Multiple optical gaps and laser with magnonic pumping in 2D Ising superconductors

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Ising superconductivity has been recently discovered in 2D transition metal dichalcogenides. We report that such superconductors have unusual optical properties controlled by the in-plane Zeeman field. First, we find several optical gaps visible as peaks of the conductivity and the Raman susceptibility. Moreover, we find that the Ising spin splitting in the spectrum of Bogolubov quasiparticles enables strong population inversion generated by the time-dependent Zeeman field. Ultimately this leads to the possibility of the superconducting laser with magnonic pumping which can be realized in the van der Waals structures consisting of the Ising superconductor and the ferromagnetic insulator layers.

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It is difficult to overestimate the importance of the optical probes for studies of superconducting materials. The existence of superconducting gap in the quasiparticle spectrum has been discovered with the help of the far-infrared optical conductivity\(^1\).\(^-\)\(^3\). Later on the superconducting gap has been observed by the inelastic Raman scattering\(^4\)\(^-\)\(^6\), which has become one of the basic tools in studying the anisotropic\(^7\)\(^,\)\(^8\) and multiband\(^5\)\(^,\)\(^8\)\(^,\)\(^9\)\(^-\)\(^11\) as well as the superconducting collective modes\(^4\)\(^,\)\(^12\)\(^-\)\(^19\).

Previous studies of the optical properties have been focused on the 3D materials. Recently the family of 2D superconducting materials has appeared\(^20\), in twisted bilayer graphene\(^21\)\(^-\)\(^23\), rhombohedral trilayer graphene\(^24\) and few-layer transition metal dichalcogenides (TMD)\(^25\)\(^-\)\(^31\). The in-plane symmetry breaking combined with the heavy transition-metal atoms leads to the extremely large spin-orbital coupling in TMD\(^32\)\(^-\)\(^36\) (SOC). It has the Ising character having the form of the effective out-of-plane Zeeman field changing the sign between electron pockets at \(K\) and \(K'\) points of the Brillouin zone\(^25\)\(^,\)\(^26\)\(^,\)\(^34\)\(^-\)\(^37\). Recently discovered so-called Ising superconductivity (IS)\(^25\)\(^,\)\(^26\)\(^,\)\(^37\)\(^-\)\(^39\) features strong enhancement of the in-plane critical field\(^26\)\(^,\)\(^30\)\(^,\)\(^37\)\(^-\)\(^42\), giant thickness-dependent transition state spin Hall signal\(^43\), electric field effect on superconductivity\(^26\)\(^,\)\(^44\), unconventional supercurrent phase\(^45\)\(^,\)\(^46\), multiple gaps\(^47\), collective modes\(^48\), and nontrivial interplay with magnetism\(^46\)\(^,\)\(^49\)\(^-\)\(^55\). Theoretically such superconductors have been studied in the context of the unconventional pairing mechanisms\(^56\)\(^-\)\(^63\), magnetic properties\(^57\)\(^,\)\(^58\)\(^,\)\(^64\), parity\(^58\)\(^,\)\(^59\)\(^,\)\(^62\)\(^,\)\(^65\)\(^,\)\(^66\) and singlet-triplet mixing superconductivity\(^58\)\(^,\)\(^59\)\(^,\)\(^61\)\(^,\)\(^62\)\(^,\)\(^65\)\(^,\)\(^66\), topological\(^37\), transport properties\(^37\)\(^,\)\(^68\) and magnetic properties\(^66\)\(^,\)\(^69\)\(^,\)\(^70\).

One can expect that 2D IS should have unusual optical properties but there have been no studies in this direction. In the present Letter we predict multiple peaks in the spectrum of the optical conductivity \(\sigma(\Omega_{ph})\) and the Raman scattering \(\chi_{Ram}(\Omega_{Ram})\) where \(\Omega_{Ram} = \Omega_{ph}^{(f)} - \Omega_{ph}^{(i)}\) is the Raman shift, see Fig.1a,b. These features are controlled by the on-plane Zeeman field which can be induced either by the external magnetic field or by the magnetic proximity effect in the Van der Waals (VdW) structure consisting of the superconductor and ferromagnetic insulator (FI) layers. Recently the VdW IS/FI structures like NbSe\(_2\)/CrB\(_3\) or other combinations has been studied experimentally\(^46\)\(^,\)\(^49\)\(^-\)\(^55\) and theoretically\(^70\). The optical response of IS pumped by magnons Fig.1c, that is by the time-dependent Zeeman field features the lasing effect. Ising spin splitting of Bogolubov spectrum allows to generate very strong quasiparticle population inversion resulting in the negative conductivity \(Re(\sigma(\Omega_{ph})) < 0\) of the IS.

Our model consists of the multiband 2D superconductor with the pairing between the electronic states situated at \(K\) and \(K'\) points of the Brillouin zone as shown in Fig.2a, with the Zeeman field is \(\mathbf{h}\). It is described by the Hamiltonian

\[
\hat{H} = \hat{T}(\xi_k - i\hat{\Lambda}) + \hat{\Lambda} = i(\beta\hat{\tau}_3\hat{\sigma}_z + \hat{\tau}_3\hat{h}\hat{\sigma}) + \hat{\tau}_1\Delta,
\]

where \(\hat{\tau}_1,2,3\) and \(\hat{\sigma}_{x,y,z}\) are the Pauli matrices in Nambu and spin spaces, respectively. The kinetic energy relative to the Fermi level is \(\xi_k = (k^2 - k_0^2)/2m = v_F k\), where the Fermi velocity \(v_F\) in general depends on the moment-
FIG. 2: (a) Schematic Fermi surface of the two-dimensional metal with Ising SOC field oriented along z causing the spin-splitting between spin-up (blue) and spin-down (red) states. The parts of Fermi surface in the second Brillouin zone are shown by dashed lines. (b) The model piecewise-constant dependence of the SOC strength $\beta(\theta r)$. (c) Bogolubov quasi-particle spectrum in the Fermi surface pocket near the $K$ point. The red and blue lines correspond to the spin-up and spin-down Fermi surfaces as in (a).

The SOC field which in general has to be the odd function of momentum direction relative to the $\Gamma$ point $\beta(k_{1}) = -\beta(-k_{1})$ is taken in the model piece-wise constant form shown in Fig.2b. The order parameter $\Delta$ is assumed to the isotropic. The in-plane Zeeman field $h_{x}z$ is induced either by the external magnetic field $h = \mu_{B}B$ or by the magnetic proximity effect with the adjacent FI. The Hamiltonian (1 ) yields Bogolubov spectrum consisting of two spin-split branches

$$E_{\uparrow(\downarrow)}(k) = \sqrt{E_{bdg}^{2} + \beta^{2} + h^{2} \pm 2 \sqrt{h^{2}E_{bdg}^{2} + \xi_{k}^{2}\beta^{2}}} \quad (3)$$

$E_{bdg} = \sqrt{\xi_{k}^{2} + \Delta^{2}}$ is the Bogolubov spectrum in the usual superconductor. For $h = 0$ the Ising SOC splits the usual Bogolubov spectrum by shifting the spin-up/down branches left/right as shown in Fig.2c. The spin-up/down branch intersection at $k = k_{F0}$ shown in Fig.2c, that is at $\xi_{k} = 0$ is removed by the Zeeman field $h \perp z$. For $h \neq 0$ the spectrum (3) has two types of gaps. The ordinary gap has the magnitude of $\Delta_{ord} = \sqrt{1 - h^{2}/\beta^{2}}$ and is located at $\xi_{k} = \sqrt{\beta^{2} - h^{2}\Delta^{2}/\beta^{2}}$. Note that the spectrum becomes gapless $\Delta_{ord} = 0$ at $h > \beta$ while the order parameter remains non-zero $\Delta \neq 0$. The additional so-called “mirage” gap$^{64}$ is located at $\xi_{k} = 0$ and separates the gap edges at $\Delta_{k} = \sqrt{(h \pm \Delta^{2})/\beta^{2}}$. The spectrum in the illustrative case $\beta = 3\Delta$, $h = 2\Delta$ is shown in Fig.3a.

The average $\langle S_{z} \rangle$ spin projection is shown by the color code of the spectrum branches in Fig.3a. Near the usual gap edge the spin state is almost $\langle S_{z} \rangle = \pm 1/2$. On the other hand, near the mirage gap edges $\xi_{k} = 0$ the spin state $\langle S_{z} \rangle = 0$, which indicates the dominating role of the spin-triplet pairing at this point.

Several van Hove singularities in the spectrum (3) results in the multiple optical gaps visible as peaks in the optical responses. In Fig.3a three most important of them are indicated by the dashed arrows I, II and III. The excitation process I corresponds to the breaking of the usual spin-singlet Cooper pairs, while II and III corresponds to the breaking of the spin-triplet Cooper pairs. The corresponding optical gaps are $\Omega_{I} = 2\Delta_{ord}$, $\Omega_{II} = \Delta_{ord} + \Delta_{-}$ and $\Omega_{III} = \Delta_{ord} + \Delta_{+}$ respectively. At such frequencies the external perturbation creates the pair of quasiparticles at the van Hove singularities which will be shown to result in peaks in the optical responses.

For $h = 0$ in Eq.(3) the Ising spin-splitting makes the minimal energy difference between empty spin-up and occupied spin-down branches to be $\min(E_{\uparrow} + E_{\downarrow}) = 2\beta$. Therefore, magnon pumping with frequency $\Omega_{m} \approx 2\beta$ creates the hot spot in the quasiparticle population in the IS with magnon pumping. The vertical black arrow shows the excitation of quasiparticles by the oscillating Zeeman field $h_{m} \cos(\Omega_{m} t)z$ generating hot spot in the quasiparticle population (pink shading). The violet arrows show photon emissions or absorptions with quasiparticle down/up transitions assisted by the impurity scattering.

In each of the six Fermi pockets in Fig.2a we introduce the quasiclassical propagator$^{71}$ $\tilde{g}(t, t', \mathbf{r}, \mathbf{v}_{F})$ which satisfies by the Eilenberger equation$^{72}$

$$i\mathbf{v}_{F} [\tilde{\gamma}_{3} \mathbf{A}, \tilde{g}]_{t} + [\mathbf{A} \tilde{\gamma}_{4} \mathbf{A}, \tilde{g}]_{t} = -\{\tilde{\gamma}_{3} \partial_{t}, \tilde{g}\}_{t} + \{\tilde{\mathbf{A}} \cdot \tilde{\gamma}_{4} \tilde{g}\}_{t} + \frac{[\tilde{\gamma}_{3} \mathbf{g}, \tilde{g}]_{t}}{2\tau_{imp}}. \quad (4)$$

In general it contains the time-dependent vector potential $\mathbf{A}(t)$ and the Zeeman field $h = h(t)$. The last term in Eq.4 is the collision integral describing the intraband...
impurity scattering with the rate $\tau^{-1}_{\text{imp}}$. We denote the commutators $[X, g] = X(t)g(t, t') - g(t, t')X(t')$ and the convolution is given by $\langle \hat{g} \rangle = \int_0^{2\pi} \hat{g}(\theta) d\theta$ where $\theta_K$ is the angle which characterises the direction of the momentum $k$ relative to the K point. Here we take into account the contribution of all three K points in Fig.2a by extending the integral over the angle $\theta_K$ from 0 to $2\pi$.

The first term in the l.h.s. of Eq.4 is the usual paramagnetic vertex which determines the conductivity and the Meissner effect. The second term is the diamagnetic vertex describing the electron density modulation by the time-dependent $A(t)$. This term comes with the coefficient $\gamma_I = \partial E/\partial k_i \partial k_i - (\partial E/\partial k_i \partial k_i)$ where the subtraction of Fermi pocket average term takes into account Coulomb interaction. The charge neutrality in Eq.4 is maintained since $\langle \hat{g} \rangle = 0$. However the direction-dependent density variation responsible for the Raman scattering is non-zero.

The equilibrium GF $\hat{g}_0(\omega) e^{i\omega(t-t')}$ is determined by the stationary Eilenberger Eq.4 in the imaginary time domain $t \rightarrow it$ with $A = 0$ and $h = \hbar \alpha x = \text{const}$. It has an analytic solution. With the equilibrium GF we can study the order parameter determined by the self-consistency equation

$$\Delta = \lambda \frac{\pi T}{4} \sum_\omega \text{Tr}[\hat{\tau}_1 \hat{g}_0(\omega)]$$

where $\omega$ runs over fermionic Matsubara frequencies. The dependence $\Delta(h, T)$ Fig.4a for $\beta = 4\Delta_0$ strongly exceeds the Chandrasekar-Clogston limit $\Delta/\sqrt{2}$. Here we use the usual units of $\Delta_0 = 1.76 T_0$, where $T_0$ is the critical temperature at $h = 0$. The density of states is given by the analytic continuation $N(\varepsilon) = \text{Re}[\text{Tr}[\hat{\tau}_1 \hat{g}_0(-i\varepsilon)]]/4$. The function $N(\varepsilon, h) = T = 0.01 T_0$, is shown in Fig.4b. It demonstrates the peaks both at the usual spectral gap and at the “mirage” gap edges in accordance with the van Hove singularities of the spectrum (3).

**Linear response conductivity.** The conductivity tensor is given by the Fermi pocket average of the combination $\sigma_{ij} = \nu e^2 \langle v_F e_i^* v_F e_j \rangle K \sigma$, where $\nu$ is the normal density of states, $e$ is the electron charge. The direction-independent conductivity amplitude $\sigma$ is determined by the correction to the GF linear in the applied ac field $A e^{i\Omega t}$ which can be written in the form $(\nu_F A) \hat{g}_A$. Then the conductivity is given by the expression in terms of the Keldysh component $\hat{g}_A^K$

$$\frac{\sigma}{\sigma_n} = \frac{1}{16 \Omega_{ph}^2} \int_{-\infty}^{\infty} d\varepsilon \text{Tr}[\tau_3 \hat{g}_A^K]$$

where $\sigma_n = \tau_{\text{imp}}/4$ is the normal state conductivity. The function $\hat{g}_A$ can be found analytically from the Eilenberger Eq.4 linearized with respect to $A$. The particularly simple expression is obtained in the limit of large impurity scattering $\tau_{\text{imp}} \Delta \ll 1$. We use this approximation to find $\hat{g}_A$ for Matsubara frequencies and and implementing the analytical continuation to the real frequencies. The resulting conductivity spectrum $\sigma(\Omega_{ph})$ is shown in the Fig.5a.

**FIG. 4:** (a) The order parameter $\Delta(T, h)/\Delta_0$. (b) The density of states $N(\varepsilon, h)$ taken at $T = 0.01$. In both panels $\beta = 4\Delta_0$.

The equilibrium GF $\hat{g}_0(\omega)e^{i\omega(t-t')}$ is determined by the stationary Eilenberger Eq.4 in the imaginary time domain $t \rightarrow it$ with $A = 0$ and $h = \hbar \alpha x = \text{const}$. It has an analytic solution. With the equilibrium GF we can study the order parameter determined by the self-consistency equation

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**FIG. 5:** (a) Conductivity $\sigma(\Omega_{ph})$ of the IS. (b) Frequency derivative of conductivity $d\text{Re}(\sigma)/d\Omega_{ph}$. (c) Raman susceptibility $\chi_{\text{Ram}}(\Omega_{Ram})$ of the IS. (d) Frequency derivative of Raman susceptibility $d\text{Im}(\chi_{\text{Ram}})/d\Omega_{Ram}$. Arrows I, II, III point to the features corresponding to the resonant pair-breaking processes shown in Fig.3a. Black solid lines in (b,d) show the optical gaps $\Omega_{II}(h), \Omega_{1I}(h), \Omega_{1II}(h)$. In all panels $T = 0.01 T_0, \beta/\Delta_0 = 4$. In (a,c) $h/\Delta_0 = \pm 2.8$ which corresponds to the crossection along dashed lines in (b,d).

**Raman susceptibility** Now let us consider the Raman susceptibility determined by the generalized density-density correlation function $\nu((e_F^\alpha e_F^\beta)^2) \chi_{\text{Ram}}$. Here the prefactor is the Raman vertex determined by the initial $A_i = e^{i\Omega t} A_i e$ and final $A_f = e^{i\Omega t} A_f e$ photon polarizations. The isotropic amplitude which we denote for brevity $\chi_{\text{Ram}}$ can be found by solving the
Eilenberger Eq.4 linearized with respect to the diamagnetic driving term \((A_i \hat{\gamma}_A - A^*_f \hat{\gamma}_A) e^{i \Omega R \text{amt}}\). The corresponding correction to the GF has the form \((A_i \hat{\gamma}_A - A^*_f \hat{\gamma}_A) g_{AA}^{R}\). It can be found analytically in the limit of large impurity scattering \(\tau_{imp} \Delta \ll 1\), see details in\(^7\). Then the Raman susceptibility is given by the expression in terms of the Keldysh component \(g_{AA}^{R}\)

\[
\frac{\chi_{\text{Ram}}^{(n)}}{\chi_{\text{Ram}}^{(e)}} = \frac{1}{16 \Omega_{\text{Ram}}} \int_{-\infty}^{\infty} d\varepsilon \text{Tr}[g_{AA}^{R}] (7)
\]

where \(\chi_{\text{Ram}}^{(n)} = \Omega_{\text{Ram}} \tau_{imp}/4\) is the normal metal Raman susceptibility amplitude in the dirty limit. Implementing the analytical continuation to the real frequencies\(^7\) \(\Omega_{\text{Ram}} \rightarrow i \Omega_{\text{Ram}}\) yields the Raman susceptibility shown in the Fig. 5c,d.

Both \(\sigma(\Omega_{ph})\) and \(\chi_{\text{Ram}}(\Omega_{Ram})\) have three distinct features marked by the arrows I, II and III in Fig.5 corresponding to excitation processes across the optical gaps with the same numbering in Fig.3a. To make these features more visible we plot the derivatives \(d \sigma/d \Omega_{ph}\) in Fig.5b and \(d \chi_{\text{Ram}}/d \Omega_{Ram}\) in Fig.5d. One can see that these derivatives have three peaks which perfectly coincide with the optical gaps shown by the black solid lines \(\Omega_I(h), \Omega_{II}(h), \Omega_{III}(h)\). Note that these dependencys are symmetric by \(h \rightarrow -h\).

**Superconducting laser with magnon pumping.** The high-frequency electromagnetic pumping generates the population inversion in the spectrum of Bogolubov quasiparticles which leads to the non-trivial effects such as the stimulated superconductivity\(^8\)–\(^8\)\(^4\). However this type of the population inversion is not sufficient for the superconducting laser, that is for the stimulated light mission due to the dipolar transitions between the different quasiparticle states.

We assume that the superconductor is pumped by Zeeman field \(\textbf{h} = h_m \cos(\Omega_{mt})\textbf{x}\) and calculate the linear response conductivity \(\sigma(\Omega_m, \Omega_{ph})\). The total conductivity can be written as the two parts \(\sigma = \sigma_{eq} + \sigma_{hh}\). The first term here \(\sigma_{eq}\) is the conductivity of the equilibrium superconductor given by the Eq.6. The second term \(\sigma_{hh}\) is determined by the correction to the GF linear in the applied ac field \(A e^{i \Omega_{ph} t}\) and of the second order in the oscillating Zeeman field amplitude \((v_F A)^2 g_{hh}\). This correction can be found analytically in the limit of large impurity scattering rate \(\tau_{imp} \Delta \ll 1\). For this we use iterative solution\(^7\) of the Eilenberger Eq.4 to get the stationary second-order correction driven by the Zeeman \(h_m^2 g_{hh}\).

We use at as a source to find the correction driven by the electromagnetic field \(g_{hh}\). Then, similarly to the usual conductivity (6), the magnon-driven correction \(\sigma_{hh}(\Omega_{ph})\) is given by

\[
\frac{\sigma_{hh}}{\sigma_n} = \frac{h_m^2}{16 \Omega_{ph}} \int_{-\infty}^{\infty} d\varepsilon \text{Tr}[\tau_{3} g_{hh}^{R}] (8)
\]

Note that the magnitude of the correction \(\sigma_{hh}\) is determined by the ratio of pumping strength and the rate of the inelastic relaxation \(\Gamma\), so that \(\sigma_{hh}/\sigma_n \sim h_m^2 \Gamma\). Since the inelastic relaxation is slow \(\Gamma/\Delta_0 \ll 1\) we can achieve large \(\sigma_{hh} \sim \sigma_{eq}\) with weak pumping \(h_m \sim \sqrt{\Gamma \Delta_0}\).

The total conductivity \(\sigma(\Omega_{ph}, \Omega_m)\) is shown in the Fig.6 for the narrow interval of magnon frequencies \(\Omega_m \sim 2 \beta\). There is a sharp negative peak at \(\Omega_m \approx 2 \beta\) and \(\Omega_{ph} \ll \Delta\) corresponding to the superconducting laser according to the mechanism shown in Fig.3b. At larger pumping frequency \(\Omega_m \approx 2 \beta\) the hot spot in quasiparticle distribution becomes larger which leads to the stronger photon absorption than emission. In result there appears a sharp positive peak \(\text{Re} \sigma(\Omega_{ph}, \Omega_m) > 0\). The peaks shown in Fig.6a at \(\Omega_m \approx 2 \beta\) are very sharp because of the peculiar resonant nesting of the branches 3. Indeed the interlevel distance is almost constant \(\min(E_1 + E_1) \approx 2 \beta\) at \(|\varepsilon_k| < \beta\) which leads to the excitation of many many states by the same pumping frequency \(\Omega_m \approx 2 \beta\).

To conclude, we predict unusual optical properties of IS resulting from the interplay of the superconductivity, SOC and ferromagnetism. Multiple optical gaps in conductivity and Raman susceptibility offer the direct way to detect the finite-energy spin-triplet pairing correlations and the “mirage” gap\(^6\)\(^8\) controlled by the in-plane magnetic field. Moreover, we have shown that the Ising spin splitting of Bogolubov quasiparticle spectrum allows for the generation of quasiparticle population inversion driven by the oscillating Zeeman field. This effect leads to the predicted superconducting laser which potentially can be realized in VdW IS/FI structures.

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Appendix A: Calculation of the equilibrium Green function (GF)

The full GF is determined by the Gor’kov equation for imaginary frequencies

\[
\hat{G}_0^{-1}\hat{G} = 1 \quad (A1)
\]

\[
\hat{G}_0^{-1} = H + i\omega \quad (A2)
\]

We will need the quasiclassical GF which can be obtained from the full GF by integrating over the kinetic energy in the vicinity of the Fermi surface

\[
\hat{g} = \frac{i}{\pi} \int d\xi \hat{\tau}_3 \hat{G} \quad (A3)
\]

The full GF determined by (A1) can be written as

\[
\hat{G} = \frac{\hat{C}_0 + \xi_2 \hat{C}_1 + \xi_2^2 \hat{C}_2 + \xi_2^3 \hat{C}_3}{(\xi_2^2 - E_1^2)(\xi_2^2 - E_2^2)} \quad (A4)
\]

where

\[
E_{1,2} = \sqrt{h^2 + \beta^2 - \Delta^2 - \omega^2} \pm 2i\sqrt{\beta^2(\Delta^2 + \omega^2) + h^2\omega^2} \quad (A5)
\]

Then the integration over \(\xi_p\) yields

\[
\int \frac{d\xi_k}{(\xi_k^2 - E_1^2)(\xi_k^2 - E_2^2)} = \frac{i\pi}{2E_1E_2(E_1 + E_2)} \quad (A6)
\]

\[
\int d\xi_k \frac{\xi_k^2}{(\xi_k^2 - E_1^2)(\xi_k^2 - E_2^2)} = -\frac{i\pi}{2(E_1 + E_2)} \quad (A7)
\]

Hence

\[
\hat{g}_0 = \frac{1}{2(E_1 + E_2)} \left( \hat{C}_2 - \frac{\hat{C}_0}{E_1E_2} \right) \quad (A8)
\]

It is instructive to check the structure of quasiclassical GF in spin-Nambu space. In general it is given by

\[
\hat{g}_0 = g_{x0}\hat{\sigma}_x\hat{\tau}_0 + g_{y0}\hat{\sigma}_y\hat{\tau}_2 + (g_{x1}\hat{\sigma}_0 + g_{x2}\hat{\sigma}_x)\hat{\tau}_1 + (g_{y3}\hat{\sigma}_0 + g_{y3}\hat{\sigma}_x)\hat{\tau}_3 \quad (A9)
\]

The last four terms in the r.h.s. are the usual ones for the superconductor with the Zeeman field. The first two terms appear due to the interplay of the Ising SOC and the Zeeman spin splitting fields.

Appendix B: Calculation of the conductivity and Raman susceptibility

To find the conductivity we need to determine the linear correction to the GF \((\mathbf{v}_F A)\hat{g}_A(\omega_+ , \omega)\) generated by the applied ac field \(\mathbf{A} e^{i\Omega m t}\) where \(\omega_+ = \omega + \Omega_{ph}\). For that we need to solve the linearized Eilenberger equation for imaginary frequencies

\[
i[\tilde{r}_3\hat{g}_0(\omega) - \hat{g}_0(\omega_+ )\tilde{r}_3] = \hat{\Lambda} \hat{g}_A + \hat{g}_A - \omega \hat{g}_A \tilde{r}_3 + \tau_{imp}^{-1}[\hat{g}_0(\omega_+ )\tilde{r}_3 \hat{g}_A - \hat{g}_A \hat{g}_0(\omega )] \quad (B1)
\]

The solution can be found analytically in the limit of \(\tau_{imp}\Delta_0 \ll 1\) in the form

\[
\hat{g}_A = \frac{i\tau_{imp}[\hat{g}(\omega_+ )\tilde{r}_3 \hat{g}(\omega) - \sigma_0 \tilde{r}_3]}{2} \quad (B2)
\]

Implementing the analytical continuation to the real frequencies we obtain the Keldysh component \(\hat{g}_A^K\) and hence the conductivity.

The Raman susceptibility is given by the linear response relation written in terms of the quasiclassical GF correction linear in the diamagnetic term in the Eilenberger equation \((\mathbf{A} A^\dagger)_\beta \hat{g}_{AA}(\omega_+ , \omega)\) where \(\omega_+ = \omega + \Omega_{Ram}\). The Eilenberger equation for the correction \(\hat{g}_{AA}\) reads as

\[
\hat{g}_0(\omega) - \hat{g}_0(\omega_+ ) = [\hat{\Lambda} \hat{g}_{AA} + \hat{g}_A \tilde{r}_3 \hat{g}_{AA} - \omega \hat{g}_{AA} \tilde{r}_3] + \tau_{imp}^{-1}[\hat{g}_0(\omega_+ )\tilde{r}_3 \hat{g}_{AA} - \hat{g}_{AA} \hat{g}_0(\omega )] \quad (B3)
\]

where we take into account that \(\langle (e_i^\gamma e_f)\rangle = 0\). The solution can be found analytically in the limit of \(\tau_{imp}\Delta_0 \ll 1\) in the form

\[
\hat{g}_{AA} = \frac{i\tau_{imp}[\hat{g}(\omega_+ )\hat{g}(\omega) - \sigma_0 \hat{r}_0]}{2} \quad (B4)
\]

Implementing the analytical continuation to the real frequencies we obtain the Keldysh component \(\hat{g}_{AA}^K\) and hence the Raman susceptibility.

Appendix C: Conductivity of the Ising superconductor with magnon pumping

We assume that the Ising superconductor is subjected to the ac electromagnetic field \(\mathbf{A} e^{i\Omega_{m1} t}\) and the Zeeman field with two tones \(h(t) = h_m(e^{i\Omega_{m1} t} + e^{i\Omega_{m2} t})\). In order to construct correctly the analytical to real frequencies we need to consider from the beginning the general case with \(\Omega_{m1} \neq -\Omega_{m2}\). We introduce the following notation for frequencies

\[
\omega_1 = \omega, \quad \omega_2 = \omega + \Omega_{m1}, \quad \omega_3 = \omega + \Omega_{m2}, \quad \omega_4 = \omega + \Omega_{m1} + \Omega_{m2}, \quad \omega_5 = \omega + \Omega_{ph}, \quad \omega_6 = \omega + \Omega_{m1} + \Omega_{m2} + \Omega_{ph} \quad (C1)
\]

First, we find the second-order correction to the GF \(\hat{g}_{hh}(41) \sim h_m^2\), where we introduce notation \(\hat{g}_{hh}(41) \equiv \hat{g}_{hh}^{(2)}(41)\). For frequencies
\( \hat{g}_{hh}(\omega_4, \omega_1), \) so that the in the time domain it is 
\( \hat{g}_{hh}(t, t') = \hat{g}_{hh}(\omega_4, \omega_1)e^{i(\omega_4 t - \omega_1 t')} \). After the analytical continuation we will put \( \Omega_m = -\Omega_m = \Omega_m \) to get the stationary correction \( \hat{g}_{hh}(t, t') = \hat{g}_{hh}(t - t') \). We search this correction by solving the Usadel equation by iterations with respect to the Zeeman field in the imaginary frequencies. We start with the first-order correction \( \hat{g}_h = h_m(\hat{g}_x \sigma_x + \hat{g}_y \sigma_x) \). For that we need to solve the linearized Eilenberger equation for imaginary frequencies
\[
i \sigma_\omega \left[ \hat{\tau}_3 \hat{g}_0(\omega) - \hat{g}_0(\omega_+^\prime) \hat{\tau}_3 \right] = \left[ \hat{\Lambda}, \hat{g}_h \right] + \left[ \omega + \hat{\tau}_3 \hat{g}_h - \omega \hat{g}_h, \hat{\tau}_3 \right]
\]
where \( \omega_+^\prime = \omega_\omega \) for \( \hat{g}_h(21) \) or \( \omega_+^\prime = \omega_3 \) for \( \hat{g}_h(31) \). The solution is given by
\[
\hat{g}_h(21) = \frac{i(s_1 + s_2)}{(s_1 + s_2)^2 + 4\beta_3^2} \left[ \hat{g}_0(2) \hat{\tau}_3 \hat{g}_0(1) - \hat{\tau}_3 \right] \\
\hat{g}_h(31) = \frac{2\beta_3^2}{(s_1 + s_2)^2 + 4\beta_3^2} \left[ \hat{\tau}_3 \hat{g}_0(1) - \hat{g}_0(2) \hat{\tau}_3 \right] 
\]
For \( \hat{g}_h(31) \) we need to replace \( 2 \rightarrow 3 \). The second order correction \( \hat{g}_{hh}(41) \) can be found by solving the equation
\[
s_4 \hat{g}_0(41) \hat{g}_{hh} - s_1 \hat{g}_{hh} \hat{g}_0(1) = \frac{i\hat{\tau}_3 \hat{g}_0(21) - \hat{g}_x(43) \hat{\tau}_3 + i\hat{\tau}_3 \hat{g}_x(31) - \hat{g}_x(42) \hat{\tau}_3 }
\]
It is supplemented by the normalization condition
\[
\hat{g}_0(41) \hat{g}_{hh} + \hat{g}_{hh} \hat{g}_0(1) + \hat{g}_x(43) \hat{g}_y(31) + \hat{g}_x(42) \hat{g}_y(21) + \hat{g}_y(43) \hat{g}_y(31) + \hat{g}_y(42) \hat{g}_y(21) = 0
\]
The solution is given by
\[
(s_1 + s_4) \hat{g}_{hh}(41) = i\hat{g}_0(4) \hat{\tau}_3(\hat{g}_x(21) + \hat{g}_x(31) - (\hat{g}_x(43) + \hat{g}_x(42) \hat{\tau}_3) \\
- s_1 \hat{g}_0(4) [\hat{g}_x(42) \hat{g}_x(21) + \hat{g}_x(43) \hat{g}_x(31)] \\
- s_1 \hat{g}_0(4) [\hat{g}_y(42) \hat{g}_y(21) + \hat{g}_x(43) \hat{g}_y(31)]
\]
To find the contribution to electric current in the presence of magnon pumping we need to find the anisotropic correction to the GF which is linear in \( A \) and of the second order in Zeeman field \( (v_F A) \hat{g}_{hh} \). We consider the limit of strong impurity scattering when the anisotropic correction driven by the field \( A \) to the GF in is general given by \( (v_F A) \hat{g}_A \)
\[
\hat{g}_A(t, t') = \frac{i\tau_{imp}}{2} \left[ \hat{g}(t, t') e^{i\Omega_{ph} t} \hat{\tau}_3 \hat{g}(t_1, t'') - \hat{\tau}_3 \hat{\sigma}_0 \hat{g}(t_1, t'') e^{i\Omega_{ph} t} \right]
\]
where \( \hat{g}(t, t') \) is in general the non-equilibrium the GF in the absence of electromagnetic field. Taking into account that in the presence of the magnon pumping \( \hat{g} = \hat{g}_0 + \hat{g}_{hh} \) and going to the Fourier representation we get from (C8) \( \hat{g}_{hh}(61) = (C8) \)
\[
\hat{g}_{hh}(61) = \frac{i\tau_{imp}}{2} \left[ \hat{g}_0(6) \hat{\tau}_3(\hat{g}_h(41) + \hat{g}_{hh}(65) \hat{\tau}_3 \hat{g}_0(1) \right]
\]
Using this expression we can use the analytical continuation to real energies to find the Keldysh component \( \hat{g}_K \) which determines the correction to the conductivity \( \sigma_{hh} \).

**Appendix D: Analytical continuation**

In order to find the real-frequency response we need to implement the analytical continuation. The analytic continuation of the sum by Matsubara frequencies is determined according to the general rule
\[
T \sum_{\omega} \hat{g}_1(\omega_1) \hat{g}_2(\omega_2) \rightarrow \int \frac{d\varepsilon}{4\pi i} n_0(\varepsilon) \left[ \hat{g}_1(-i\varepsilon^R) - \hat{g}_1(-i\varepsilon^I) \right] \hat{g}_2(-i\varepsilon^A) + \int \frac{d\varepsilon}{4\pi i} n_0(\varepsilon) \hat{g}_1(-i\varepsilon^A) \left[ \hat{g}_2(-i\varepsilon^R) - \hat{g}_2(-i\varepsilon^I) \right]
\]
where \( n_0(\varepsilon) = \tanh(\varepsilon/2T) \) is the equilibrium distribution function. In the r.h.s. of (D1) we substitute \( \varepsilon_1 = \varepsilon, \varepsilon_2 = \varepsilon + \Omega \) and \( \varepsilon^A = \varepsilon \pm i\Gamma \). Here the term with \( \Gamma > 0 \) is added to shift of the integration contour into the corresponding half-plane.

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Optical signatures of Ising superconductivity
(Dated: November 8, 2021)

We report that Ising superconductivity in 2D superconductors shows up as peaks in both the linear conductivity and Raman scattering amplitude induced by the external in-plane magnetic field.

PACS numbers:

I. INTRODUCTION

II. RESULTS

A. Spectrum and order parameter

The full GF is determined by the Gor’kov equation

\[ \hat{G}_0^{-1} \hat{G} = 1 \]  
\[ \hat{G}_0^{-1} = \tau_3 (\xi_p + \beta \sigma_3) + \tau_1 (i\omega + h\sigma_x) \]  

The Zeeman field is induced by the external magnetic field \( h = \mu_B B \).

We will need the quasiclassical GF which can be obtained from the full GF by integrating over the kinetic energy in the vicinity of the Fermi surface

\[ \hat{g} = i \pi \int d\xi_p \hat{r}_3 \hat{G} \]  

The equilibrium quasiclassical GF is determined by the Eilenberger equation

\[ [\hat{A}, \hat{g}] = 1 \]  
\[ \hat{A} = \beta \sigma_3 + \tau_1 \Delta + \tau_3 (i\omega + h\sigma_x) \]  

The order parameter is determined by the self-consistency equation

\[ \Delta = \pi T \sum_\omega \text{Tr}[\tau_1 \hat{g}(\omega)] \]  

The density of states is \( N(\varepsilon) = \text{Tr}[\tau_3 \hat{g}(-i\varepsilon)] \). It is shown in Fig.1.

B. Conductivity: Usadel theory

The conductivity is given by

\[ \frac{\sigma}{\sigma_n} = \frac{\pi T}{\Omega} \sum_\omega \text{Tr}[\tau_3 \hat{g}(\omega + \Omega) \tau_3 \hat{g}(\omega)] \]  

where \( \sigma_n \) is the normal state conductivity.

FIG. 2: (Color online) Conductivity and Raman response of the Ising superconductor, \( T = 0.01 T_{c0} \).

C. Raman scattering: dirty limit

Raman scattering vertex does not break the local charge neutrality, i.e., it does not change the total electron density. Therefore it can be calculated using quasiclassical theory with the time-dependent diamagnetic term as the source \( \gamma(p) A^2(t) \), where the coefficient \( \gamma(p) \) takes into account corrections to the electron mass due
to the interband transitions\textsuperscript{7}. The charge neutrality is maintained by adding the isotropic electric potential term equal to $\langle \gamma \rangle_{FS} A^2(t)$ where $\langle \gamma \rangle_{FS}$ is the average over the Fermi surface. In the non-stationary situation the diamagnetic term does not disappear from the Eilenberger equation which then in the dirty limit reads as

$$\frac{e^2}{c^2} [\hat{\gamma} A^2, \hat{g}]_t = \frac{\langle \hat{g} \rangle_{\Omega} \hat{g}}{2\tau_{imp}}$$

(8)

where $\hat{\gamma} = \gamma - \bar{\gamma}$. The solution can be written in the form

$$\hat{g} = A_{\Omega_2} A_{-\Omega_1} \hat{g}_{AA}$$

(9)

$$g_{AA} = \frac{e^2}{c^2} \tau_{imp} \hat{\gamma} \hat{g}(\omega + \Omega) \hat{g}(\omega)$$

(10)

The Raman scattering amplitude is given by

$$I_R = \nu \pi T \sum_{\omega} \text{Tr} [\langle \hat{g}_{AA} \rangle_{FS}]$$

(11)

D. Analytical continuation

In order to find the real-frequency response we need to implement the analytical continuation of Eq.(??,??).

These third-order responses are obtained by the summation of expressions which depend on the four shifted fermionic frequencies such as $g = g(\omega_1, \omega_2, \omega_3, \omega_4)$. The analytical continuation of the sum by Matsubara frequencies is determined according to the general rule\textsuperscript{7}

$$T \sum_{\omega} g(\omega_1, \omega_2) \rightarrow$$

$$\int \frac{d\varepsilon}{4\pi i} n_0(\varepsilon_1) \left[ g(-i\varepsilon_1^R, -i\varepsilon_2^R) - g(-i\varepsilon_1^A, -i\varepsilon_2^R) \right] +$$

$$\int \frac{d\varepsilon}{4\pi i} n_0(\varepsilon_2) \left[ g(-i\varepsilon_1^A, -i\varepsilon_2^R) - g(-i\varepsilon_1^A, -i\varepsilon_2^A) \right]$$

(12)

where $n_0(\varepsilon) = \tanh(\varepsilon/2T)$ is the equilibrium distribution function. In the r.h.s. of (12) we substitute in each term $\omega_{k<l} = -i\varepsilon_k^R$ and $\omega_{k>l} = -i\varepsilon_k^A$ for $k = 1, 2, 3, 4$, denote $\varepsilon_k = \varepsilon + (3 - k)\Omega$ and $\varepsilon^R = \varepsilon + i\Gamma$, $\varepsilon^A = \varepsilon - i\Gamma$. Here the term with $\Gamma > 0$ is added to shift of the integration contour into the corresponding half-plane.