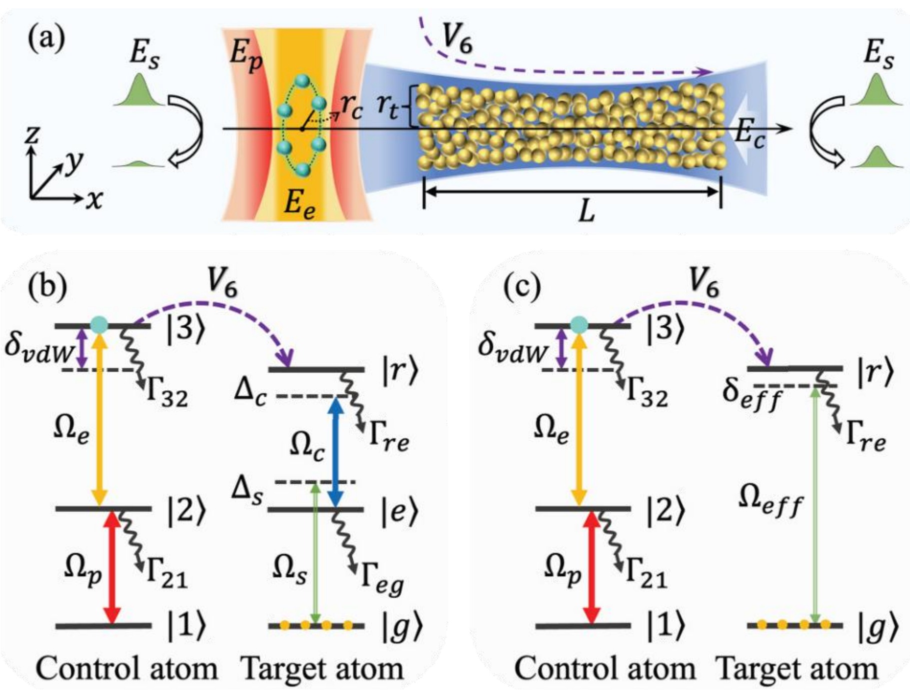
控制原子： $\Delta_p = \Delta_e + \delta_{vdW} = 0$

$$\Omega_p \geq \Omega_e > \gamma_{21} \gg \gamma_{31}$$

$$\rho_{33} \simeq \frac{(\gamma_{21}+\gamma_{31})\Omega_p^2\Omega_e^2}{\gamma_{21}\Omega_e^4 + (\gamma_{21}+3\gamma_{31})\Omega_p^2\Omega_e^2 + \gamma_{21}^2\gamma_{31}\Omega_e^2}$$

$$\simeq \Omega_p^2/(\Omega_p^2+\Omega_e^2) \Rightarrow \mathbf{1}$$

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射



控制原子和目标原子的哈密顿量：

$$H_c/\hbar = -\Delta_p\sigma_{22} - \left(\Delta_p + \Delta_e + \delta_{vdW}\right)\sigma_{33} - \Omega_p\sigma_{21} - \Omega_e\sigma_{32} - \Omega_p^*\sigma_{12} - \Omega_e^*\sigma_{23}$$

$$H_t/\hbar = -\Delta_s\sigma_{ee} - (\Delta_s + \Delta_c)\sigma_{rr} - \Omega_s\sigma_{eg} - \Omega_c\sigma_{re} - \Omega_s^*\sigma_{ge} - \Omega_c^*\sigma_{er} + \mathcal{V}_6\sigma_{33}\sigma_{rr}$$

主方程： $\partial_t\rho = -i[H/\hbar, \rho] + \mathcal{L}(\rho)$

$$\mathcal{L}(\rho) = \sum \Gamma_{\mu\nu}\left[\sigma_{\nu\mu}\rho\sigma_{\mu\nu} - \frac{1}{2}(\rho\sigma_{\mu\nu}\sigma_{\nu\mu} + \sigma_{\mu\nu}\sigma_{\nu\mu}\rho)\right]$$

$$\mathcal{V}_6 = \sum_{k=1}^{6} \frac{C_6}{[(x - x_0)^2 + |\vec{r} - \vec{r}_k|^2]^3}$$

目标原子：

$$\Delta_s \simeq -\Delta_c$$

$$|\Delta_s| \gg \gamma_{eg} \gg \Omega_s$$

$$|\Delta_c| \gg \Omega_c \gg \gamma_{re}$$

$$\rho_{rr} = \frac{2\gamma_{rg}\Omega_{eff}^2}{\Gamma_{re}\left[\gamma_{rg}^2 + (\delta_{eff} + \mathcal{V}_6\rho_{33})^2\right] + 4\gamma_{rg}\Omega_{eff}^2}$$

$$\rho_{gg} = \frac{\Gamma_{re}\left[(\delta_{eff} + \mathcal{V}_6\rho_{33})^2 + \gamma_{rg}^2\right] + 2\gamma_{rg}\Omega_{eff}^2}{\Gamma_{re}\left[(\delta_{eff} + \mathcal{V}_6\rho_{33})^2 + \gamma_{rg}^2\right] + 4\gamma_{rg}\Omega_{eff}^2}$$

$$\rho_{eg} = -(\Omega_c^*/\Delta_s)\rho_{rg} - (\Omega_s/\Delta_s)\rho_{gg}$$

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射

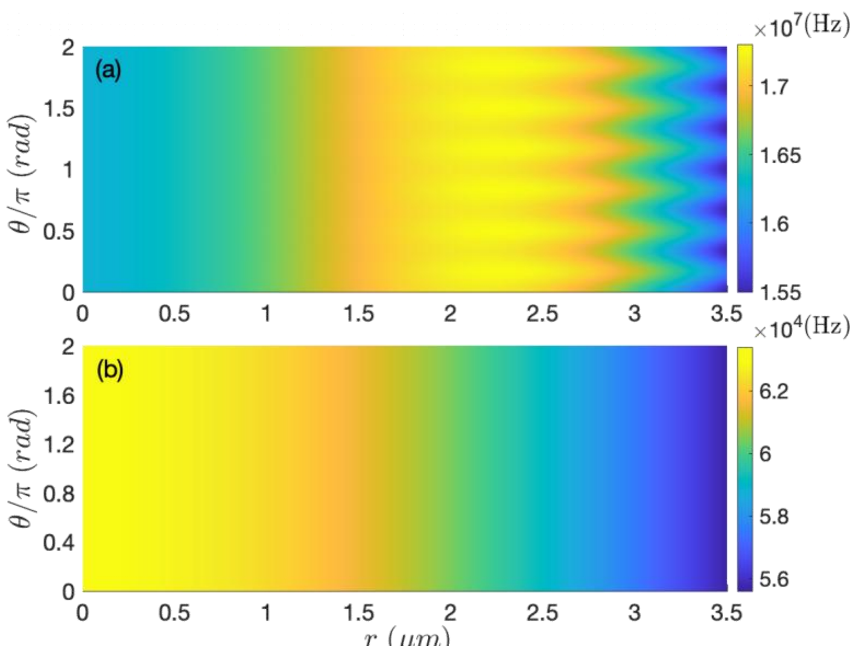
采取近似：$\gamma_{rg}\Gamma_{re} \gg 4\Omega_{eff}^2$

信号场的极化率：

$$\chi_s = \frac{N_0\wp_{eg}^2}{\varepsilon_0\hbar}\frac{\rho_{eg}}{\Omega_s} = \frac{N_0\wp_{ge}^2}{\hbar\varepsilon_0}\left[\frac{\Omega_c^2}{\Delta_s\Delta_c}\frac{\delta_{eff}+\mathcal{V}_6\rho_{33}-i\gamma_{rg}}{\gamma_{rg}^2+\left(\delta_{eff}+\mathcal{V}_6\rho_{33}\right)^2}-\frac{1}{\Delta_s}\right]$$

$$\delta_{eff} = \Delta_s + \Delta_c - \Delta_{e1} - \Delta_{e2}$$

$$\Delta_{e1} = \Omega_c^2/\Delta_s \quad \Delta_{e2} = \Omega_s^2/\Delta_c$$



$$\frac{s_{max}-s_{min}}{s_{max}+s_{min}} \sim 6\%$$

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射

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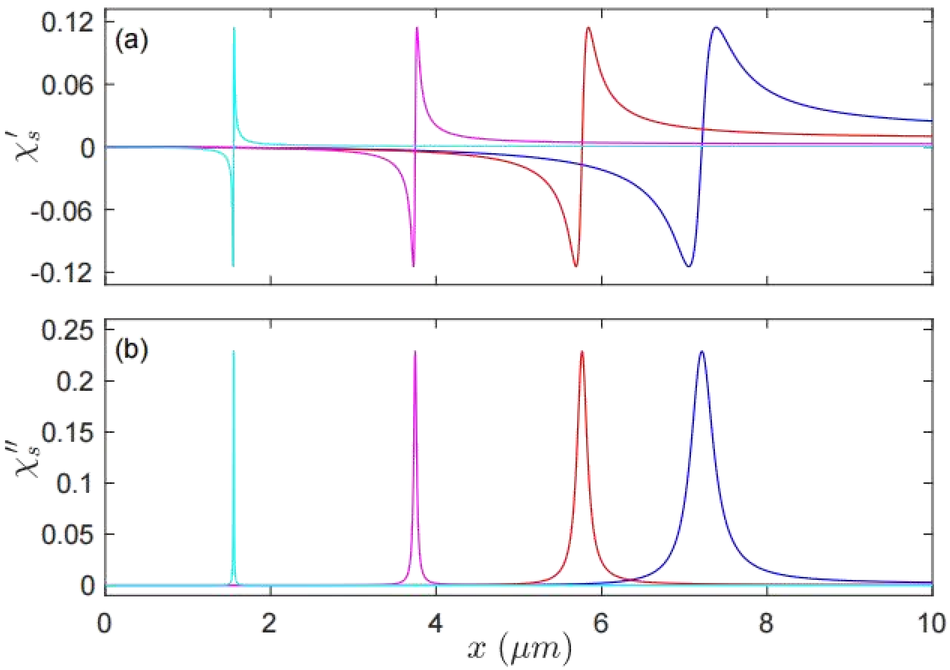

$$D_{kk}^{I}=\frac{\int_0^L\left[|\chi_s''(x)|-\left|\frac{1}{\pi}\mathrm{P}\int_0^L\frac{\chi_s'(\xi)}{\xi-x}d\xi\right|\right]dx}{\int_0^L|\chi_s''(x)|dx}$$

$$D_{kk}^{R}=\frac{\int_0^L\left[|\chi_s'(x)|-\left|\frac{1}{\pi}\mathrm{P}\int_0^L\frac{\chi_s''(\xi)}{x-\xi}d\xi\right|\right]dx}{\int_0^L|\chi_s'(x)|dx}$$

➢ $D_{kk}^{I,R}\rightarrow 0$ **完美空间KK关系**

$\delta_{eff}/2\pi$
$=-5.0\mathrm{MHz},-1.2\mathrm{MHz},-0.4\mathrm{MHz},-0.2\mathrm{MHz}$

$$x_c=x_0+\left(-C_6/\delta_{eff}\right)^{1/6}$$

$$\delta x_{\pm}=x_0-x_c+\left[-C_6/\left(\delta_{eff}\mp\gamma_{rg}\right)\right]^{1/6}$$

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射

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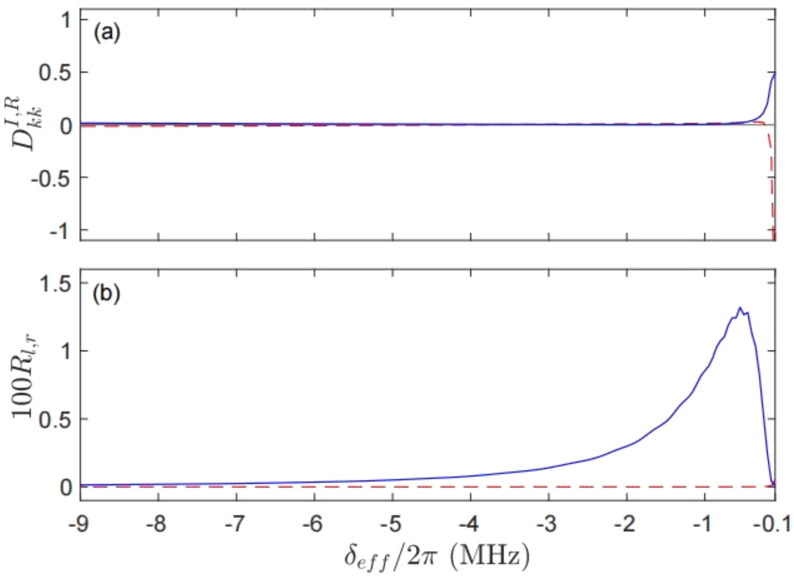
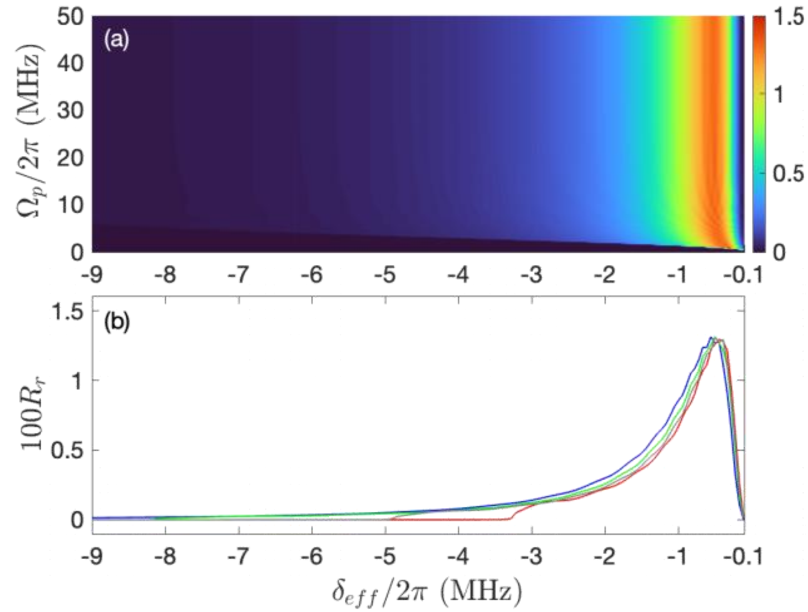
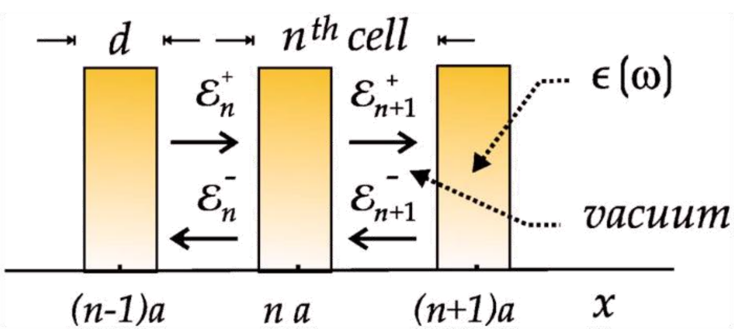


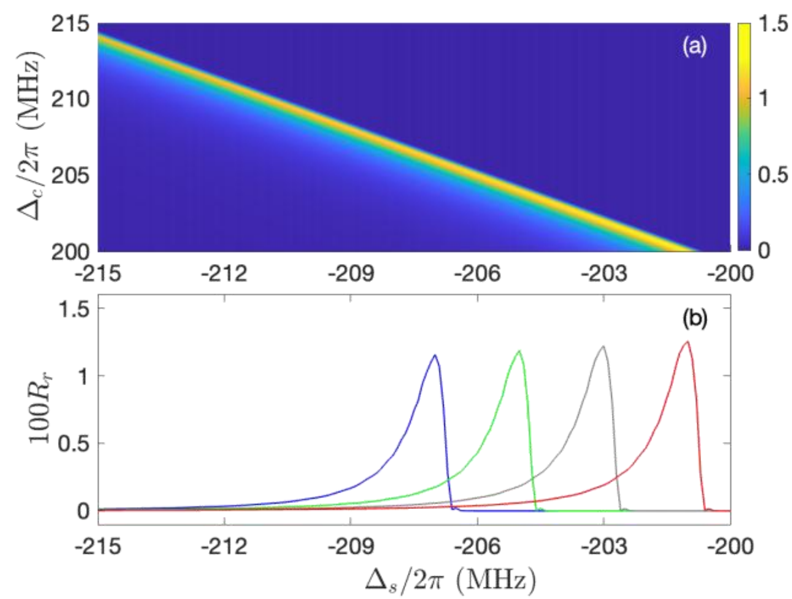


Ω_p



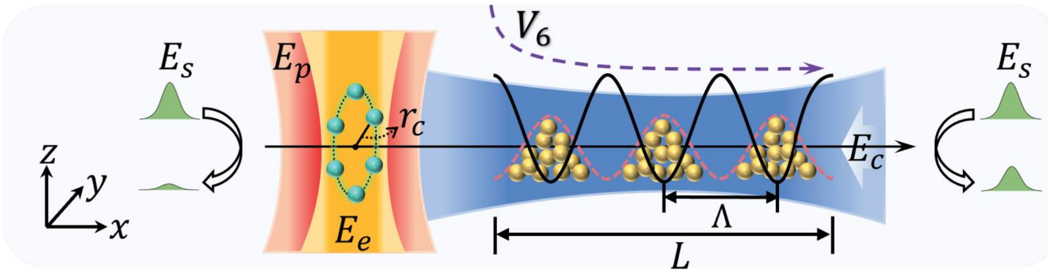
传输矩阵法是处理光场在周期介质中传播的问题中所使用的的一种常见的方法。

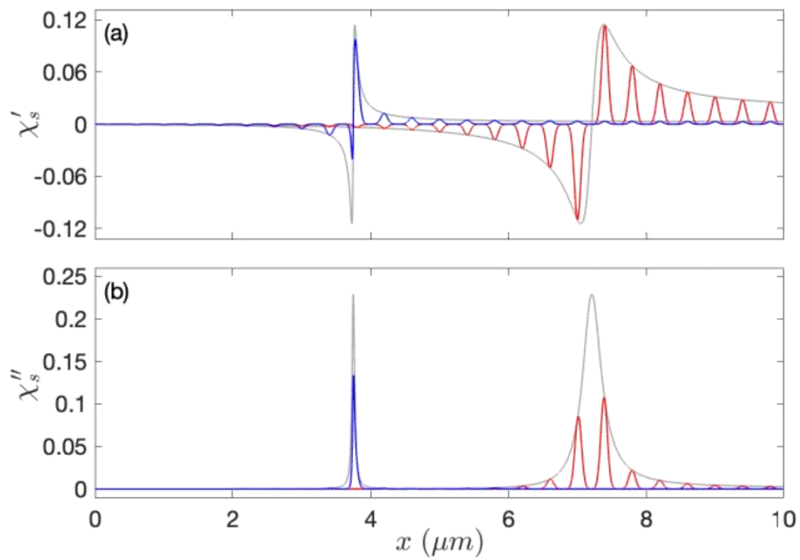
Phys. Rev. E **72**, 046604 (2005)



Δ_c

里德堡相互作用引起的空间Kramers-Kronig关系和单向反射





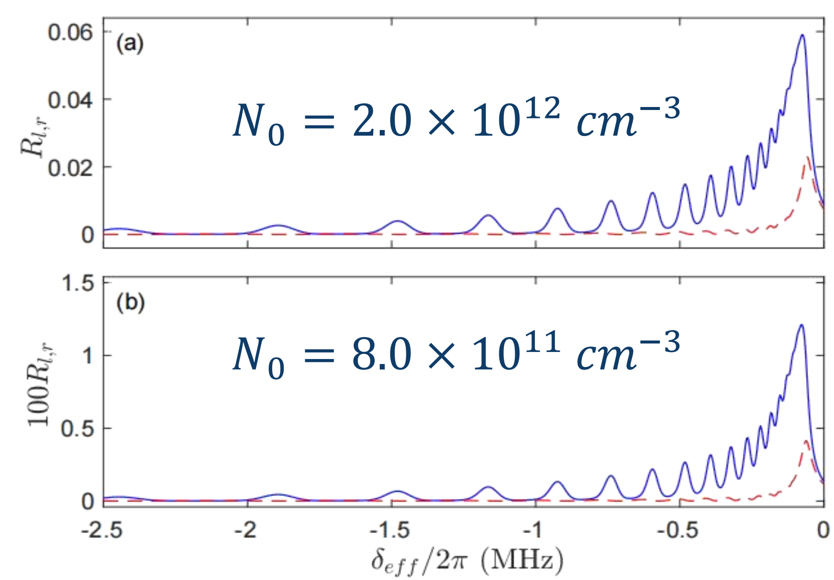
目标原子分布在周期晶格中

$$N(x) = \sum_{k=1}^{K} N_k(x)$$

$$= \sum_{k=1}^{K} \frac{\Lambda N_0}{\delta x\sqrt{\pi}} e^{-(x-x_k)^2/\delta x^2}$$

$$x_k = (k - 1/2)\Lambda$$

$$\Lambda = 400\ nm \qquad L/\Lambda = 25$$



里德堡原子气体中由原子关联增强的 Goos-Hänchen横向位移

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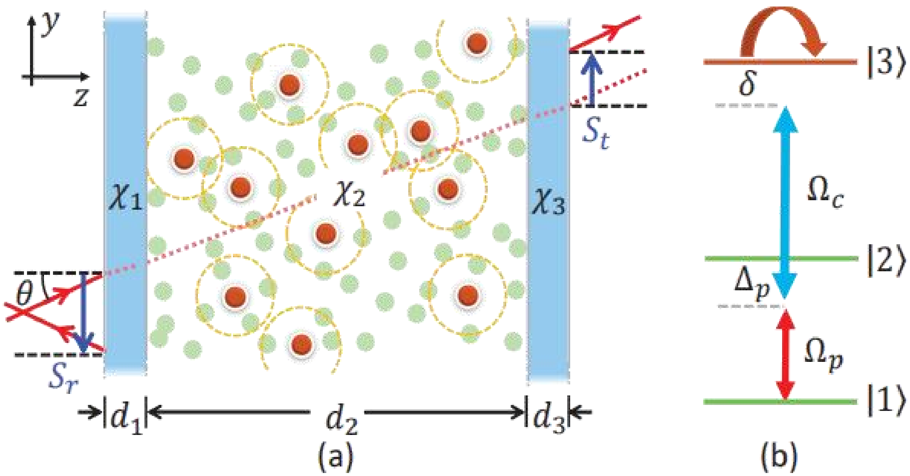


- ^{87}Rb原子

$$|1\rangle \equiv |5S_{1/2}, F=2\rangle$$
$$|2\rangle \equiv |5P_{3/2}, F=3\rangle$$
$$|3\rangle = |60S_{1/2}\rangle$$
$$d_1 = d_3 = 5\ \mu\mathrm{m}$$
$$\chi_1 = \chi_3 = 1.22$$

传输矩阵法计算反射系数r和透射系数t：

$$r = \frac{q_0(M_{22} - M_{11}) - (q_0^2 M_{12} - M_{21})}{q_0(M_{22} + M_{11}) - (q_0^2 M_{12} + M_{21})}$$

$$t = \frac{2q_0}{q_0(M_{22} + M_{11}) - (q_0^2 M_{12} + M_{21})}$$

$$M_j = \begin{bmatrix} cos(k_z^j d_j) & \dfrac{isin(k_z^j d_j)}{q_j} \\ iq_j sin(k_z^j d_j) & cos(k_z^j d_j) \end{bmatrix}$$

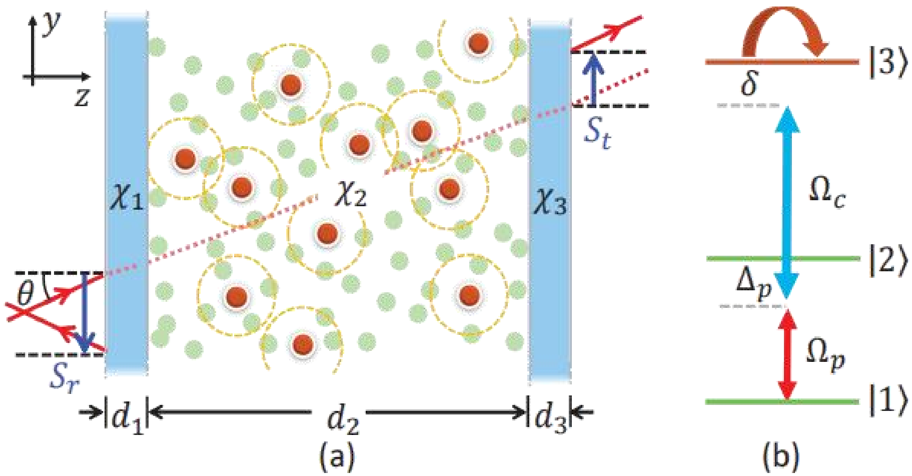
$$k_z^j = k_p(\epsilon_j - \sin^2\theta)^{1/2} \qquad \epsilon_j = 1 + \chi_j$$

$$S_{r,t} = -\frac{c}{\omega_p cos\theta}\frac{\partial\phi_{r,t}}{\partial\theta}$$

$\phi_{r,t}$：r和t的相位

$\theta(\omega_p)$：探测场的入射角（角频率）

里德堡原子气体中由原子关联增强的 Goos-Hänchen横向位移



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$$|3\rangle = |60S_{1/2}\rangle$$
$$d_1 = d_3 = 5\ \mu\text{m}$$
$$\chi_1 = \chi_3 = 1.22$$

里德堡原子气体层的哈密顿量： $\hat{H} = N_a \int d^3 r \hat{\mathcal{H}}(\mathbf{r})$

$$\hat{\mathcal{H}}(\mathbf{r})/\hbar = \Delta_p \hat{S}_{22}(\mathbf{r}) + \delta \hat{S}_{33}(\mathbf{r}) - [\Omega_p \hat{S}_{12}(\mathbf{r}) + \Omega_c \hat{S}_{23}(\mathbf{r}) + \mathrm{H.c.}] + N_a \int d^3 r' \hat{S}_{33}(\mathbf{r}') V(\mathbf{r}' - \mathbf{r}) \hat{S}_{33}(\mathbf{r})$$

$$\delta = \omega_{31} - \omega_c - \omega_p$$
$$V(\mathbf{r} - \mathbf{r}') = C_6 / |\mathbf{r} - \mathbf{r}'|^6$$

主方程： $i\hbar \partial_t \langle \hat{S}_{\alpha\beta}(\mathbf{r}) \rangle = \langle [\hat{S}_{\alpha\beta}(\mathbf{r}), \hat{\mathcal{H}}(\mathbf{r})] \rangle + \mathcal{L}(\rho)$

$$\mathcal{L}(\rho) = \sum \Gamma_{\mu\nu} [\sigma_{\nu\mu} \rho \sigma_{\mu\nu} - \frac{1}{2} (\rho \sigma_{\mu\nu} \sigma_{\nu\mu} + \sigma_{\mu\nu} \sigma_{\nu\mu} \rho)]$$

里德堡原子气体中由原子关联增强的Goos-Hänchen横向位移

$$\partial_t \rho_{21}(\mathbf{r}) = -g_{21}\rho_{21}(\mathbf{r}) + i\Omega_c^* \rho_{31}(\mathbf{r}) + i\Omega_p[\rho_{11}(\mathbf{r}) - \rho_{22}(\mathbf{r})]\#$$

$$\partial_t \rho_{31}(\mathbf{r}) = -g_{31}\rho_{31}(\mathbf{r}) + i\Omega_c \rho_{21}(\mathbf{r}) - i\Omega_p \rho_{32}(\mathbf{r}) \; - iN_a \int d^3 r' V(\mathbf{r}' - \mathbf{r}) \rho_{33,31}(\mathbf{r}', \mathbf{r})$$

两体动力学方程：$\partial_t \langle \hat{S}_{\alpha\beta}(\mathbf{r}')\hat{S}_{\mu\nu}(\mathbf{r})\rangle = \langle \partial_t \hat{S}_{\alpha\beta}(\mathbf{r}')\hat{S}_{\mu\nu}(\mathbf{r})\rangle + \langle \hat{S}_{\alpha\beta}(\mathbf{r}')\partial_t \hat{S}_{\mu\nu}(\mathbf{r})\rangle$

$$\boxed{\begin{aligned}\partial_t \rho_{33,31}(\mathbf{r}', \mathbf{r}) = &-[g_{33,31} + iV(\mathbf{r}' - \mathbf{r})]\rho_{33,31}(\mathbf{r}', \mathbf{r}) + i\Omega_c[\rho_{33,21}(\mathbf{r}', \mathbf{r}) + \rho_{31,23}(\mathbf{r}', \mathbf{r})] \\ &- i\Omega_c^* \rho_{32,31}(\mathbf{r}', \mathbf{r}) - i\Omega_p \rho_{33,32}(\mathbf{r}', \mathbf{r}) \cancel{- iN_a \int d^3 r'' V(\mathbf{r}' - \mathbf{r}) \rho_{33,33,31}}(\mathbf{r}'', \mathbf{r}', \mathbf{r})\,\#\end{aligned}}$$

微扰法求解：

$$\rho_{\alpha\beta}(\mathbf{r}) = \sum_{n=0}^{\infty} \rho_{\alpha\beta}^{(n)}(\mathbf{r}) \qquad \rho_{\alpha\beta,\mu\nu}(\mathbf{r}', \mathbf{r}) = \sum_{n=0}^{\infty} \rho_{\alpha\beta,\mu\nu}^{(n)}(\mathbf{r}', \mathbf{r})$$

单体一阶解：

$$\chi_2^{(1)} = \frac{N_a \mu_{21}^2}{\epsilon_0 \hbar} \frac{\rho_{21}^{(1)}}{\Omega_p} = \frac{N_a \mu_{21}^2}{\epsilon_0 \hbar} \frac{i g_{31}}{g_{21} g_{31} + |\Omega_c|^2}$$

$$g_{31} = \gamma_{31} + i\delta$$
$$g_{21} = \gamma_{21} + i\Delta_p$$

单体三阶解：

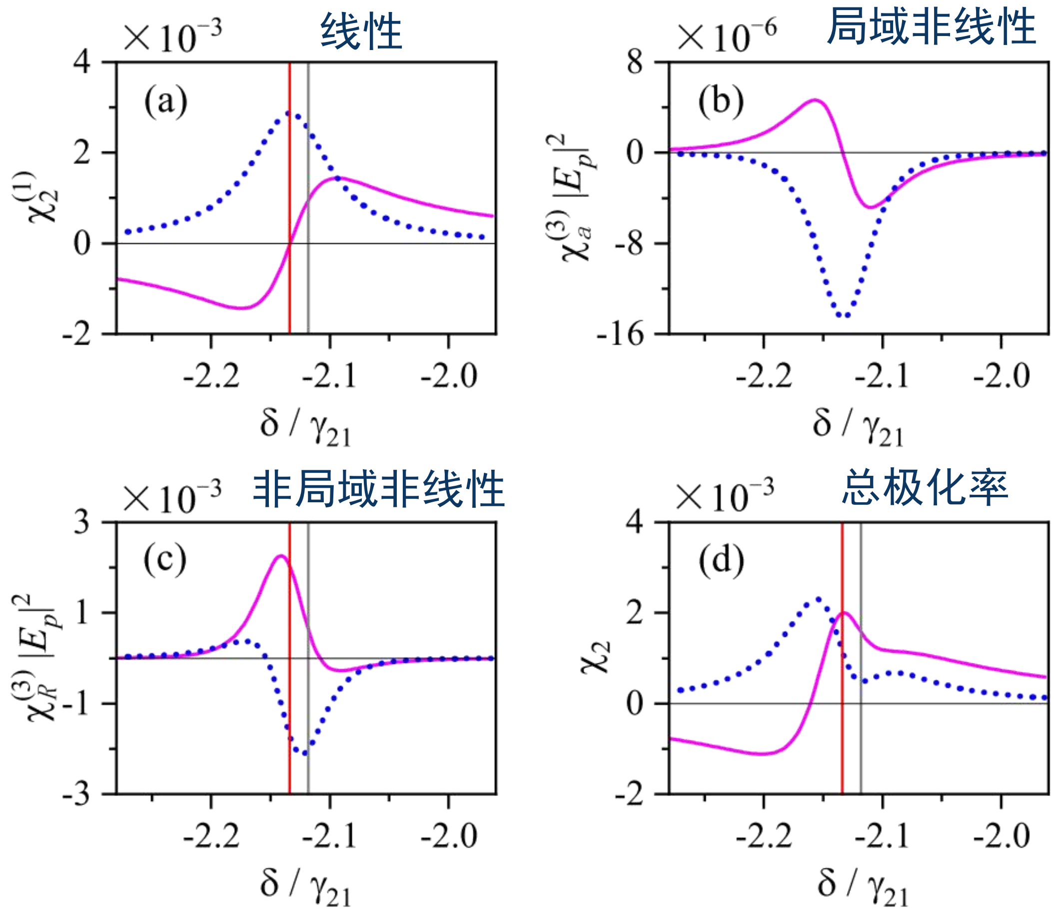
$$\chi_2^{(3)} = \frac{N_a \mu_{21}^4}{4\epsilon_0 \hbar^3} \frac{\rho_{21}^{(3)}}{|\Omega_p|^2 \Omega_p} = \chi_a^{(3)} + \chi_R^{(3)}$$

局域项：$\chi_a^{(3)}$

非局域项：$\chi_{\mathrm{R}}^{(3)}$

探测场总光学响应： $\chi_2 = \chi_2^{(1)} + \chi_a^{(3)}|E_p|^2 + \chi_R^{(3)}|E_p|^2$

里德堡原子气体中由原子关联增强的 Goos-Hänchen横向位移



1、红色竖线$\delta = -2.13\gamma_{21}$ $\chi_2 \simeq 0.002 + 0.001i$

2、灰色竖线$\delta = -2.12\gamma_{21}$ $\chi_2 \simeq 0.002 + 5 \times 10^{-4}i$

里德堡原子气体中由原子关联增强的 Goos-Hänchen横向位移

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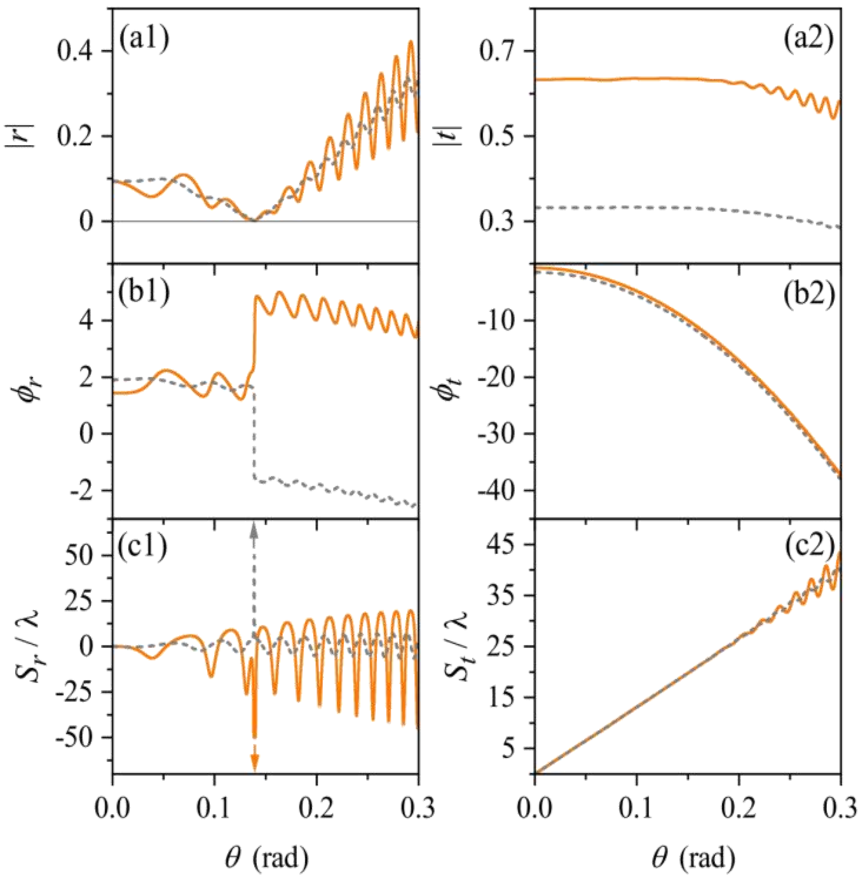
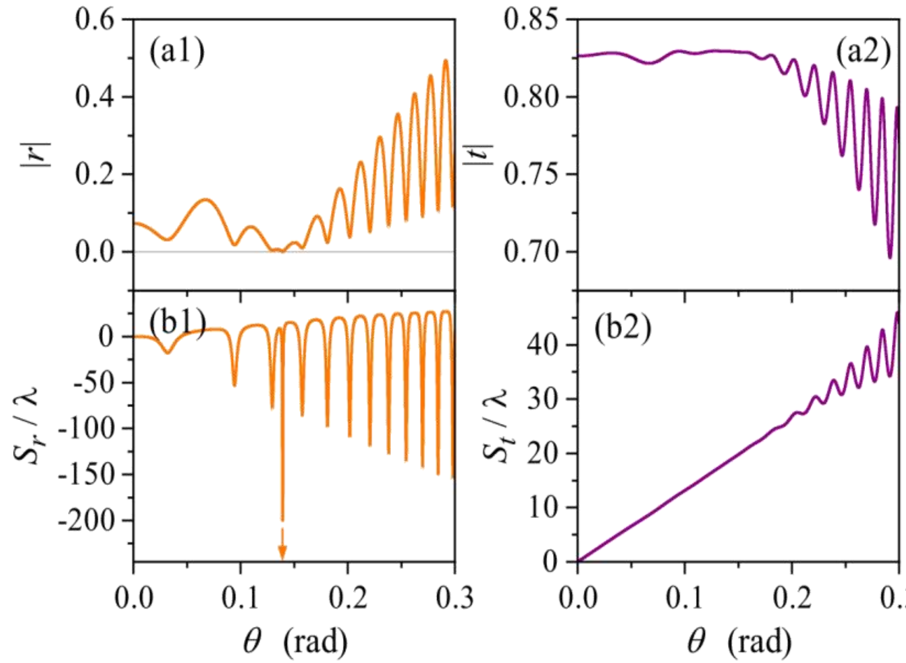


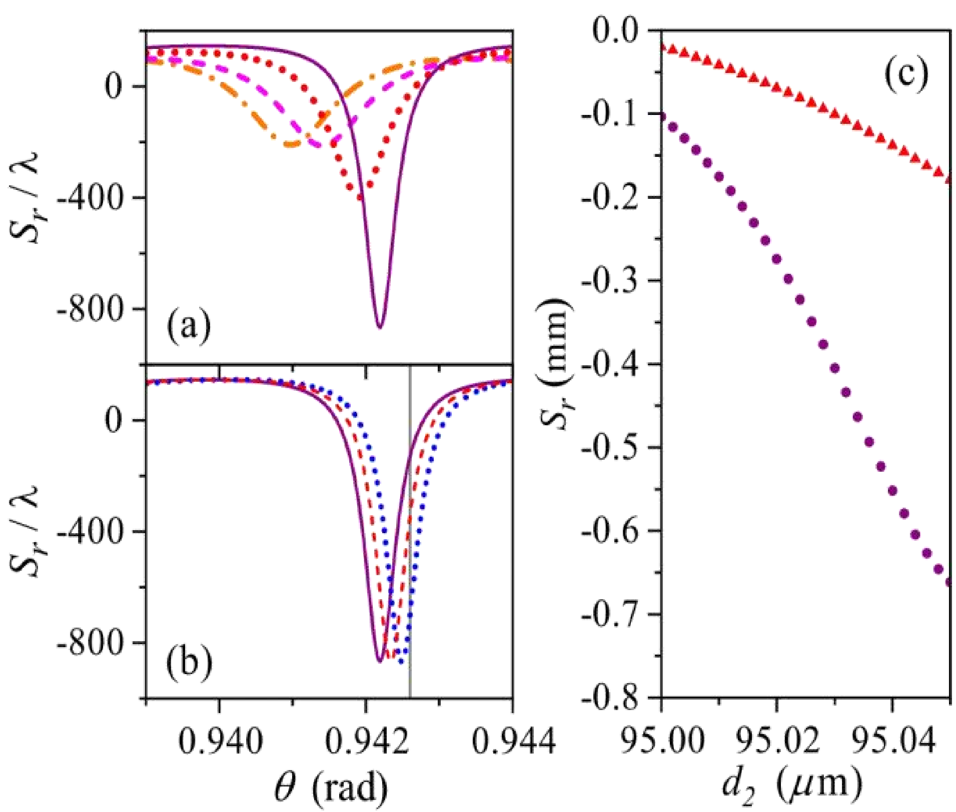


橙色实线：包含里德堡原子关联项
灰色虚线：不包含里德堡原子关联项

里德堡原子气体中由原子关联增强的 Goos-Hänchen横向位移

$$\chi_R^{(3)} = \frac{N_a^2 \mu_{21}^4}{4\epsilon_0 \hbar^3} \frac{\Omega_c^*}{\mathcal{P}_1} \int d^3 r' V(\boldsymbol{r}' - \boldsymbol{r}) \frac{\rho_{33,31}^{(3)}(\boldsymbol{r}', \boldsymbol{r})}{|\Omega_p|^2 \Omega_p}$$

灰色竖线$\delta =- 2.12\gamma_{21}$　$\chi_2 \simeq 0.002 + 5 \times 10^{-4} i$



(a) $N_a = 2N_0$、$3N_0$、$4N_0$、$4.5N_0$

(b) $N_a = 4.5N_0$
$d_2 = 95.00\ \mu m$, $95.02\mu m$, $95.04\mu m$

(c) $N_a = 4.0 \times 10^{10}\ \mathrm{cm}^{-3}$
$N_a = 4.5 \times 10^{10}\ \mathrm{cm}^{-3}$

总结与展望

里德堡原子系统

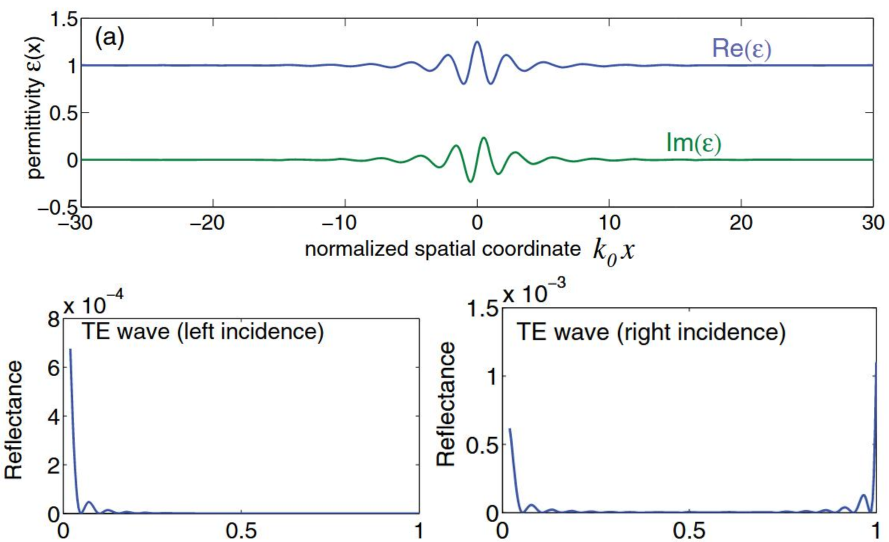
长程可调控非局域偶极-偶极相互作用

空间KK介质

实现了可调控的光场不对称反射和单向无反射行为，并通过协同周期原子晶格实现了单侧反射的增强

克尔非线性介质

实现了反射和透射增强的GH位移，并通过调节原子密度和介质长度实现了高灵敏位移传感器



Opt. Lett. 41, 3727 (2016)

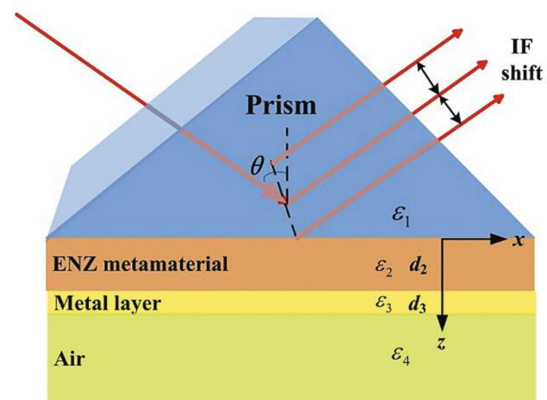


Fig. 1. Illustration of IF effect of reflected light from prism-coupling waveguide with ENZ metamaterial.

Superlattices Microstruct. 120, 766-770 (2018)

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Thank you!