Multiple optical gaps and lasing in 2D Ising superconductors

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PRB 99, 224511 (2019) arXiv:2111.03623

## **Properties on demand in quantum materials**



Basov, Averitt & Hsieh, Nat. Mat. 16, 1077 (2017)

## **Properties on demand in quantum materials**



Basov, Averitt & Hsieh, Nat. Mat. 16, 1077 (2017)

In typical superconductors (Nb) Tc=10 K  $\rightarrow \Delta \approx 0.5$  THz

#### Outline:

Interaction of THz light with superconductors can

- Excite collective modes
- Probe unconventional pairing in Ising SC

## Interaction of superconductors with light

#### **IR conductivity of superconductors**

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# Conductivity of Superconducting Films for Photon Energies between 0.3 and $40kT_c^*$

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AND

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#### **Absorption mechanism**



 $H_{BdG}\Psi = E\Psi$ 

 $p-p_F$ 

7

#### **Absorption mechanism**





Electron-hole symmetry prohibits vertical transitions.

We need impurity scattering

#### Theory of the Anomalous Skin Effect in Normal and Superconducting Metals\*

D. C. MATTIS<sup>†</sup> AND J. BARDEEN Department of Physics, University of Illinois, Urbana, Illinois (Received February 24, 1958)

Impurity scattering is introduced phenomenologically into the electron density matrix



$$\mathbf{j}(\mathbf{r},t) = \sum_{\omega} \frac{e^2 N(0) v_0}{2\pi^2 \hbar c} \int \frac{\mathbf{R} [\mathbf{R} \cdot \mathbf{A}_{\omega}(\mathbf{r}')] I(\omega,\mathbf{R},T) e^{-\mathbf{R}/l} d\mathbf{r}'}{\mathbf{R}^4}$$

"Dirty" limit  $I \ll \xi$ : Re ( $\sigma$ )  $\frac{\sigma_1}{\sigma_N} = \frac{2}{\hbar\omega} \int_{\epsilon_0}^{\infty} [f(E) - f(E + \hbar\omega)]g(E)dE$   $+ \frac{1}{\hbar\omega} \int_{\epsilon_0 - \hbar\omega}^{-\epsilon_0} [1 - 2f(E + \hbar\omega)]g(E)dE,$ 

#### **Quasiclassical Green's function approach**



Abrikosov, Gor'kov, Dzyaloshinskiy, Methods of Quantum Fields Theory in Statistical Physics

#### **Quasiclassical Green's function approach**



#### **Quasiclassical Green's function approach**



**Non-linear optics in superconductors** 

## Light-induced Higgs mode

#### **Higgs field**

High-energy: scalar field providing the gauge symmetry breaking mass of electroweak W and Z bosons, electrons and other particles (Higgs, PRL 1964) Superconductors: Meissner effect





<u>Higgs mode / boson:</u> Oscillations of the order parameter Higgs field amplitude

Way to observe HM: Excitation of usual superconductor with light- nonlinear optics.



## Light-induced Higgs mode

NbTiN Tc=15K,  $\Delta 0=0.75$  THz

THz pump-probe spectroscopy of Higgs mode

THz pumps  $\Omega$ =1.75 THz < 2 $\Delta$ (0)

Matsunada et al PRL 111, 057002 (2013)





Volkov, Kogan, JETP 38, 1018 (1974)].

## **Light-induced Higgs mode**

Resonant 3<sup>rd</sup> harmonic generation in transmission spectroscopy

Matsunaga, et al, Science 345, 1145 (2014).



NbN Tc=15K, Δ0=0.75 THz



Resonant behavior of  $3^{rd}$  harmonic  $\Omega = \Delta(T)$ 

Clean limit: no coupling



This can be interpreted as a Galilean invariance:



Clean limit: no coupling



This can be interpreted as a Galilean invariance:



Correlation-based mechanisms

Tsuji, Aoki, PRB 92, 064508 (2012)



Cea, Castellani, Benfatto, PRB, 93, 180507 (2016)



## Mechanism of Higgs mode excitation by light



**Editors' Suggestion** 

#### Nonlinear electromagnetic response and Higgs-mode excitation in BCS superconductors with impurities

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#### Mechanism of Higgs mode excitation by light

## Third harmonic current



Silaev, PRB 99, 224511 (19)

#### **Correlation effects masked by disorder**





$$\tau_{\rm imp}T_c > 10^{-3} \left(\frac{E_F}{T_c}\right)^2$$

## **Other collective modes**

## Collective Modes in Single-layer NbSe<sub>2</sub>



## Wan W. et al. Advanced Materials (2022)





Interband tunneling produces phase excitation – the Leggett mode Leggett, PTP (1966);

## **Other collective modes**



Interband tunneling produces phase excitation – the Leggett mode *Leggett, PTP (1966);* 

# Leggett mode in MgB<sub>2</sub>



Giorgianni, et al Nat. Phys. 15, 341 (2019)

## Decay of Higgs boson



## Decay of superconducting Higgs mode



Figure from https://msneubauer.github.io/projects/4\_project/

Main message:

# Because of disorder light is directly coupled to the collective modes in superconductors

## Theory:

Eliashberg, JETP Lett. 11, 114 (1970)]

Experiment:

Gap enhancement

Kommers and Clarke, PRL, 38, 1091 (1977)

# I<sub>c</sub> enhancement

Dayem and Wiegand, Phys. Rev. **155**, 419 (1967). Wyatt, et al., PRL **16**, 1166 (1966). Klapwijk, Van Den Bergh,. Mooij, JLTP 26, 385 (1977).

## Stationary corrections to order parameter:



Correction to the quasiparticle distribution function:





Correction to the quasiparticle distribution function:





Correction to the quasiparticle distribution function:



holes  $2\Delta$ *p-p*<sub>*F*</sub>

 $\rightarrow$  Superconductivity enhancement

Correction to the quasiparticle distribution function:





Correction to the quasiparticle distribution function:

Ω=2Δ ;T=0.91  $\Omega$ =3 $\Delta$  ;T=0.11 \_\_\_0.5 0.5 f 0 0 -5 5 0 -5 5 0  $E/\Delta(T)$  $E/\Delta(T)$ Effective cooling Population inversion  $\rightarrow$  $\rightarrow$  Superconductivity lasing effect ? enhancement



## **Opical properties of Ising superconductors**

## Introduction to Ising SC in Transition Metal-Dichalcogenides



transition-metal dichalcogenides (TMD) MoS<sub>2</sub>, NbSe<sub>2</sub>, WS<sub>2</sub>, WSe<sub>2</sub>, MoTe<sub>2</sub>

Inversion symmetry breaking in single layer
+ Spin-orbit coupling
→ spin -valley locking

Ising SOC

 $\rightarrow$  Opposite Zeeman fields in K/-K points

 $H_{so} = s \beta \sigma_z$  with  $s = \pm 1$  for K/-K

D. Xiao, et al., Coupled Spin and Valley Physics in Monolayers MoS2 and other group-VI dichalcogenides, PRL (2012).

## **Evidence of Ising SC : enhanced critical field**

In usual superconductors there is Pauli critical field given by Chandrasekhar-Clogston limit

$$H_p = \frac{\Delta}{\sqrt{2}\mu_B}$$



Superconductivity in crystals without symmetry centers Belaevskii, Guseinov, Rusinov, JETP, 44, 1243 (1976)

 $\lambda$ : impurity scattering rate  $\alpha$ : SOC strength

 $H_c$  strongly exceeds  $H_P$ 

## **Evidence of Ising SC : enhanced critical field**

Ising pairing in superconducting NbSe2 atomic layers, Xi, et al., Nat. Phys. (2016)



#### See also:

Luo, et al, Evidence for two-dimensional Ising SC in gated MoS2, Science (2015) Saito, et al, SC protected by spin-valley locking in ion-gated MoS2, Nat.Phys. (2016) Lu, et al, Full SC dome of strong Ising protection in gated monolayer WS2, PNAS (2018) Sohn, , et al, An unusual continuous paramagnetic-limited SC phase transition in 2D NbSe2, Nat. Mat. (2019) Li, , et al, Printable 2D SC monolayers, Nat. Mat. (2020)

Tunnelling spectroscopy of superconducting  $NbSe_2$ ,

Kuzmanović, et al arXiv:2104.00328

## **Ising superconductivity +** ferromagnetism





#### Two-fold symmetric superconductivity in few-layer NbSe<sub>2</sub>

 $\mu_0 H = 3 T$ 

G/G ...

180

 $\theta$  (°)

270

360

90

Alex Hamill<sup>1,8</sup>, Brett Heischmidt<sup>1,8</sup>, Egon Sohn<sup>2,3,8</sup>, Daniel Shaffer<sup>1</sup>, Kan-Ting Tsai<sup>1</sup>, Xi Zhang<sup>1</sup>, Xiaoxiang Xi<sup>4</sup>, Alexey Suslov<sup>5</sup>, Helmuth Berger<sup>6</sup>, László Forró<sup>6</sup>, Fiona J. Burnell<sup>1</sup>, Jie Shan<sup>3,7</sup>, Kin Fai Mak<sup>3,7</sup>, Rafael M. Fernandes<sup>1</sup>, Ke Wang<sup>1</sup> and Vlad S. Pribiag<sup>1</sup>

> Modulation of local conductance by the direction of H is the signature of magnetic proximity



## "Mirage" superconducting gap

$$E_{\pm}(k) = \sqrt{E_{bdg}^2 + \beta^2 + h^2 \pm 2\sqrt{h^2 E_{bdg}^2 + \xi_k^2 \beta^2}} \qquad E_{bdg} = \sqrt{\xi_k^2 + \Delta^2}$$



Tang, Bruder, Belzig, PRL 126, 237001 (2021)

## "Mirage" superconducting gap



Tang, Bruder, Belzig, PRL 126, 237001 (2021)

#### Density of states



## **Optical probes of finite-energy pairing**



Optical Gaps visible in conductivity and Raman scattering

$$\Omega_{I} = 2\Delta_{ord}$$
$$\Omega_{II} = \Delta_{ord} + \Delta_{-}$$
$$\Omega_{II} = \Delta_{ord} + \Delta_{+}$$

$$\Delta_{ord} = \Delta \sqrt{1 - h^2 / \beta^2}$$
$$\Delta_{\pm} = \sqrt{(h \pm \Delta)^2 + \beta^2}$$

Tc ≈ 6.5 K β ≈ 70 -140 K

Luo, et al, Evidence for 2D Ising SC in gated MoS2, Science (2015)  $\mathbf{i}$   $\Omega_{\mathrm{I}}, \Omega_{\mathrm{II}} \approx 1.5 \mathrm{THz}$ 

# **Optical conductivity**

Quasiclassical calculation, dirty limit,  $\beta$ =4 $\Delta_{0,} \Delta_{0}$ =1.76 T<sub>c0</sub>





# **Optical conductivity**

Quasiclassical calculation, dirty limit,  $\beta$ =4 $\Delta_{0,} \Delta_{0}$ =1.76 T<sub>c0</sub>

![](_page_41_Figure_2.jpeg)

![](_page_41_Figure_3.jpeg)

## **Raman scattering**

Dirty limit,  $\beta = 4\Delta_{0} \Delta_0 = 1.76 T_{c0}$ 

![](_page_42_Figure_2.jpeg)

![](_page_42_Figure_3.jpeg)

#### Technical details: arXiv:2111.03623

## **Direct spin- flip transitions**

![](_page_43_Figure_1.jpeg)

![](_page_43_Figure_2.jpeg)

 $\boldsymbol{h} = h_m \cos(\Omega_m t) \boldsymbol{x}$ 

## **Non-equilibrium distribution function**

![](_page_44_Figure_1.jpeg)

![](_page_45_Figure_0.jpeg)

## Conclusions

- THz spectroscopy and Raman response can be used to detect Ising superconductivity
- > Lasing effect is possible in Ising superconductors

Thank you for your attention!