



中国科学院半导体研究所

Institute of Semiconductors, CAS



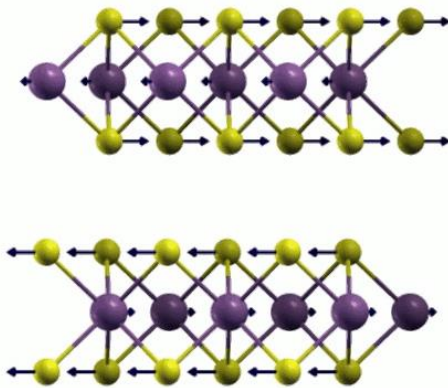
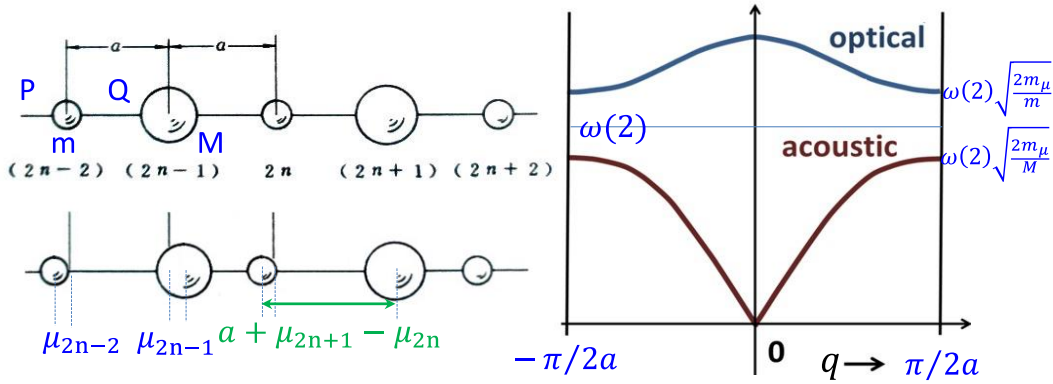
# 电声子耦合的光学探测和调控

张俊

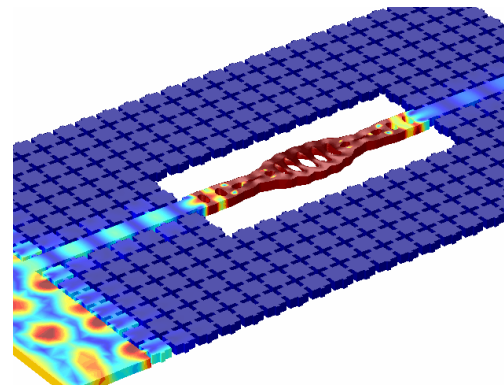
State Key Laboratory of Superlattices and Microstructures  
Institute of Semiconductors, CAS

计科中心-2024-05-13

# 声子



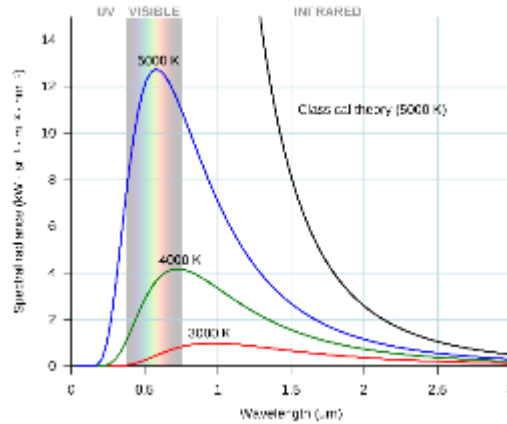
Lattice Vibration



Mechanical Oscillator

# 温度、声子

温度是表示物体冷热程度的物理量，微观上来讲是物体分子热运动的剧烈程度。



$$I(\nu, T) = \frac{2h\nu^3}{c^2} \frac{1}{e^{\frac{h\nu}{kT}} - 1}$$



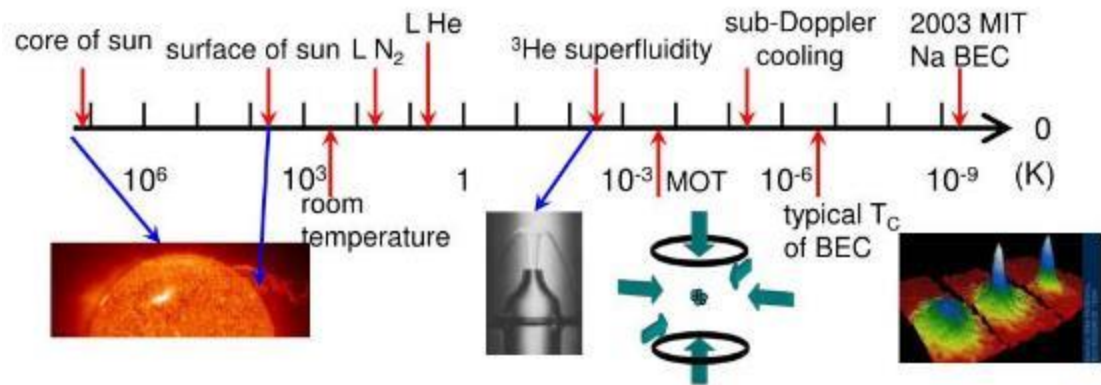
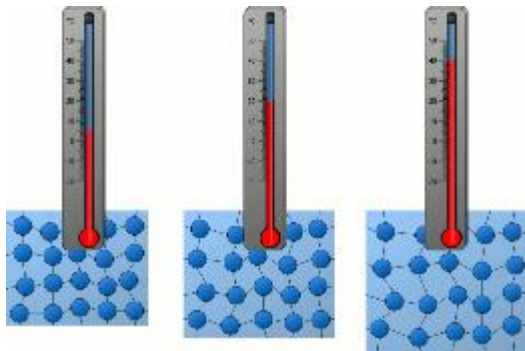
普朗克



爱因斯坦



德拜



➤ 声子是原子/分子热振动的量子，是一种准粒子，是波色子。

# 热能是生命之源

- ▶ 地球在与太阳合适的距离和大气环境下，拥有了适宜孕育生命的温度环境，太阳的热量源源不断的为地球生命提供能量
- ▶ 原始人自从学会了钻木取火，知道了如何获取热能，促进了人类的进化和文明的发展
- ▶ 人类（动物）消耗食物转化为热量，保持适宜的体温，对抗熵增

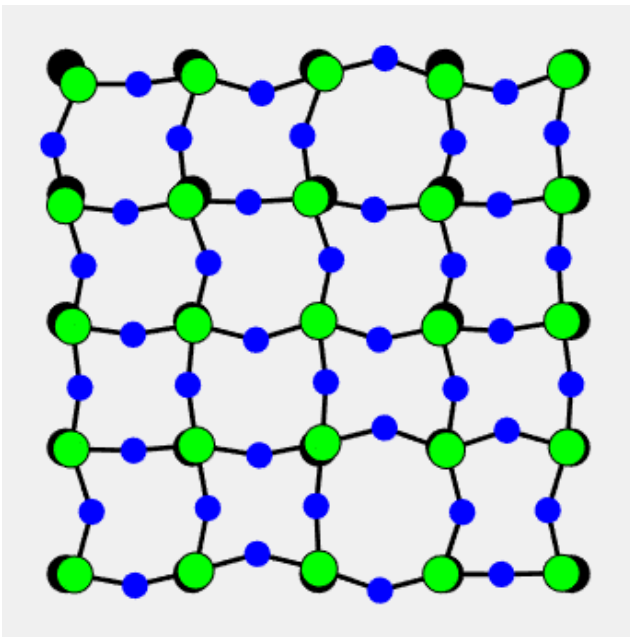


# 声子是简单而完美的新世界

- 声子是原子/分子热振动的量子，是一种准粒子，是波色子。
- 随着温度的降低，原子或分子会形成晶体或者超流体，在极低温下，唯一的低能激发就是声子，声子是“简单而完美”的新世界！（文小刚，量子多体理论，科学出版社）

$$E_n = \left(n + \frac{1}{2}\right) h\nu$$

零点能



## Zero Point Energy

*(Emerging science, 1948...)*

**What?**

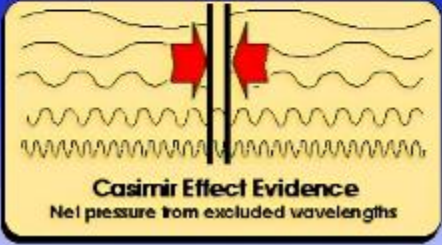
- Random Electromagnetic waves remain after all energy is removed
- Enormous energy density:  $10^{24}$  to  $10^{50}$  Joules/m<sup>3</sup>
- Theorized to indirectly cause gravity and inertia

**Why?**

- As an energy source?
- As a reactive medium?

**Evidence?**

- Casimir Effect
- Planck blackbody spectrum
- quantum effects

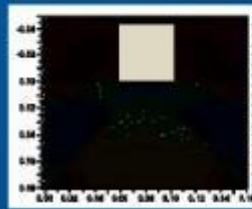


**Casimir Effect Evidence**  
Net pressure from excluded wavelengths

# 声子成为后摩尔器件关键科学问题

## Optimizing Choices for Transistors on Multiple Fronts

Increasing  
MOBILITY



Strain



Ge



III-V



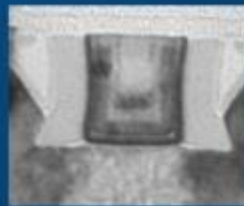
CNT



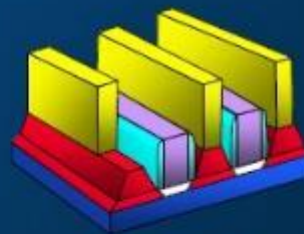
Graphene

The

Increasing  
COUPLING



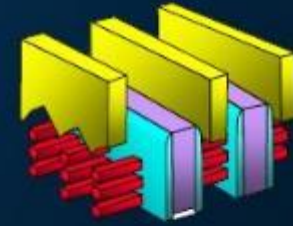
Planar  
With High K



UTB SOI  
(or QW)



Fins



Wires/Ribbons

of feature size,  $a$ , at different temperatures.

Transistor size

# 声子是一种重要的准粒子

HOME > CUSTOM PUBLISHING > BOOKLETS > 125 QUESTIONS: EXPLORATION AND DISCOVERY

Produced by the Science/AAAS  
Custom Publishing Office

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Science | AAAS

## 125 questions: Exploration and discovery

14 MAY 2021

### Physics

#### What are the limits of heat transfer in matter?

Materials science research may help us address the challenge of high-temperature energy transfer.

#### What is the microscopic mechanism for high-temperature superconductivity?

#### What are the fundamental principles of collective motion?

Despite differences in the length scales and

- 物质的传热极限 (声子-声子散射)
- 高温超导微观机理 (电子-声子耦合。。。)
- 集体激发的基本原理 (声子是原子振动的集体激发)

### Information Science

#### Is there an upper limit to computer processing speed?

One of the most well-known principles of computing design and power is Moore's Law, named after the co-founder of Intel Corporation, Gordon Moore. He predicted in the 1950s that the number of transistors on a chip would double every two years. Moore had built a plan to achieve a consistent exponential growth in the number of transistors on a chip.

- 计算机处理速度上限 (电子-声子散射)

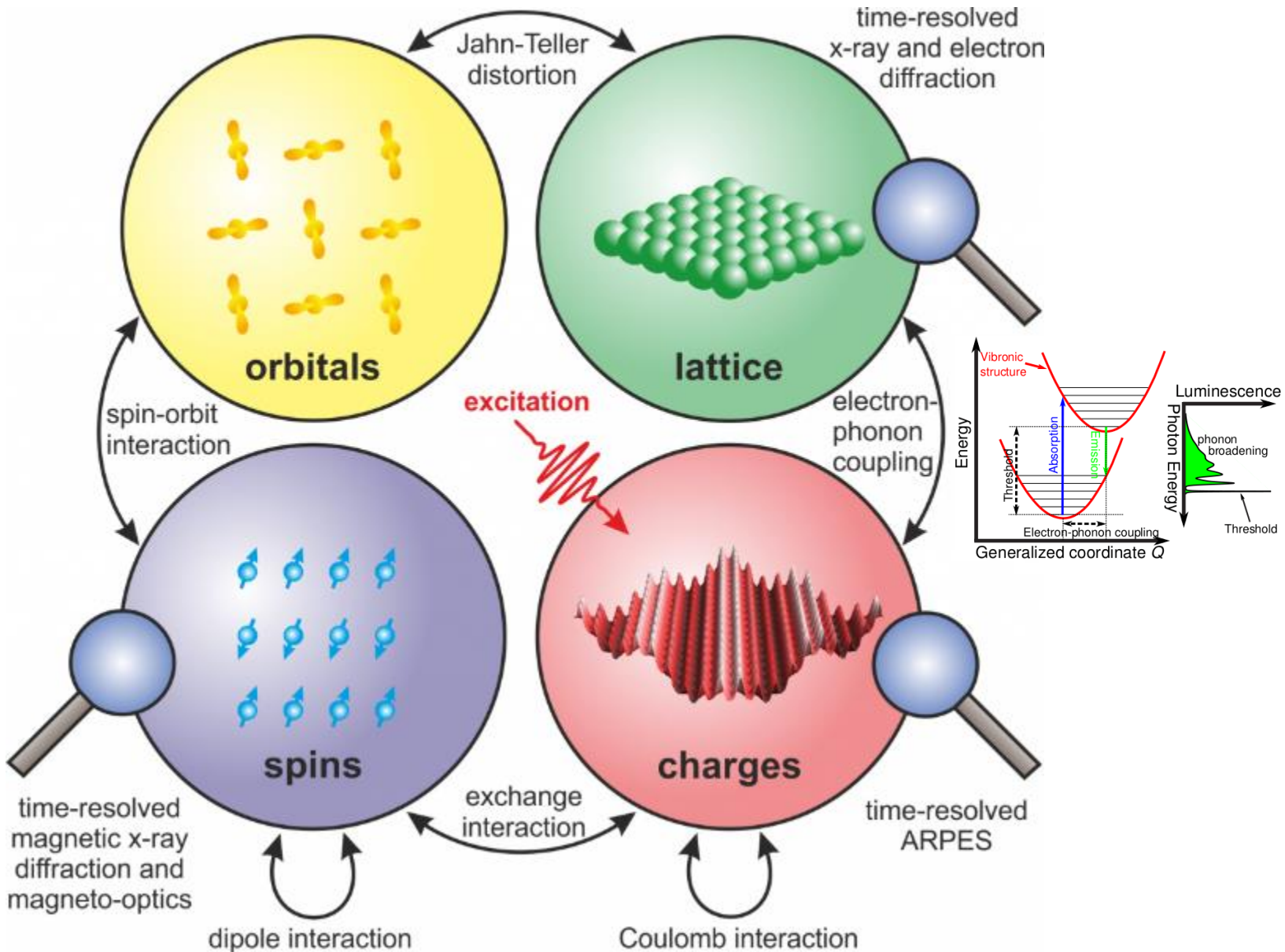
### Engineering & Materials Science

#### How can we break the current limit on energy-conversion efficiencies?

Energy-conversion efficiency is the central metric in determining the success of an energy system. It simply takes the useful energy output (benefit), and divides it by the energy input (cost). Scientists in academia, industry and government are experimenting with different

- 如何突破能量转换效率的当前极限 (电子弛豫发射声子)

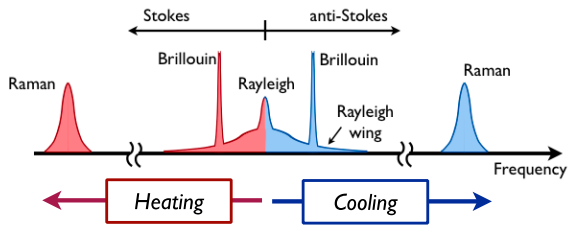
# 固体中的元激发和相互作用







# Our Researches



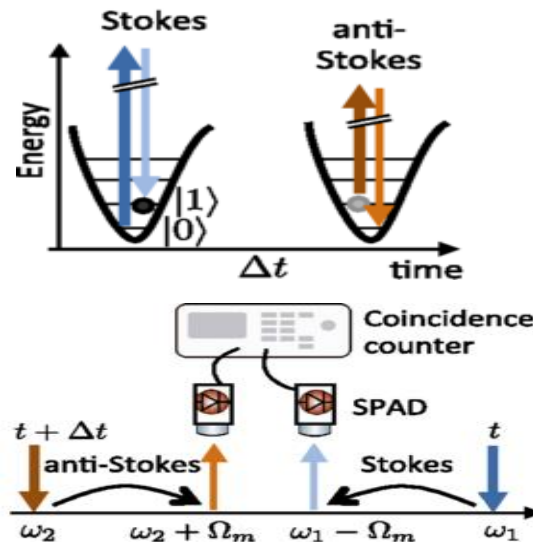
**Brillouin scattering**

Light interacts with sound

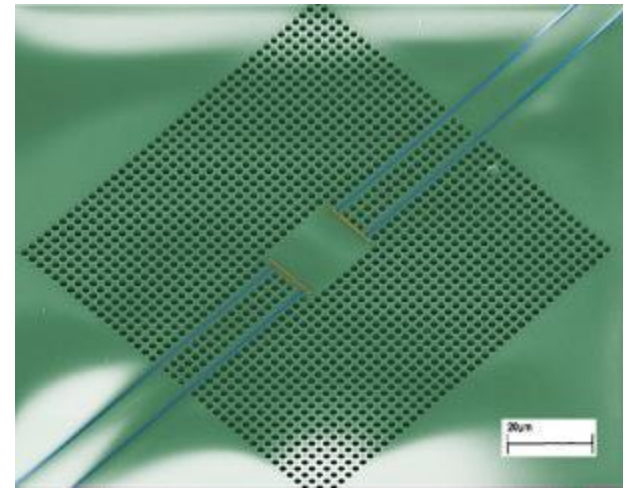
**Raman scattering**

Light interacts with molecular or atomic vibrations

**Phonon Properties**

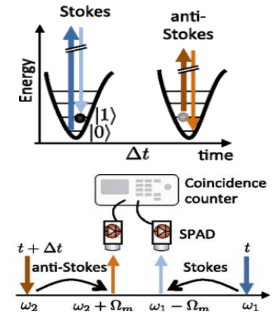
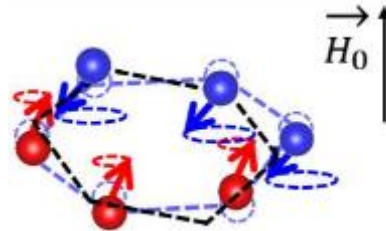
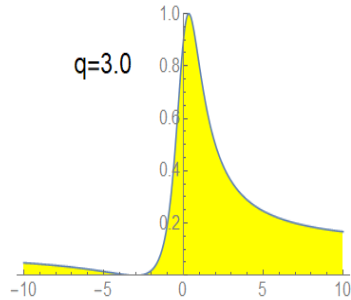
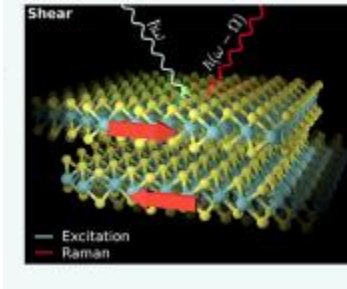


**Phonon Control**



**Phononic Devices**

# Selected Publications



*Nano Lett.* 11, 2407 (2011)

*Nano Lett.* 13, 3, 1007 (2013)

*Nano Lett.* 14, 8, 4724 (2014)

*Nano Lett.* 22, 1331 (2022)

*Nano Lett.* 22, 1233 (2022)

*Nano Lett.* 22, 13, 5385 (2022)

*Nano Lett.* 22, 7129 (2022)

*Phys. Rev. B* 85, 085418 (2012)

*Phys. Rev. B* 88, 195313 (2013)

*Phys. Rev. B* 88, 075320 (2013)

*Phys. Rev. B* 90, 245428 (2014)

*Phys. Rev. M.* 4, 034002 (2020)

*Phys. Rev. B* 106, 085205 (2022)

*Adv Mater.* 27, 4502 (2015)

*ACS Nano* 8, 4, 3796 (2014)

*ACS Nano* 8, 10, 10931 (2014)

*ACS Nano* 11, 11777 (2017)

*ACS Photonics* 9, 1605 (2022)

*J. Phys. Chem. Lett.* 9, 6656 (2018)

*J. Phys. Chem. Lett.* 10, 3087 (2019)

*J. Phys. Chem. Lett.* 13, 1533 (2022)

*Nano Res.* 14, 239 (2021)

*Nano Res.* 14, 1711 (2021)

*Nature* 493, 504 (2013)

*Nature Photon* 10, 115 (2016)

*Nature Photon* 10, 600 (2016)

*Nature Photon* 17, 1 (2023)

*Nat Commun.* 10, 2419 (2019)

*Nat Commun.* 12, 3048 (2021)

*Nat Commun.* 14, 88 (2023)

*Nat Commun.* Accepted (2023)

# 发展了几种声子探测方法

**Raman: > 5 cm<sup>-1</sup> ( 160 GHz )**

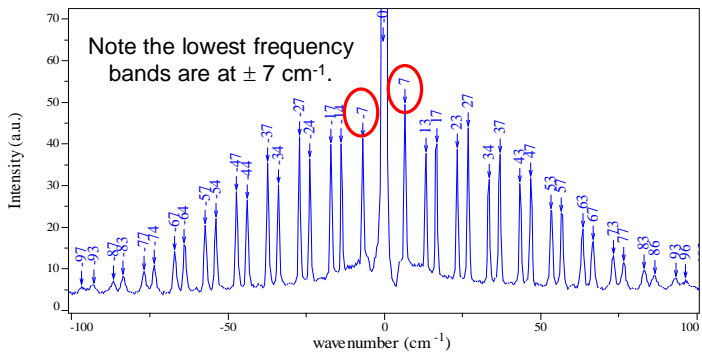
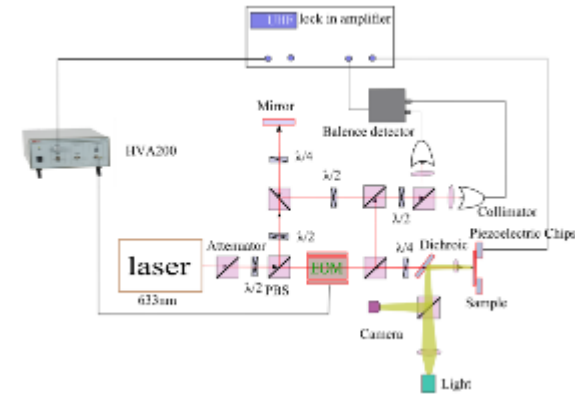
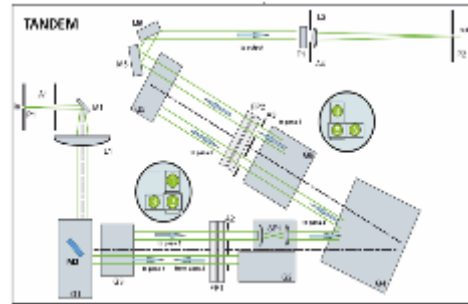
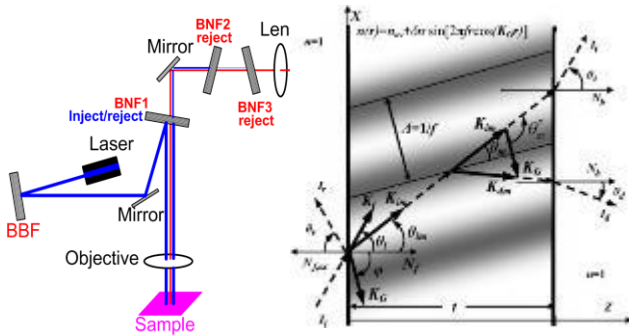
**BLS : 1-300 GHz**

**外差干涉 : < 1 GHz**

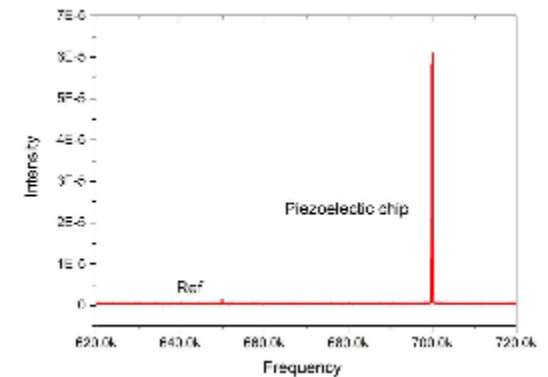
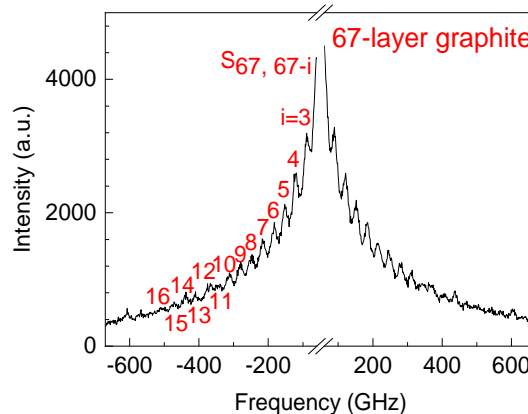
◆ 低温, 磁场, 多波长激发

◆ 低温, 磁场, 波矢/相位/时间分辨

◆ 灵敏度: ~0.1 fm × Hz<sup>-1/2</sup>



Data courtesy of P. H. Tan, State Key Laboratory for SL and Microstr., Institute of Semiconductors, Beijing, P. R. China, and K. Brunner, University Wurzburg, Germany



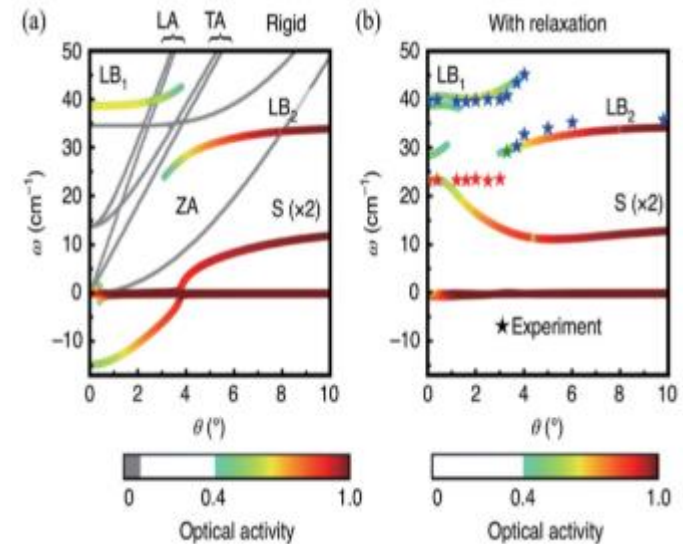
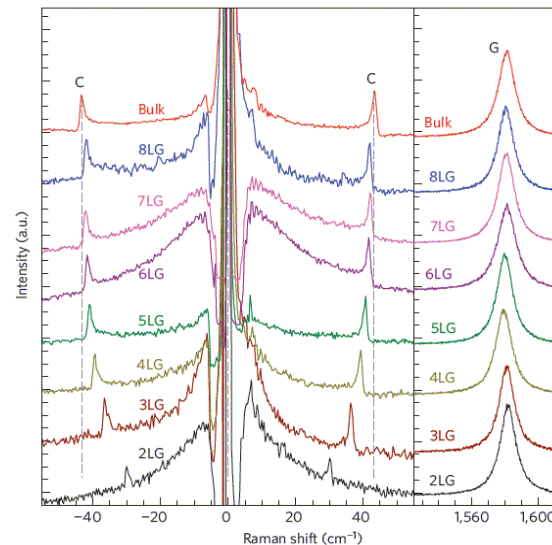
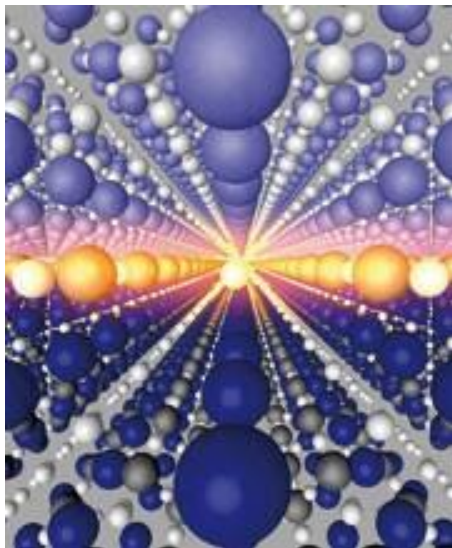
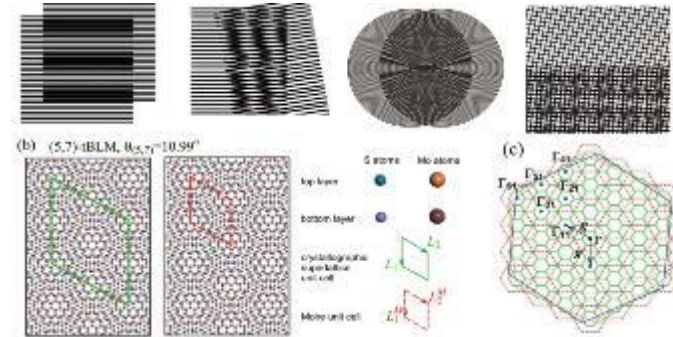
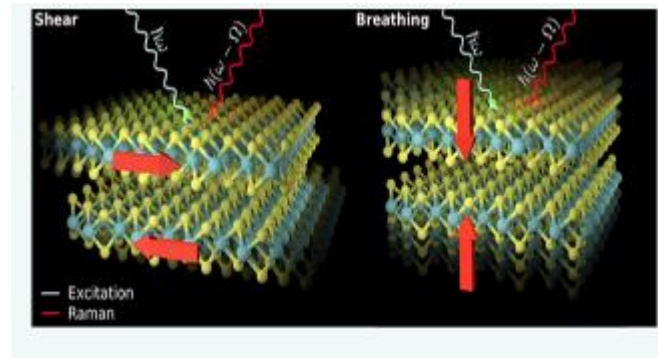
# 二维材料原子界面层间耦合探测

The Interface is the device

层间呼吸和剪切振动

莫尔异质结层间振动

The Interface is still the device



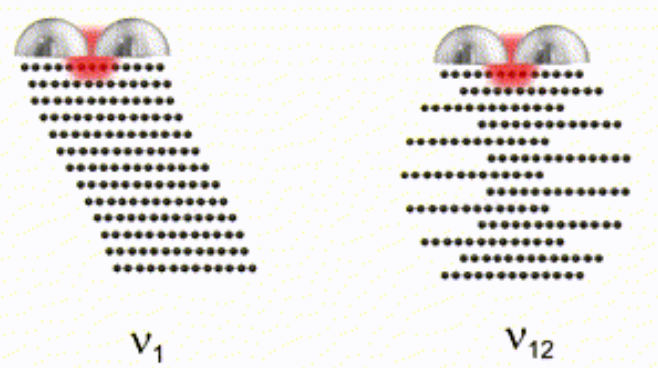
Nature Materials (2012)

PH Tan\* et al.,  
Nature Materials (2012)

PH Tan\*/F. Libisch\*/XQ Li\* et al.,  
Nature Materials (2021)

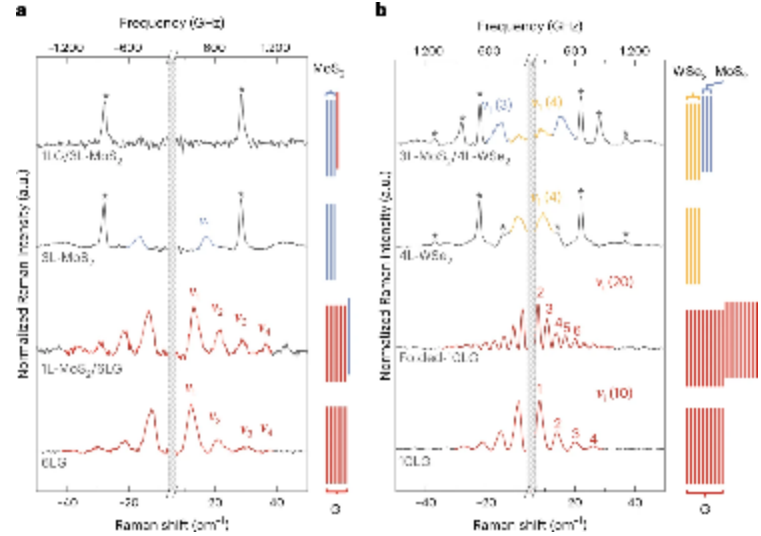
# 极低频隐藏界面振动的探测

## 宏观力学振动调制等离激元

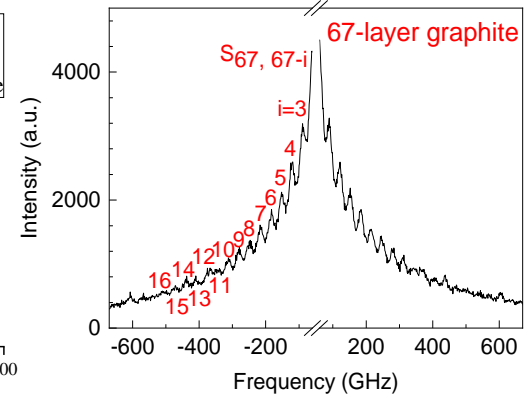
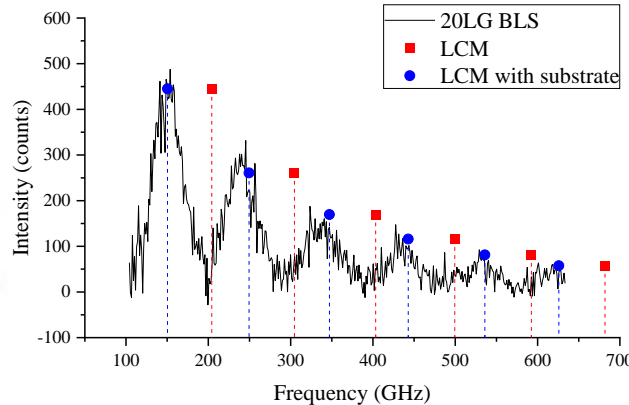
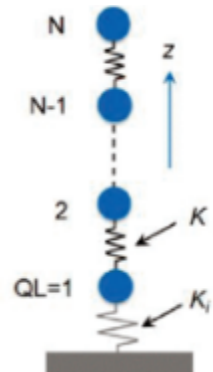
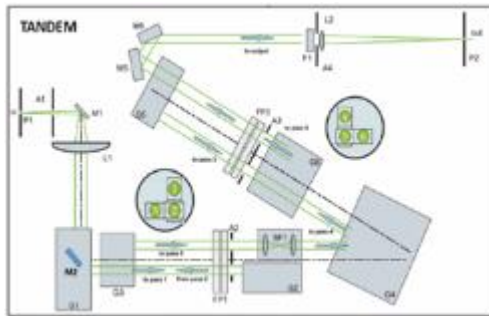


WG Xu\* et al.,  
Nature Photonics (2023)

## 力学振动的探测



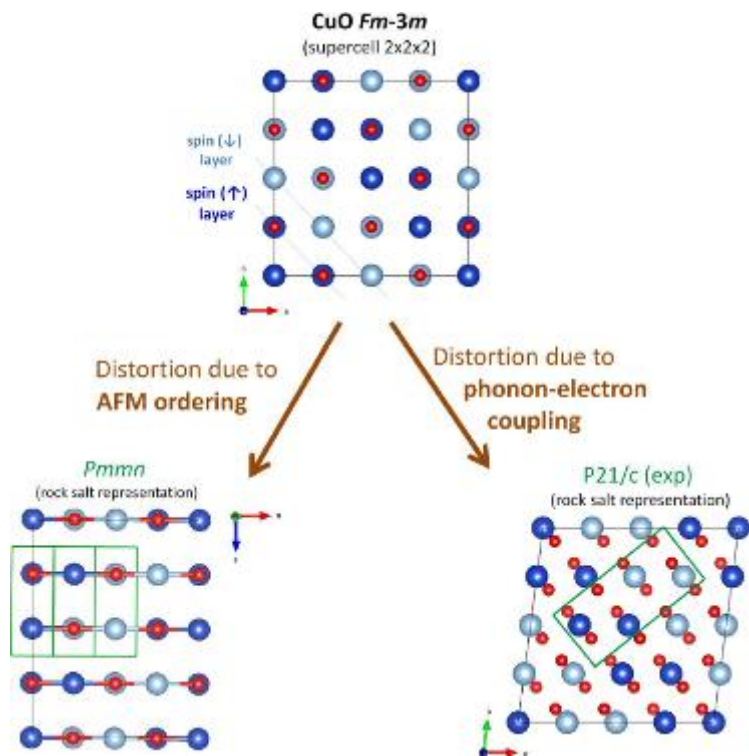
## 布里渊散射: 极低频隐藏界面振动的探测



YPLv/J. Zhang\* et al.,  
In Preparation (2023)

# 声子与电子、自旋耦合

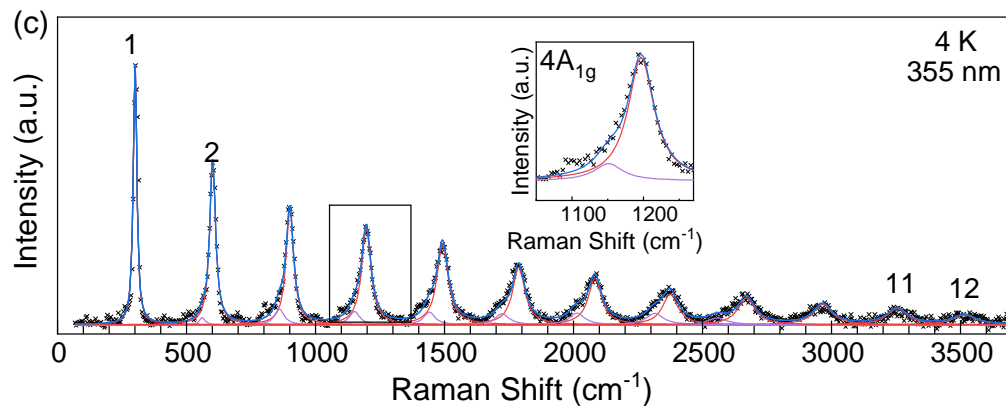
## 声子与电子、自旋耦合机制



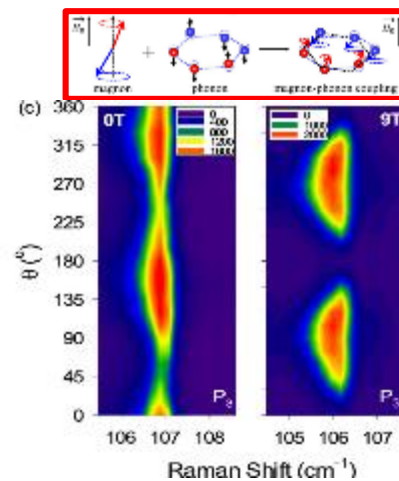
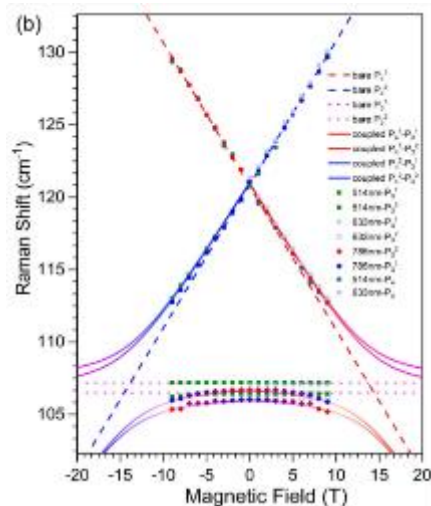
KX Xu/J. Zhang\* et al.,  
PRB (2022)

YJ Sun/J. Zhang\* et al.,  
JPCL (2019)

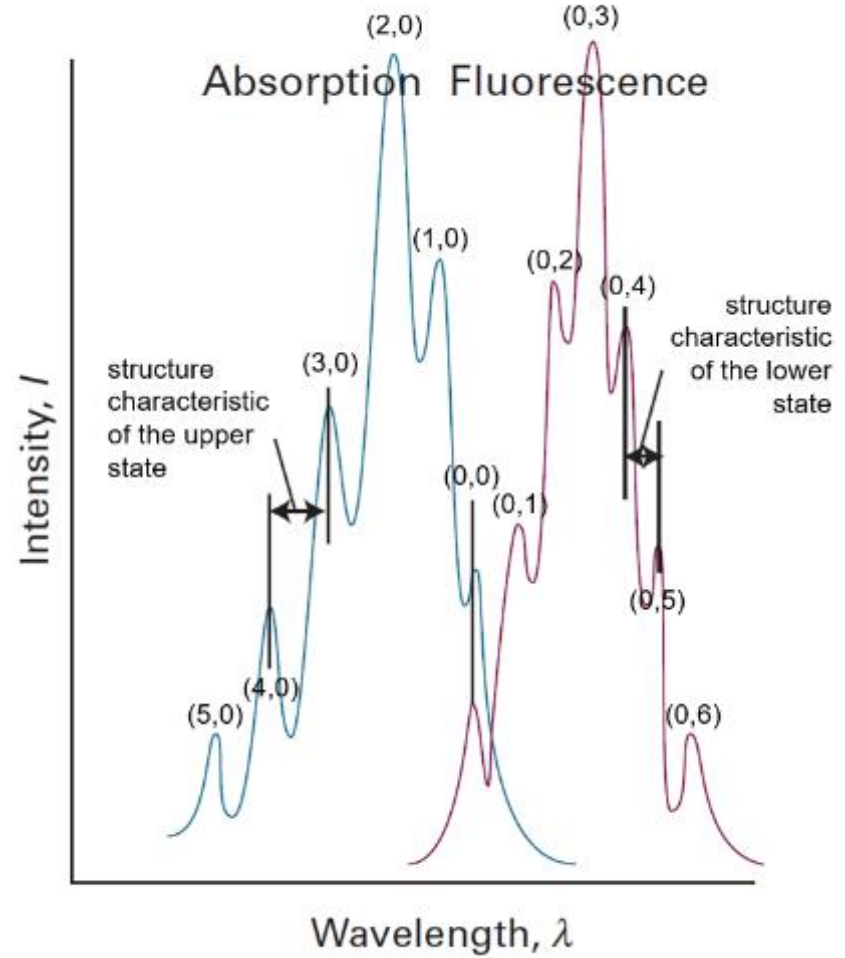
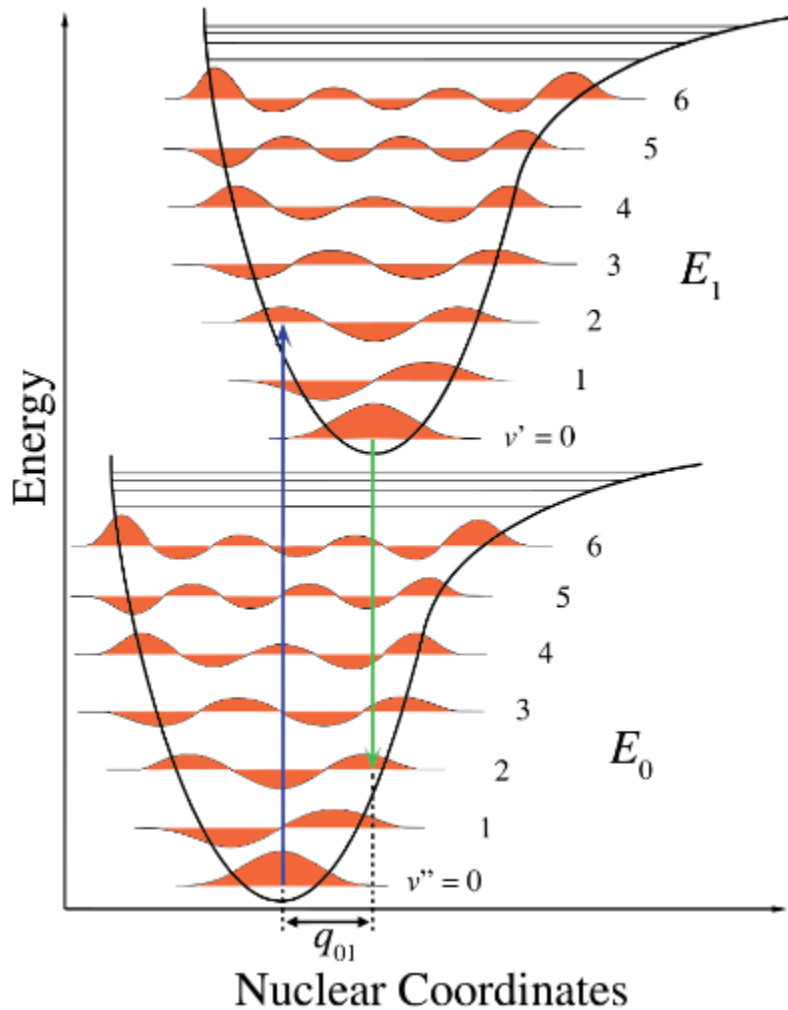
## 强电声子耦合导致的 THz 频率梳



## 声子-自旋波耦合测量: 能量、角动量交换



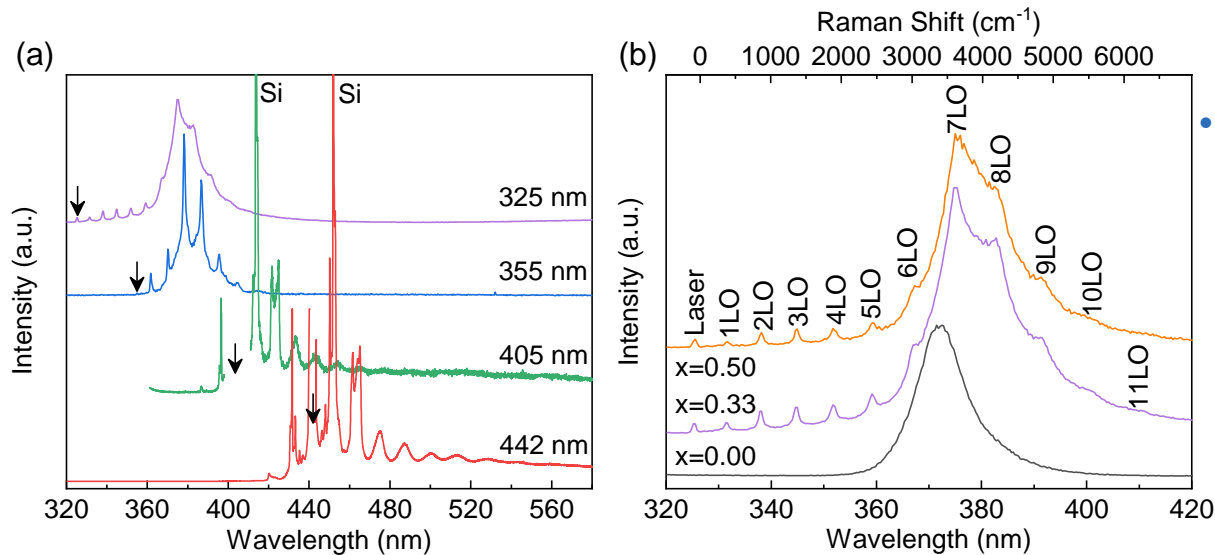
# Electron-Phonon Coupling



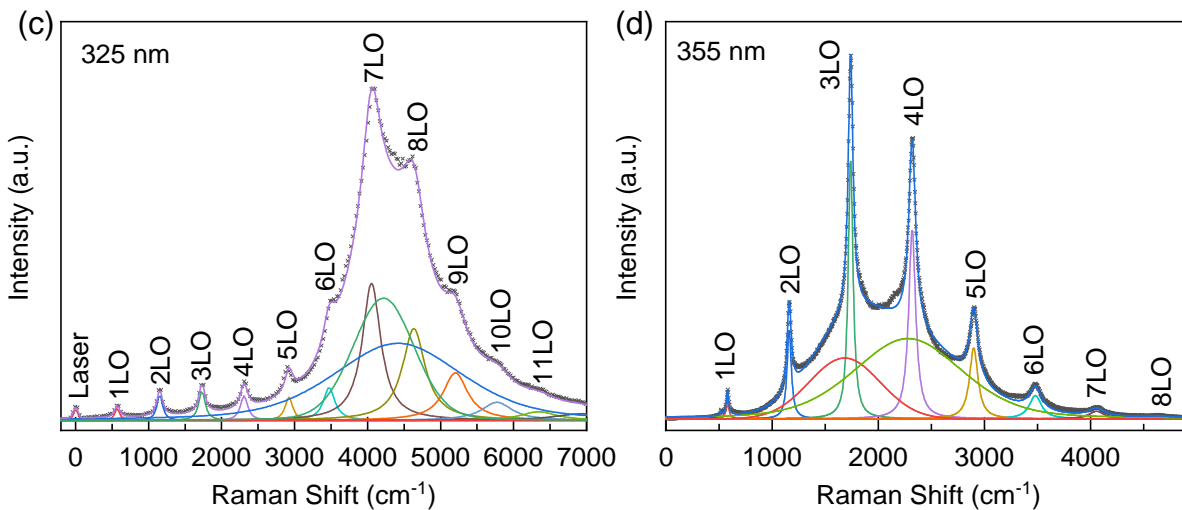
Franck-Condon 原理



# Multiphonon process

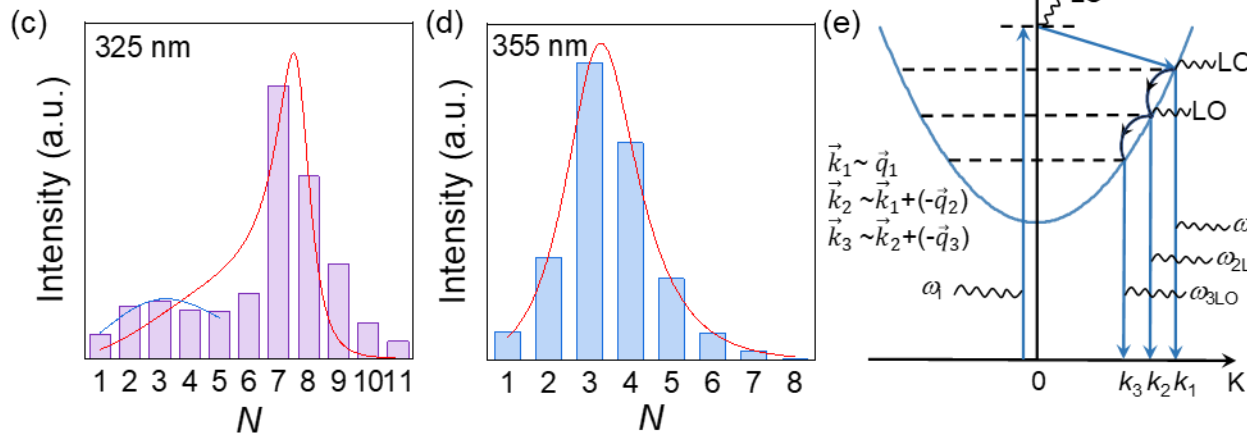
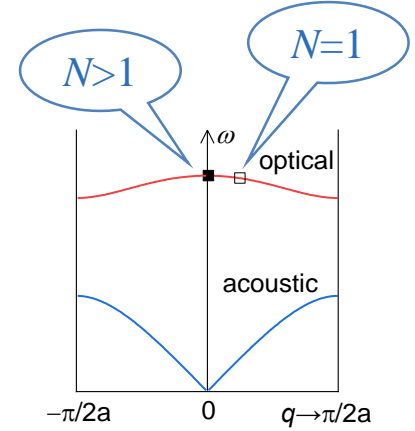
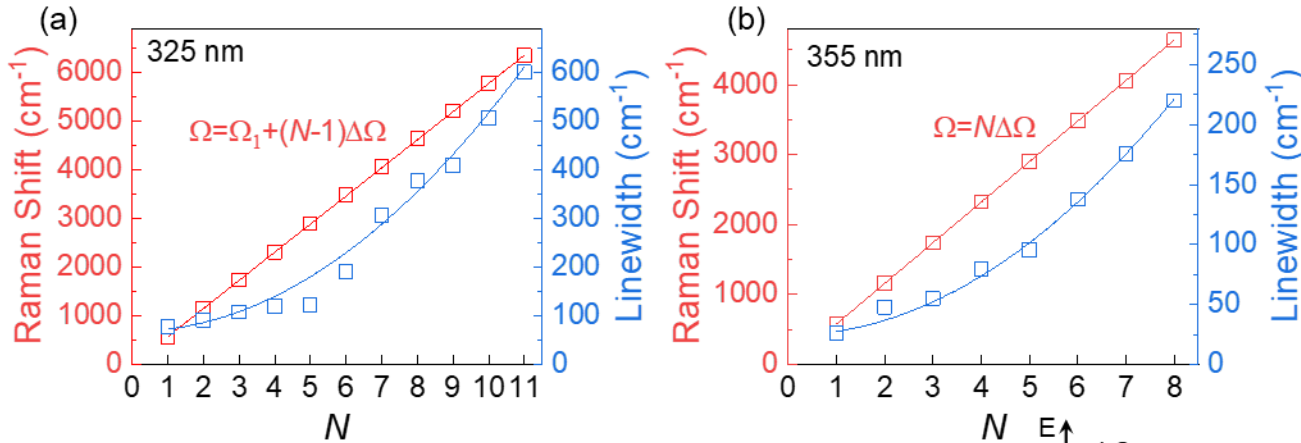


- LO声子-电子: Fröhlich相互作用  
**ZnO  $E_g=3.37$  eV (368 nm)**  
**Mn-doped ZnO**  
 **$E_g=3.26$  eV (380 nm)**



# Hotluminescence

## ZnO中的多声子参与的热荧光过程



峰位:  $\Omega = \Omega_1 + (N-1)\Delta\Omega$

$\Omega_1 = 571 \pm 3 \text{ cm}^{-1}$   
 $\Delta\Omega = 578 \pm 0.7 \text{ cm}^{-1}$

线宽:  $\Gamma \propto \Gamma_0 + \alpha N^2$

强度:

$$I_N = I_0 e^{-S} \frac{S^N}{N!} \frac{1}{(E_i - E_{ex} - N E_{LO})^2 + \kappa^2}$$

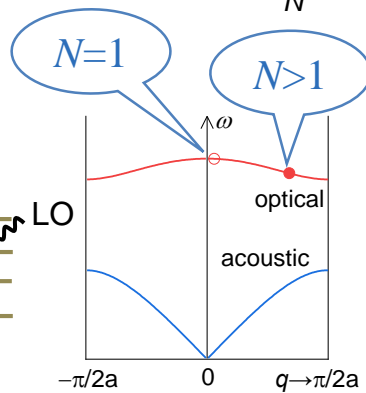
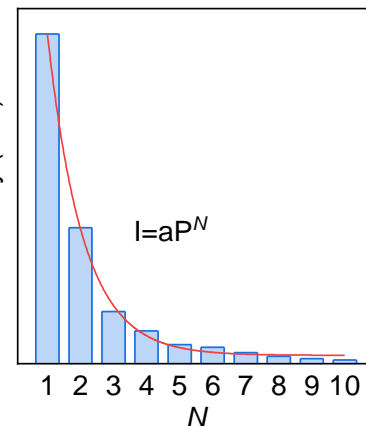
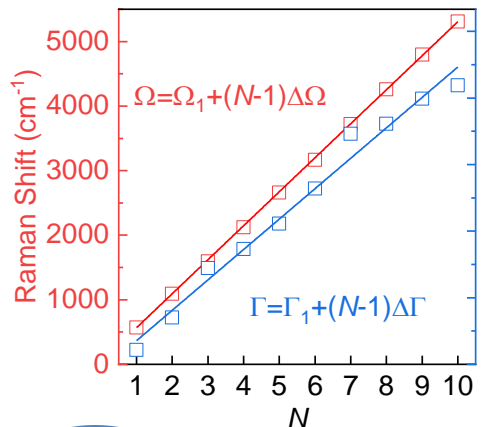
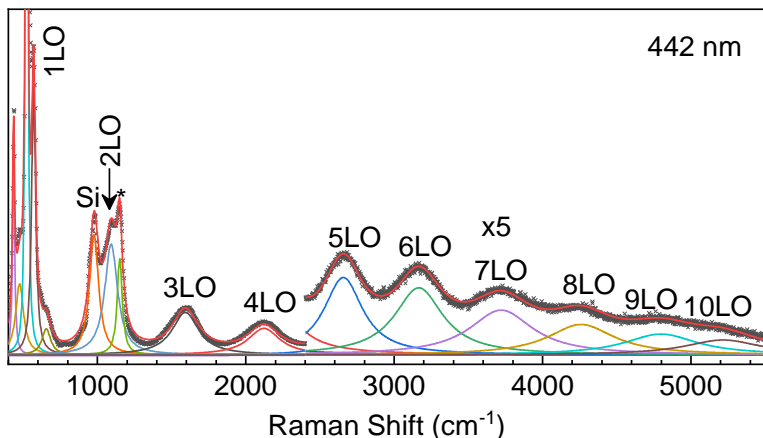
- $\Omega_1 < \Delta\Omega$ .
- 线宽随阶数二次依赖关系。
- 强度泊松分布+共振因子。

多声子发射的级联模型

355 nm激发,  $S = 4.2 \pm 0.1$ ;  
 325 nm激发,  $S = 3.7 \pm 0.2$ .

# High-order Scattering

## ZnO中的高阶拉曼散射过程



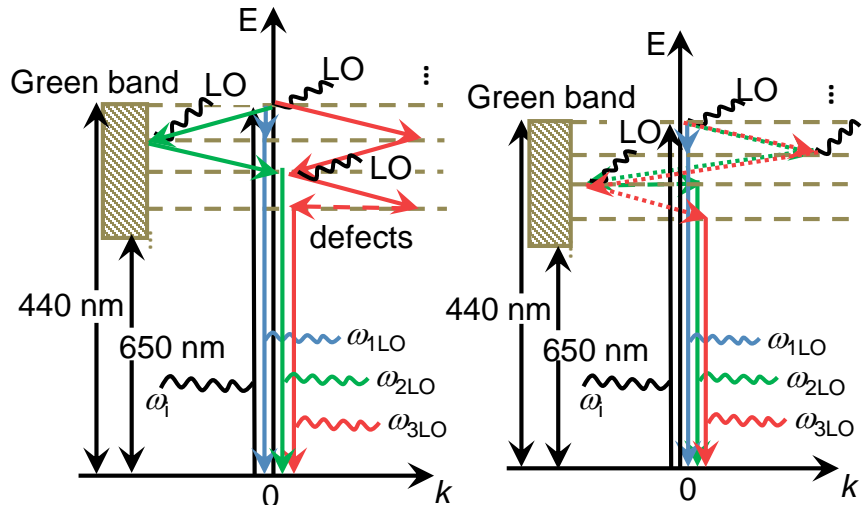
峰位:  $\Omega = \Omega_1 + (N-1)\Delta\Omega$

$\Omega_1 = 568 \pm 6 \text{ cm}^{-1}$   $\Delta\Omega = 526 \pm 2 \text{ cm}^{-1}$

线宽:  $\Gamma = \Gamma_1 + (N-1)\Delta\Gamma$

$\Gamma_1 = 53 \pm 8 \text{ cm}^{-1}$   $\Delta\Gamma = 68 \pm 3 \text{ cm}^{-1}$

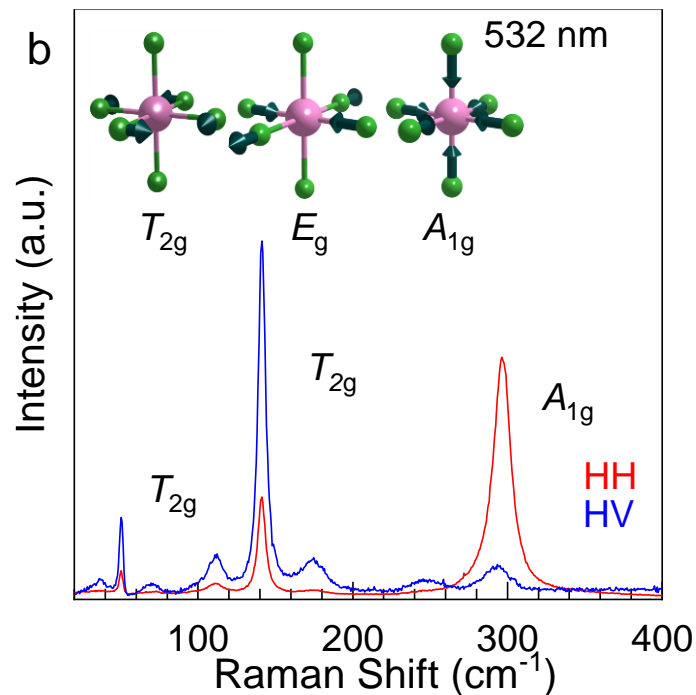
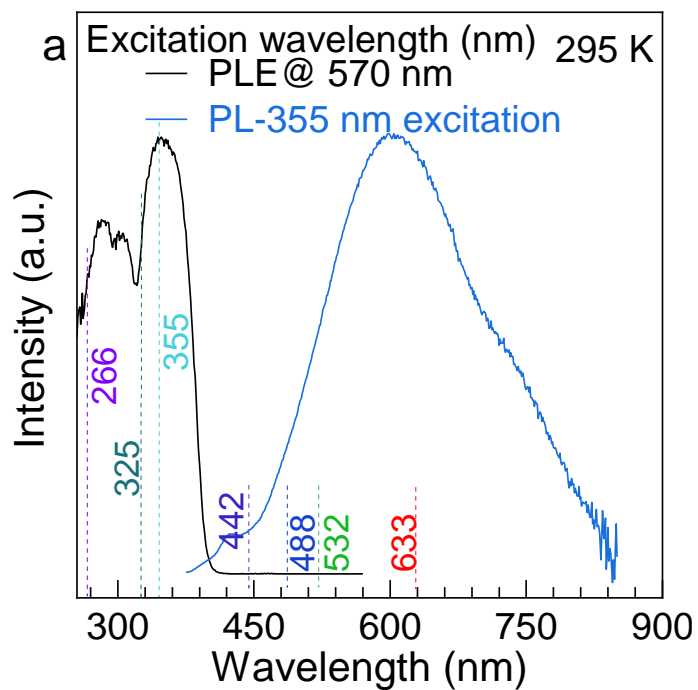
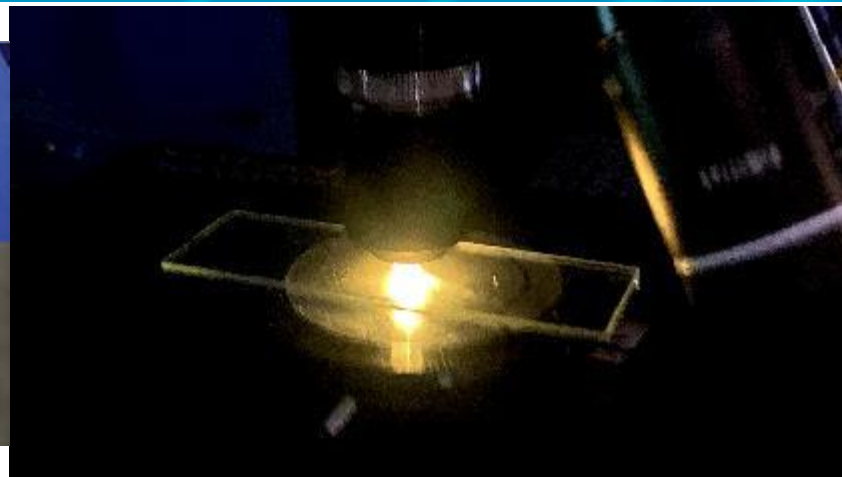
强度:  $I = aP^N$   $P = 0.34 \pm 0.01$



- $\Omega_1 > \Delta\Omega$ 。
- 线宽线性增加。
- 强度取决于散射几率，随阶数指数衰减。
- 442 nm 激发光低于带隙，缺陷来源于氧原子缺陷引起的绿光带。

# High-Order Scattering in STE

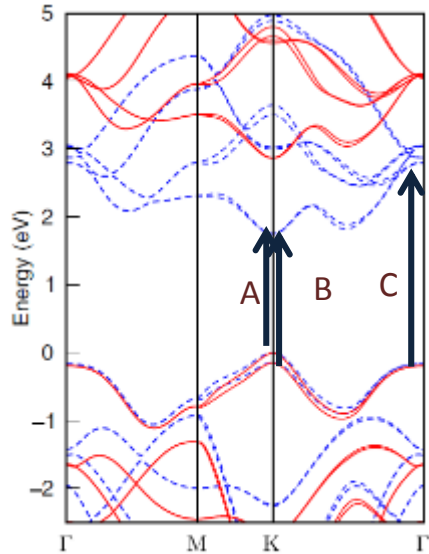
双钙钛矿半导体  
STE : 自陷激子





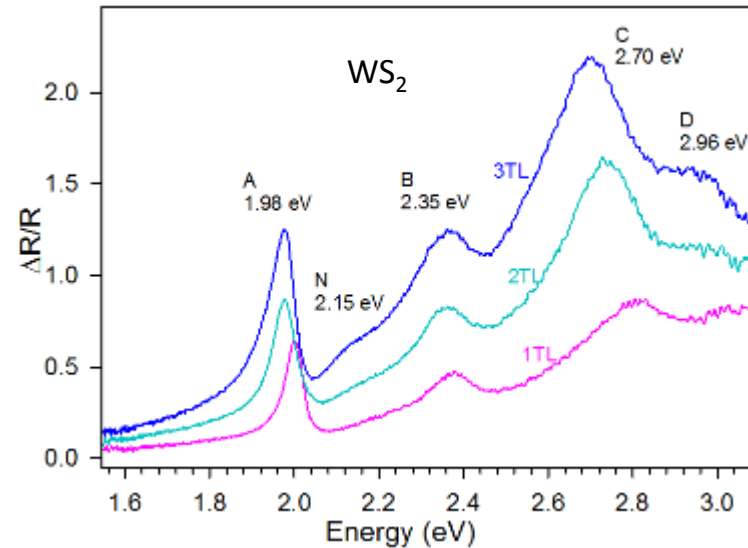
# The multiple exciton states in transition metal dichalcogenides

## ❖ Theory



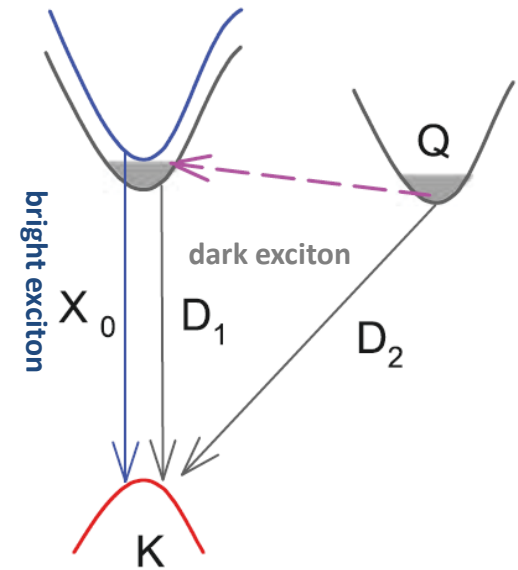
Steven G. Louie, et al, PRL, 2013

## ❖ Observation of bright excitons



QH Tan/J Zhang, 2D Mater, 2017

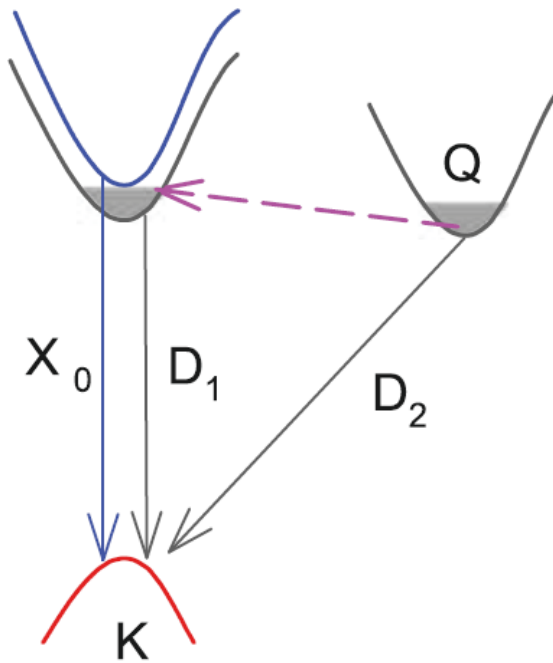
## ❖ Dark exciton



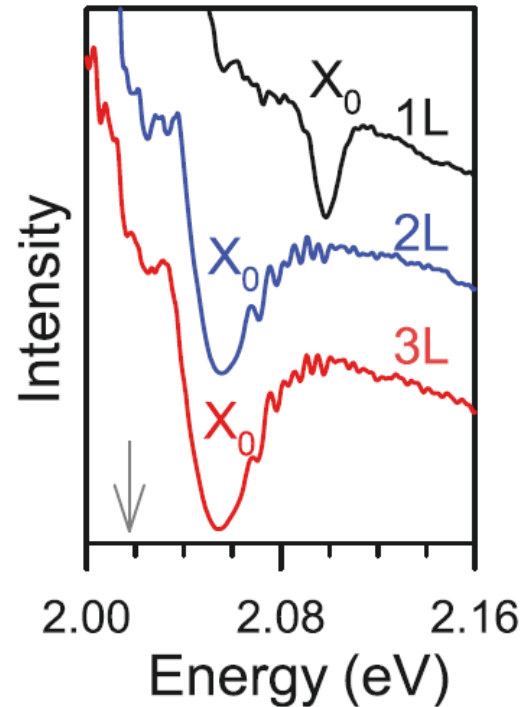
- Strong light-matter interaction in transition metal dichalcogenides (TMDs).
- Dark exciton
- Optical and Optoelectronics properties are dominated by exciton, phonon and exciton-phonon coupling.

# Observation of dark exciton in bilayer $\text{WS}_2$

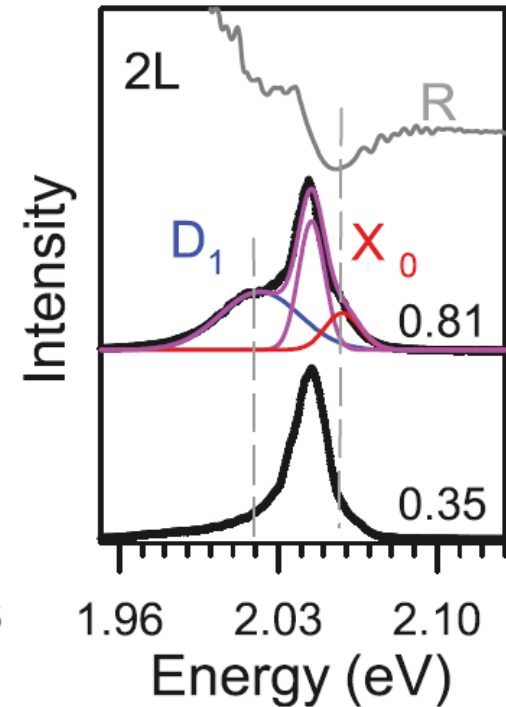
- ❖ Spin-forbidden dark exciton ( $D_1$ ) and momentum-forbidden dark exciton ( $D_2$ ).



- ❖ Reflection spectra



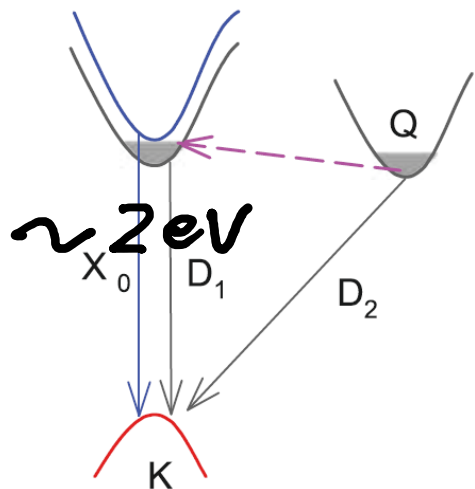
- ❖ PL spectra



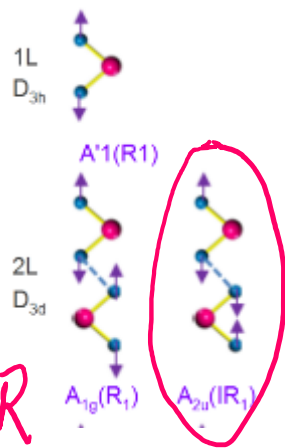
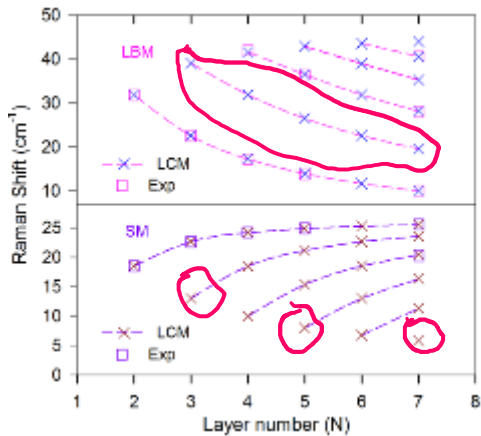
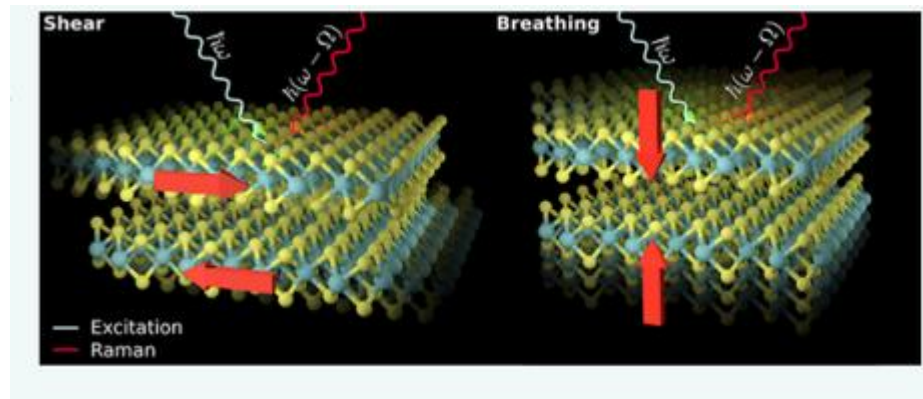
- By using high numerical apertures (NA) objective, the dark A exciton is observed with PL spectra.

# Parity, Dark Exciton, Forbidden Phonon

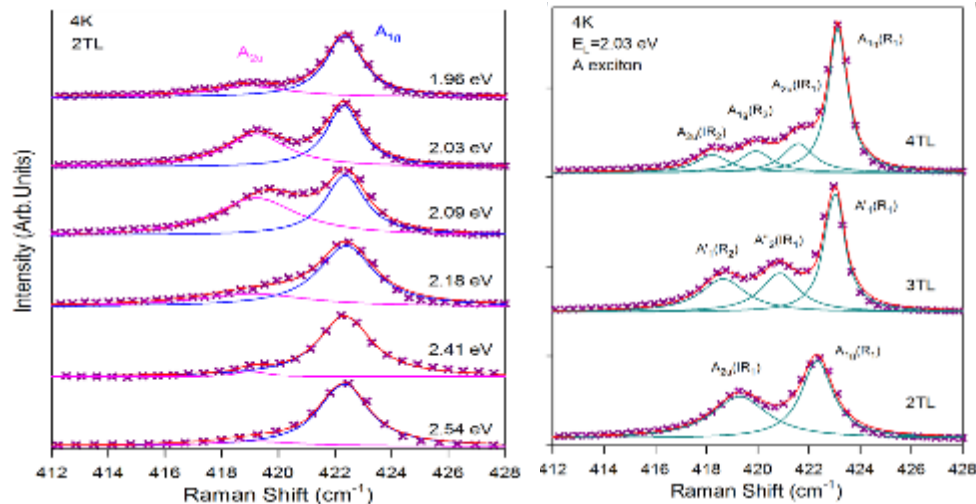
$W_p: 0.5 - 60 \text{ meV}$



Symmetry  
odd  
 $S(-Q) \approx -S(Q)$   
 $S(-Q) \approx S(Q)$   
Even



IR





# Parity , Dark Exciton , Forbidden Phonon

$$P \propto \left| \frac{\langle i | H_{eR}(\omega_s) | m' \rangle \langle m' | H_{eL} | m \rangle \langle m | H_{eR}(\omega_i) | i \rangle}{(E_{m'} - \hbar\omega_i - i\gamma_{m'})(E_m - \hbar\omega_s - i\gamma_m)} + \frac{\langle i | H_{eR}(\omega_i) | m \rangle \langle m | H_{eL} | m' \rangle \langle m' | H_{eR}(\omega_s) | i \rangle}{(E_m - \hbar\omega_i - i\gamma_m)(E_{m'} - \hbar\omega_s - i\gamma_{m'})} \right|^2$$

$\omega_i, \omega_{ph}, \omega_s$  分别是入射光子, 声子以及散射光子频率,  $m'$ 和 $m_s$  中间态,  $\gamma_m, \gamma_{m'}$  是线宽(与寿命有关),  $H_{eL}$ 是激子与声子作用项,  $H_{eR}$ 激子-辐射场相互作用,  $i$ 是初态,  $m/m'$ 是中间态

拉曼活性—偶宇称

红外活性—奇宇称

亮态激子—奇宇称, 偶极跃迁允许

暗态激子—偶宇称, 偶极跃迁禁戒

$P \neq 0, H_{eL}$  (声子)宇称必须和  $H_{eR}(\omega_i) \otimes H_{eR}(\omega_s)$  一致

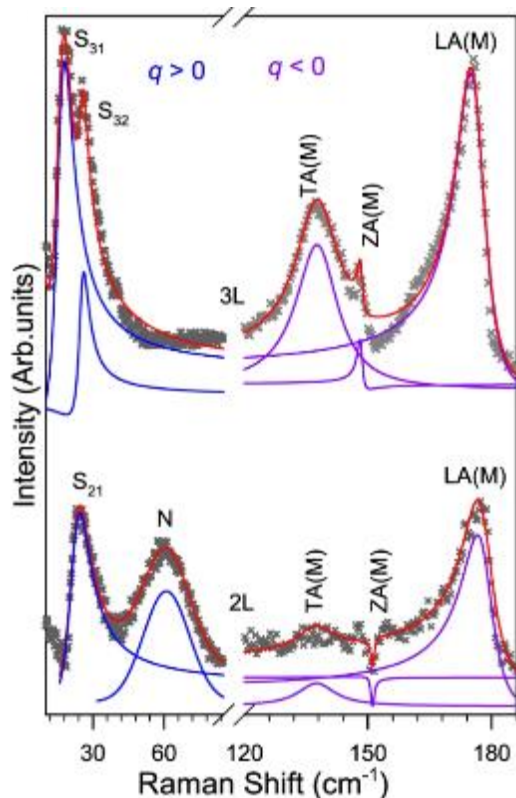
亮态	}	奇	奇	}	偶宇称 → 偶宇称 (拉曼活性)	}	$H_{eL}$ (声子)
		$H_{eR}(\omega_i) \otimes H_{eR}(\omega_s)$					
暗态	}	偶	奇				

暗态的参与, 使得红外活性模式可以被拉曼观测到

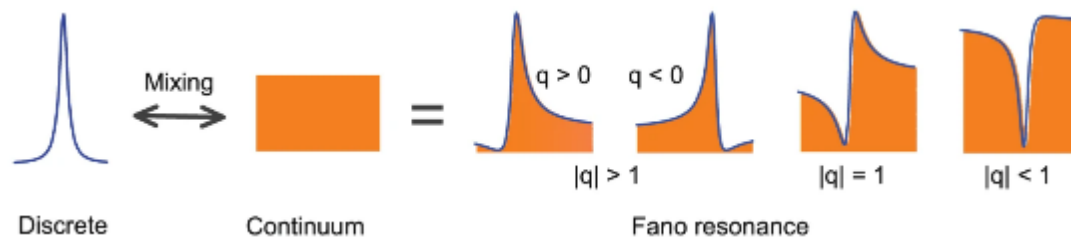
# Quantum interference between dark exciton and phonon

$$I = I_0 \frac{(1 + (\omega - \omega_0)/q\gamma)^2}{1 + ((\omega - \omega_0)/\gamma)^2}$$

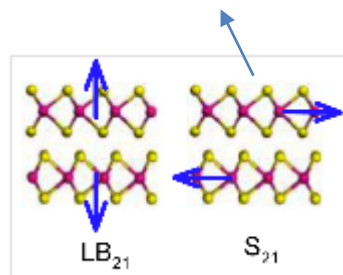
## ❖ Experimental results



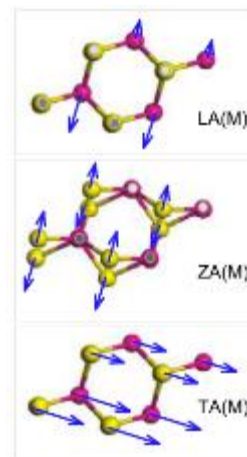
## ❖ Fano resonance



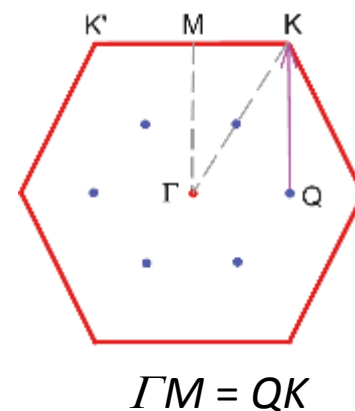
## ❖ Shear phonon



## ❖ Acoustic phonons



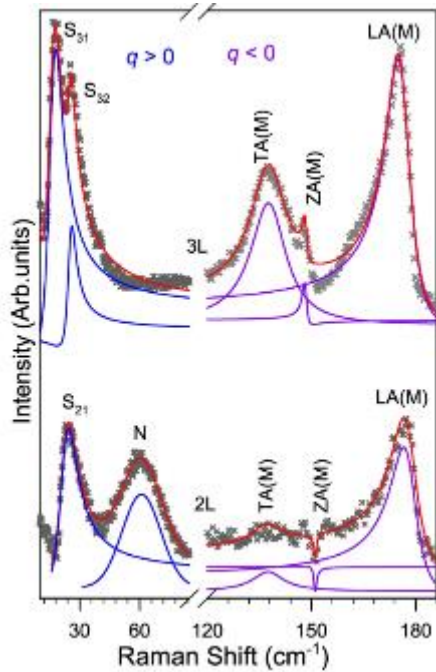
## ❖ Momentum match



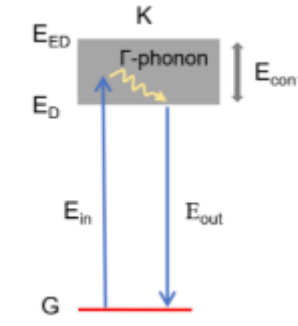
$$H_{ep} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, \mathbf{q}, mn\nu} g_{mn\nu}(\mathbf{k}, \mathbf{q}) \hat{a}_{m, \mathbf{k}+\mathbf{q}}^\dagger \hat{a}_{n, \mathbf{k}} (\hat{b}_{\mathbf{q}\nu} + \hat{b}_{-\mathbf{q}\nu}^\dagger), \quad g_{mn\nu}(\mathbf{k}, \mathbf{q}) \propto g_0 \mathbf{q} \cdot \mathbf{e}_{\mathbf{q}\nu}$$

# Quantum interference between dark exciton and phonon

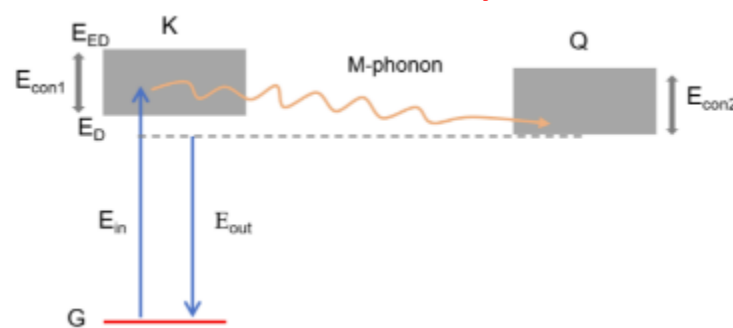
## ❖ Experimental results



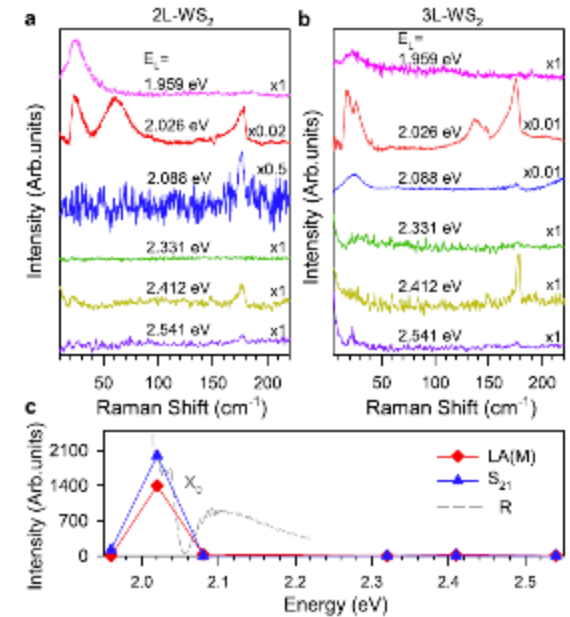
## ❖ Quantum Interference between shear phonon and dark exciton in K valley



## ❖ Quantum Interference acoustic phonon and dark exciton in Q valley



## ❖ Fano resonant profile



# Breakdown of Raman Select Rule

$$\sigma = \sigma_0 \frac{\omega_s}{\omega_i} \left| \sum_{\alpha\beta} \epsilon_i \mathbf{R}^{\alpha\beta} \epsilon_s \right|^2$$

$$A_{1g} : \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix}, \quad \epsilon_i = (\sin\beta, \cos\beta, 0), \quad \epsilon_s = (0, 1, 0)$$

$$I \propto |\epsilon_s \cdot \mathbf{R} \cdot \epsilon_i|^2 = a^2 \cos^2 \beta,$$

$$E_{2g} : \begin{pmatrix} 0 & d & 0 \\ d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \begin{pmatrix} d & 0 & 0 \\ 0 & -d & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad I \propto |\epsilon_s \cdot \mathbf{R}_1 \cdot \epsilon_i|^2 + |\epsilon_s \cdot \mathbf{R}_2 \cdot \epsilon_i|^2 = d^2,$$

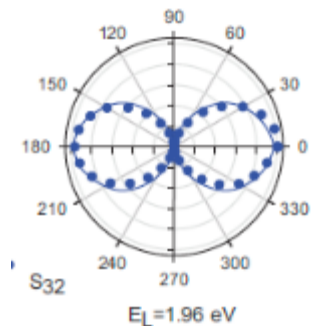
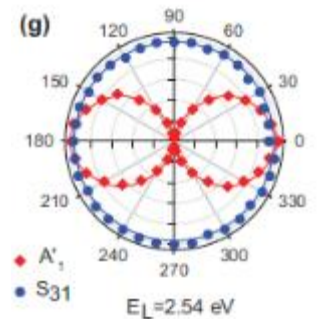
$$\mathbf{R}^{\alpha\beta} = \sum_{ij} \frac{\mathbf{P}_{0j}^\alpha \mathbf{M}_{ij} \mathbf{P}_{i0}^\beta}{(E_j - \omega_i + \omega_0)(E_i - \omega_s)}$$

$$\begin{aligned} |\langle 1s | \mathbf{H}_{(F,q)} | 1s \rangle| &= \mathbf{M}^F(q) \\ &= \left( \frac{C_F}{q} \right) \left[ \left( \frac{1}{1 + (p_h \mathbf{q}r/2)^2} \right)^2 - \left( \frac{1}{1 + (p_e \mathbf{q}r/2)^2} \right)^2 \right] \quad (3) \end{aligned}$$

where  $p_e = \frac{m_e}{m_e + m_h}$ ,  $p_h = \frac{m_h}{m_e + m_h}$ . For small but non-vanishing values of  $\mathbf{q}$ , this magnitude expression can be expanded as

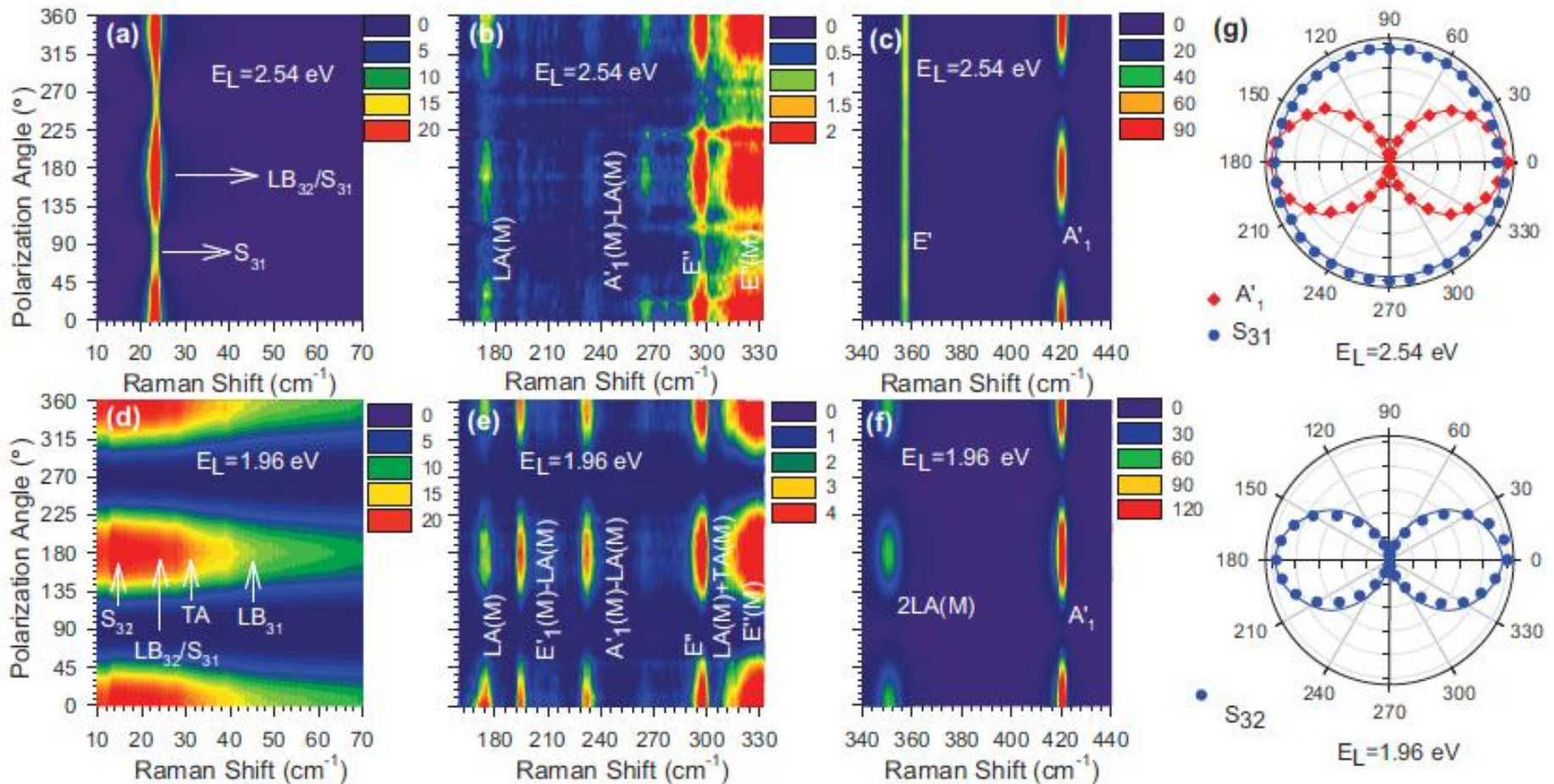
$$\mathbf{M}^F(q) \simeq C_F \mathbf{q}r \frac{m_e - m_h}{m_e + m_h} \quad (4)$$

$$I \propto (\mathbf{q}r)^2 \cos^2 \beta$$



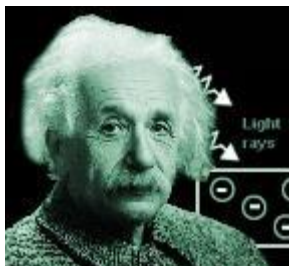
# Breakdown of Raman Select Rule

❖ 488 nm excitation close to C exciton



❖ 633 nm excitation (close to dark A exciton)

# anti-Stokes上转换激发



1905年《关于光的产生与转化的启发式观点》爱因斯坦文集第二卷，(Annalen der Physik 17, 132-148, (1905))

## 7. Stokes规则

$$\frac{R}{N}\beta\nu_2 \leq \frac{R}{N}\beta\nu_1, \quad \nu_2 \leq \nu_1.$$

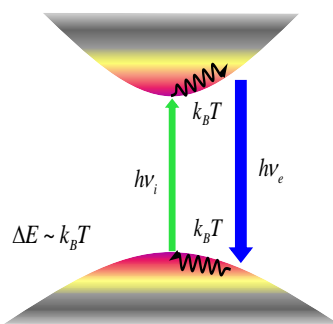
假设单频光由光激发光转化为多频光，并且，依照刚刚得到的结果，让我们假设入射光和发射光都是由能量量级为  $(R/N)\beta\nu$  的能量子构成的，这里  $\nu$  指相关的频率。那么转化过程可以被如下内

根据针对这里展示的现象的设想，Stokes规则在以下情况下出现例外是可能的：

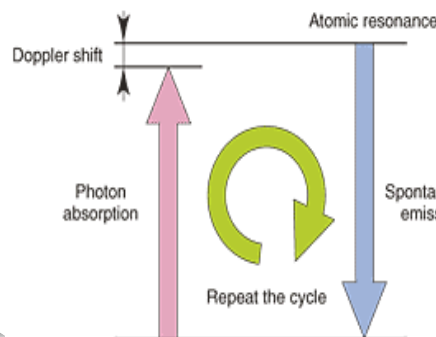
1. 若每单位体积同时被转化生成的能量子数量极大，以至于发射光的一个能量子可以从很多入射能量子中吸收能量。
2. 当在Wien定律适用范围内，入射（或发射）光并不具有和黑体辐射相同的能量分布；若，举个例子，入射光是由一个温度那样高的物体产生的，以至于Wien定律不再适用于与之相关的波长。

后一种可能情况需要特殊关注。根据上述构建的设想，在Wien定律适用范围内，若我们考虑辐射的能量，甚至处于密度很低的“非Wien辐射”也将有不同于黑体辐射的行为——这确实不是不可能的。

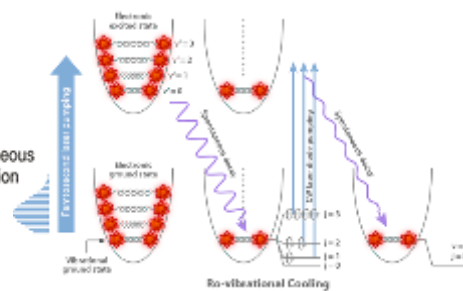
## 声子辅助荧光上转换



## 激光冷却原子



## 分子振动和转动冷却



## 宏观物体振动的冷却



P. Pringsheim, *Z. Phys.* **A 57**, 739, (1929)  
L. Landau, *J. Phys.* **10**, 499, (1946)

Proposed in 1975 by Wineland, Dehmelt et al.;  
realized in 1978 by Wineland, Drullinger, and Walls

*Science* **357**, 1002 (2017)

*Science* **372**, 1333 (2021)

# 光对晶格声子的辐射压力

## Heating and cooling of local and quasilocal vibrations by a nonresonance field

M. I. Dykman

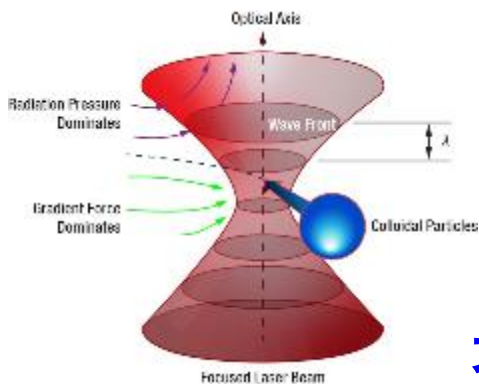
*Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR, Kiev*

(Submitted January 25, 1978)

*Fiz. Tverd. Tela (Leningrad) 20, 2264-2272 (August 1978)*

The dynamics of an anharmonic oscillator interacting with phonons in a nonresonant quasimonochromatic field is studied. The field-induced decay effects alter considerably the effective oscillator temperature. When an induced decay corresponds to simultaneous excitation of the oscillator and creation of a phonon, a runaway effect occurs for strong pumping, i.e. the oscillator energy increases exponentially with time.

$$U = -\frac{1}{2} \vec{P} \cdot \vec{E} = -\frac{1}{2} \chi |\vec{E}(\vec{r})|^2$$



光场空间分布不均匀

$$\vec{\nabla} |\vec{E}(\vec{r})|^2$$

VOLUME 56, NUMBER 17

PHYSICAL REVIEW LETTERS

28 APRIL 1986

## Light-Pressure Cooling of a Crystal

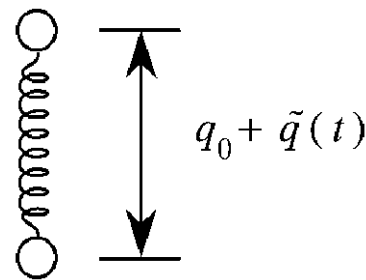
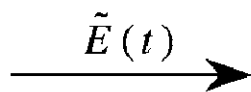
Juha Javanainen

*Max-Planck-Institut für Quantenoptik, D-8046 Garching, Federal Republic of Germany*

(Received 10 January 1986)

We investigate laser cooling of phonons in a low-density crystal of ions. If the linewidth of the optical transition covers the range of phonon frequencies, the rate of decrease of mechanical energy per one ion in the crystal is found to be essentially the same as in an ideal gas of two-level systems, but the cooling is concentrated on a subset of the phonon modes. A broad phonon spectrum tends to counteract the cooling.

$$\begin{aligned} \vec{F} &= -\vec{\nabla} U \\ &= \frac{1}{2} \chi \vec{\nabla} |\vec{E}(\vec{r})|^2 + \frac{1}{2} |\vec{E}(\vec{r})|^2 \vec{\nabla} \chi \end{aligned}$$



$$\vec{\nabla} \chi$$

极化率分布不均匀

$$P_{oe} = \frac{du}{dq}$$

$$= \frac{1}{2} \epsilon_0 \left( \frac{\partial a}{\partial q} \right)_0 E^2$$

Ashkin, Trapping of Atoms by Resonance Radiation Pressure, *Phys. Rev. Lett.* **40**, 729 (1978)

Nobel Prize in Physics (2018)

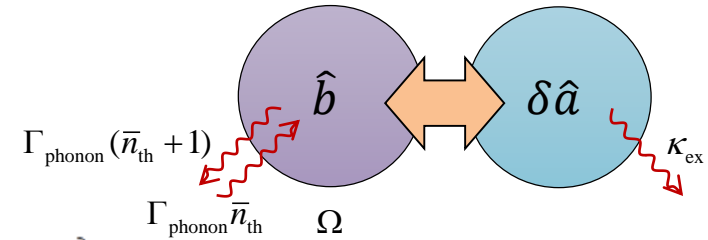
J. Zhang\*/QH Xiong\* et al., *Nature Photonics* 2016; JM Lai/J. Zhang\* et al., *Nano Lett.* 2022

# Exciton Optomechanics

## 可分辨边带拉曼冷却——单个声子模式的调控

### 光场作用下的激子-声子耦合量子理论

激子-声子耦合体系  $\hat{H} = \hbar\omega_{\text{ex}}\hat{a}^\dagger\hat{a} + \hbar\Omega\hat{b}^\dagger\hat{b} + \hbar g_0\hat{a}^\dagger\hat{a}(\hat{b}^\dagger + \hat{b})$   $g_0 = \frac{\partial\omega_{\text{ex}}}{\partial x}x_{\text{ZPF}}$



频率为  $\omega_i = \omega_{\text{ex}} + \Delta$  的驱动光场作用下  $\hat{H} \rightarrow \hat{U}^\dagger \hat{H} \hat{U} - \hat{A}$   $\hat{A} = \hbar\omega_i\hat{a}^\dagger\hat{a}$ ,  $\hat{a} = \bar{\alpha} + \delta\hat{a}$

$$\hat{H} = -\hbar\Delta\hat{a}^\dagger\hat{a} + \hbar\Omega\hat{b}^\dagger\hat{b} + \hbar g(\delta\hat{a}^\dagger + \delta\hat{a})(\hat{b}^\dagger + \hat{b})$$

激子-声子耦合强度  $g = g_0\bar{\alpha} = g_0\sqrt{\bar{n}_{\text{ex}}}$

红失谐激发  $\Delta = -\Omega$   $\hat{H}_{\text{ex-ph}} = \hbar g(\delta\hat{a}^\dagger\hat{b} + \delta\hat{a}\hat{b}^\dagger)$  **“beam-splitter” interaction**

蓝失谐激发  $\Delta = \Omega$   $\hat{H}_{\text{ex-ph}} = \hbar g(\delta\hat{a}^\dagger\hat{b}^\dagger + \delta\hat{a}\hat{b})$  **“two-mode squeezing” interaction**

零失谐激发  $\Delta = 0$   $\hat{H}_{\text{ex-ph}} = \hbar g(\delta\hat{a}^\dagger + \delta\hat{a})(\hat{b}^\dagger + \hat{b})$  **implementing QND detection**

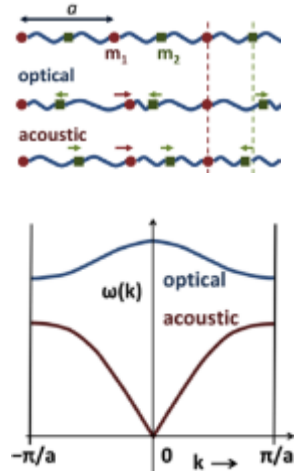
“腔”内光子寿命决定振子能量不确定度  $\Delta E \sim \hbar/\tau$

$\kappa_{\text{cx}} < \Omega$  可分辨条件

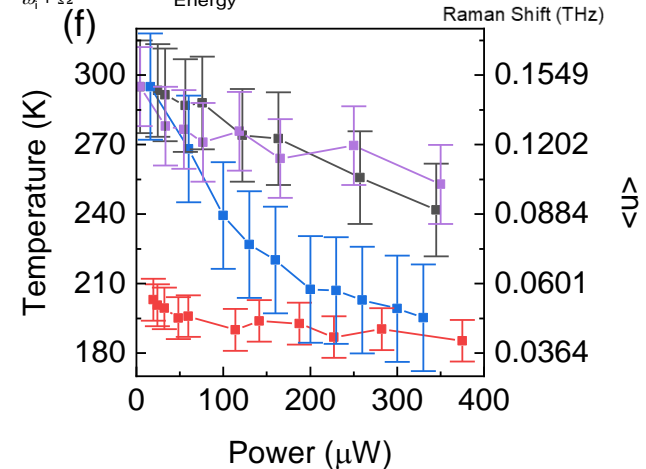
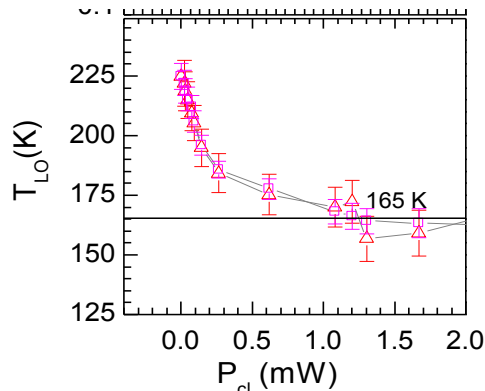
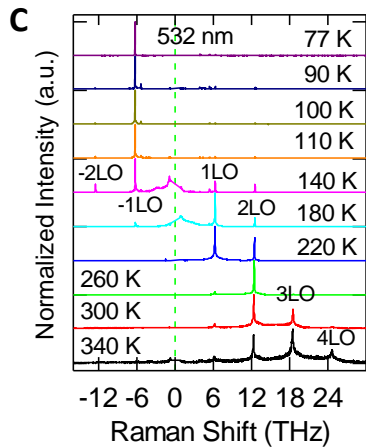
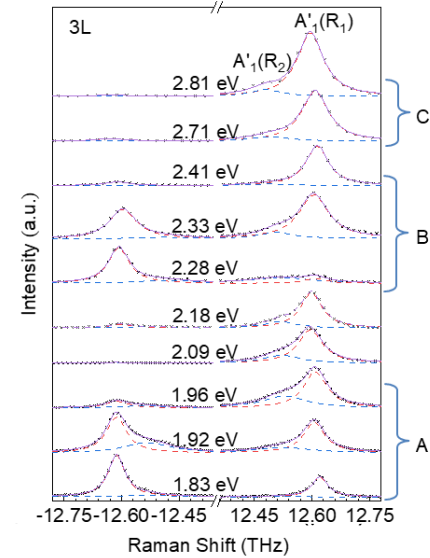
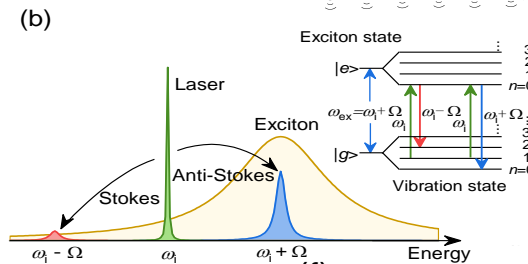
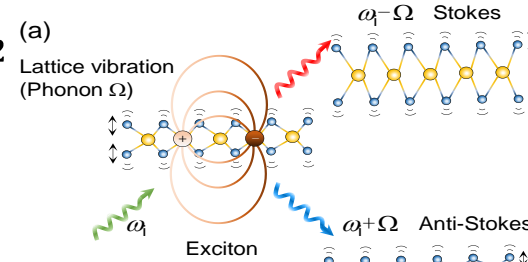
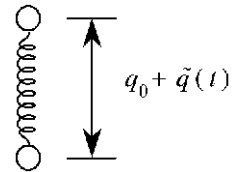


# Exciton Optomechanics

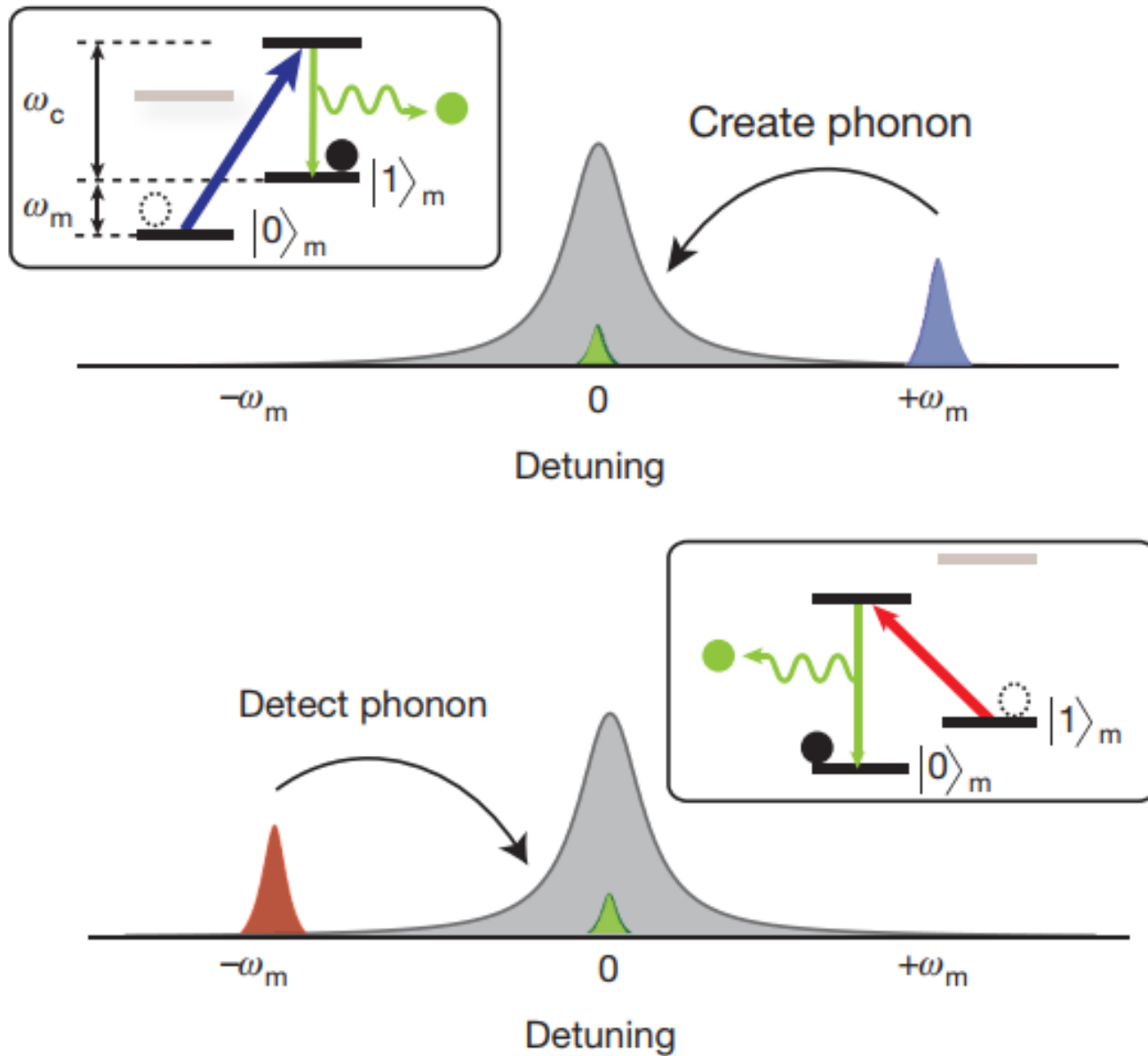
光场对晶格声子产生  
“辐射压力”



$$p_{oe} = -\frac{1}{2} \epsilon_0 \left( \frac{\partial \chi}{\partial \Delta r} \right) E^2$$



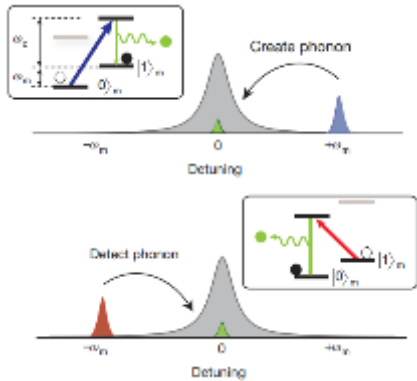
# Control of Phonon State



# 缺陷态与声子耦合

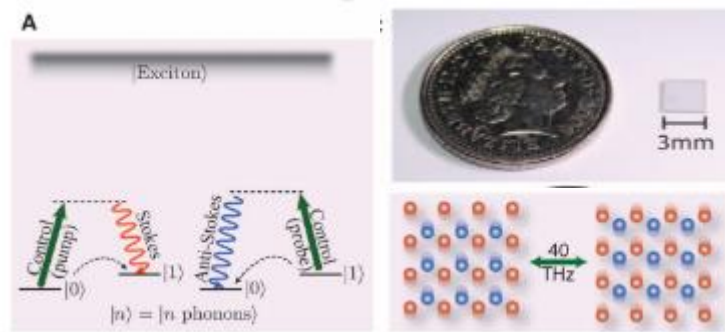
## 单光子-声子耦合

单光子-声子非经典关联



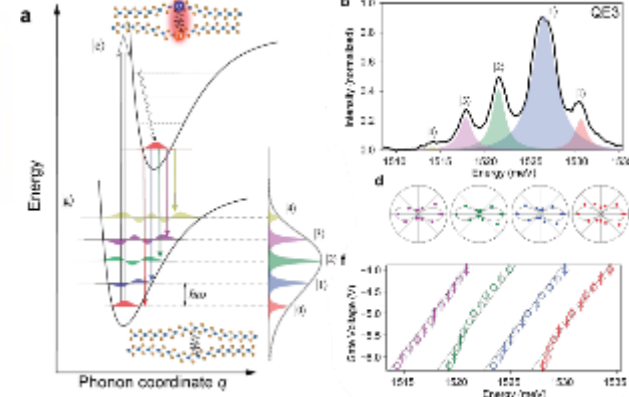
Riedinger, R., *Nature*, 2016, 530, 313.

空间分离的振动态之间的纠缠



Lee K. C., *Science*, 2011, 334, 1253

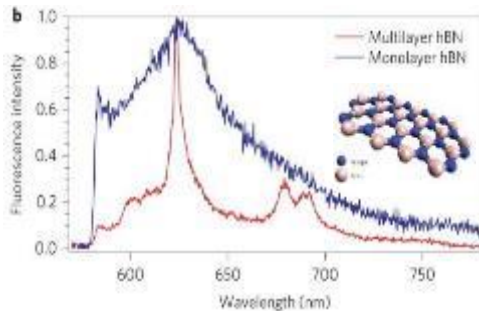
WSe<sub>2</sub>中的单光子-声子耦合



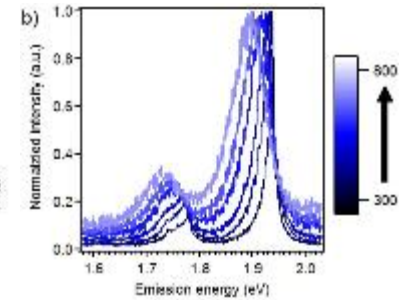
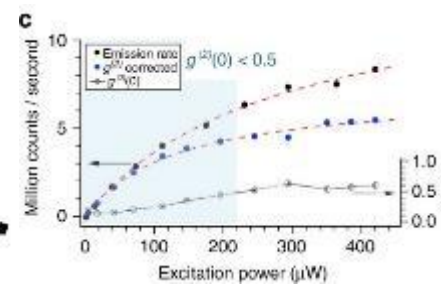
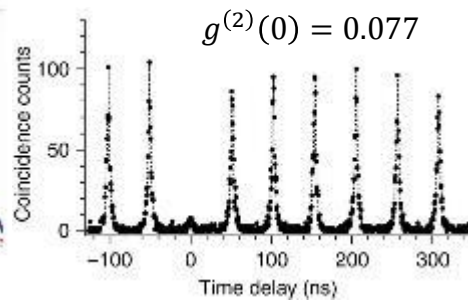
Ripin, A., 2023, arXiv:2302.13484

单光子-声子耦合较弱，或需要低温。

## hBN中的单光子发射



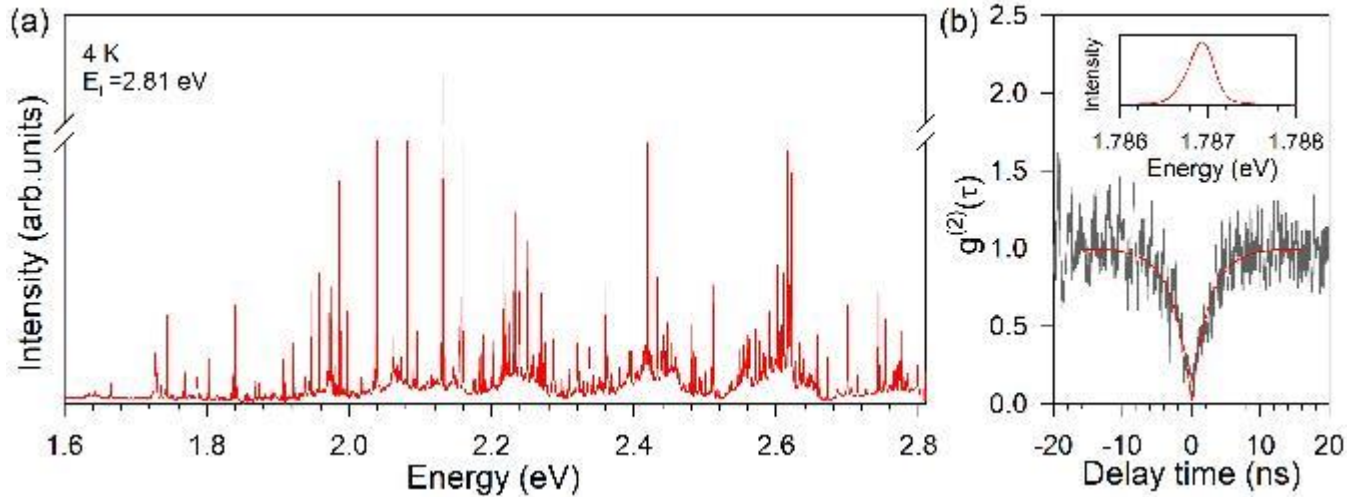
T. T. Tran, et al. *Nat. Nanotech.*, 2016, 11, 37  
 T. T. Tran, *ACS Nano*, 2016, 10, 7331  
 M. Kianinia, *ACS Photonics*, 2017, 4, 768  
 Grosso, G. et al. *Nat. Commun.*, 2017, 8, 105



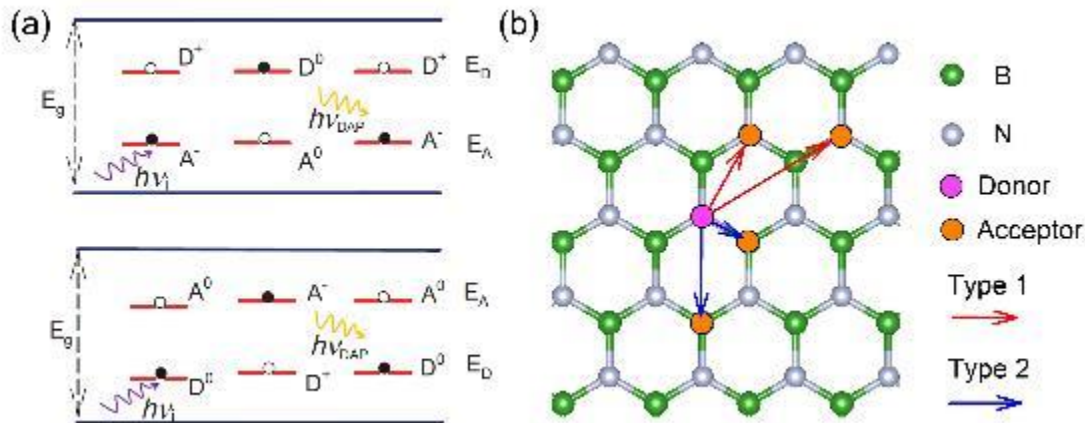
高纯度，高亮度，高温发射 (800 K)，UV~NIR，电场、压力可调.....

# DAP Quantum Emitters in hBN

4 K



linewidth  $\sim 75 \mu\text{eV}$

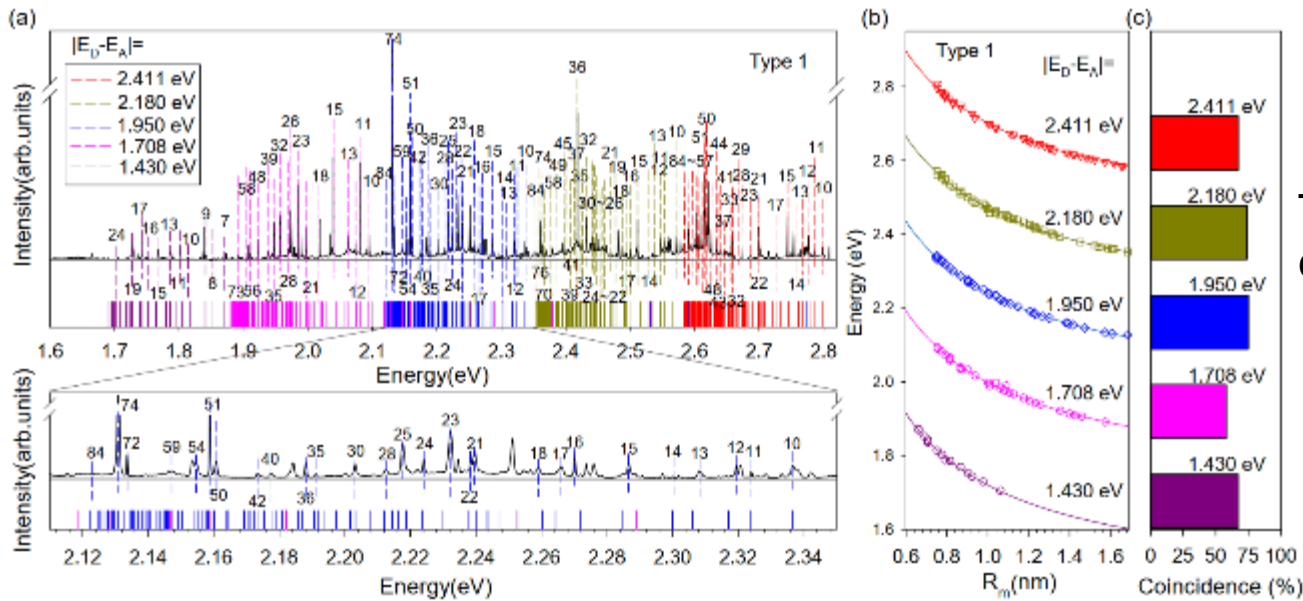


$$E(R_m) = |E_A - E_D| + \frac{e^2}{4\pi\epsilon\epsilon_0 R_m}$$

**Type 1:** Donors and acceptors are located on the same sublattice.

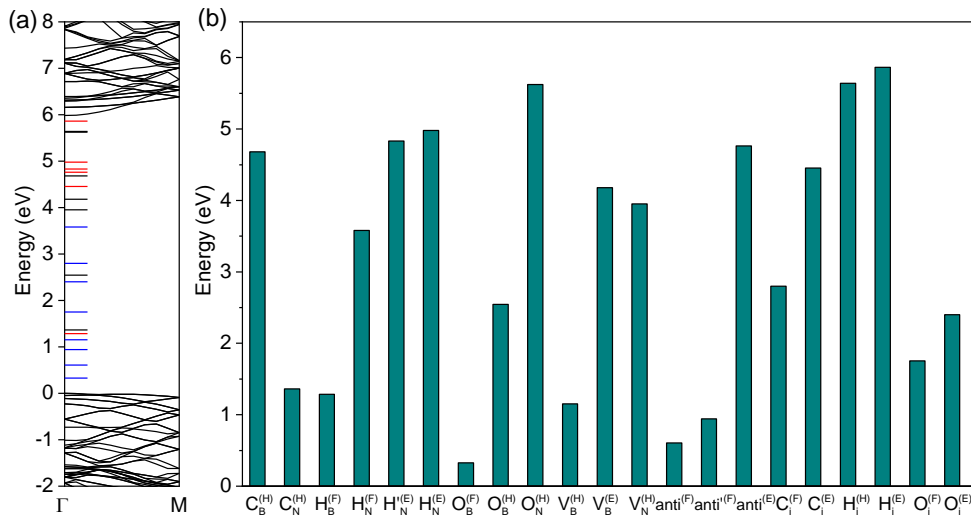
**Type 2:** Donors and acceptors occupy different sublattices.

# DAP Quantum Emitters in hBN

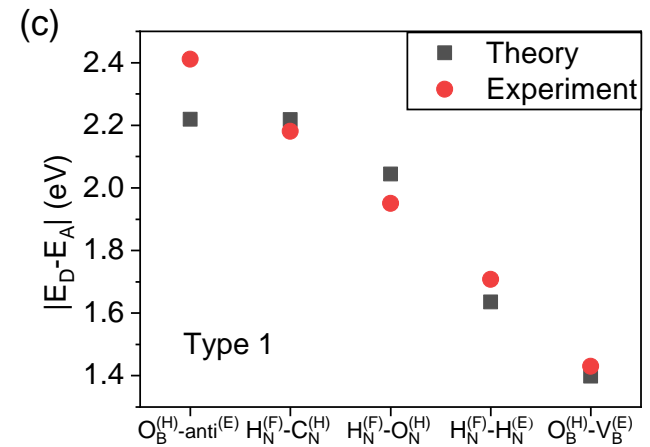


The calculated spectral distribution of type1 DAPs.

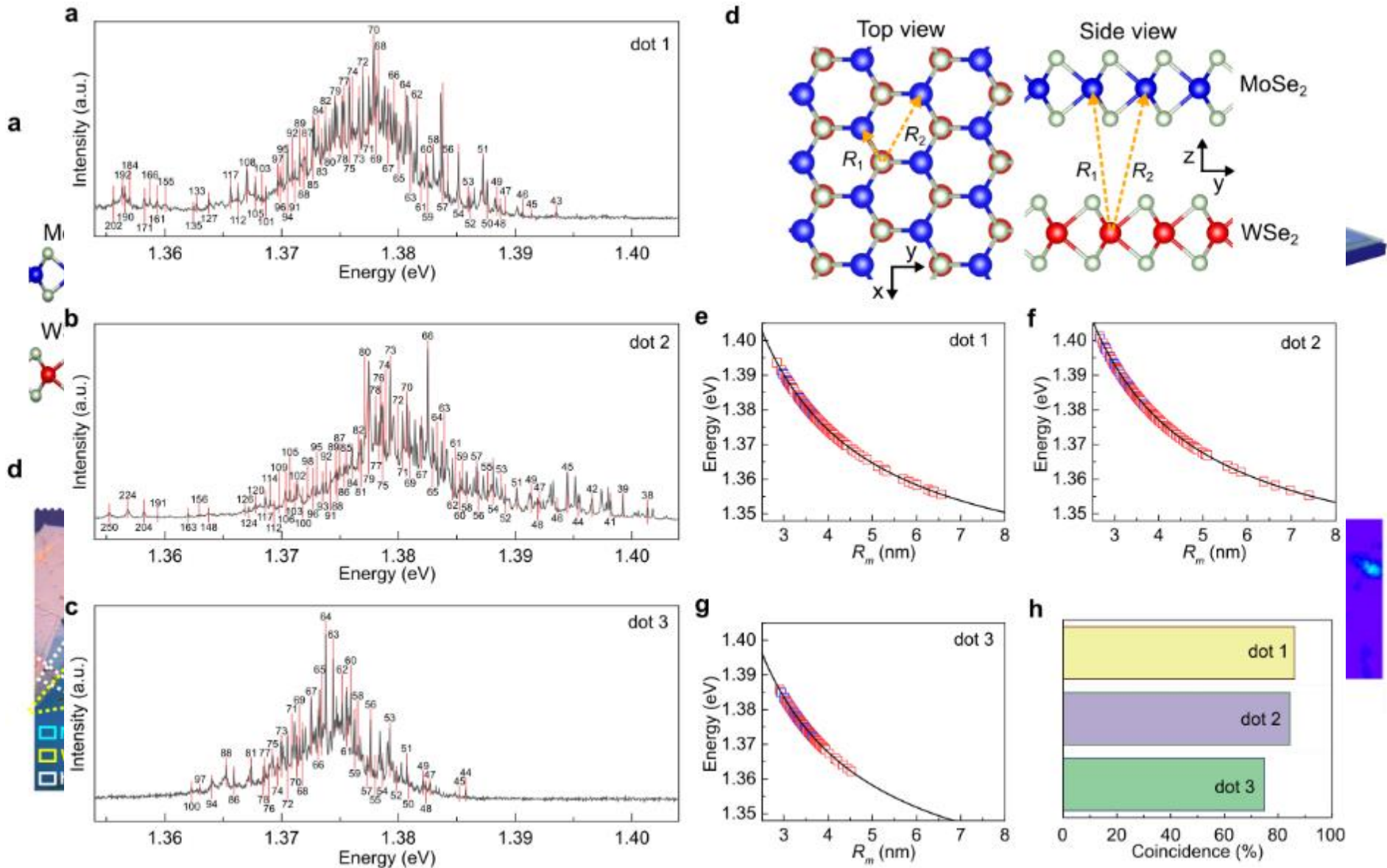
## DFT Calculation



The possible defects in hBN sample.

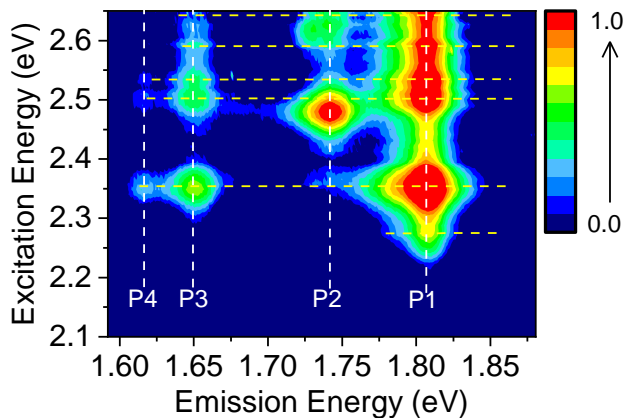


# DAP Quantum Emitters in Moire Hererobilayer

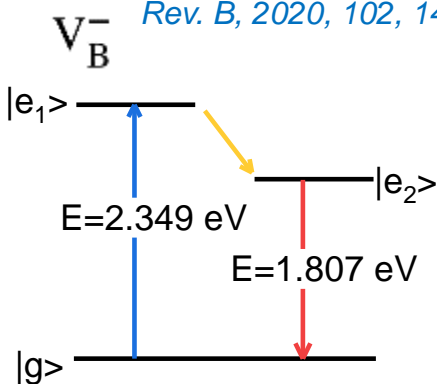


# single photon-phonon coupling in hBN

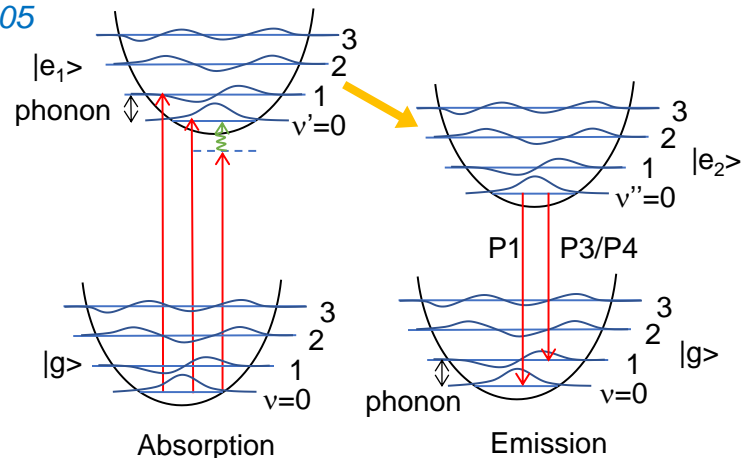
*J. R. Reimers, et al. Phys. Rev. B, 2020, 102, 144105*



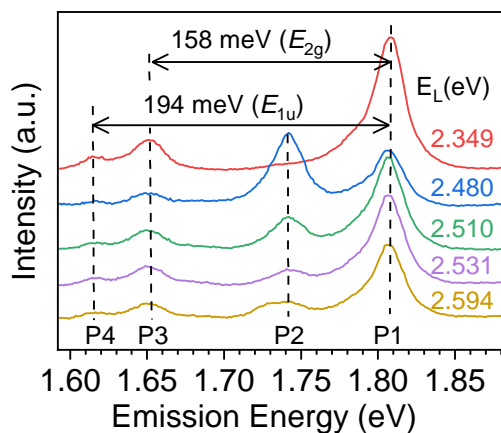
P1 :  $g^{(2)}(0)=0.09$



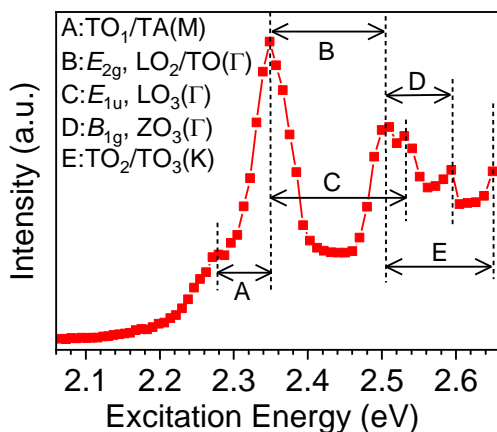
The three-level model of electron transition



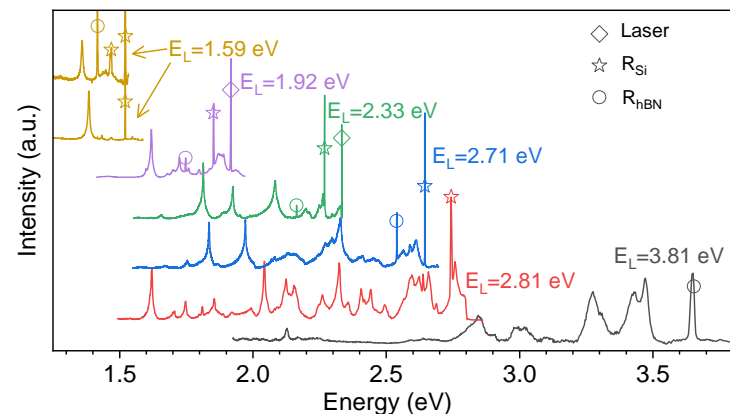
The generation of phonon Fock state



P1: Zero phonon line  
P3, P4: Phonon replica



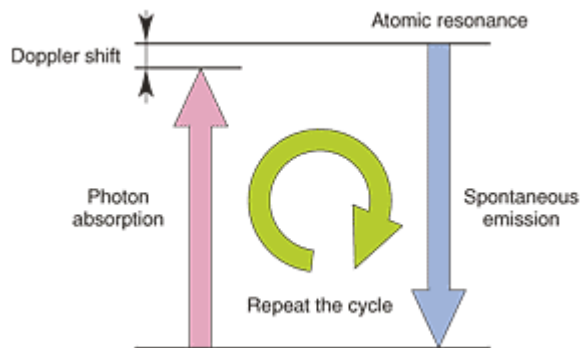
Phonon-assisted selectively enhanced excitation



UV to NIR hBN quantum emitters

# ASPL & Laser Cooling

## 激光冷却原子



Proposed in 1975 by Wineland, Dehmelt et al.;

realized in 1978 by Wineland, Drullinger, and Walls

## 与激光冷却原子相关的诺贝尔物理奖

1989: N. F. Ramsey, H. G. Dehmelt, W. Paul  
原子钟，离子阱，

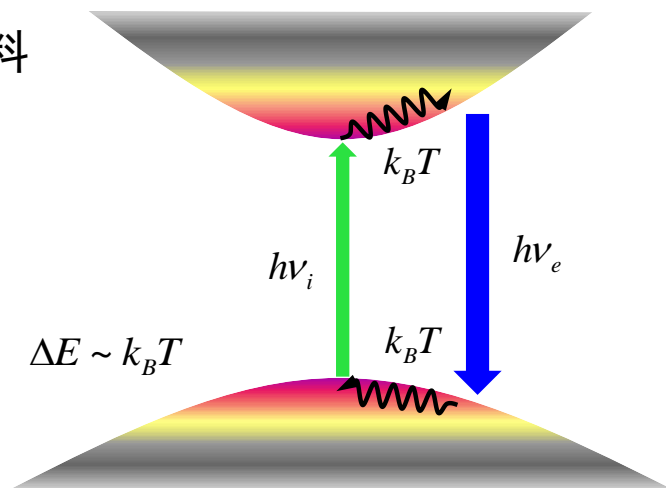
1997: S. Chu, C. C. Tannoudji, W. D. Phillips.  
激光冷却和捕获原子

2001: E. A. Cornell, W. Ketterle and C. E. Wieman

## 玻色-爱因斯坦凝聚

2012: S. Haroche and D. J. Wineland  
单个量子系统的测量和操控

## 凝聚态材料



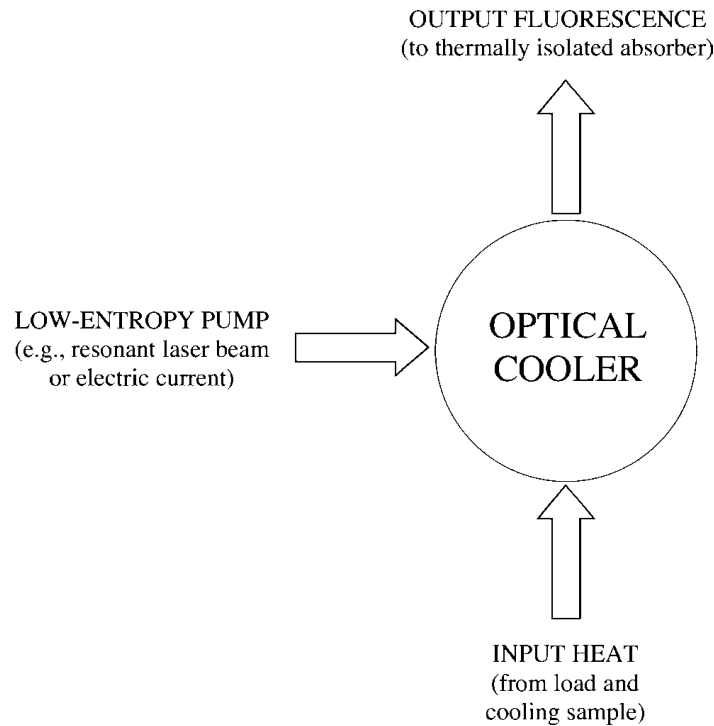
P. Pringsheim, *Z. Phys. A* **57**,  
739-746, (1929).

Note: In 1957, [C. H. Townes](#) and [A. L. Schawlow](#) invented laser at [Bell Labs](#)

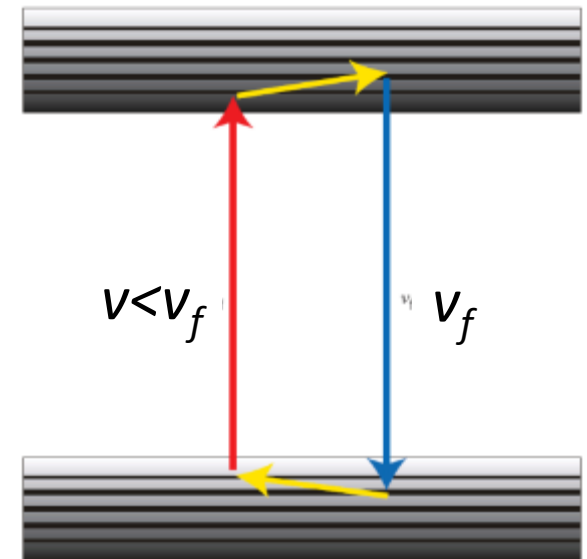


# ASPL & Laser Cooling

- Laser cooling of solids was proposed by Pringsheim in 1929, more than 30 years before the invention of laser.
- The principle of laser cooling is based on the upconversion luminescence:  $\nu < \nu_f$
- Thermodynamics



Peter Pringsheim



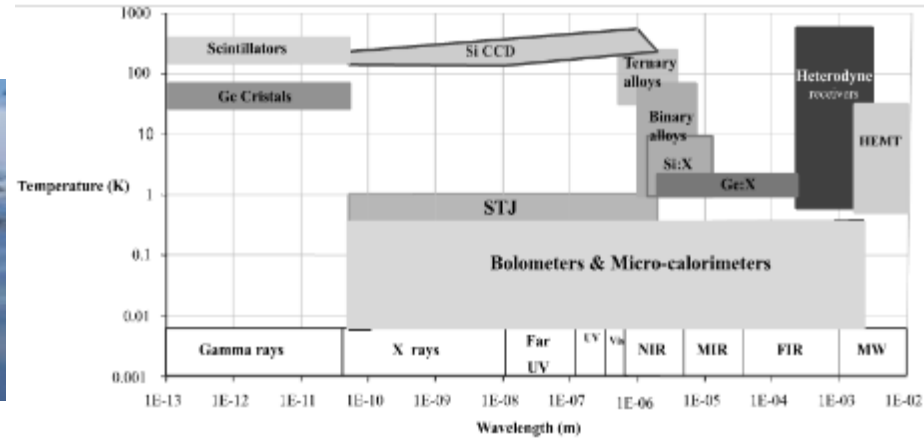
S. Vavilov, *J. Phys.* **9**, 68-72, 1945;

P. Pringsheim, *J. Phys.* **10**, 495-498, 1946;

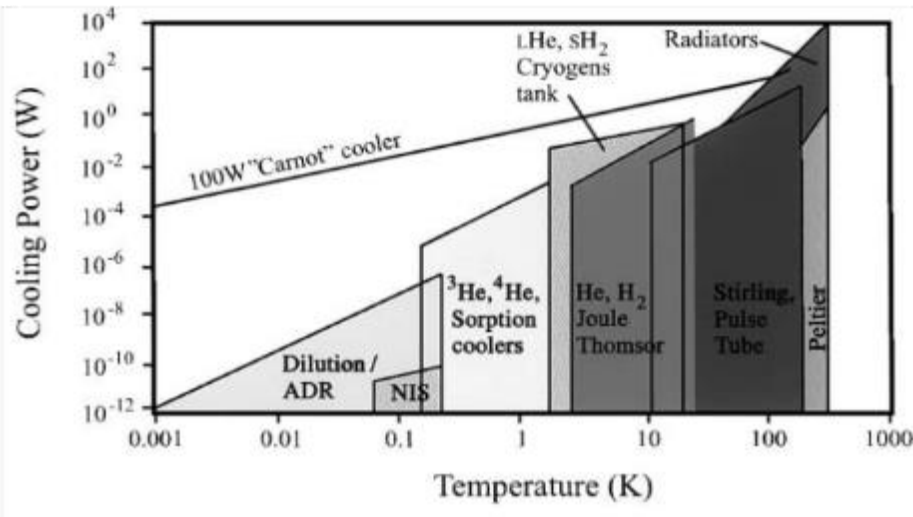
L. Landau, *J. Phys.* **10**, 499-502, 1946

C.E. Mungan, *J. Opt. Soc. Am. B* **20**, 1075, 2003.

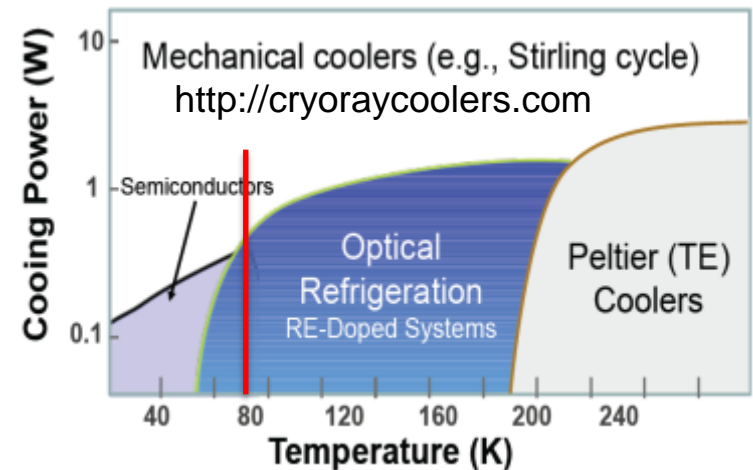
# Potential Applications



航空航天探测器，高灵敏红外探测器 绝大多数探测器需要工作在 100 K 以下



传统制冷器：压缩机，噪声，机械部分，制冷剂，笨重，寿命短

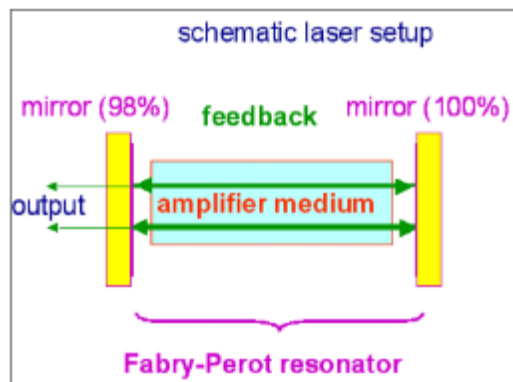


热电和光学制冷器：无机械振动，无制冷剂，轻便，可集成，长寿命

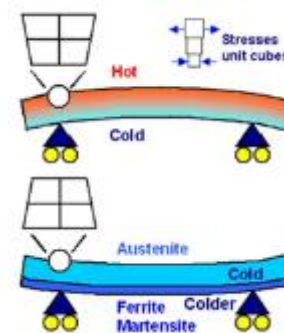
# Potential Applications



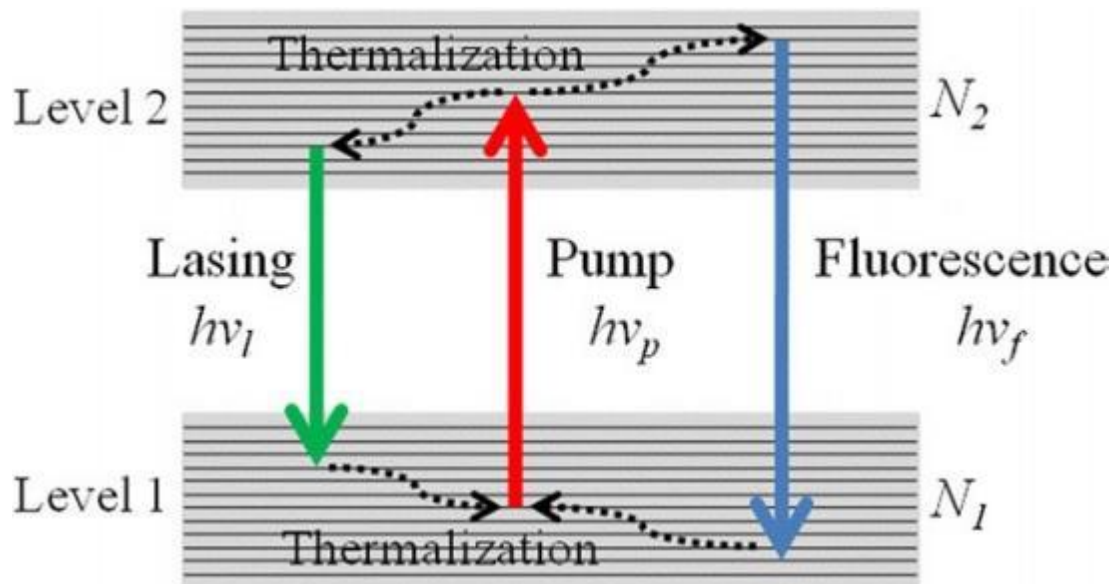
激光武器



需要对增益介质进行冷却降温

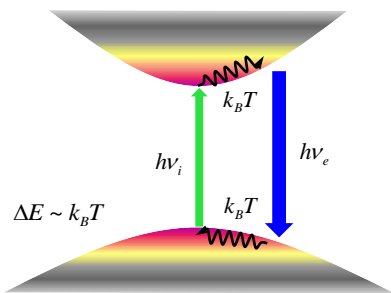


常规冷却方式，如水冷、液氮冷却造成晶体热分布不平衡，产生应力并断裂

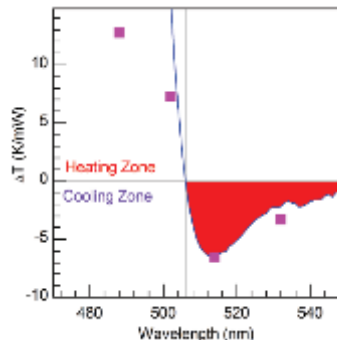
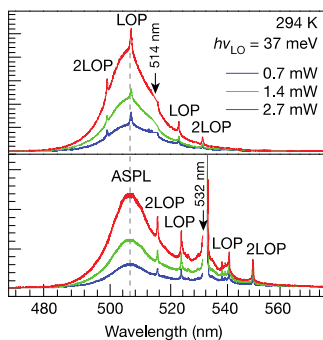


解决方案：激光冷却，辐射平衡激光器

# 整个样品的光学冷却，ASPL



声子辅助荧光上转换



**1995**  
R. Epstein, LANL  
Yb<sup>3+</sup>-doped glass

**2010**  
Sheik-Bahae, UNM  
Yb<sup>3+</sup>-doped crystal

**2013**  
张俊等  
II-VI Semiconductors

**2016**  
张俊等  
LO phonon Cooling

**2018**  
M. P. Hehlen  
et al., LANL  
YLF:Yb optical  
cryocooler

**2020**  
彭笑刚等  
浙江大学  
CdS/CdSe量子点

**2021**  
M. Sheldon et al.,  
Texas A&M  
University  
CsPbBr<sub>3</sub> QDs

**“冷却极限？  
其他方案？”**

*0.3 K cooling  
Nature 1995*

*190 K cooling  
Nature Photonics  
2010*

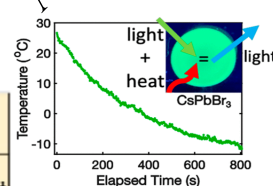
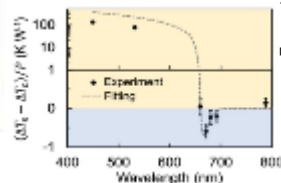
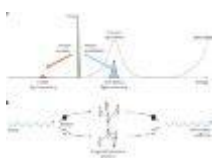
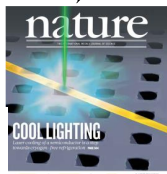
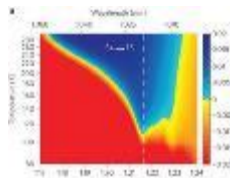
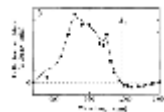
*Nature, 2013*

*Nature Photonics,  
2016*

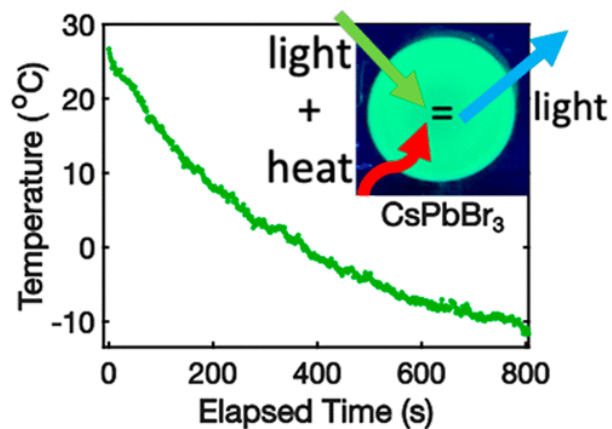
*Optical Cryocooler  
Light: S. & A.  
2018*

*Nature Commun. 2020*

*Nano Lett., 2021*



# 其他半导体材料的激光冷却进展



*Nano Lett.* 2020, 20, 12, 8874–8879

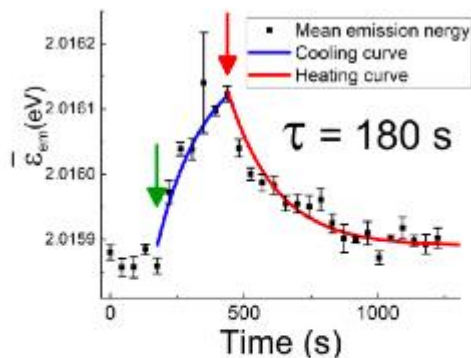


Figure 4.13. Mean emission energy of the sample's PL spectra during the experiment.  $\tau_c = 30$  s was used in the experiment, where the pump laser ( $\epsilon_{ex} = 1.941$  eV) was introduced right after the fifth temperature measurement (green arrow) and turned off right after the eleventh measurement (red arrow).

Purdue University的CdS/CdSe量子点。

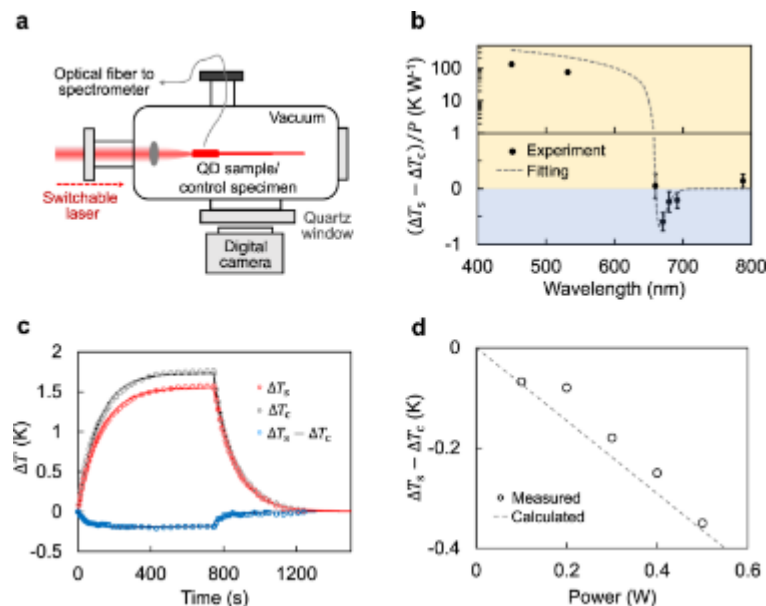
[Submitted on 13 Jan 2021]

## Laser Cooling of Germanium Semiconductor Nanocrystals

Manuchehr Ebrahimi, Wei Sun, Amr S. Helmy, Nazir P. Kherani

Laser cooling of matter through anti-Stokes photoluminescence, where the emitted frequency of lig successfully realized in condensed media, and in particular with rare earth doped systems achieving potential of achieving temperatures down to  $\sim 10$ K and that its direct integration can usher unique f semiconductors has been reported recently, laser cooling of indirect bandgap semiconductors such observation of dominant anti-Stokes photoluminescence in germanium nanocrystals. We attribute t electron-hole plasma, the inherent degeneracy of longitudinal and transverse optical phonons in nc intensities, laser cooling with lattice temperature as low as  $\sim 50$ K is inferred.

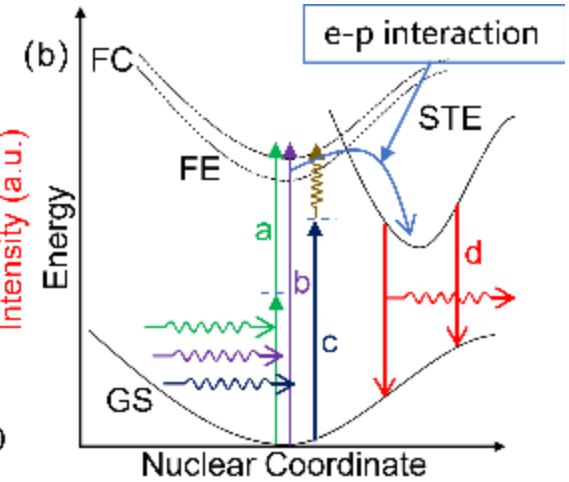
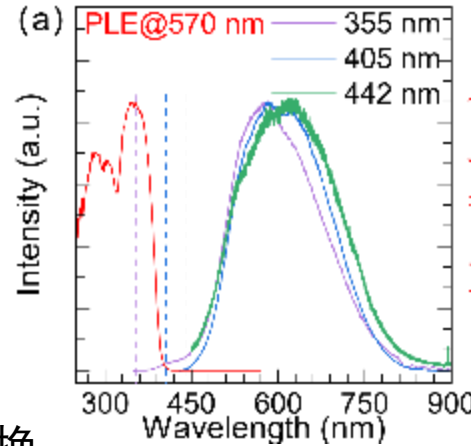
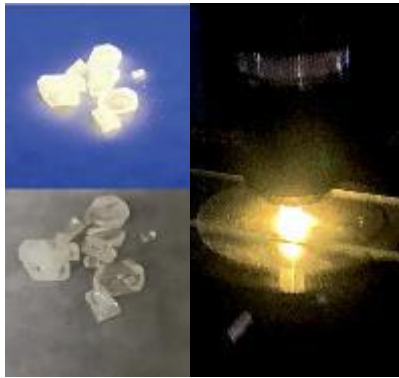
加拿大Ge纳米晶



Ye, Z., Lin, X., Wang, N. *et al.* *NatCommun* **12**, 4283 (2021)

# 光学冷却极限—零点振动

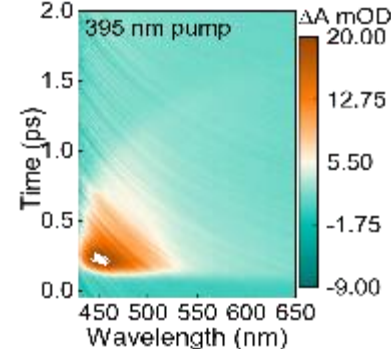
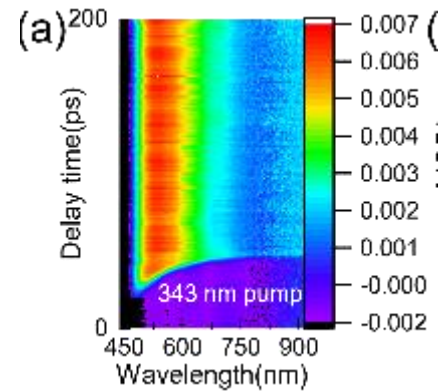
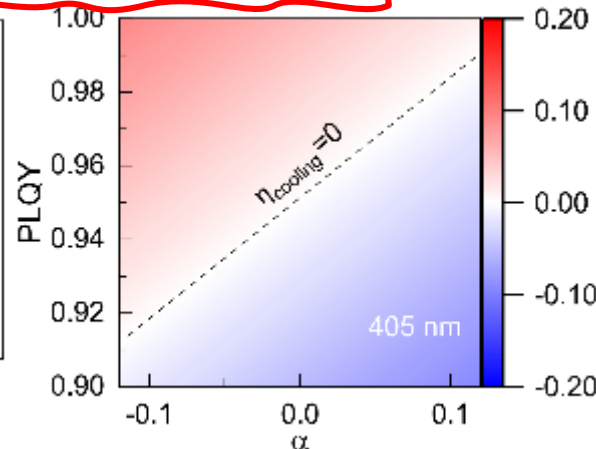
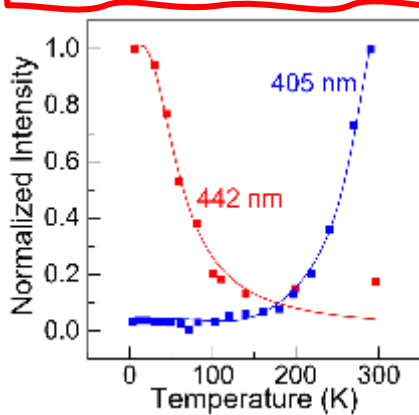
强电声相互作用-自陷态-高荧光量子效率-高光子逃逸效率



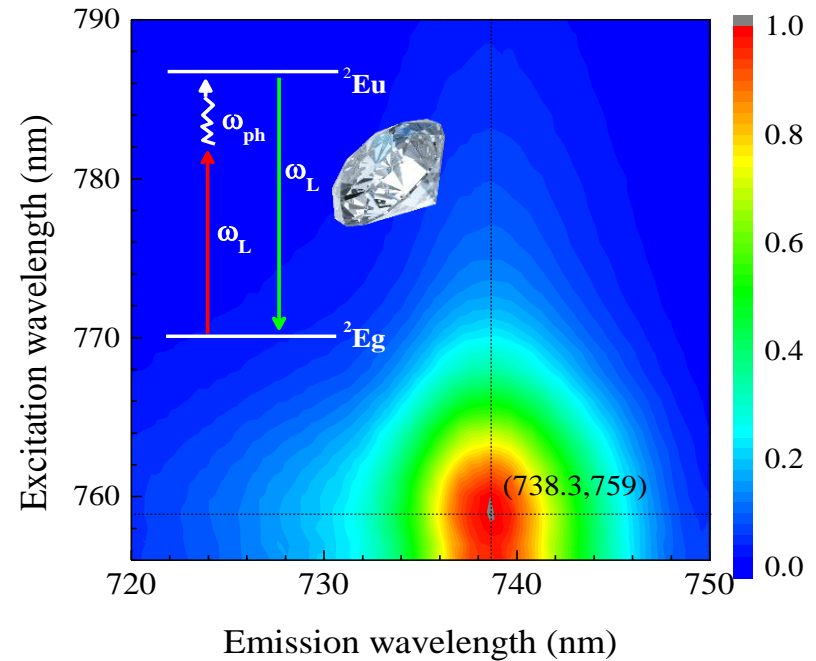
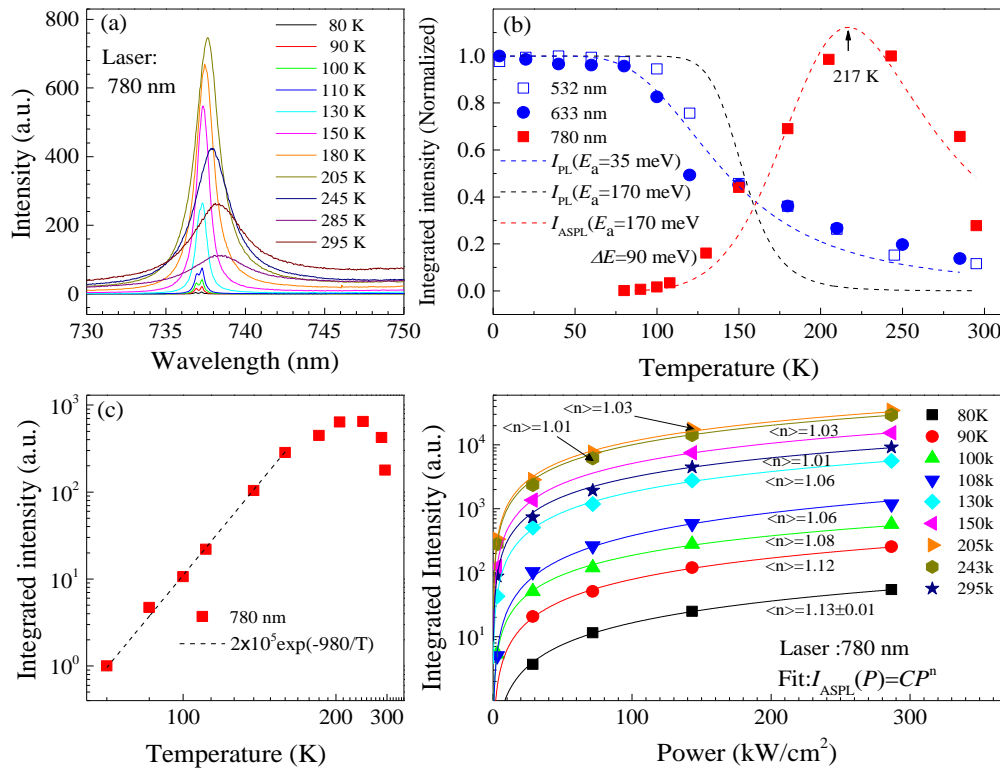
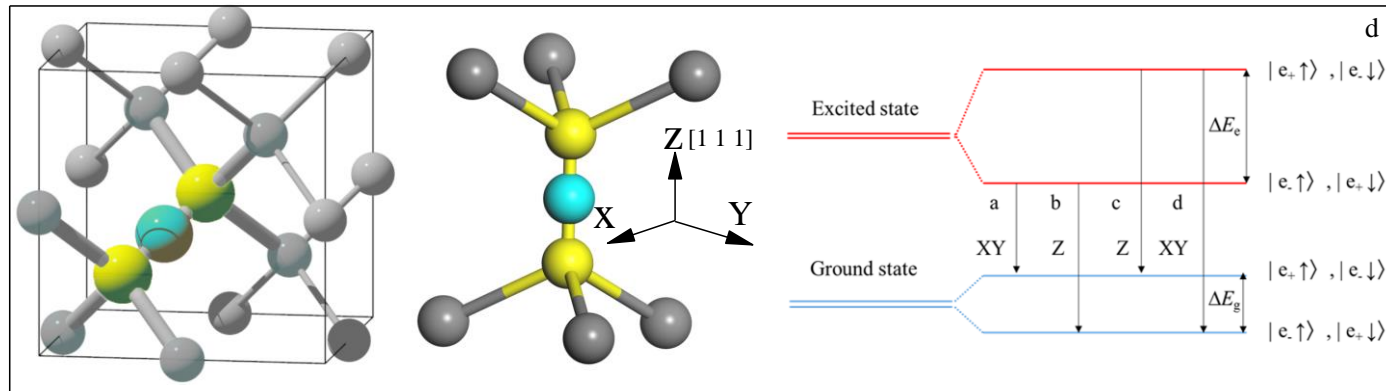
可以实现4-300K声子辅助荧光上转换

$$I_{UCPL}(T) = \frac{I_0 \eta_e A T^{-3/2}}{\eta_e A T^{-3/2} + B e^{-E_a/k_B T}} \left( \frac{1}{\exp\left(\frac{h\nu}{k_B T}\right) - 1} + 1 \right)^{CT+1}$$

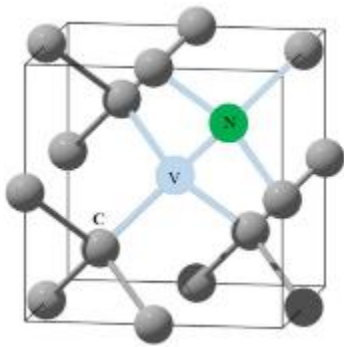
上转换激发下，光场和声子作用形成Floquet态，加速上转换发光



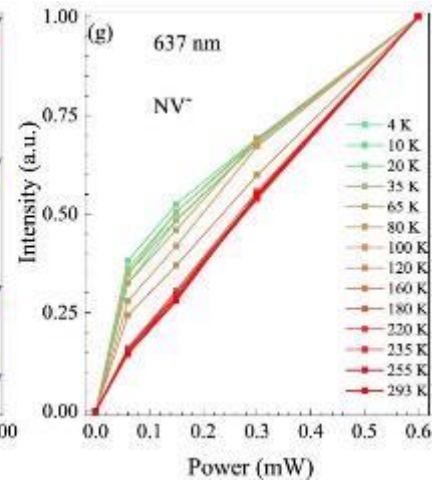
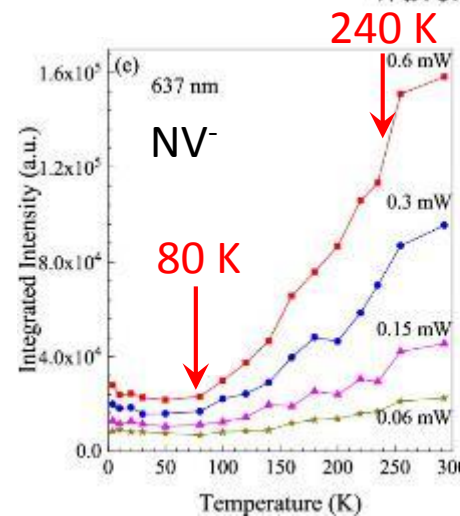
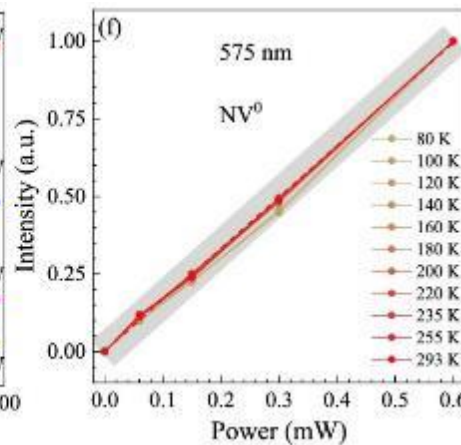
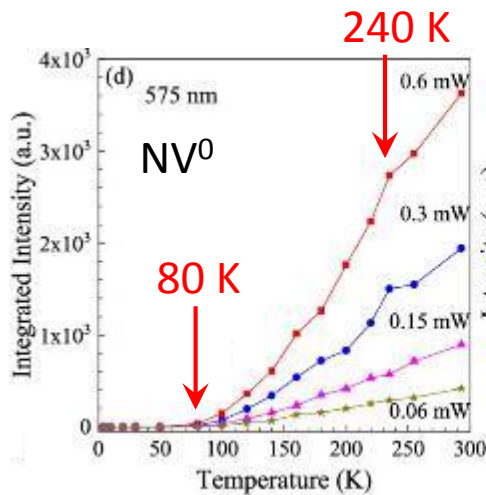
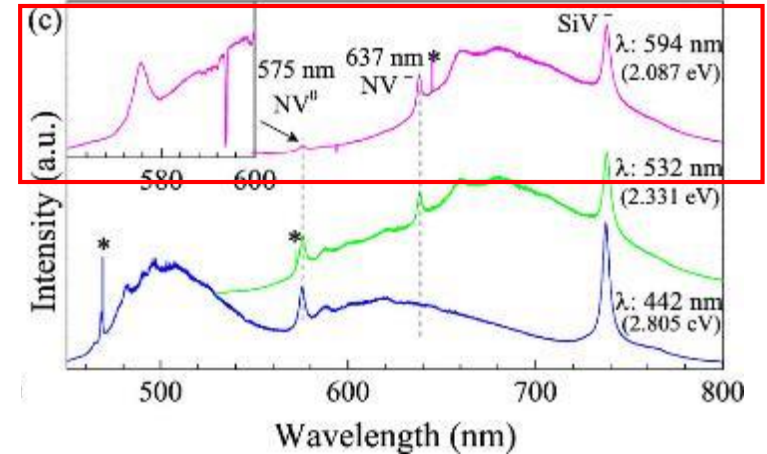
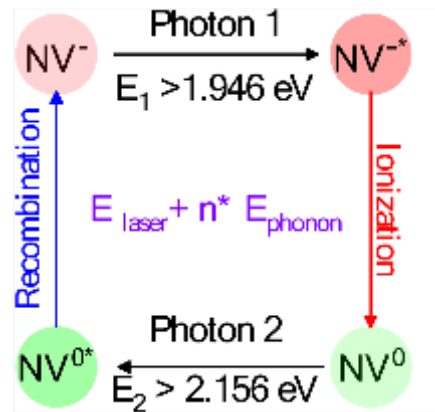
# Diamond SiV-ASPL



# Charge state manipulation by ASPL



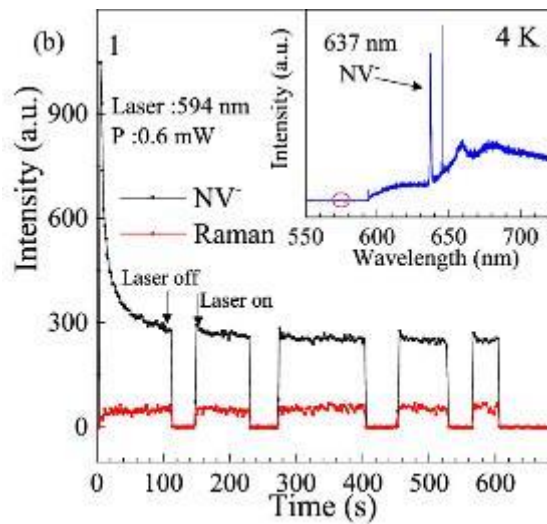
NV in diamond



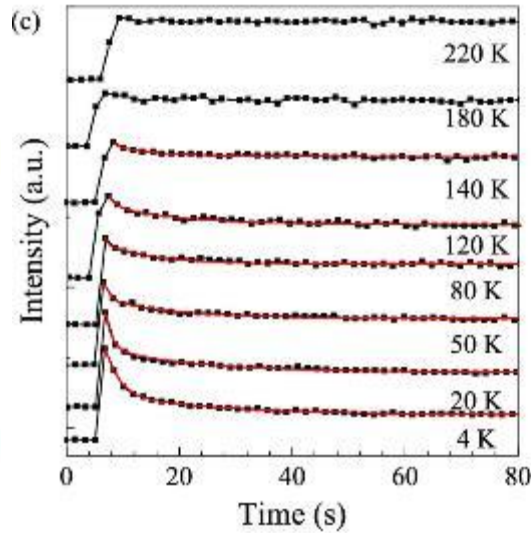
- < 80 K, unidirectional process,  $\text{NV}^- \rightarrow \text{NV}^0$ ;  $\text{NV}^-$  saturation phenomenon.
- 80-240 K, phonon-assisted PL upconversion, closed-loop conditions,  $\text{NV}^- \rightleftharpoons \text{NV}^0$ ;  $\text{NV}^-$  saturation phenomenon disappears.
- > 240 K, external quantum efficiency is significantly reduced.



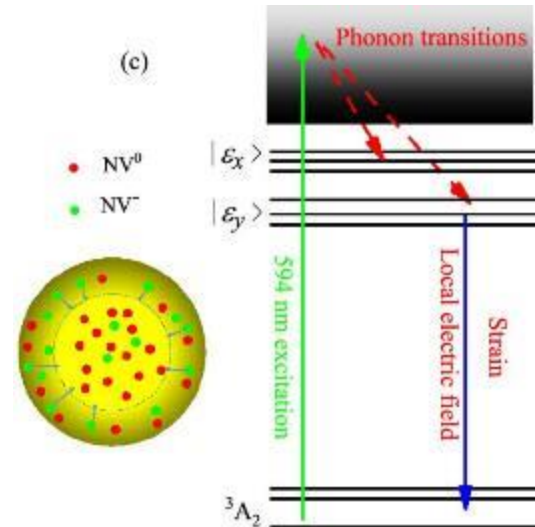
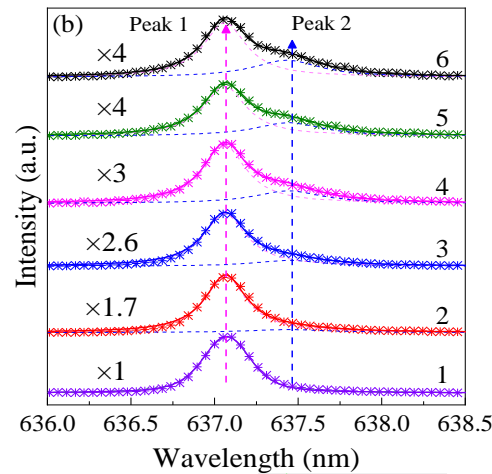
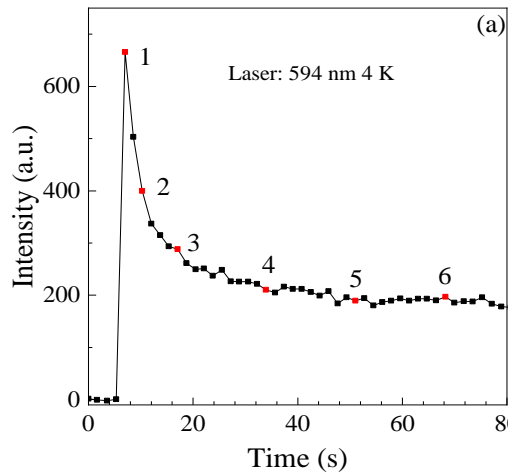
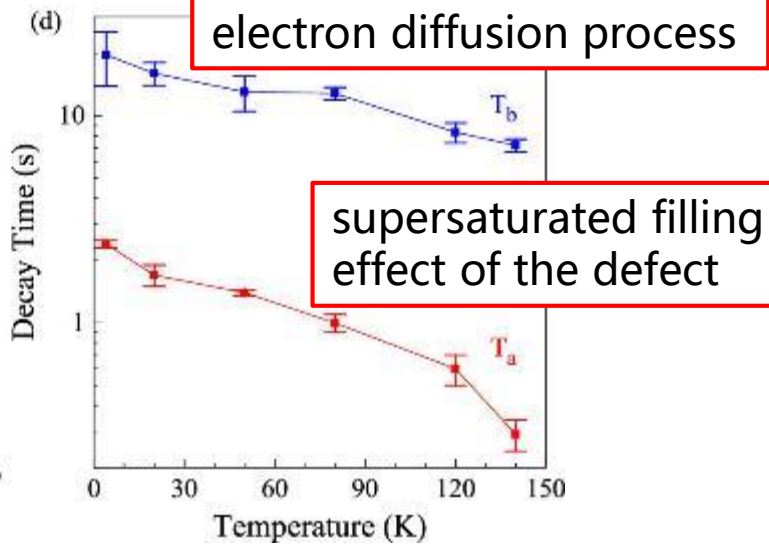
# Evolution of the PL of the NV-



4 K, NV<sup>-</sup> → NV<sup>0</sup>

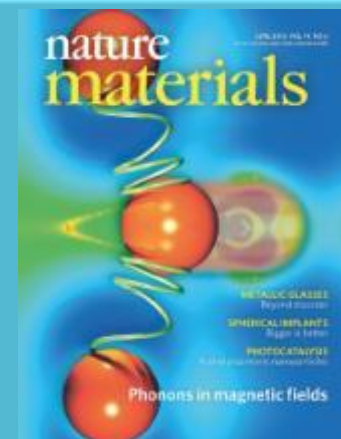
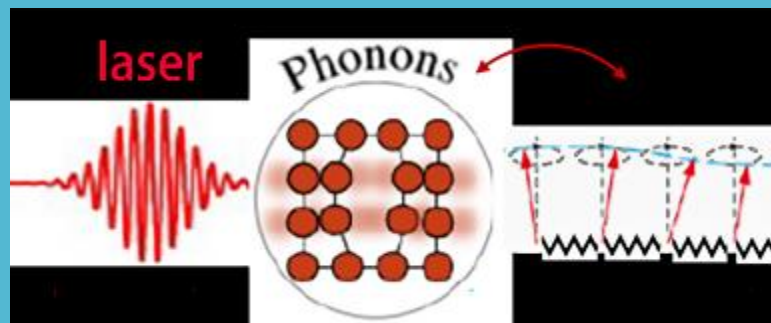
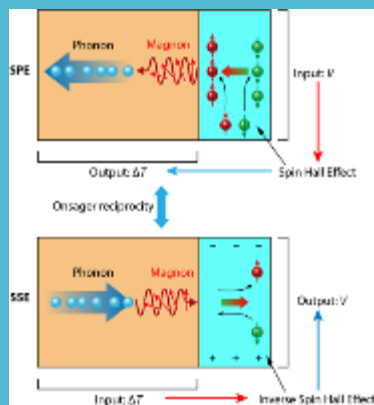


> 80 K, NV<sup>-</sup> ↔ NV<sup>0</sup>



# 自旋晶格相互作用

如何用拉曼散射探测自旋声子耦合？



自旋 Peltier /Seebeck 效应

Ref : Nature 455, 778 (2008)

声子磁响应

Ref : Nature Materials, June (2015)

问题1：磁相变如何影响声子特性？

→ 提供自旋涨落与晶格相互作用物理

问题1：电子态共振激发下自旋-声子耦合？

→ 提供电子-声子-自旋相互作用信息

问题3：如何自旋波-声子混合量子态？

→ 新量子体系：半声子-半自旋波准粒子

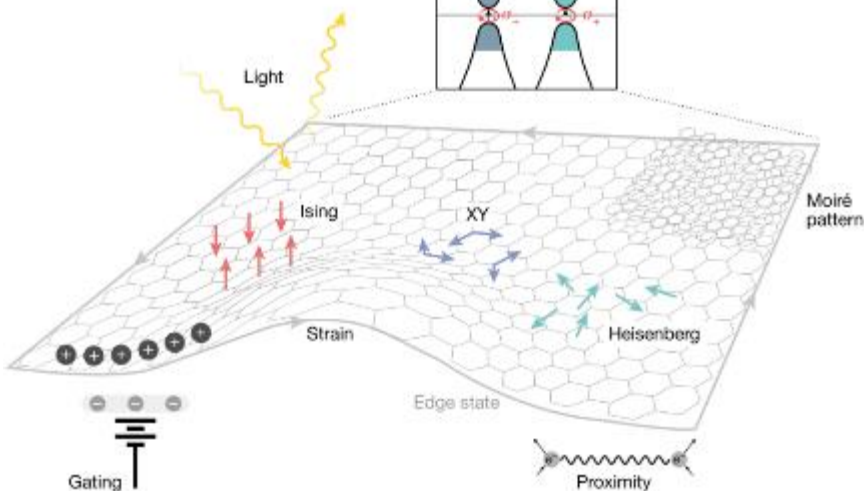
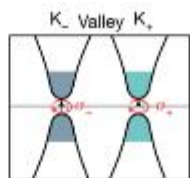
# 二维磁性材料

Chalcogenides	$\text{Cr}_2\text{Ge}_2\text{Te}_6$ , $\text{Cr}_2\text{Si}_2\text{Te}_6$ , $\text{Fe}_3\text{GeTe}_2$ , $\text{VSe}_2^*$ , $\text{MnSe}_x^*$	$\text{Fe}_2\text{P}_2\text{S}_6$ , $\text{Fe}_2\text{P}_2\text{Se}_6$ , $\text{Mn}_2\text{P}_2\text{S}_6$ , $\text{Mn}_2\text{P}_2\text{Se}_6$ , $\text{Ni}_2\text{P}_2\text{S}_6$ , $\text{Ni}_2\text{P}_2\text{Se}_6$ , $\text{CuCrP}_2\text{Se}_6^*$ , $\text{AgVP}_2\text{S}_6$ , $\text{AgCrP}_2\text{S}_6$ , $\text{CrSe}_2$ , $\text{CrTe}_3$ , $\text{Ni}_3\text{Cr}_2\text{P}_2\text{S}_9$ , $\text{MnBi}_2\text{Te}_4^*$ , $\text{MnBi}_2\text{Se}_4^*$	$\text{CuCrP}_2\text{S}_6$
Halides	$\text{CrI}_3^*$ , $\text{CrBr}_3$ , $\text{GdI}_2$	$\text{CrCl}_3$ , $\text{FeCl}_2$ , $\text{FeBr}_2$ , $\text{FeI}_2$ , $\text{MnBr}_2$ , $\text{CoCl}_2$ , $\text{CoBr}_2$ , $\text{NiCl}_2$ , $\text{VCl}_2$ , $\text{VBr}_2$ , $\text{VI}_2$ , $\text{FeCl}_3$ , $\text{FeBr}_3$ , $\text{CrOCl}$ , $\text{CrOBr}$ , $\text{CrSBr}$ , $\text{MnCl}_2^*$ , $\text{VCl}_3^*$ , $\text{VBr}_3^*$	$\text{CuCl}_2$ , $\text{CuBr}_2$ , $\text{NiBr}_2$ , $\text{NiI}_2$ , $\text{CoI}_2$ , $\text{MnI}_2$
			$\alpha\text{-RuCl}_3$
Others	$\text{VS}_2$ , $\text{InP}_3$ , $\text{GaSe}$ , $\text{GaS}$	$\text{MnX}_3$ ( $X = \text{F}, \text{Cl}, \text{Br}, \text{I}$ ), $\text{FeX}_2$ ( $X = \text{Cl}, \text{Br}, \text{I}$ ), $\text{MnSSe}$ , $\text{TiCl}_3$ , $\text{VCl}_3$	$\text{SnO}$ , $\text{GeS}$ , $\text{GeSe}$ , $\text{SnS}$ , $\text{SnSe}$ , $\text{GaTeCl}$ , $\text{CrN}$ , $\text{CrB}_2$

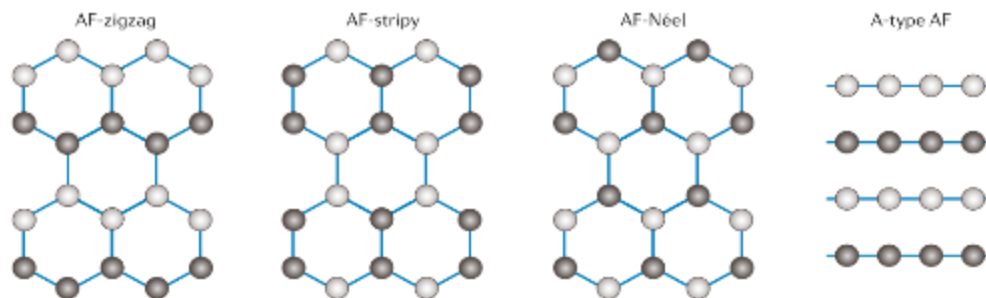
G. Cheng, Science, (2019)

## 基本磁性模型

$$H = -\frac{1}{2} \sum_{i,j} \left( JS_{i,j} \cdot S_j + \Lambda S_i^z S_j^z \right) - \sum_i A (S_i^z)^2$$



K.S. Burch, Nature, (2018)



X.Z. Wang, 2D Mater., (2016)

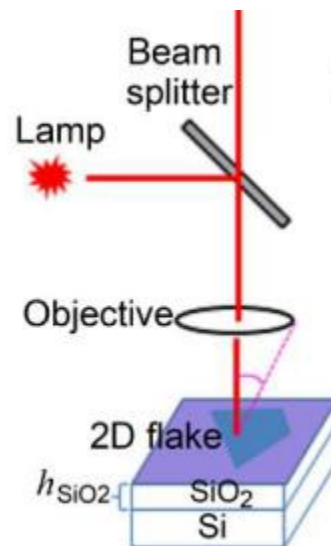
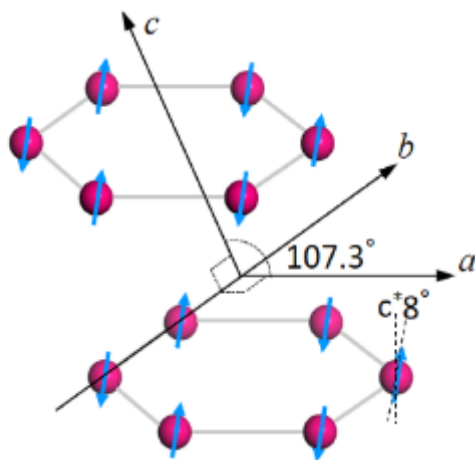
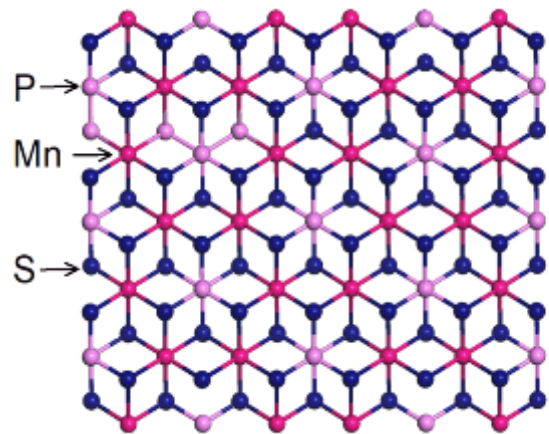
K.F. Mak, Nat. Rev. Phys., (2019)

## 二维反铁磁材料的优势：

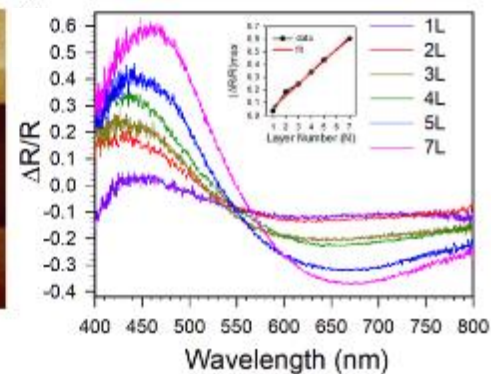
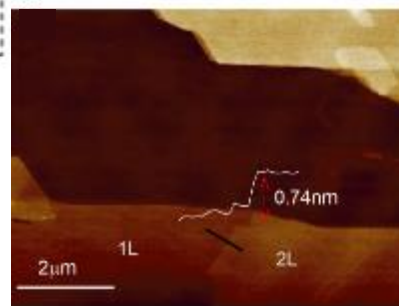
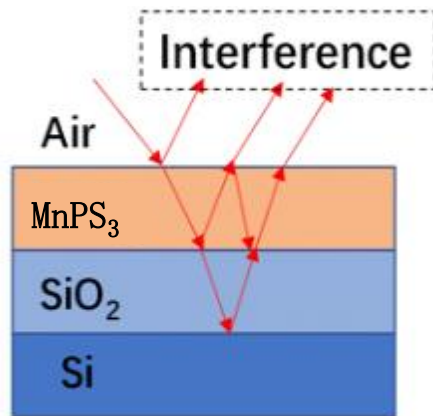
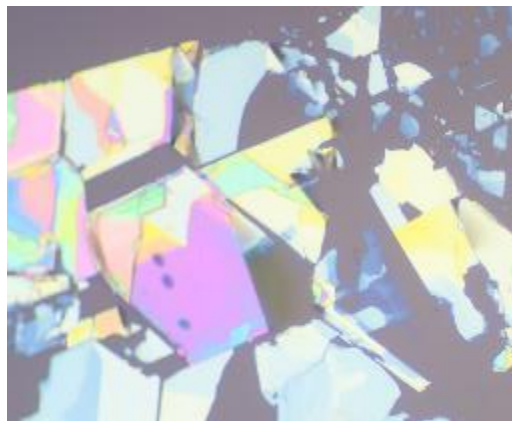
1. 磁共振频率在太赫兹范围
2. 弥散场为零
3. 对外磁场扰动不敏感
4. 更丰富的磁结构

# MPX3的性质介绍

## MnPS<sub>3</sub>的结构特点和光学衬度谱



$$OC(\lambda) = 1 - R_{2dm+Sub}(\lambda) / R_{Sub}(\lambda)$$

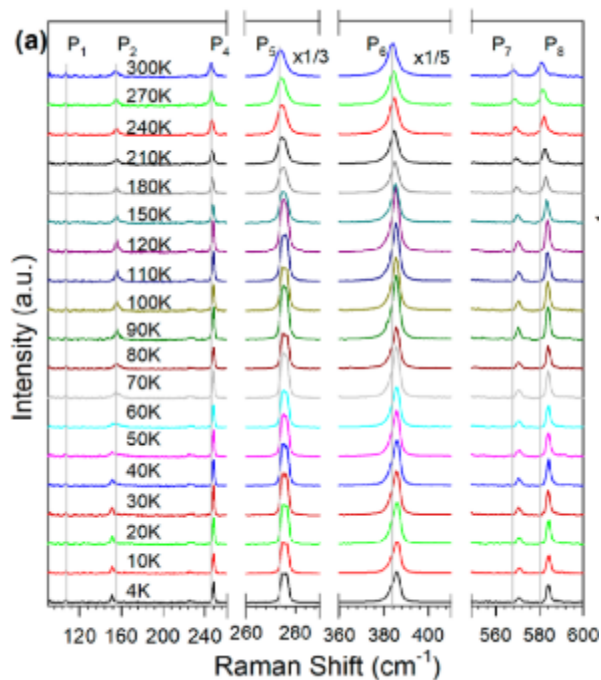


$$0.1 \times (N - 1) < (\Delta R/R)_{max} < 0.1 \times N (N \leq 7)$$

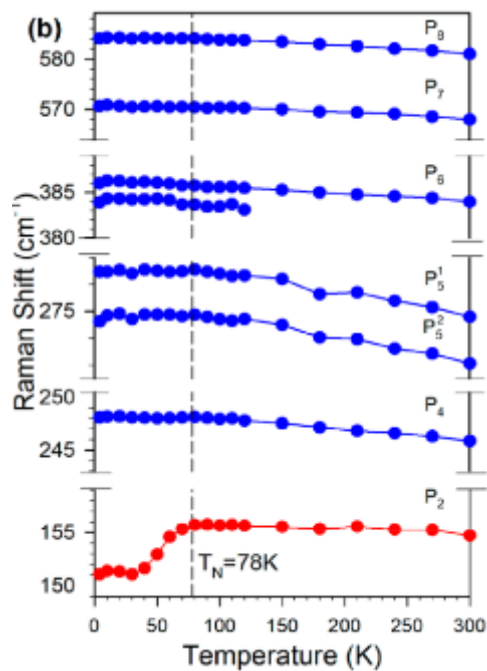
结合原子力显微镜和光学衬度法确定少层MnPS<sub>3</sub>样品的厚度

# MnPS3的相变研究

变温拉曼光谱

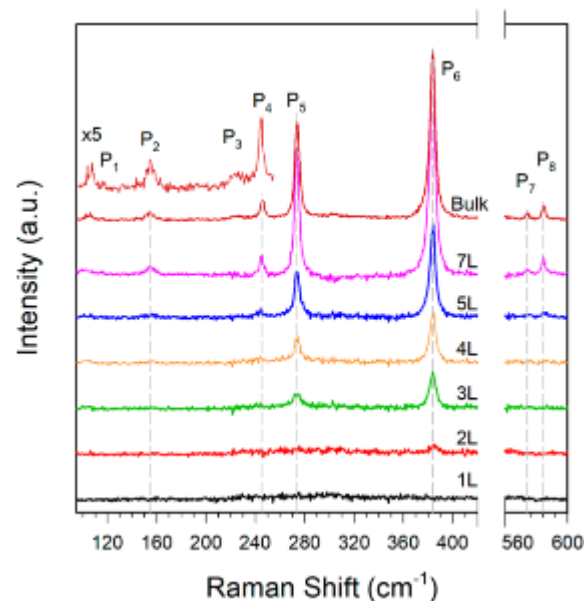


$P_2$ 模式频率与温度的关系

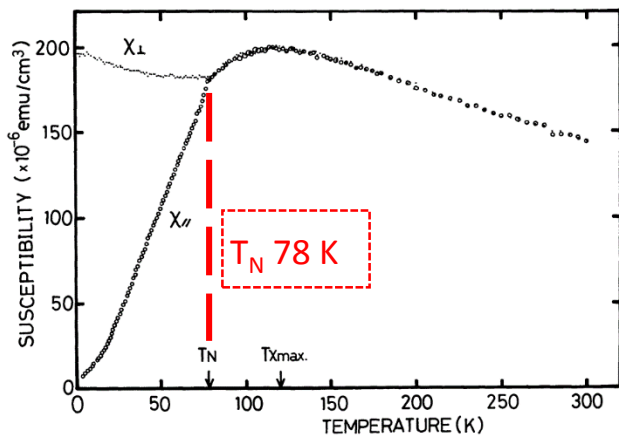
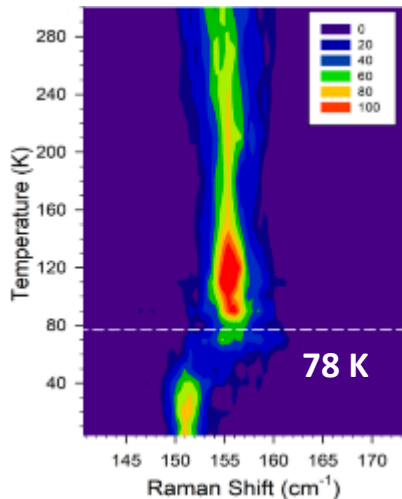
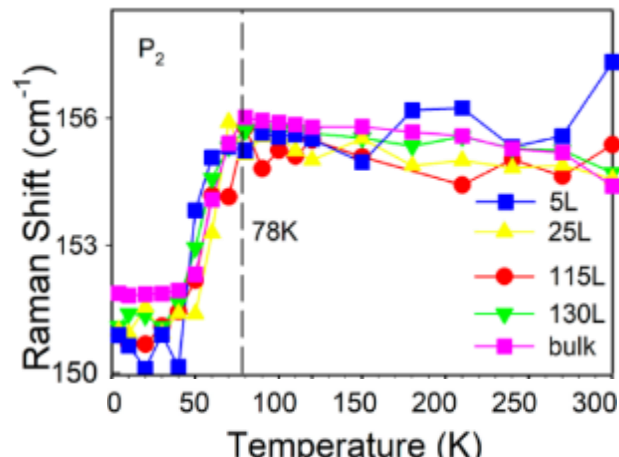


层间耦合

不同厚度MnPS<sub>3</sub>的拉曼光谱

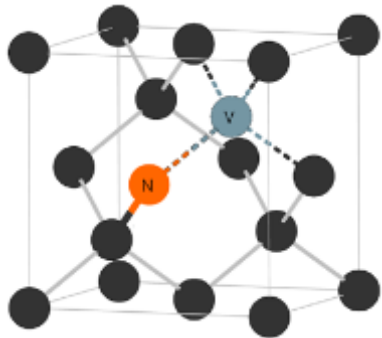


层间耦合较弱  $T_N$ 不依赖于层数

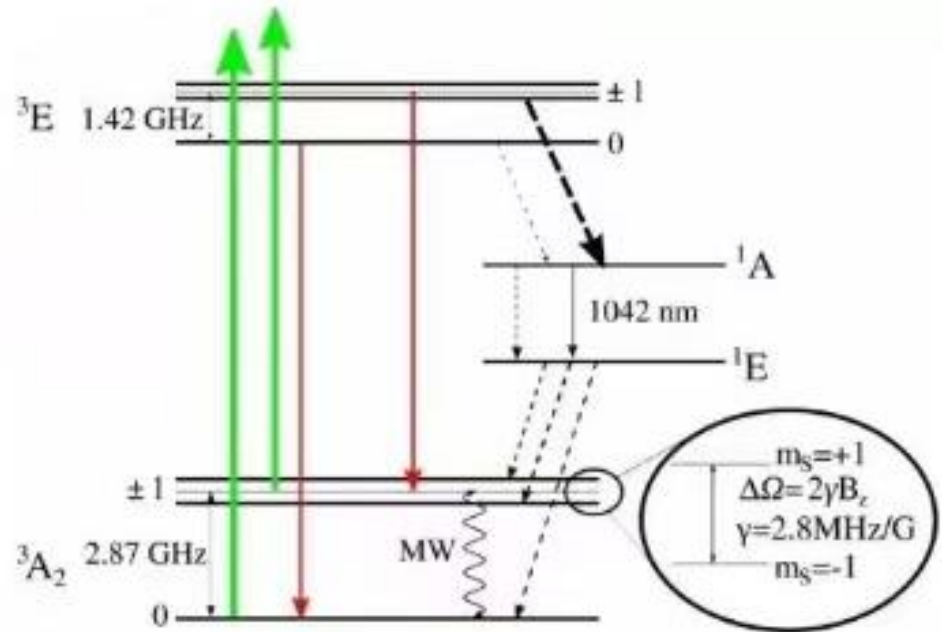


# 光探测磁共振 (ODMR)

实验原理：以金刚石中NV色心为例



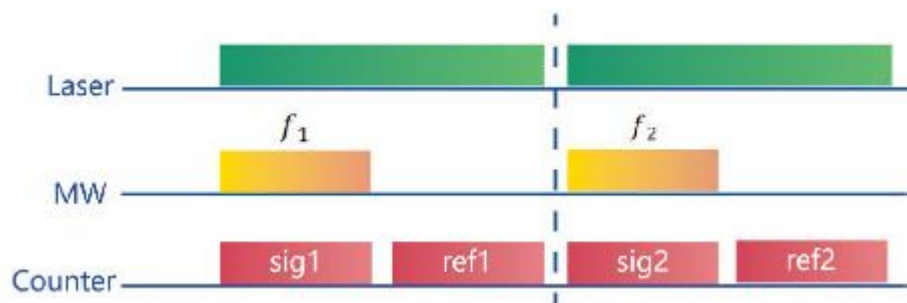
金刚石中NV色心的结构



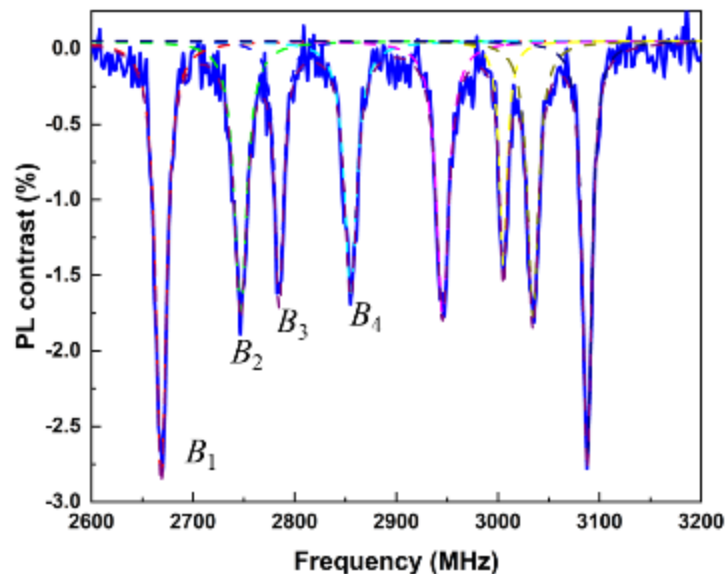
金刚石中NV色心的能级

# 光探测磁共振 (ODMR)

## 实验1: CW实验



测量序列



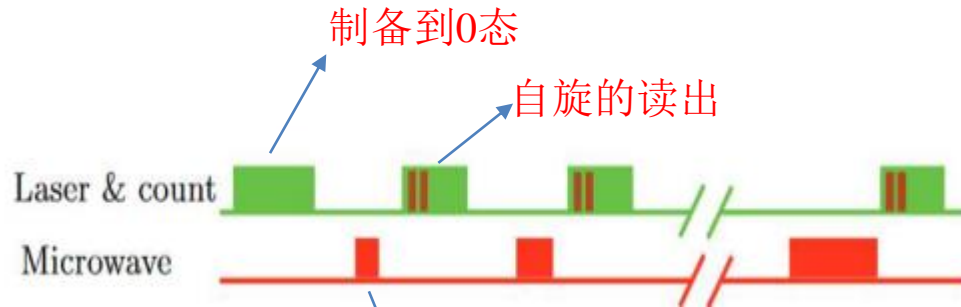
磁场下的光探测磁共振的CW谱

作用:

- 根据共振微波的频率确定能级之间的间隔
- 根据加磁场后塞曼劈裂的大小确定加的磁场的大小  
(对于矢量磁场, 可以确定平行于NV轴方向的磁场大小)

# 光探测磁共振 (ODMR)

## 实验2: Rabi 实验



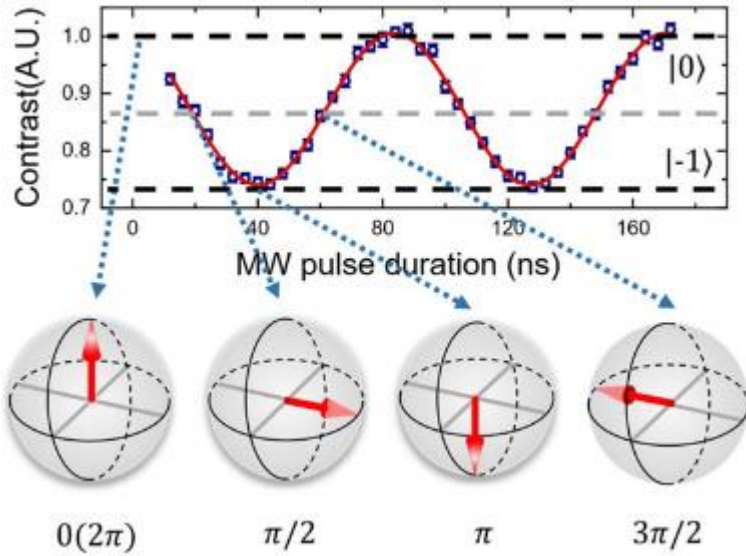
加一定时间的微波，使自旋在布洛赫球上转动

测量序列

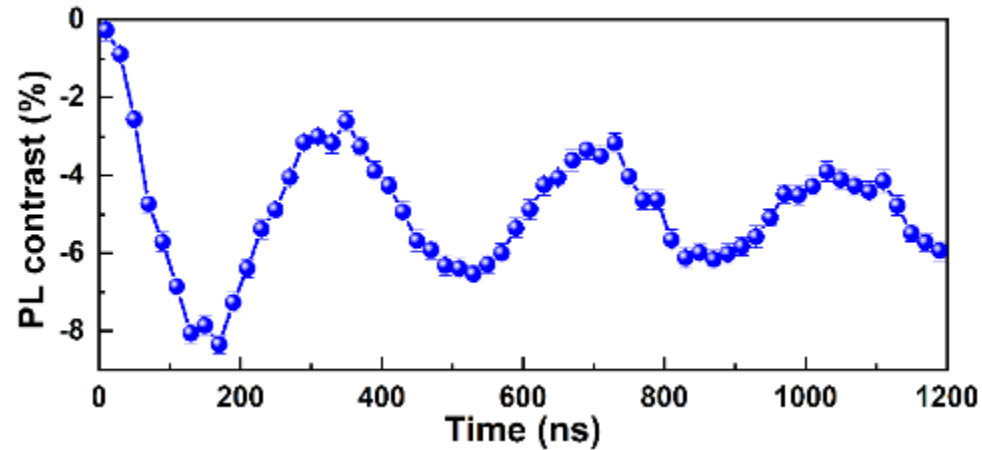
作用:

基于自旋拉比振荡，可以方便地定义量子态的操控脉冲

在量子传感领域，可以用来定量得知GHz频率的微波强度



自旋在布洛赫球上的转动

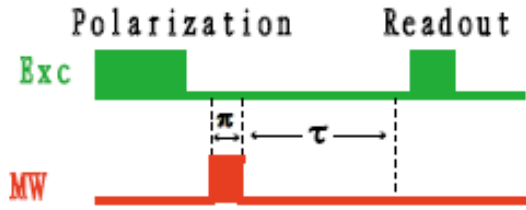


实验测量结果



# 光探测磁共振 (ODMR)

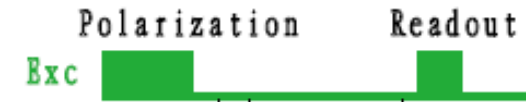
## 实验2: T1 (纵向弛豫) 实验



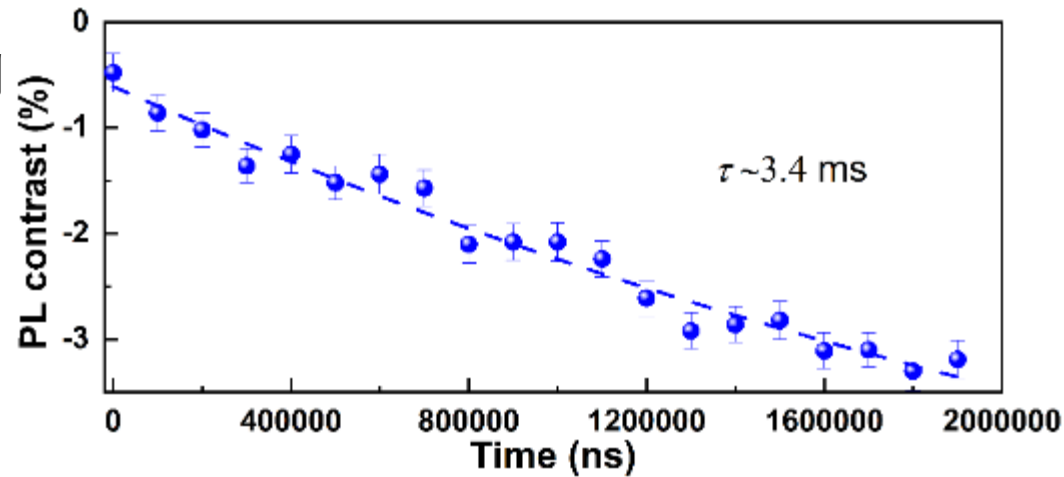
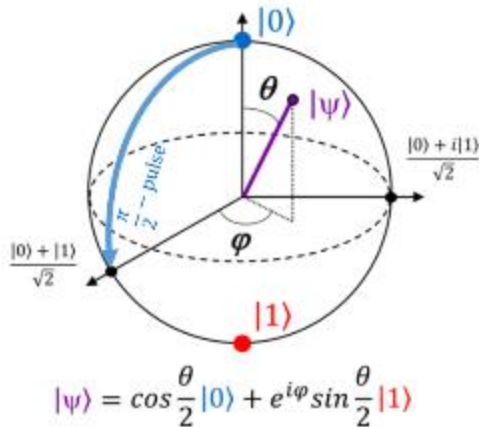
作用:

确定自旋系统从激发态返回平衡态所需的时间  
了解自旋系统与其周围环境之间的相互作用和动力学过程

从  $|+1\rangle$  态自旋纵向弛豫的时间测量序列



从  $|0\rangle$  态自旋纵向弛豫的时间测量序列

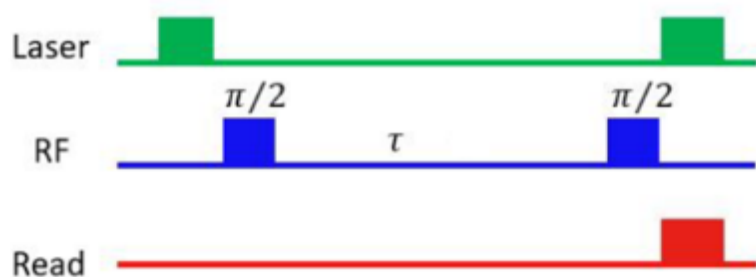


实验测量结果

自旋从布洛赫球的一极弛豫到另一极(纵向弛豫)

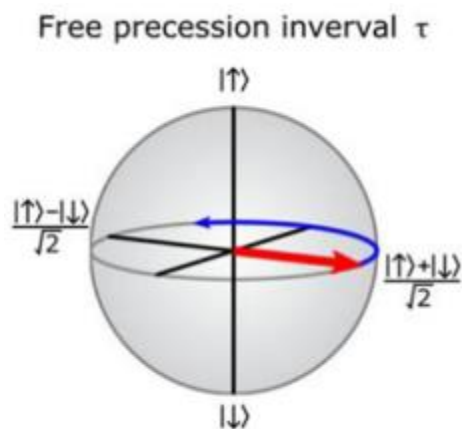
# 光探测磁共振 (ODMR)

## 实验3: $T_2^*$ (横向弛豫) 实验

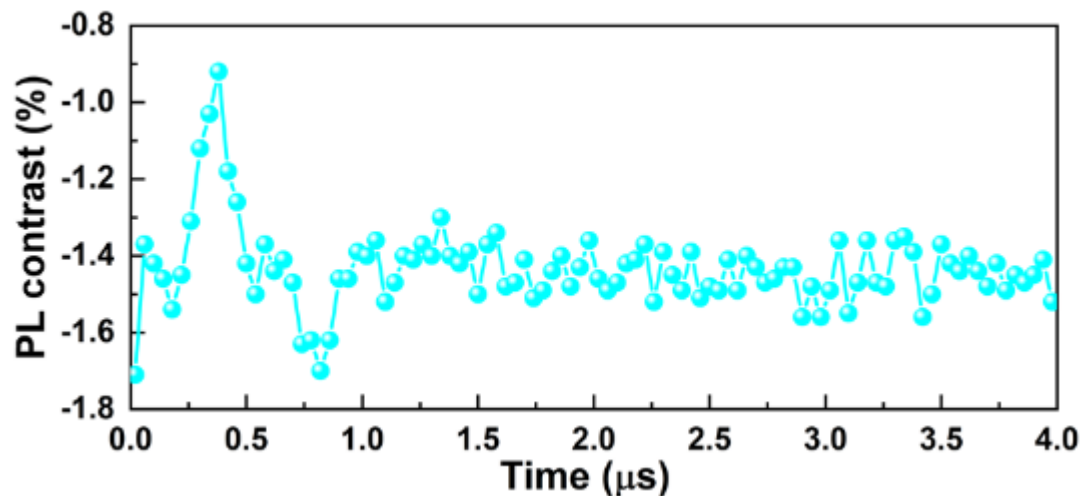


测量序列

作用：  
得到环境的磁涨落信息。  
研究NV色心与其他原子的耦合



自旋沿着赤道面演化(纵向弛豫)

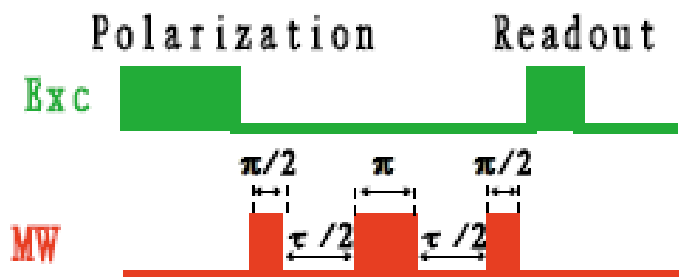


实验测量结果

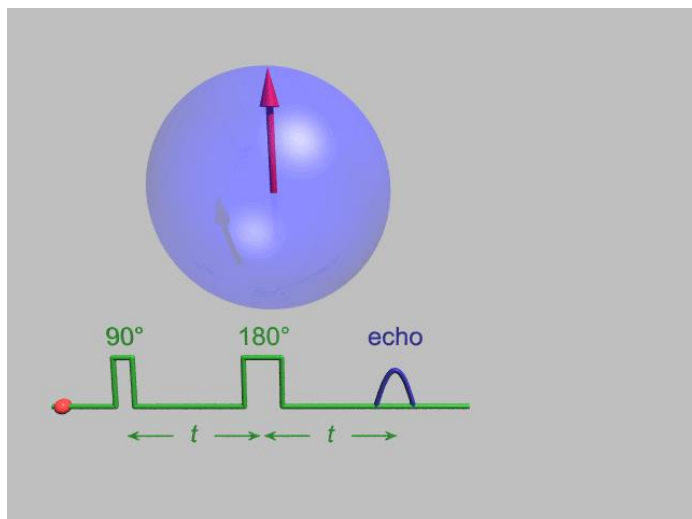
$T_2^* \sim 500 \text{ ns}$

# 光探测磁共振 (ODMR)

## 实验4: T2 (自旋回波) 实验

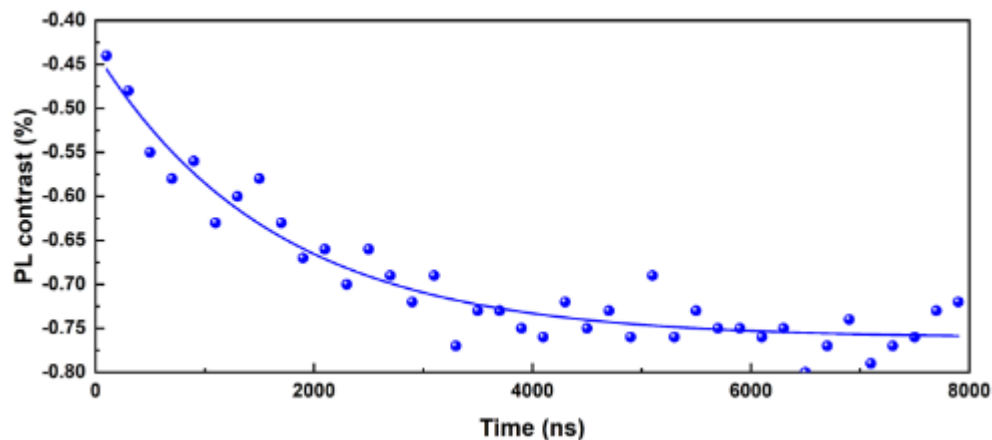


测量序列



自旋回波实验过程中自旋演化

作用：  
利用回波排除磁场不均匀对于自旋弛豫的影响，研究自旋本征的横向弛豫时间



实验测量结果

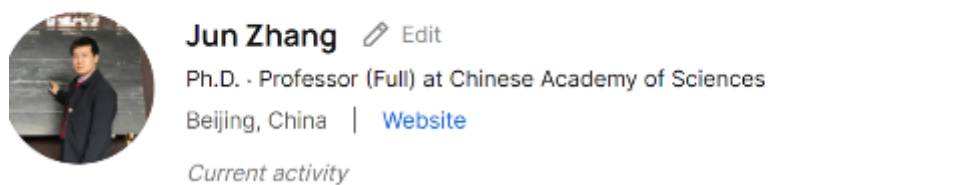
$T_2 \sim 1.6 \mu s$


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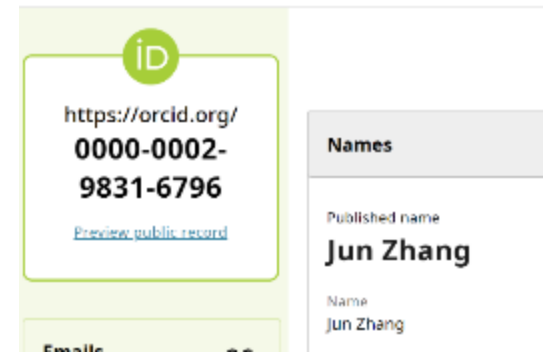
Student: Yuanfei Gao, Qing-Hai Tan, Yu-Jia Sun, Jia-Ming Lai, Kaixuan Xu, Feilong Song, Yanpei Lv,

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