Proposal for THz lasing from a topological quantum dot: Supplementary material

Marie S. Rider*
The Blackett Laboratory, Imperial College London, SW7 2AZ

Vincenzo Giannini†
Instituto de Estructura de la Materia (IEM-CSIC),
Consejo Superior de Investigaciones Científicas, Serrano 121, 28006 Madrid, Spain
Technology Innovation Institute, Building B04C, Abu Dhabi P.O. Box 9639, United Arab Emirates and
Centre of Excellence ENSEMBLE3 sp. z o.o., Wolczynska 133, Warsaw, 01-919, Poland
(Dated: July 27, 2021)

S1: Model Hamiltonian and topological surface states

The four band low energy Hamiltonian for topological insulator materials and resulting topological surface state physics has been obtained and discussed extensively in References [1, 2], but we summarise here the general method of obtaining the topological surface states for spherical TI nanoparticles used in the main text of this work. Using either symmetry principles or a $\mathbf{k}\cdot\mathbf{p}$ method, the low-energy, four band effective Hamiltonian for topological insulator materials is given by [1]

$$\begin{aligned}\mathbf{H}(\mathbf{k}) = \epsilon_0(\mathbf{k})\mathbb{1}_2 \otimes \mathbb{1}_2 + m(\mathbf{k})\mathbb{1}_2 \otimes \boldsymbol{\sigma_3} \\ + A_0 k_x \boldsymbol{\sigma_1} \otimes \boldsymbol{\sigma_1} + A_0 k_y \boldsymbol{\sigma_2} \otimes \boldsymbol{\sigma_1} \\ + B_0 k_z \boldsymbol{\sigma_3} \otimes \boldsymbol{\sigma_1},\end{aligned} \tag{1}$$

where $m(\mathbf{k}) = m_0 + m_1 k_z^2 + m_2(k_x^2 + k_y^2)$ and $\epsilon_0(\mathbf{k}) = C_0 + C_1 k_z^2 + C_2(k_x^2 + k_y^2)$. For Bi_2Se_3, $A_0 = 3.33$ eVÅ, $B_0 = 2.26$ eVÅ, $C_0 = -0.0083$ eV, $C_1 = 5.74$ eVÅ^2, $C_2 = 30.4$ eVÅ^2, $m_0 = -0.28$ eVÅ^2, $m_1 = 6.86$ eVÅ^2 and $m_2 = 44.5$ eVÅ^2.

Various simplifications can be made to this Hamiltonian to make the solution of the surface states analytical in the case of the spherical particle. The linear terms may be simplified by approximating spin-orbit coupling to be isotropic, such that $A = (A_0 + 2B_0)/3$ as used in the main text. The term $\epsilon_0(\mathbf{k})$ is often neglected to make the solution for the surface more tractable, enforcing particle-hole symmetry. However if this term is reintroduced then the leading k^2 terms in the surface state energies are given by $C_1 k_z^2 + C_2(k_x^2 + k_y^2)$. Averaging over the three Cartesian coordinates, for Bi_2Se_3 we arrive at the value $A_1 = (C_1 + 2C_2)/3 = 22.18$ eVÅ^2 used in this work when considering k^2 contributions to the surface state dispersion relation.

To find the analytical expression of the surface states, the $\epsilon_0(\mathbf{k})$ term is neglected. It is also usual to use that $m_1 = m_2$ (giving a symmetric dispersion relation) without sacrificing much accuracy, as terms involving either parameter can be found to be negligible. These assumptions then lead to the simplified bulk Hamiltonian

$$\begin{aligned}\mathbf{H}(\mathbf{k}) = m(\mathbf{k})\mathbb{1}_2 \otimes \boldsymbol{\sigma_3} + Ak_z \boldsymbol{\sigma_3} \otimes \boldsymbol{\sigma_1} \\ + Ak_x \boldsymbol{\sigma_1} \otimes \boldsymbol{\sigma_1} + Ak_y \boldsymbol{\sigma_2} \otimes \boldsymbol{\sigma_1},\end{aligned} \tag{2}$$

stated in the main text. This Hamiltonian can be rewritten in spherical coordinates and split into components perpendicular and parallel to the surface, $\mathbf{H} = \mathbf{H}_\perp + \mathbf{H}_\parallel$. For the surface state wavefunction Ψ, we can write down an eigenvalue equation

$$\mathbf{H}_\perp \Psi = E\Psi, \tag{3}$$

where E gives the position of the Dirac point. Projecting $\mathbf{H}_\parallel$ onto the resulting form of Ψ gives the final form of the topological surface states, and the effective surface Hamiltonian which has the form of a Dirac Hamiltonian.

In this protocol, the Dirac point is usually placed at $E = 0$ for mathematical convenience. The surface termination of topological insulators has been shown to have an important effect on the position of the Dirac point [3, 4] in thin films, and so experimental or numerical *ab initio* studies on the dispersion relation of spherical nanoparticles would be useful in ascertaining if doping would be required to produce TI nanoparticles that are suitable for lasing. Topological insulator materials are generally n-doped, resulting in a Fermi level close to the conduction band.

The approximations of isotropic spin-orbit coupling and parabolic dispersion relation allow for the analytical solution of the surface states of a spherical nanoparticle. Lifting these conditions would result in the breaking of spherical symmetry, and a lifting of the degeneracy of the energy levels used in the lasing scheme. The extent to which this degeneracy is lifted depends on the specific material used, and will result in a finite linewidth of the THz laser.

The topological surface states of topological insulators exist due to strong spin-orbit coupling in the bulk of the material, and are protected by time-reversal symmetry. As such, the surface states are remarkably robust when subjected to perturbations which do not break the time-reversal symmetry. The protection due to time-reversal symmetry persists in nanoparticles [5], and so the surface

* marie.rider@ic.ac.uk
† www.GianniniLab.com

states are expected to be robust against (non-magnetic) perturbations.

S2: Jacobi polynomials

Class of orthogonal polynomials $J_n^{\alpha,\beta}(x)$, orthogonal w.r.t the weight $(1-x)^\alpha(1+x)^\beta$ on the interval $x \in [-1,1]$, where $\alpha, \beta > -1$, such that

$$\int_{-1}^{1}(1-x)^\alpha(1+x)^\beta J_n^{\alpha\beta}(x)J_{n'}^{\alpha\beta}(x)dx = \frac{\delta_{nn'}}{N_{n\alpha\beta}^2} \qquad (4)$$

where

$$N_{n\alpha\beta}^2 = \frac{(2n+\alpha+\beta+1)!(n+\alpha+\beta)!n!}{2^{\alpha+\beta+1}\Gamma(n+\alpha)!(n+\beta)!}. \qquad (5)$$

S3: Matrix element

Some explicit values of the E1 matrix element $V_{\mathrm{i,f}}/R^2$ for LH polarized light (such that $\epsilon = (1,-i,0)/\sqrt{2})$ are given below.

$\lvert\mathrm{i}\rangle \to \lvert\mathrm{f}\rangle$	$V_{\mathrm{i,f}}/R^2$
$\lvert+,0,-1/2\rangle \to \lvert+,1,1/2\rangle$	1/9
$\lvert+,0,-1/2\rangle \to \lvert-,0,1/2\rangle$	2/9
$\lvert+,0,-3/2\rangle \to \lvert+,0,-1/2\rangle$	1/3
$\lvert+,0,-3/2\rangle \to \lvert-,1,-1/2\rangle$	2/75

[1] Chao-Xing Liu, Xiao-Liang Qi, HaiJun Zhang, Xi Dai, Zhong Fang, and Shou-Cheng Zhang. Model Hamiltonian for topological insulators. *Physical Review B*, 82(4):045122, 2010.

[2] Haijun Zhang, Chao-Xing Liu, Xiao-Liang Qi, Xi Dai, Zhong Fang, and Shou-Cheng Zhang. Topological insulators in Bi_2Se_3, Bi_2Te_3 and Sb_2Te_3 with a single Dirac cone on the surface. *Nature physics*, 5(6):438–442, 2009.

[3] Deepnarayan Biswas and Kalobaran Maiti. Exceptional surface states and topological order in bi2se3. *Journal of Electron Spectroscopy and Related Phenomena*, 208:90–94, 2016.

[4] Xie-Gang Zhu, Yun Zhang, Wei Feng, Bing-Kai Yuan, Qin Liu, Rui-Zhi Qiu, Dong-Hua Xie, Shi-Yong Tan, Yu Duan, Yun Fang, et al. Electronic structures of topological insulator Bi_2Te_3 surfaces with non-conventional terminations. *New Journal of Physics*, 18(9):093015, 2016.

[5] Gleb Siroki, Peter D Haynes, Derek KK Lee, and Vincenzo Giannini. Protection of surface states in topological nanoparticles. *Physical Review Materials*, 1(2):024201, 2017.